Process Integration to Increase Woody Biomass Utilization for Energy Purposes

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Abstract

Woody biomass is an abundant renewable energy resource in Sweden, and the Swedish government has been promoting research and development programs for the exploitation of this resource as a means to meet the targets on the reduction of the carbon dioxide emissions from the industrial, energy and transportation sectors.

This thesis aims at expanding the knowledge on the efficient utilization of the available woody biomass, so that a larger amount of this renewable resource can be used for energy purposes. The thesis presents a collection of studies following the main two policies that have been identified for the reduction of carbon dioxide emissions, i.e. the implementation of measures improving energy conservation and efficiency and a deep decarbonization of the energy sector. Process integration and optimization techniques are applied to forest industry sites in order to improve the resource and energy efficiency, the benefits of the integrated design configurations being evaluated by both technical and economic analyses. The integration of woody biomass with intermittent renewable energy sources is also studied in order to enable a large share of non-fossil sources in the energy mix.

The results of the investigations show a significant potential for improving biomass resource utilization in the forest industry sites strictly from the energetic point of view. Optimizing the process integration in sites including Kraft pulp and paper mills and/or sawmills and a dedicated common CHP system can lead to a much greater power generation for the same input biomass and for the same production volume, or to a large amount of excess heat to be used in nearby processes or district heating, or even to the re-routing of part of the input biomass to other conversion processes (e.g. lignin separation and hemicellulose fermentation to produce biofuels). The operational profit of the site is consequently increased, but, when the investment costs are considered, some form of subsidies to the “green” byproducts are usually still required to make the integrated design configurations economically viable. The integration of woody biomass with intermittent renewable energy sources can result in an increased efficiency of hybrid power generation plants (e.g. with concentrated solar thermal collectors), and on a large scale it could facilitate the decarbonization
of the energy sector with the fundamental contribution from power-to-X technologies in order to produce chemical fuels from the excess intermittent electricity. These technologies would be clearly incentivized by a carbon tax, but the benefit deriving from the large volumes of captured CO$_2$ that are required for the synthesis of chemical fuels through co-electrolysis should also be taken into account.

**Keywords:** Forest industry, process integration, pinch analysis, HEATSEP method, optimization, CHP system, techno-economic, biorefinery, intermittent renewables.
List of appended papers

This thesis is composed of the following papers. The papers are referred in the thesis according to the assigned roman numerals.


Related work not included in the thesis


Technical reports


Conference contributions


Co-authorship statement

Papers I-II
Mesfun is the lead author. Mesfun and Toffolo developed the process integration models and performed the optimizations. Mesfun planned and wrote the papers under the supervision of Toffolo.

Paper III
Mesfun is the lead author responsible for developing of the process simulation model. Mesfun performed all the calculations with input data provided from Smurfit Kappa, a chemical pulp mill located in Piteå, Northern Sweden. Kudahettige-Nilsson provided experimental data regarding butanol fermentation. Mesfun planned and wrote the paper. Toffolo, Lundgren, Grip and Rova supervised the work.

Paper IV
Mesfun is the lead author responsible for performing the process integration, optimization and economic analyses. Mesfun and Umeki developed the process integration models and Anderson provided data regarding Nordic sawmills. Mesfun planned and wrote the paper under the supervision of Umeki and Toffolo.

Paper V
Paper V is a joint effort of Tanaka, Mesfun and Umeki. Mesfun performed the process integration and optimization analyses and contributed to paper writing under the supervision of Toffolo.

Paper VI
Mesfun is the lead author responsible for enhancing the Alps BeWhere model. Mesfun performed the simulations and wrote the first draft of the paper. Sanchez processed the input data for intermittent renewable energy sources and contributed to paper writing. Biberacher helped Sanchez with input data processing. Leduc is the original model developer and closely supervised the work. Wetterlund supervised the work in depth and contributed to paper writing. Lundgren and Kraxner supervised the work.
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**Nomenclature**

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Acknowledgments
1.1 Background

The largest share of the global primary energy supply, including the industrial, residential and transportation sectors, is currently met with carbon-based fossil fuels, mainly oil (31%), coal (29%) and natural gas (20%) [1]. Uncontrolled utilization of carbon-based fossil fuels via conventional conversion technologies (e.g. combustion of natural gas and gasified/pulverized coal in boilers/gas turbines, or combustion of petroleum-derived liquid fuels in motor vehicles) results in a big amount of CO$_2$ released to the atmosphere. CO$_2$ is by far the largest contributor (76% in 2010 [2]) to the accumulation of greenhouse gases (GHG) in the atmosphere, leading to global warming and other detrimental consequences for the environment.

A recent report from the Intergovernmental Panel on Climate Change (IPCC) indicates that the concentration of CO$_2$ in the atmosphere has reached a level that is 40% higher than that of the pre-industrial period (290 ppmv in the mid 1800s against 394 ppmv in 2012 [4], see Figure 1.1), primarily because of fossil related emissions [5]. About 42% of the global CO$_2$ emissions in 2011 were the result of electric power and heat generation, and nearly half of it came from the industrial sector [4,6]. The transportation sector is the second largest emitter, accounting for about 22% of the global CO$_2$ emissions [4].

Different models have been used to predict CO$_2$ emissions and concentration in short, medium and long-term scenarios. These models, called representative concentration pathways [5], have been also used to investigate the effects of mitigation policies aiming at stabilizing GHG concentration at levels that would prevent dangerous interactions with the climate. Measures improving energy conservation and
efficiency [2,7] and a deep decarbonization of the energy sector [2] are considered to be among the most important policies (see Figure 1.2).

Measures improving energy conservation and efficiency in energy intensive industries can play a significant role in achieving emissions targets, since the sector contributes about 36% of the global CO$_2$ emissions [8,9]. Accordingly, energy intensive industries have turned their focus into maximizing the process-to-process internal heat recovery, increasing the efficiency of component operation, integrating innovative process pathways, and recycling the waste streams. These actions resulted into new and more complicated process configurations with a larger number of design parameters that need to be investigated. The complex task of minimizing energy usage would require the development of representative models of the industrial sites that include the utility network, and the formulation of appropriate algorithms that are able to solve the synthesis/design optimization problem and are interfaced with the developed models.

A deep decarbonization of the energy sector through a large scale exploitation of renewable energy sources (RES) can significantly reduce CO$_2$ emissions from the energy sector. Low- or zero-carbon energy resources (e.g., sustainable biomass, hydro, solar, wind, geothermal and wave) can play a transformative role in enabling carbon-neutral economies, with potential applications in power, heat, chemical and transportation sectors. However, solar, wind and wave technologies are characterized by the intermittent nature of the energy source, as well as by the mismatch
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Figure 1.2: Cumulative emission reduction due to the four different representative concentration pathways in [7].

between demand and supply in both time and space, and therefore their integration in the grid present significant challenges. A system-level approach combining long-term development programs with an adequate representation of spatial and temporal features of RES is absolutely essential to provide a sufficient insight to address these issues.

1.2 Aim and objectives

The overall aim of this work is to expand the knowledge about the efficient utilization of woody biomass, one of the most important RES in Sweden, as a way to contribute to the reduction of GHG emissions. The content of the work follows two main “tracks” according to the policies and the related challenges identified in the previous section. Process integration measures for the improvement of resource and energy efficiency are considered in existing forest industry plants, in the perspective of the transformation of such plants into future biorefinery sites. Moreover, the integration of woody biomass utilization with other RES, namely solar, hydro and wind power, is investigated in the context of low-carbon energy systems.

Within this broad research framework, specific objectives of this work are to:

- apply advanced process integration and optimization strategies to achieve feasible process configurations leading to improved resource and energy efficiency in the forest industry;

- quantify the impact of such strategies, in terms of potential for biomass resource saving, increased productivity and/or improved energy efficiency;
Introduction

- perform techno-economic analyses about the integration of innovative technologies in existing forest industry (e.g. biofuel production pathways) and in power grids (e.g. power-to-gas technologies);
- integrate woody biomass utilization with intermittent RES to enable low-carbon energy systems featuring a large scale exploitation of renewable sources.

Table 1.1 summarizes how these objectives are related to the papers appended to the thesis.

1.3 Overview of appended papers

The work presented here considers different scales of energy system analysis. The boundaries of the investigated systems are progressively extended from a single process unit within a plant to a cluster of industries, and then further to a complete regional energy system. Insights from a broader economic perspective are also provided in some of the studies.

The mathematical model of a Kraft pulp and paper mill has been developed based on the data from an actual mill in order to establish a reference case for investigating the potential for improving energy efficiency in the chemical pulping process as well as to set a basis for exploring biorefinery concepts based on chemical pulp mills. The details of the model mill along with the results of an energy optimization study that consider three process integration system boundaries (the multiple effect evaporator (MEEV), the mill subprocesses and the total site) are documented in paper I.

A process integration model with even larger system boundaries is developed for an industrial site featuring a Kraft pulp and paper mill, its supply chain (log choppers and sawmills), a wood pellet plant and a shared CHP utility. The model is used to investigate the potential for improving biomass resource utilization and/or increasing energy efficiency as an outcome of the integrated site operation by comparison with the corresponding standalone plant configurations. Different scenarios are considered according to the technologies of the shared CHP utility, and their distinct characteristics are presented in paper II.

The hemicellulose fraction of pulping wood is an underutilized resource in Kraft pulp mills. The hemicellulose ends up in the black liquor, which is simply combusted in a recovery boiler to satisfy the thermal energy demand of the mill as well as to facilitate the recovery of the pulping chemicals. A process integration model has been developed to describe the extraction of the hemicellulose and part of the lignin fraction in pulping wood, and the conversion of the hemicellulose into bio-butanol via a conventional fermentation process. The model is used to assess the impact
of the extraction of hemicellulose and lignin on mill operation and on the energy balance of total site, including the fractionation plant, as it is reported in paper III.

Sawmills produce large quantities of biomass byproducts, about 53% of the input timber on a dry basis. Currently, most of the byproducts are exported to other process industries such as chemical pulp mills and wood pellet plants. The concept of integrating innovative substitute natural gas (SNG) plant with a typical Nordic sawmill is investigated from an energetic and economic perspectives. In order to assess economy of scale effects, different sawmill sizes are considered and the results are summarized in paper IV.

A process integration model featuring a hybrid biomass gasification - concentrated solar thermal (CST) power plant is proposed and studied in the context of the large scale integration of woody biomass with intermittent RES. The synergy in such configuration is investigated by considering a case study in which the biomass resource covers the base load power of the plant and the CST contributes a variable thermal input. The effects of different proportions between the input contributions from the two integrated sources are discussed in paper V.

The impact of the integration of woody biomass with temporally and spatially intermittent RES in power grids has also been investigated, the utilization of excess intermittent power being exploited using power-to-X processes. The power-to-X processes considered in this work assume that co-electrolysis of steam and CO₂ using high-temperature solid oxide electrolytic cells (SOECs) is a mature technology that can be deployed on a large scale. In these cells, the over-generated power from the grid is first used for the electrochemical reduction of gas-phase H₂O and CO₂. The resulting reduced gas, essentially similar in composition to a syngas, can then be used for the synthesis of higher quality liquid/gas fuels. This energy system solution is investigated using the Alpine region as a test case. Its engineering and economic potential both as a storage for intermittent RES and as a low-carbon source of energy for different sectors (heating, transportation and power) is discussed in paper VI.
Table 1.1: Objectives in the appended papers

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<th>Appended papers</th>
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<tr>
<td><strong>Paper I.</strong> Optimization of process integration in a Kraft pulp and paper mill-Evaporation unit and CHP system</td>
<td>- Apply advanced process integration and optimization strategies</td>
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<td>- Quantify the potential for resource saving, production increase and/or efficiency improvement</td>
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<td>- Perform techno-economic analyses to assess the benefits from the integration of innovative technologies</td>
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<td>- Integrate woody biomass utilization with intermittent RES</td>
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<tr>
<td><strong>Paper II.</strong> Integrating the process of a Kraft pulp and paper mill and its supply chain</td>
<td>- Apply advanced process integration and optimization strategies</td>
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<td>- Quantify the potential for resource saving, production increase and/or efficiency improvement</td>
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<td><strong>Paper III.</strong> Black liquor fractionation for biofuels production – a techno-economic assessment</td>
<td>- Apply advanced process integration and optimization strategies</td>
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<td>- Quantify the potential for resource saving, production increase and/or efficiency improvement</td>
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<td><strong>Paper IV.</strong> Integrated substitute natural gas (SNG) production in a typical Nordic sawmill</td>
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<td><strong>Paper VI.</strong> Power-to-gas and power-to-liquids for managing renewable electricity intermittency in the Alpine Region</td>
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A brief summary of literature based on relevant previous work in relation to the specific objectives of this thesis is presented in this chapter. Particular attention was given to studies about the forest industry and the large scale exploitation of RES implementing approaches that are similar to those used in this work.

2.1 Forest industry

Sweden has a large forest industry with an annual production capacity of about 8 million tonnes of chemical wood pulp and about 18 million cubic meter of sawn wood in 2014 [10].

The pulp and paper sector is one of the major energy intensive industries (it is the sixth largest energy consumer in Europe [11] and absorbs about 50% of the energy consumed by the industrial sector in Sweden [12]) and by far the largest woody biomass consumer. A large portion of the energy demand in pulp and paper mills is covered from biomass resources (mainly black liquor, a byproduct of the pulping process in the case of chemical pulping), and, consequently, anthropogenic GHGs emissions from the sector are not significant compared to the other energy intensive industries. However, fine tuning the pulp and paper manufacturing processes in order to improve their efficiency could result in better resource utilization. Saved biomass can be used to replace fossil-derived resources in other industrial sectors or can be utilized to produce biofuels for the transportation sector through innovative thermo/bio-chemical conversion technologies in the so-called biorefineries.
Previous work and motivation

Sawmills convert harvested timber into lumber. In so doing, large quantities of byproducts that significantly contribute to the biomass market are generated in the form of woodchips, bark and sawdust. The pulp and paper sector is the largest consumer of sawmill byproducts, mainly woodchips. Due to the sizable material flow connection between sawmills and pulp and paper mills, it is considered important to study the entire productive chain in the context of industrial symbiosis, in order to assess the potential for increasing the availability of biomass for other purposes.

2.1.1 Energy efficiency measures in forest industry

Energy efficiency measures in the forest industry in general, and in chemical pulp mills in particular, have been the subject of several studies in the literature, since the overall conversion process is composed of energy intensive subprocesses.

The heat integration opportunities for improving the energetic performance of a Scandinavian Kraft pulp mill is investigated in [13] using Pinch Analysis techniques. A conventional approach to save live steam and a process-integrated evaporation approach are implemented, both resulting in about the same steam surplus (52 and 56 MW) with similar investments (10 and 12 M€).

The pre-evaporation of effluents in a Swedish board mill to utilize the excess heat within the mill itself has been investigated as a case study in [14] using combined Pinch technology and the MIND method. Results showed that this is a cost-effective energy efficiency measure.

An energy audit of an existing pulp mill has been reported in [15]. The results showed that the considered pulp mill can be energy self-sufficient, with a potential in electricity saving equal to 22% of the total electricity use (during the period of time in which the audit was made), provided that all the machines were upgraded to or replaced by the most efficient counterparts available.

Energy conservation opportunities for Kraft pulp mills have been discussed in [16] using a systematic approach based on energy audits and benchmarking against existing and new mills. The mill data in the study included typical Nordic mills as well as mills in Northern and Southern America.

The potential for improving the efficiency and reducing GHG emission in US pulp and paper mills has been discussed in detail in [17]. The results showed that a primary energy saving potential of 31% can be achieved (16% in a cost-effective manner) and CO₂ emission reduction potential is 25% (14% in a cost-effective manner).

The benefits of using Pinch analysis about process steam and water usage in a
Previous work and motivation

Canadian Kraft paperboard mill is investigated in [18]. Results showed a saving potential of 15% of the steam generated in the boilers and a reduction of 6000 m$^3$/day in process water consumption.

Several researchers also investigated the concept of industrial symbiosis in relation to forest industry, see e.g. [19,20]. In an attempt to create a systematic map of the material and energy flows in an industrial sector, Wolf et al. [19] made an inventory of existing material and energy exchange in the Swedish forest industry. The results illustrate the existence of a significant degree of industrial symbiosis within the forest industry in the form of by-product exchange, as one third of the studied sites have some kind of material and energy exchange with adjacent sites.

The symbiosis within the Swedish forest industry is further discussed in [20] with a case study aimed at assessing the total cost benefit of integrating forest industry plants (a sawmill, a wood-pellet plant and a chemical pulp mill). The study concluded that industrial symbiosis can have economic benefits, although their magnitude needs to be evaluated on a case-by-case basis.

2.1.2 Biorefinery concepts based on forest industry

Future biorefinery concepts often have links with existing forest industry sites in order to exploit existing infrastructure. In particular, Kraft pulp and paper mills are regarded as existing examples of biorefineries because the chemical transformations that occur in the process of converting lignocellulosic biomass into pulp and other intermediate products can be readily reconfigured, so that value added chemicals can be produced along with (or instead of) the ordinary core products. Moreover, the increased global competition in the pulp and paper sector has driven the traditional pulp producers to focus on the production of value added products [21]. Several authors have accordingly investigated the Kraft pulp processes in relation to biorefinery concepts.

Andersson [22] performed techno-economic evaluations of different innovative downstream configurations based on pressurized-entrained flow gasification technology for the production of biofuels and green chemicals, namely methanol [23–25] and ammonia [26]. The study further investigated co-gasification of black liquor and pyrolysis oil, produced from biomass resources, in order to benefit from economy of scale effects.

A techno-economic assessment of the integration of substitute natural gas (SNG) from biomass process in a thermo-mechanical pulp mill is performed in [27]. This study identified the market price of SNG and the size of the pulp mill as the main parameters that determine the commercial feasibility of the considered processes.
Previous work and motivation

The technical and commercial feasibility of dimethyl-ether (DME) and/or methanol synthesis from the gasification of black liquor has been investigated and reported in detail in [28]. The same topic has been further discussed in [29] by investigating the integration of those production pathways in chemical pulp mills and assessing their conversion efficiency and biofuel production potential. Moreover, the impact on mill energy utilization from the integration of black liquor gasification with the subsequent production of biofuels or electricity is studied in [30] by considering different mill types, various future scenarios for energy prices, policy instruments, economic performance [31] and climate benefits in terms of CO$_2$ emissions [31,32].

Comparison of methanol synthesis using pressurized and dry (with direct causticization) gasification of black liquor has been studied in [33]. The paper further discussed the comparison of methanol production against other biorefinery products (hydrogen, DME and bio-methane). Results showed that the production of methanol is more than doubled in the case of pressurized gasification of black liquor. In addition, a potential global reduction of CO$_2$ emissions of 117 million-tonnes/year can be achieved assuming that all the black liquor available globally is used to produce methanol according to the proposed method.

The integration of a hydrogen production plant supplied by black liquor gasification in a Kraft pulp mill has been explored in [34]. The results of that investigation showed a significant potential for the reduction of CO$_2$ emission and concluded that if the proposed process is integrated in all the chemical pulp mills in Sweden, it can result in a reduction of about 8% of the total CO$_2$ emission of the country.

Fornell [8] has investigated the techno-economic and process integration feasibility aspects in different biorefinery concepts based on a model Kraft pulp mill reconfigured as an ethanol production plant (see [35–37] for further reading). A techno-economic analysis about to a pulp mill converted into an ethanol plant is performed in [38] using two approaches, namely conventional or process integration measures to increase the energy efficiency. When both kinds of measures are implemented 54 to 62 MW of steam are saved, which correspond to a decrease in the steam demand decrease of about 36 to 42%.

The integration of a process to extract hemicellulose and produce ethanol in a conventional Kraft pulping process is investigated in [39] from an energetic point of view. Pinch analysis is used to investigate the potential for improving the internal heat transfer. The study also illustrates a way to improve the water utilization within the integrated mill using water composite curves that include all water sinks and sources. The results indicate that the implementation of the proposed efficiency measures would reduce the steam consumption of the Kraft pulping process by about 5.04 GJ/Adt.
A techno-economic analysis about the use of surplus steam from heat integrated Kraft pulp mills is documented in [40]. Two alternatives, lignin separation and increased power generation, are investigated in a model mill. With the first alternative 326 to 500 GWh/year of lignin could be extracted, resulting in a reduction in power generation of 19 to 30% compared to the base-case. With the second alternative 97 to 127 GWh/year of extra power could be generated, which correspond to an increase of 50 to 65% of the base-case power generation.

A comparison between two strategies to increase the capacity (“debottlenecking”) of Kraft pulp mills is investigated in [41], i.e. integrating a lignin separation process versus upgrading the recovery boiler. Lignin and electricity prices are identified as the key parameters for the decision makers. Lignin separation, combined with steam saving measures, show a better profitability for lignin prices above 15 €/MWh, and up to 346 GWh/year of lignin can be exported. However, higher electricity prices combined with a long-term investment strategy make the upgrade of the recovery boiler more profitable, and if a new turbine is also installed 109 GWh/year of extra power can be generated (a 56% increase compared to the base-case).

2.2 Integration of woody biomass with intermittent RES

Wise utilization of low- or zero-carbon energy sources could play a crucial role in the deep decarbonization of the power, heating and transportation sectors, thereby clearing the path towards achieving short and long-term climate change mitigation goals. Electricity storage is not an issue as long as the intermittent RES, such as wind, solar and wave power, only account for a minor share of the energy supply. However, when the intermittent power capacity is significantly increased, electricity storage becomes a need in order to fully utilize RES potential. There will be times when a large surplus of electricity is produced, and large scale storage systems, such as batteries, compressed-air, flywheel and pumped-hydro solutions could help to balance the supply-demand mismatch.

Two approaches at very different system scale are considered in this thesis. The first approach is the hybridization of the concentrated solar thermal (CST) with biomass power generation, so that the integration of the intermittent RES can be realized without compromising the overall plant availability. The second approach is the large scale deployment of power-to-gas and power-to-liquid technologies for enabling low-carbon energy systems. A brief summary of the state of the art about both technologies is presented in the following subsections.
2.2.1 Concentrated solar-biomass hybrid systems

Major drawbacks with standalone concentrated solar power (CSP) systems are the high specific investment cost and the intermittency of power output. Peterseim et al. [42] discussed several CSP hybrid options with different energy sources, such as biomass, coal, natural gas and geothermal, and estimated that the cost of hybrid CSP can be up to 50% lower than that of standalone CSP plants.

The techno-economic assessment of a hybridization case study based on an annual woody biomass availability of 100 kilotonnes is reported in [43]. Seventeen different plant configurations are evaluated featuring different combinations of biomass conversion technologies (fluidized bed and grate combustion, gasification) and CSP collectors (parabolic trough, Fresnel and solar tower). In addition, three different working fluids are considered, namely thermal oil, steam and molten salt. The results show that solar tower-biomass gasification hybrids reach the highest efficiency (32.9%), but Fresnel-fluidized bed hybrids have the lowest specific investments. The article concluded that the latter benefit outweighs the additional power generation in the former case.

The concept of boosting CSP efficiency using externally superheated steam generated with biomass energy is discussed in [44]. The CSP is based on solar parabolic trough technology, with a capacity of 50 MWel and 7.5 h molten salt thermal storage, raising steam at 380°C and 100 bar. The study is conducted using four types of biomass fuels to externally superheat the steam from 380°C and 100 bar to 540°C and 130 bar. The results show that the peak solar-to-electricity efficiency can be increased by 10.5%, the gross efficiency reaching above 30%. The corresponding reduction in CSP investment is estimated to be up to 23.5%.

Another case study with a similar concept of solar (parabolic trough) aided biomass power plant, with a capacity of 12 MWel, is reported by Hou et al. [45]. In this configuration the solar thermal energy is used to preheat the feed water replacing the steam extractions from the turbine. The resulting annual solar-to-electricity efficiency is 21%, while the standalone solar plant efficiency is about 14%.

A tri-generation configuration (i.e. with production shift among electricity, heat and cooling) for a hybrid CSP-biomass power plant, with a capacity of 2-10 MWth, is investigated in [46]. The study concluded that, for a small to medium scale of the biomass plant capacity, tri-generation is feasible in India provided that the existing solar capital subsidies remain in place (i.e. 30% grid connected and 60% off-grid). However, at the scale of the considered plant hybrid electricity generation plants are not feasible.

Other studies (see, e.g., [47–53]) also focus on a different hybridization approach,
Previous work and motivation

i.e. supplying the thermal energy demand of the biomass gasification process with solar energy. Such configurations boost the yield of syngas and the conversion efficiency of the gasification step. However, the operational instability due to intermittent solar radiation may overcomplicate process control.

2.2.2 Power-to-X technologies

Technologies exploiting RES are typically designed to produce electricity (such as wind, solar and hydro power), so that the share of fossil fuel consumption can be reduced in the power generation sector and, to a lesser extent, in the transportation sector thanks to electric vehicles. This would however limit the role of RES in the large scale de-carbonization of energy systems, which are responsible for CO$_2$ emissions from a wide-range of sources in addition to the power generation sector. Under such circumstances, electrochemistry can add operational flexibility by establishing a direct link between the chemical energy in the bonds of liquid/gas fuel molecules and electric energy. Power from intermittent RES can be converted to synthetic fuels via water electrolysis for the production of hydrogen [54,55] or, even better, via high temperature co-electrolysis of steam and CO$_2$ for direct production of syngas, see e.g. [56–59].

Varone and Ferrari [60] made a preliminary assessment of RES power generation and production of synthetic fuels in 2050 Germany. Their assessment is based on the assumption that the share of RES in the electricity mix of 2050 Germany is in the range from 66% to 94%, as the outcome of a projection from the situation in year 2012. The 94% renewable share scenario results in an overflow of about 84%, i.e. the period of time during the year with excess production of electricity amounts to 7343 h. This corresponds to 1052 TWh of surplus power, which, assuming a power-to-methanol conversion efficiency of 70%, results into 737 TWh of methanol with a production cost of 81.4 €/MWh.

A comprehensive review on the power-to-gas plants that have already been realized or are being planned is presented in [61]. The review includes information about the components, the installed capacities and their operating experience. Furthermore, an overview of power-to-gas technologies is reported in [62], including the systems analysis and the economic assessment for a case study in Germany.

Jentsch et al. [63] investigated the optimum power-to-gas capacity and its spatial distribution in an 85% renewable energy scenario for Germany. The analysis is based on a simplified power transmission network for the entire country. The results indicate that the optimum power-to-gas capacity for the single plants falls in the range of 6 to 12 MW, depending on the share of the excess power converted to heat.
2.3 Concluding remarks on literature

Most of the works regarding forest industry summarized in Section 2.1 focus on the integration of innovative processes utilizing biomass resources and excess energy that are already available in chemical pulp mills. In some cases, new conversion technologies may also be considered as alternative routes to existing subprocesses in chemical pulp mills, e.g. gasification of BL is often considered as an alternative technology to the combustion in the recovery boiler. However, the studies tend to focus on the mill sections that are directly involved in the innovative process to be integrated, and often leave unaltered the configuration and the operational parameters of the rest of mill subprocesses without considering further potential synergic effects. Other studies focus on improving the energy efficiency in specific sections of the mill, possibly overlooking energy saving potentials that can be realized by considering wider boundaries.

The investigation of energy efficiency measures and innovative biorefinery concepts in forest industry plants are objectives of this thesis as well. The fundamental approach considered here, however, is based on the concept of fully integrated design solutions that are identified by using advanced integration and optimization tools. System boundaries may range from a specific subprocess within a plant to the limits of the plant and beyond, including other plants from the same or a different industrial sector in the framework of integrated industrial sites. In these fully integrated design solutions the plants and their processes complement each other by exchanging both material and energy flows, and in particular the configuration and the design parameters of the common CHP utility are always completely redesigned according to the optimization criteria.

Several studies have already investigated the way in which intermittent RES should be introduced on a large scale into the energy mix. Most of them lack a simulation approach that combines an energy system model in which the temporal and spatial characteristics of RES are adequately described with a model of the power transmission grid. The wide system-level view adopted in this thesis work is essential to assess the technical feasibility and the economic implication of long term development programs at regional/national scale, in order to facilitate the penetration of RES in the energy sector and enable low-carbon energy systems. Moreover, to the best of the author’s knowledge, the integration of power-to-X technologies into a high-resolution energy system model for decision support at regional level is something not yet investigated in the literature. This integration represents an important step for the utilization of excess power coming from intermittent RES in other sectors than the power generation one, in the perspective of an even deeper de-carbonization of energy systems.
Overview of the processes

A brief technical overview of the main technologies considered in the process integration studies reported in the appended papers is presented in this chapter. For each of them, a short description is given about the most important design and operating features of the related processes.

3.1 Kraft pulp and paper mill

Fibers in wood are glued together by lignin, and the pulping process basically releases the fibers by removing the lignin. There are different techniques for pulp production depending on the nature of the delignification process: mechanical, semi-mechanical, and chemical. The mill type considered in this thesis implements a sulphate delignification technique, and the description below refers to the so-called Kraft pulp process.

The Kraft pulp mill is an energy intensive process by which wood chips are converted into pulp. Pulp is the intermediate product from which different types of finished paper products and other materials are made. The heart of the Kraft pulp process is the digester (delignification step), where lignocellulosic wood fibers are separated to form pulp with the help of cooking chemicals. The cooking chemicals are in this case mainly composed of sodium hydroxide (NaOH) and sodium sulfide (Na$_2$S) dissolved in water, and this solution is known as white liquor. Following the chemical delignification step the digestion liquor is separated into two streams, the spent liquor stream and the cellulosic wood fiber stream. The wood fiber stream is washed, bleached, pressed, and dried in subsequent stages to produce market quality...
Overview of the processes

Figure 3.1: Schematic of the processes in a Kraft pulp and paper mill

pulp. The Kraft pulp mill may also have an integrated paper machine for on-site paper production.

The relevant substances in the spent liquor stream are the cooking chemicals with degraded sodium carbohydrate and the lignin component of the wood, which jointly form a high water content solution known as black liquor (BL). The BL, which exits the digester having 14-18% dry content, is concentrated in a multiple effect evaporator up to 70-75% dry content. The thick BL is subsequently burned in a recovery boiler (RB) to generate high pressure (HP) steam required for producing electricity and process heat as well as to recover the spent chemicals in the form of hot molten ashes (or smelt, mainly composed of Na₂CO₃ and Na₂S). The HP steam is expanded in a steam turbine to generate power, but some of it is extracted at intermediate stages to fulfill process steam demand at medium and low pressure levels. In order to satisfy the overall heat demand of the mill, additional HP steam is produced in a biomass fired boiler, in which biomass in the form of bark or residues as well as oil are burnt.

The spent chemicals collected from the bottom of the RB undergo a chemical recovery loop that has several stages. The smelt is dissolved in water or weak white liquor to form green liquor, which is mainly composed of water, Na₂CO₃, NaHS, NaOH and traces of Na₂SO₄. The green liquor reacts with lime (CaO) in the causticization process to produce white liquor as main product (which is rich of NaOH) and calcium carbonate (CaCO₃) as byproduct. Finally, the white liquor is sent back
to the delignification process whereas CaCO$_3$ is fed to an on-site lime kiln to recycle the lime. Figure 3.1 presents a schematic of a Kraft pulp and paper mill, while Figure 3.2 shows a block diagram of plant flowsheet in which the processes that generate or consume process heat are highlighted.

3.2 Typical Nordic sawmill

Sawmills produce wooden boards used in construction and manufacturing industries from forest trees, according to the sequence of processes shown in Figure 3.3. Sawmills also produce large quantities of byproducts (about 53% dry wt. of input timber) in the form of bark, woodchips and sawdust, see Figure 3.4.

Sawmills consume considerable amounts of electricity and heat for their processes. In particular, about 70% of the total energy demand is in the form of heat [64], which is mainly required for drying the sawn wood (lumber) boards. The initial moisture content of lumber is about 55-60%, and is reduced to about 18% in the drying kilns by circulating heated air as drying medium. The drying process is driven by the humidity difference between the drying air and lumber moisture content. Outdoor air is continuously supplied to the drying kilns, after being heated to a temperature of about 75 °C, to displace the humidified air inside the kilns. The heat to raise the temperature of the outdoor air usually comes from the combustion of part of the byproducts in a dedicated furnace. Accordingly, a typical sawmill
Overview of the processes

Figure 3.3: Schematic of the processes in a typical Nordic sawmill [68]

consumes about 23% dry wt. of the byproducts for internal use [65]. The remainder of the byproducts are exported to other industries such as chemical pulp mills and wood-pellet plants or sold as solid fuels. Although bark is the least economically attractive byproduct, the dedicated furnace is fueled with a mixture of the byproducts (bark 85%, sawdust 9%, and woodchips 6% wt. on dry basis) in order to avoid the problems that may arise from the combustion of bark alone (the combustion of bark is problematic due its high moisture content, high ash content and low calorific value, see e.g. [66,67]).

A typical material balance in a Nordic sawmill is presented in Figure 3.4, as described in [65].
3.3 Woody biomass gasification

In this thesis, gasification technology is considered for converting biomass to supply the energy demand of the integrated industrial sites as an alternative to conventional combustion in boilers or as an intermediate process for upgrading biomass into higher quality biofuels. Gasification adds flexibility to the utilization of biomass by streamlining diverse biomass feedstocks into a homogenous product.

The term “gasification” refers to the process of converting solid or liquid fuels into synthetic gas in an oxygen deficient reactor at elevated temperatures. Part of the chemical energy in the solid or liquid feedstock is recovered in the conversion as chemical energy in the synthetic gas, often referred as syngas.

There are different types of gasification technologies, broadly categorized according to the type of bed and flow configuration. A gasifier can be a fixed/moving-bed reactor [69–71], a fluidized-bed reactor [71,72] or an entrained flow reactor [73,74]. The details of these technologies such as feed quality, operating conditions and capacity ranges, along with their advantages and disadvantages, are well documented in the literature, see e.g. [75,76]. In this thesis fluidized bed and pressurized entrained flow gasifiers are considered for woody biomass and black liquor gasification, respectively. The typical configurations of these gasifiers are shown in Figure 3.5.

Depending on the choice of oxidizing agent (for e.g. air, oxygen, steam or CO₂) the composition of the syngas can be optimized to suit end-use. In the case of air-blown reactors the syngas, often referred as product gas, is diluted with nitrogen, which makes it unfavorable for synthesis of high grade biofuels. On the other hand, in oxygen and/or steam-blown reactors the syngas is primarily composed of carbon
Overview of the processes

monoxide (CO) and hydrogen (H₂), which is often preferable for synthesis of high quality biofuels, see e.g. [29,34,77].

![Diagram of gasification technologies](image)

**Figure 3.5:** The reactor configuration for two types of gasification technologies, circulating-fluidized-bed gasifier (a) and Chemrec entrained-flow gasifier (b).

### 3.4 Solid-oxide electrolysis cells (SOECs)

Electrolysis is the fundamental process in the operation of power-to-X technologies. It is an electrochemical process in which direct electric current is passed between two electrodes through a ionized medium (electrolyte) to deposit positively and negatively charged ions onto their respective electrodes. There are different types of electrolytic cells according to the nature of the electrolyte material and cell operating conditions. Alkaline water electrolysis, with a liquid alkaline electrolyte, and acidic proton exchange membrane (PEM) electrolysis, with a proton-conducting polymer electrolyte, are relatively mature technologies and most existing pilot power-to-gas plants employ either of them or both [61], while the high-temperature electrolysis with a solid oxide electrolyte is an emerging technology.
The overall conversion efficiency is defined as the ratio between output chemical energy in the final product (hydrogen) and input power, and depends on the operating pressure and cell current density [61,63]. It varies between 60-71% (based on the HHV of H₂) for alkaline type cells, and between 65-83% for PEM cells. Alkaline electrolyzers use an aqueous alkaline electrolyte, and are operated at temperatures in the range from 70 °C to 140 °C, pressures from 1 bar to 200 bars [61] and low current densities (0.2 – 0.4 A/cm²) [62]. PEM electrolyzers are limited to a maximum temperature of 80 °C due to the polymeric electrolyte, and are operated at pressures from 1 bar to 100 bars and high current densities (0.5 – 2 A/cm²) [62]. A detailed comparison between these two electrolysis processes, which are also categorized as low-temperature electrolysis, can be found in [62].

The high-temperature SOECs are of particular significance because they are operated in the range 700–1000 °C. In this way part of the energy required to electrochemically dissociate H₂O (in the case of water electrolysis) or H₂O(g) and CO₂ (in the case of co-electrolysis) is supplied as heat, so that the required electric input is minimized [78]. Thus, the performance of high-temperature SOECs has the advantage of both higher conversion efficiency and faster reaction rates [79,80]. The heat required to keep the cell at such high temperatures can be externally supplied via a heat exchanger when current density is low, or it can be internally generated due to the ohmic resistance of the cell itself when the SOEC is operated at high current densities.

The co-electrolysis process in SOECs is of substantial importance in the work presented in this thesis. Co-electrolysis achieves products that can be readily upgraded in a one-step process into liquid or gas fuels having an existing market infrastructure. In principle, synthetic gas (syngas) can be produced in a two-step process, i.e. electrolysis of H₂O to produced H₂ followed by hydrogenation of CO₂ into syngas through reverse shift water-gas reaction. Then the syngas can be catalytically upgraded into methane (Sabatier process) or higher quality hydrocarbons [62,78]. In this regard, the co-electrolysis process reduced the number of stages by directly depositing high quality syngas (mainly H₂ and CO) on the cathode thanks to the simultaneous electrochemical reduction of H₂O and CO₂. In so doing, the gas deposited on the anode is pure O₂, which could also bring additional value to the process. Moreover, the operation mode of SOECs allows to obtain a flexible composition of the syngas produced. For instance, the quality of the syngas can be tailored to enhance its catalytic conversion into synthetic fuels in the later stages of the process by controlling the composition of the feed stream to the SOEC [79].

Recent developments and performance improvements have demonstrated the effectiveness of high-temperature co-electrolysis of H₂O(g) and CO₂ in SOECs. Ohmic resistance as well as cell degradation rates and mechanisms are rather similar of
Overview of the processes

those in the electrolysis of steam alone [81,82]. In the light of such developments of SOECs, the overall efficiencies that can be expected are demonstrated to be about 70% for power-to-liquid conversion (the ratio of the calorific value of the liquid fuel produced, such as methanol, to power input) [60,83] and 80% for power-to-gas conversion (the ratio of the calorific value of the methane produced to power input) [84]. Figure 3.6 presents a conceptual power-to-X process configuration for balancing and long-term storage of intermittent power from RES.

![Figure 3.6: Conceptual scheme for balancing and long-term storage of intermittent power from RES with power-to-X technologies](image)

3.5 CHP system

This section presents a summary of the configurations of the shared CHP systems that are used in the appended papers and of the related decision variables that are considered in the optimization problems set for the total site integrations.

The two technologies that are considered for supplying heat to the steam Rankine cycle of the common CHP systems in the integrated industrial sites are:

- boilers (papers I, II, IV and V)
- gas turbines in a bIGCC configuration (papers II, IV and V)

The main purpose of the CHP system in papers I, II, IV and V is to cover the thermal requirements of the industrial processes and to generate power, which can be also used to partially or totally cover the electric requirement of the industrial processes. In some cases, steam is also directly consumed in process units and, as a consequence, part of the steam generated in the Rankine cycle of the CHP system
Overview of the processes

has to be extracted for this purpose.

If the hot utility demand were to be supplied directly from the combustion of the available biomass fuels, the exergy destruction would be too large, not to mention the technical constraints that could prevent the exchange of heat between the high temperature combustion gases and the process streams. The steam Rankine cycle of the CHP system is therefore designed to exploit the heat pockets generated by the hot thermal streams of the boilers (or the gas-turbine exhaust gases in the case of bIGCC) and those of the integrated processes, while providing process heat at the required temperature levels using condensing steam. The size of the heat pocket in the total site grand composite curve (GCC) is limited by the availability of the biomass fuel burnt in the boilers (or converted in the gasifiers in the case of bIGCC), which also limits the amount of power produced by the steam turbine.

In papers I and II, the steam generated by the boilers (or by the HRSGs in the bIGCC configuration in paper II) has three usages:

- part of the steam extracted at 4 and 10 bar and all the steam extracted at 30 bar are directly consumed (either mixed with the fiber line or used for soot blowing in the boilers) in the Kraft pulp and paper mill processes;
- part of the steam extracted at 4 and 10 bar is condensed to supply process heat;
- the remaining part is used for electricity generation through expansion in a steam turbine.

An amount of make-up water (1 bar, 5°C) equal to that of the steam directly consumed must therefore be supplied continuously to the steam cycle. The thermal streams associated with the generation of the directly consumed steam are accounted for with the process thermal streams in the integrated GCC representations (see Section 4.2).

The following changes are introduced (or considered) to the configuration of the base-case steam cycle in the Kraft pulp and paper mill CHP system:

- the maximum cycle pressure is raised from 60 (as in the base-case) to 100 bar;
- a reheat is introduced at 30 bar to the maximum cycle temperature (450°C);
- in the base-case, the steam is expanded to 4 bar in a back-pressure turbine; however the perspective of expanding the steam further down to 0.05 bar in a condensing turbine is explored in some of the scenarios evaluated for the integrated design cases.
Overview of the processes

In paper IV, the SNG process requires steam that is directly consumed at 15 and 30 bars during the gasification and methanation steps, respectively. Consequently, a biomass fired utility boiler is considered for generating the process steam. Accordingly, the following decision variables are used during the optimization of total site integration:

- the maximum cycle pressure and temperature of the steam Rankine cycle
- the fraction of biomass fuel mix (BFM) that goes to the utility boiler

The bIGCC configurations in paper IV generate high temperature streams that result in a heat surplus larger than the demand of the bIGCC and that of the sawmill, as such no utility boiler is needed for this configuration. As a result, the decision variables in the bIGCC configurations are:

- the maximum cycle pressure and temperature of the steam Rankine cycle
- the steam mass flow rates that circulates in steam Rankine cycle (part of the steam is directly consumed in the gasifier)

The bIGCC without the DH output option has an additional constraint that limits the steam condensed at 1.9 bar to be just enough to satisfy the sawmill heat demand. The shared CHP system of the hybrid bIGCC and solar thermal power unit investigated in paper V is designed to exploit the thermal energy of the exhaust gases from the gas turbine and the thermal input from the concentrated solar thermal system. Accordingly, the steam Rankine cycle is configured such that:

- the upper bounds of the maximum cycle pressure and temperature are limited to 100 bar and 540°C
- the steam demand of the gasification process is continuously satisfied with steam extraction from the steam turbine
Process integration and optimization in the field of energy intensive industries usually require representative mathematical models that are able to simulate the actual plant behavior as close as possible. The emphasis on and level of modeling details is somewhat predetermined by the intended objectives set for the integration and optimization investigations. The main focus of the models developed in this work is to simulate the considered industrial processes according to their mass and energy balances. This chapter briefly presents the simulation software, the process integration and optimization tools as well as the process evaluation methods that are used in this work.

4.1 Process simulation tools

The model for the Kraft pulp and paper mill is developed in the Simulink environment. Simulink is a graphical extension to MATLAB for modeling and simulation of generic systems and uses a block diagram user interface with blocks and arrows. The different processes occurring in the mill are represented by blocks, which can be regarded from a pure mathematical point of view as transfer functions. These blocks can be put in a sequence, to represent for instance a series of processes occurring sequentially, or one within the other hierarchically, to represent internal sub-units, according to the complexity of the represented processes. The arrows/signals that connect the blocks are actually travelled by numerical vectors containing thermodynamic state variables (temperature and pressure), physical quantities (mass flow rate and other properties) and chemical quantities (mass fractions defining stream composition).
The model of the conceptual process for lignin separation and butanol production presented in paper III is developed in Aspen Plus. Aspen Plus is a commercial software with a graphical interface for developing models and performing simulations of chemical processes. In addition, it is equipped with a wide range of built-in models to simulate the behavior of commonly used components and with a comprehensive database of substance properties. The property database and the built-in models make Aspen Plus convenient for simulating processes containing mixture streams that need separation and purification. Moreover, Aspen Plus allows the user to customize the behavior of the built-in component models using FORTRAN and/or Excel calculator blocks.

4.2 Process integration

Process integration (PI) is a broad subject dealing with the way in which different sections of industrial processes can interact within an overall flowsheet. A system perspective on this interaction should hopefully lead to an improved exchange/utilization of the available resources in a techno-economically feasible manner in order to fulfill a particular objective. Examples of the objective(s) could be the minimization of energy usage, the reduction of environmentally hazardous waste and the minimization of total cost (the sum of investment and operation costs related to a specific production). PI can be roughly categorized into mass integration and energy integration. Mass integration deals with the generation, separation and routing of species and streams throughout the process [85]. Energy integration deals with the allocation, generation and exchange of different forms of energy (in particular thermal energy) throughout the process [85]. The focus in this thesis is on energy integration using Pinch Analysis techniques and therefore a brief description of the method is presented here. The reader is referred to the literature in the field (e.g. [86–89]) for further details.

4.2.1 Pinch Technology

Pinch Analysis (PA) is a systematic approach that was initially developed to synthesize efficient heat exchanger networks. Given a list of the thermal streams in the process/plant under consideration, the strength of PA relies on the identification of the energy recovery potential and of the minimum energy requirement (MER) from external utilities prior to the detailed design of the heat exchangers. The method was first proposed in the late 70s by Linhoff and co-workers and, over the years, pinch-based approaches have been developed into standard methodologies for energy targeting and the design of thermal and chemical processes with the associated utilities.

The first step to energy targeting consists in the systematic construction of the
so-called composite curves (CCs). The CCs are obtained by plotting the cumulative enthalpy flows of the hot/cold streams according to the temperature ranges in which they are released/required, as shown in Figure 4.1a. When both curves are plotted on the same temperature-heat load (T-H) diagram, they show the opportunity for heat recovery as well as the heating and cooling duties required. In fact, the overlap between the horizontal projections of hot and cold CCs in the abscissas of the T-H diagram represents the heat recovery potential/target, while the non-overlapping horizontal projections of the hot and cold CCs represent the cold and hot utility targets, respectively. The relative position of the hot and cold CCs is dictated by the definition of the minimum temperature difference ($\Delta T_{\text{min}}$) that is allowed for any possible heat transfer among the streams, and the point of closest approach between the two CCs is called the process pinch. The pinch location divides the overall process into two distinct thermal regions [86,87,90];

- A net heat sink above the pinch temperature
- A net heat source below the pinch temperature

The following rules can be applied to the identified regions as a thermodynamic corollary in order to achieve the maximum energy recovery potential [87,91];

- No cold utilities must be placed above the pinch
- No heat transfer must occur across the pinch
- No hot utilities must be placed below the pinch

In fact, it is easy to demonstrate that the violation of one of these rules would result in an increase of the requirements from the external utilities and, at the same time, in a reduction of the internal heat recovery.

The other important tool in PA is the grand composite curve (GCC), which visualizes the overall heat transfer problem as a thermal cascade that is obtained through the Problem Table algorithm [87]. The information required by the algorithm is the same used for the calculation of the CCs, but the temperature ranges are now defined according to a shifted temperature scale and in each range algebraic sum of the thermal capacities of both the hot and cold streams is considered (Figure 4.1b). The shifted temperature scale is obtained by adding $\Delta T_{\text{min}}/2$ to the cold stream temperatures and subtracting $\Delta T_{\text{min}}/2$ from the hot stream temperatures, so that the heat transfer driving force within each shifted temperature range is at least $\Delta T_{\text{min}}$. Thanks to the visualization of the temperature ranges in which cumulated heat surpluses are made available and cumulated heat deficits are to be covered, the GCC readily identifies the possibilities for heat recovery through process to process exchange, as well as guides the selection and placement of utilities [87,90]. In fact, a distinct feature of the GCC is that it provides fundamental information about the
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temperature levels at which the utilities can be supplied.

**Figure 4.1:** Hot and cold composite curves (a) and grand composite curve (b)
More advanced integration opportunities can also be investigated by the so-called integrated grand composite curve (IGCC) representation, which extends the concept of GCC to allow for the integration of other thermal streams such as those belonging to the systems supplying the external utilities to the process. In the example shown in Figure 4.2, the IGCC visualizes how well the steam cycle streams from a CHP system are exploiting the heat pockets of the GCC obtained from the combined list of process streams and boiler/HRSG streams.

Taking advantage of the aforementioned PA tools, targeting procedures can be easily implemented in the design procedure of both heat exchanger network and, on a broader level, process flowsheets. In particular, useful design guidelines and algorithms (both ad-hoc and traditional) for the synthesis and optimization of energy intensive industrial processes have been developed and codified in the literature, see e.g. [92,93]. The HEATSEP method [94], which has been used extensively in this thesis, is among the notable and versatile methods that have been developed.
4.2.2 The HEATSEP method

In order to assist the development of an automated procedure for the synthesis and optimization of energy systems, the HEATSEP method suggests to separate the heat transfer section of a system by virtually cutting the thermal links between the basic system components. In this way, the temperature at the inlet of a component is free to vary independent of the temperature at the outlet of the preceding component, and the heat transfer section comprising all the thermal cuts can be represented by a ‘black box’ in which the actual heat exchanger network configuration is left undefined. However, the Problem Table algorithm has to be run for the potential hot and cold thermal streams generated by the thermal cuts in order to check the feasibility of the heat transfer. When the configuration and the design parameters of the basic system components are optimized, the mass flow rates and the temperatures of the thermal streams (i.e. the temperatures at the boundaries of the ‘black box’) can then be included among the decision variables.

As an example, an illustration of the heat transfer black box containing the thermal streams from a combined cycle power plant is shown in Figure 4.3.

![Figure 4.3: Example of a heat transfer black box for a combined cycle power plant with two pressure levels for steam generation.](image)

4.3 Process optimization

The optimizations are carried out using a single-objective Evolutionary Algorithm (EA) routine based on Genetic Diversity Evaluation Method (GeDEM), which is
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described in [95], and the Sequential Quadratic Programming (SQP) script “fmincon”, which is available in the MATLAB optimization tool box. The choice of the optimization routine depends on the mathematical nature of the optimization problems (see [95,96] for further details).

The single-objective EA is used to optimize the MEEV and the hot utility requirement of the Kraft pulp and paper mill. This is due to the fact that small changes in the decision variables may cause the pinch point(s) of the Problem Table to move from one temperature level to another resulting in discontinuities in the first derivative of the objective function (traditional optimization algorithms are not able to deal with this kind of discontinuities [96]). Moreover, the objective function is expected to have a multi-modal behavior because the different combinations of potential pinch points at the temperature levels of the different effects may result into several local optima.

A traditional optimization algorithm (“fmincon”) is chosen to solve the optimization problems set in all the total site integration scenarios considered in papers I, II, IV and V since no discontinuities are expected due to the mathematical nature of the problems. The temperature levels of all the thermal streams are fixed, so the heat transfer feasibility constraint is expressed by a set of inequalities (one at each temperature level, see [96]) that are linear functions of the mass flow rates of the thermal streams. If the objective function (e.g. the net power from the CHP system) can be calculated as an algebraic sum of terms in which specific enthalpy differences, derived from fixed steam conditions, multiply the steam mass flow rates, then it is linear in the decision variables as well.

The optimization routines are interfaced with a PA routine that retrieves the information about the thermal streams from the developed Simulink models. The PA routine checks the heat transfer feasibility within the ‘black box’ by solving the Problem Table algorithm according to the procedure set by PA [87]. A brief description of the sequence of steps performed during the optimization procedure is offered in the following text and depicted in Figure 4.4

The starting values of the chosen decision variables are initialized by the optimization algorithm, i.e. the SQP “fmincon” MATLAB function or the single-objective EA routine (as a population of solutions). These values are passed to the Simulink model and from that the quantities related to the hot and cold thermal streams are extracted and sent to the PA routine, which checks the condition according to which the accumulated heat made available by the hot streams has to be greater than or equal to the accumulated heat demand of the cold streams at all temperature intervals in the thermal cascade. The objective function value is then calculated and, on the basis of this information, the optimization algorithm proposes new values for
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the decision variables (in a search space limited by lower and upper bounds) that correspond to the solution(s) that will evaluated in the following iteration of the search procedure.

Figure 4.4: Scheme of the search procedure in the optimization problems about hot utility minimization (a) and steam Rankine cycle net power maximization (b)

4.4 BeWhere model

BeWhere is a geographically explicit cost optimization model, based on mixed integer linear programming (MILP), written in GAMS and uses CPLEX as solver. BeWhere minimizes the total cost of an energy supply chain in order to meet a known energy demand while providing information on optimal localization of new plant installations. Information such as data about resources, demand and locations for new plant installations are implicitly introduced into the model according to the
Methodology

geographic resolution used. The objective function is to minimize the total cost the energy supply chain and that of carbon emissions, as shown in Equation 4.1.

\[
\min f = \text{cost}_{\text{supplychain}} + \text{emissions}_{\text{CO}_2} \times \text{cost}_{\text{CO}_2}
\]  
(4.1)

The BeWhere model is initially developed at IIASA and Luleä University of Technology, with earlier applications focusing on bioenergy systems. Its application have been demonstrated by several researchers, for instance methanol via biomass gasification [97–99], second generation biofuels on a EU scale [100,101], cost-effective \( \text{CO}_2 \) emission reduction through bioenergy [102,103], polygeneration in different locations [104–107]. A detailed description of the BeWhere bioenergy model on a European scale can be found in [108].

In paper VI, BeWhere is used to study the impact of temporal and spatial intermittency of RES when investigating synchronized decarbonization of energy supply system in the Alpine Region.

The model takes into account the cost of the entire supply chain, according to the nature of the resource, that is:

- biomass (cost of biomass harvesting, biomass transportation to plants, bioenergy production, final energy commodity (i.e., bio-electricity to the power grid, biofuel to gas stations and bio-heat to the consumers);
- hydropower (cost of operation and transmission to the consumers);
- solar energy (cost of power production via state-of-the-art solar photo-voltaic technology and cost of transmission to the consumers);
- wind energy (cost of power production and cost of transmission to the consumers);
- fossil based energy (cost of fossil based power, heat and transportation).

and in relation power-to-X technologies, it takes into account the:

- cost of transmitting over-generated power-to-X plants;
- cost of converting the transmitted power into liquid or gas fuels;
- cost of transportation of produced fuels to gas stations or existing natural gas distribution hubs.

The power plants already in operation are also included, such as:

- biomass based CHP, pulp and paper mills etc. for the biomass feedstock allocation. First the feedstock demand from these industries is met, if there is enough feedstock left, new bio-energy production plants can be set-up;
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- hydropower stations (if a hydropower is in operation in a specific catchment area, no additional hydropower plant can be set up in the same catchment).

In addition, the model takes into account cost of CO$_2$ emissions in relation to:

- emissions from transporting biomass to processing plants and biofuels as well as liquid and gas fuels generated at power-to-X plants to gas stations;

- emissions from fossil fuels used to cover deficit in energy supply in all sectors (power, transport and heating). Any deficit in energy supply is assumed to be satisfied from fossil resources.

4.5 Economic evaluation

Several of the integrated industrial sites investigated in this thesis are assessed according to some economic indicator in order to emphasize their economic viability.

In paper II the optimal design solutions of the integrated industrial site are evaluated from an economic perspective using investment opportunity (IO) as an economic indicator. IO is a measure of the profitability of a design solution based on the algebraic difference between the product revenues and resources cost:

$$IO = \sum (\text{product revenues}) - \sum (\text{resource costs}) \quad (4.2)$$

IO offers an overall estimation of the operational profitability without the knowledge of the detailed investment cost of a design solution. The higher is the IO potential the larger is the margin for investing in a design solution while the project cash flow history remains positive at the end of its economic life time.

In paper III, the Study Estimates method, combined with the Hand method [109], and Aspen Icarus (now Aspen Plus Economic Analyzer) are used to estimate the investment costs. The sets of equipment which are required by the conceptual process are sized according to the mass and energy balance flowsheet developed in Aspen Plus. The evaluation of the equipment cost is performed by estimating the purchase cost of the components, on the basis of published data as well as in-house database information, and by multiplying them with their respective Hand factor to account for piping, insurances, installations etc. The capital cost is estimated according to the following expression:

$$\text{Capital cost} = \sum (\text{Equipment purchase cost} \times h_f \times f_m) \times f_i \times f_b \times f_p \quad (4.3)$$

where:

$h_f$ - Hand factor, $f_m$ - material factor, $f_i$ - instrumentation factor, $f_b$ - building factor and $f_p$ - place factor.
The cost of equipment for the components involved in the process are initially estimated using correlations and data available in literature [109]. The initial estimates are corrected to match the pressure and material requirement of the current process using factors reported in the same source.

In paper IV, the capital investment for the integrated configurations producing SNG or power is estimated by summing up all the installed equipment cost according to the process flowsheets. The equipment cost estimates are in turn calculated either by using module costing method [110] whenever possible or by scaling against published cost data using the power law of capacity if module costing is not applicable. The overall capital cost is then expressed by the following formula:

\[
\text{Capital cost} = [(1 + f_1) \sum_{i=1}^{n} C_{BM,i} + f_2 \sum_{j=1}^{n} C_{0BM,j}] + \sum_{j=1}^{m} f_j C_{E,j} \quad (4.4)
\]

Where:
- \( f_1 \) - a coefficient to account for contingency and fee costs depending on the reliability of cost data and completeness of flowsheet.
- \( f_2 \) - a coefficient to account for costs related to site development, auxiliary building and utilities.
- \( f_j \) - a coefficient to account for overall installation cost of equipment.
- \( i \) - set of equipment for which cost estimates are made based on module costing method.
- \( j \) - set of equipment for which cost estimates are recalculated based on published cost data.
- \( C_{BM,i} \) - bare module cost of equipment calculated at the actual process conditions.
- \( C_{0BM,j} \) - bare module cost of equipment calculated at the base rating.
- \( C_{E,j} \) - cost of equipment scaled from published cost data.
This section presents the most significant results in relation to the research objectives outlined in the introduction. Further discussions and more detailed information about the results can be found in the appended papers.

5.1 Process integration in the forest industry and quantification of its benefits

Advanced process integration and optimization techniques are applied to the model of a Kraft pulp and paper mill (paper I) and to a process integration model featuring a Kraft pulp and paper mill with its supply chain (paper II) in order to assess the potential for saving energy and resources.

In paper I a stationary model that reproduces the operation of a Kraft pulp and paper mill is developed in the Simulink environment. The process integration study is performed by selecting the decision variables that most affect the thermal energy utilization in the mill and by formulating some optimization problems that consider different system boundaries related to:

- the energy usage in the most energy intensive sub-process of the mill (the MEEV);
- the overall hot utility requirement of the mill;
- the integration of the total site with a common CHP utility, see Section 3.5.

It should be noted that the model of Kraft pulp and paper mill developed in paper I is also used as a basis for the analyses presented in papers II and III. A snapshot
Results and discussion

of the block diagram of the Simulink model is presented in Figure 5.1.

Figure 5.1: Simulink model of the base-case Kraft pulp and paper mill

The results of the optimization of the MEEV as a standalone process showed that about 21% of the base-case live steam demand can be saved thanks to the new configuration and design parameters. In terms of thermal load, the hot utility requirement of the optimized MEEV is reduced from 56.4 MW in the base-case to 44.7 MW and that of the cold utility from 64.9 MW in the base-case to 53.7 MW (see the comparison between the grand composite curves of the base-case and the optimized MEEV in Figure 5.2).

When the MEEV is optimized with the rest of the mill subprocesses, the hot utility requirement of the overall pulp and paper mill is reduced by 15%, from 78.9 MW in the base-case to 66.88 MW. In addition, a reduction of about 25% in the cold utility requirement is achieved in comparison to the base case. The chosen decision variables for the optimization are the pressures and the target DM contents of the evaporation units. The optimal values of the decision variables have resulted in an evener distribution of the thermal loads in the MEEV, which in turn has resulted in multiple pinch points at the temperature levels of the evaporation units (Figure 5.3).
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Figure 5.2: GCCs of the MEEV in the base-case (red solid line) and in the optimized case (blue dashed line)

Consequently, the GCC of the new case has moved closer to the ordinate axis showing a reduction in both the hot and cold utilities. Table 5.1 shows a summary of the total site integration scenarios evaluated in paper I, which differ according to the purpose for the utilization of the available biomass resources (BL and BFM) after the thermal requirements of the mill processes are covered.

Table 5.1: Summary of the total site integration scenarios evaluated in paper I

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Steam thermal power (heat to process/excess)</th>
<th>Electric power</th>
<th>Resources saving&lt;sup&gt;a&lt;/sup&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MP(MW)</td>
<td>LP(MW)</td>
<td>(MW)</td>
</tr>
<tr>
<td>1</td>
<td>8.3</td>
<td>58.1</td>
<td>62.1</td>
</tr>
<tr>
<td>2</td>
<td>7.8</td>
<td>55.3</td>
<td>51.6</td>
</tr>
<tr>
<td>3</td>
<td>5.2</td>
<td>54.3/80</td>
<td>45.5</td>
</tr>
<tr>
<td>4</td>
<td>5.3</td>
<td>50.7/62</td>
<td>39.2</td>
</tr>
<tr>
<td>5</td>
<td>3.5</td>
<td>46.1</td>
<td>19.2</td>
</tr>
</tbody>
</table>

<sup>a</sup>The amount of available resources in the base-case KPP mill is 25.43 kg/s of BL and 4.28 kg/s of BFM.
Results and discussion

Figure 5.3: GCCs of the KPP mill processes in the base case (red solid line) and in the optimized case (blue dashed line)

If power production is prioritized, scenario 1 presents the best option with all the available biomass resources used to generate about 62 MW\textsubscript{el} (about 88% more than the base-case mill operation) in a CHP system that includes a condensing steam turbine. On the other hand, the CHP system in scenarios 3 and 4 does not include the condensing turbine, so the condensation of LP steam produces a large amount of excess heat that can be utilized in other nearby industrial processes or in district heating networks. If resource saving is prioritized, scenario 5 shows that the combustion of about only half of the BL in the recovery boiler is sufficient to cover the thermal requirements of the mill processes, so all the BFM and the other half of the BL can be saved. In this case, the saved BFM and BL can be utilized in innovative downstream processes and transformed into high value products, such as “green” chemicals and biofuels.

Paper II investigates the potential for resources saving and/or increased power production as the result of having a Kraft pulp and paper mill, its supply chain and a wood-pellet plant all integrated in the same industrial site, which is assumed to
be served with a common CHP utility. The woodchip input to pulp and paper mills come from sawmills (as a byproduct) and from the chopping of wood logs (those that are not good enough for the production of lumber). Therefore, the industrial processes included in the site as the supply chain of the Kraft pulp and paper mill are those of sawmills and wood-log choppers. A diagram of the material flow connections within the integrated site is shown in Figure 5.4. The domestic wood harvested in Sweden is considered to be formed by equal shares, on a dry mass basis, of timber and wood logs [111], i.e. the two inputs in Figure 5.4. Figure 5.5 shows a Sankey diagram of the mass balance in the supply chain to pulp and paper mills in Sweden (see [64,111] for further details), showing the proportions between the output biomass flows from the “Logs chopper” and “Sawmill” blocks in Figure 5.4.

The production of pulp and paper in the integrated industrial site is kept the same as that of the reference standalone pulp and paper mill. The main difference in the utility requirements of the standalone Kraft pulp and paper mill and those of the integrated industrial site are highlighted in Figure 5.6, which shows the GCCs of the two cases.

As it is apparent from the comparison between the two GCCs, the expansion of the boundaries from the standalone Kraft pulp and paper mill to the integrated
industrial site has resulted in an increase of about 17.5% in the hot utility requirement. On the other hand, the cold utility has been reduced by 62% compared to the standalone Kraft pulp and paper mill case.

In order to emphasize the benefits of the integrated industrial site compared to the stand-alone plants, Table 5.2 reports a summary of the scenarios evaluated in paper II, which again mainly differ according to the purpose for the utilization of the available biomass resources after the thermal requirements of the processes in the site are covered (it should be noted that the amount of available BL remains the same as that of the stand-alone Kraft pulp and paper mill, whereas the amount of available BFM increases due to the additional byproducts from the sawmill).

<table>
<thead>
<tr>
<th>Scenario</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Net power [MW]</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Integrated (I)</td>
<td>77.76</td>
<td>121.91</td>
<td>2.56</td>
<td>-39.50</td>
<td>60.80</td>
</tr>
<tr>
<td>Stand-alone (SA)</td>
<td>36.72</td>
<td>60.60</td>
<td>-24.40</td>
<td>-24.40</td>
<td>-16.54</td>
</tr>
<tr>
<td>Resources saving[a] [kg/s, dry]</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Integrated (I)</td>
<td>BL 0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>18.05</td>
<td>13.61</td>
</tr>
<tr>
<td></td>
<td>BFM 0.00</td>
<td>0.00</td>
<td>15.96</td>
<td>9.21</td>
<td>0.00</td>
</tr>
<tr>
<td>Stand-alone (SA)</td>
<td>BL 0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>18.05</td>
<td>18.05</td>
</tr>
<tr>
<td></td>
<td>BFM 0.00</td>
<td>0.00</td>
<td>11.48</td>
<td>1.05</td>
<td>0.00</td>
</tr>
</tbody>
</table>

[a]The amount of available resources 18.05 dry kg/s of BL and 15.96 dry kg/s of BFM.
The results show that scenarios 1I and 2I are the best options if power generation is prioritized. In scenario 1 the CHP system is based on biomass boilers, while in scenario 2 the CHP system adopts a bIGCC configuration, but in both scenarios the integrated operation of the site achieves a power generation that is more than double of the one in the corresponding stand-alone cases. Of course, the bIGCC configuration in scenario 2I results in a much higher power generation (+58%) compared to that of scenario 1I. If resource saving is prioritize, scenario 4I shows that burning only 43% of the available BFM in a bark boiler is sufficient to cover the thermal requirements of the integrated site, so that all the BL and the remaining 57% of the BFM can be utilized in other processes. This, however, results in a large deficit of power that must be supplied from the grid. Alternatively, scenarios 3I and 5I show the power generation and the resource saving levels that can be obtained by intentionally gasifying all the available amount of either of the two biomass re-

Figure 5.6: GCCs of the integrated industrial site in paper II (red solid line) and of the stand-alone Kraft pulp and paper mill (blue dashed line)
Results and discussion

sources (all the BL in scenario 3I and all the BFM in scenario 5I, although in the latter case some of the BL has to be gasified as well in order to cover the thermal requirements of the integrated site). It is interesting to note that in scenarios 4 and 5 there is a trade-off between the amounts of saved biomass and generated power when the integrated site and the stand-alone plants cases are compared.

5.2 Biorefinery concepts based on forest industry plants - energetic performance

A conceptual process for extracting lignin and hemicellulose from hardwood black liquor and for converting the hemicellulose into butanol is proposed and studied in paper III.

In chemical pulp mills the hemicellulose fraction of the wood is an underutilized resource, so upgrading it through a techno-economically feasible process could generate an additional revenue for the industry. The conceptual process considered in paper III produces mainly lignin (a solid biomass fuel) and biobutanol (a motor fuel replacement/blend in Otto engines) from a stream of BL extracted from the digester of the Kraft pulp and paper mill. Part of the organic substances in the extracted stream are precipitated and filtered out to produce lignin and biobutanol through the conventional hydrolysis and fermentation pathway. The fermentative butanol process is often referred as the Acetone-Butanol-Ethanol (ABE) process (see e.g. [112–115]), as the involved micro-organisms usually produce considerable amounts of acetone and ethanol along with butanol.

This concept alters the operation of both the RB and the MEEV in the Kraft pulp and paper mill. On one side, the RB is often referred as the bottleneck against the increase of pulp production in Kraft pulp mills [116,117], then extracting part of the organic substance from the BL could help debottlenecking in such situations. On the other side, the same extraction could result in an increase of the energy demand in the evaporation unit of the mill for a given dry matter in the concentrated BL. Therefore, it is considered important to assess the impact of the extraction of organic substances from the BL on the configuration and the energy demand of the MEEV and on the thermal output of the RB. These effects are investigated in detail using the mathematical model of the Kraft pulp and paper mill developed in paper I, and the results are presented in Figure 5.7. The live steam demand of the MEEV can be higher or lower than that of the base-case (about 6.75 kg/s as indicated by the dashed line in Figure 5.7a) depending on the choice of the operating parameters, such as the dry matter content (60 to 70%) and the mass flow rate of the organic substances recirculated from the precipitated stream, the mixing temperature (80 to 120°C) of the extracted and recirculated streams of organic substances, and the dry matter content of the recycled alkali stream (12 to 23%). Moreover, a reduction
of the organic content in the BL decreases its energy content resulting in a lower thermal output from BL combustion in the RB. Figure 5.7b shows the impact of the extraction of organic substances from BL on the thermal output of the RB and the corresponding mass flow rate of the exhaust gases. The useful thermal energy output is found to vary between 53 and 30 MW, corresponding to an extraction of organic substances from BL in the range from 0 to 30%. The study of the proposed process assumes that the stream extracted from the digester contains about 10-30% of the organic substances in the BL. However, extraction of organic substances above 10% result in operating issues in some of the subprocesses of the Kraft pulp and paper mill, such as firing problems in the RB.

![Figure 5.7](image)

**Figure 5.7:** Impact of the 10% extraction of organic substances from BL on the live steam demand of the MEEV (a) and on the thermal output of the RB (b).

**Paper IV** investigates the potential for upgrading the byproducts of a typical Nordic sawmill into SNG or electricity on-site.

The alternative configurations of the sawmill with the integrated SNG process or with the integrated bIGCC process (with optional output to DH) are shown in Figure 5.8.

The results of the thermodynamic performance assessment show that 6 to 45 MW \(_{\text{LHV}}\) of SNG can be produced in sawmills having a production capacity of 50 to 400 thousand cubic meters of lumber per year, as reported in Figure 5.9. The net electric power exported from the CHP system in the site is also shown in the same figure, and it ranges from 0.5 to 7 MW \(_{\text{el}}\) depending on the sawmill size. The energy and exergy efficiencies of the different integrated configurations (bio-SNG process and
Results and discussion

The bIGCC process with and without the output to the DH are compared as a function of sawmill size in Figure 5.10. The integrated bIGCC configuration with DH output has the highest energy efficiency and the lowest exergy efficiency because of the low temperature heat output to DH, which is a big amount in terms of energy but not significant in terms of exergy (the exergy of the electric output from the condensing turbine is higher).

Figure 5.8: System boundaries of the alternative configurations for integrating a sawmill with a bio-SNG process (a) or a b-IGCC process (b)
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Figure 5.9: Energy input and output streams for the integrated SNG production in a sawmill

Figure 5.10: Overall energy (a) and exergy (b) efficiencies of the different integrated configurations in paper IV
5.3 Economic analyses

In paper II, the considered integrated and stand-alone configurations are evaluated from an economic viewpoint using investment opportunity (IO) as an indicator. In all the configurations, the integrated systems have resulted in higher IO potential as compared to their corresponding stand-alone cases, with annual margins in the range between 155 and 395 MSEK. In particular, the integrated cases with bIGCC configurations performed better due to the large amount of power produced by the gas turbines (see also Section 5.1).

The findings of the investigations about lignin extraction in paper III show that the process can be economically feasible for lignin selling prices above 36 USD/MWh. This result does not take into account any income from the other value added product resulting from the integration of a lignin extraction plant, that is the hydrolyzate that can be obtained on site from the extracted hemicellulose fraction. In view of that, an allocation of the production costs between the lignin and black liquor hydrolyzate indicate that lignin can be produced in the cost range between 0 and 36 USD/MWh while the corresponding cost range of BBLH is from 28 to 0 USD/tonne.

In addition, a butanol production plant that utilizes the black liquor hydrolyzate on site via a conventional ABE fermentation process is investigated, showing that butanol can be produced in the cost range between 7.1 and 3.6 kUSD/ton-butanol for lignin prices that are in the range from 30 to 50 USD/MWh. If acetone and ethanol are also considered to be sold to the market, the combined production cost for the solvents is lowered to the range between 5.6 and 3.6 kUSD/ton-ABE. The resulting price of butanol is rather high compared to its market value (1.03-1.61 kUSD/ton-butanol at the time paper III was written). The main reason for such high production cost is the low yield of solvents (acetone, butanol and ethanol) due to the toxic nature of black liquor hydrolyzate, which inhibits the growth of the bacterial culture of Clostridium acetobutylicum ATCC 824 [118]. In order to assess economy of scale effect, the 30% extraction of organics case is also investigated. Accordingly, the corresponding production costs per ton of butanol or ABE reduces by about 42% as compared to the 10% extraction of organics case for a lignin price in the range 30 to 50 USD/MWh.

In paper IV, IRR is used as an indicator to investigate the economic aspects in the integrated SNG and/or power production in a typical Nordic sawmill. Economically viable market prices of natural gas or electricity, depending on the process configuration, are calculated for three imposed IRR values (4, 8 and 12%) and are reported in Figure 5.11. The results show that market prices of natural gas in the ranges 77-123 €/MWh (4% IRR), 87-146 €/MWh (8%) and 99-171 €/MWh (12%) are required depending on sawmill size, larger mills corresponding of course to the lower
end of the cost range. Under the 2013 economic scenario, a biofuel support policy in the range of 28-52 €/MWh is required to obtain a break-even SNG production at 4% IRR for sawmills with a capacity larger than 106 thousand cubic meter of lumber per year. For the bIGCC configuration with DH output option, the ranges of the economically viable electricity market prices are found to be 74-191 €/MWh (4% IRR), 85-234 €/MWh (8%) and 98-282 €/MWh (12%). The bIGCC configuration with DH output option requires lower electricity market prices compared to the alternative without DH output, due to the large amount of thermal energy sold to DH (see Figure 5.11).

Figure 5.11: Economically viable market prices of natural gas and electricity in order to meet the IRR values imposed in the economic analysis in paper IV

5.4 Integration with intermittent RES

A hybrid power generation system based on the integration between a biomass gasifier and concentrated solar thermal collectors is investigated in paper V. The integrated bIGCC/CST hybrid is examined over a range of gasifying agent compositions and temperatures. The results show that when the gasifier is oxidized with
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enriched air at a temperature of 300 °C the overall electricity efficiency of the hybrid plant slightly decreases from 36.7% to 35.9% as the oxygen fraction is increased in the range between 21% and 40% vol. (see Figure 5.12a). This is because a higher O₂ content in the enriched air requires a higher power demand for oxygen production. On the contrary, the overall electricity efficiency slightly increases from 32.7% to 33.7% for the oxygen and steam gasification case as the oxygen fraction in the gasifying agent is increased from 20% to 40% vol. The lower electricity efficiency of the oxygen and steam gasification case is due to the additional energy demand for steam generation.

![Figure 5.12: Effect of composition and temperature of the gasifying agent on the overall electric efficiency with a 100 MW heat input from CST collectors: gasifying agents at temperature of 300 °C with varying oxygen fractions (a), and varying gasifying agent temperatures for oxygen fractions of 20% and 40% vol. (b)](image)

The potential benefit of the hybrid bIGCC/CST configuration is evaluated by comparing it with the corresponding stand-alone bIGCC and CSTP plants of the same capacity. The electric efficiencies of the stand-alone bIGCC and CSTP plants are 28.1% and 30%, respectively. A marginal power output term, defined as the difference in power output between the hybrid system and the two stand-alone plants, is used to quantify the synergy as a result of integrated operation, and is reported in Figure 5.13a. A marginal power efficiency term, which is defined as the ratio of the marginal power output term to the sum of power outputs of the two stand-alone plants, is also evaluated to highlight the effect of the capacity of the thermal input from integrated CST collectors, and it is reported in Figure 5.13b.

The marginal increase in electricity production follows two distinct gradients for the considered range of thermal input from the CST collectors. The gradient is much steeper with 0 to 10-15 MW CST input than in the rest of the range, as it appears in Figure 5.13a. As the CST input is further increased beyond 100 MW,
this gradient is eventually expected to fall to zero, as the fixed contribution from
the bIGCC would become proportionally negligible.

![Figure 5.13](image)

**Figure 5.13:** Effects of thermal energy input from CST collectors on the marginal power output (a) and efficiency (b) for different compositions and temperatures of the gasifying agent

**Paper VI** explores the impact of the integration of woody biomass and other intermittent RES in power dispatch systems, where the utilization of excess intermittent power is exploited using power-to-X technologies as low-carbon energy source for different sectors (heating, transportation and power).

In order to emphasize the economic benefits of power-to-X technologies both as a link between power and gas/liquid energy carriers and as an energy storage option (see Chapters 2 and 3), it is necessary to analyze the power supply mix and the energy demand for a geographical region at a reasonably high temporal resolution.
Results and discussion

Figure 5.14: Aggregated power dispatch at the sampled hours for carbon tax in the range of 0-200 €/ton and base-case fossil fuel prices (FFPs). Over-generation, i.e. the available power for power-to-X technologies, is represented by the area above the demand curve.
Results and discussion

The Alpine region is used as a case study in this work because of the opportunity to include different RES, such as biomass, solar, wind and hydropower, and 192 sample hours are used to represent the time history of energy supply and demand during an entire year. The BeWhere tool is used to find the least expensive power generation mix for a given set of values assigned to carbon tax and fossil fuel prices (FFPs). In this work, the base-case scenario ($S\{1,1\}$) assumes a carbon tax of 100 €/ton-CO$_2$ and the market FFPs at the time considered in economic evaluations. In addition, the impact of a high penetration of intermittent RES in the power generation sector is discussed in relation to the reduction of CO$_2$ emissions.

Figure 5.14 presents the evolution of the least expensive power generation mix for a carbon tax in the range from 0 to 200 €/ton-CO$_2$ and for different FFPs. It should be noted that the contribution of base-load coal plants (which is set to cover 10% of the demand in each region) and existing hydropower plants (which cover about 18% of the total demand of the entire region) remains constant in all the cases. Consequently, the variations in carbon tax and FFPs mainly affect the contribution from intermittent RES (in this case solar and wind energy) and, to a much lesser extent, the contribution from new hydropower and biomass plants. For instance, at zero carbon tax and base-case FFPs the power generation is dominated by natural gas with minor contributions from new hydropower (about 9%), biomass (1.3%) and wind energy (0.25%), Figure 5.14-S{0, 1}. When the carbon tax is increased in steps of 50 €/ton-CO$_2$, the share of intermittent RES in the mix (particularly solar) starts to become significant and increases progressively to cover about 17% of the power demand (specifically, the power fed to the grid) for a carbon tax of 200 €/ton-CO$_2$, see Figure 5.14-S{2,1}.

![Figure 5.14: Annual amount of CO$_2$ recycled in power-to-X technologies (a) and annual CO$_2$ emissions avoided due to the substitution of fossil fuels with RES (b).](image-url)
Results and discussion

Another important aspect is that power-to-X technologies provide the opportunity to recycle large volumes of captured CO\textsubscript{2} for biofuel production. Figure 5.15a presents the annual amount of recycled CO\textsubscript{2} by assuming that a mole of carbon dioxide is consumed to produce a mole of methanol or methane. In the considered ranges of carbon tax and FFPs 0.15 to 15 Mtons of captured-CO\textsubscript{2} per year can be recycled. Recycling only affects the storage requirements for the captured CO\textsubscript{2} [119], which could be crucial in countries where geological CO\textsubscript{2} storage is not permitted. It is worth mentioning that the major benefit in terms of decarbonization is still due to the substitution of conventional fossil sources with renewable ones. In fact, Figure 5.15b shows the annual amount of CO\textsubscript{2} emissions avoided due to the high penetration of RES which is in the range between 22 and 103 Mtons/year depending on the carbon tax and FPPs.
Conclusions

CHAPTER 6

Conclusions

This thesis has investigated the benefits of process integration and optimization applied to different scales of energy system analysis in order to increase woody biomass utilization for energy purposes. The boundaries of the considered systems are progressively extended from a single process unit within a plant to a cluster of plants, and then further to a complete regional energy system. Insights from a broader economic perspective are also provided in some of the studies. The main conclusions drawn in the appended papers are summarized in the following text.

6.1 Process integration and optimization in forest industry

The application of process integration and optimization techniques to the design of integrated site configurations in the forest industry has shown that there are significant margins for improving resource and energy utilization from a purely energetic perspective.

In the investigations about the Kraft pulp and paper mill, the potential for reducing the thermal energy demand of the MEEV is mainly due to the optimized operating parameters, while the contribution of the configuration changes that have been introduced is minor. The optimization of the hot utility requirement of the Kraft pulp and paper mill also showed a margin for the potential reduction similar to that of the MEEV (see Figures 5.2 and 5.3).

The application of the HEATSEP method made it possible to dissolve any predefined thermal stream match between utility steam and process streams (with this method the feasibility of the heat transfer interaction among the mill streams is
checked within the black box only). As a result, MP and LP steam no longer have predefined thermal load matches in the real plant. Thus, the integration of the Kraft pulp and paper mill with the streams of the CHP system features an excess of steam generation when the recovery and bark boilers are run at full load. In fact, the combustion of about 50% of the black liquor is enough to cover the thermal requirements of the mill processes. In this situation two feasible actions are recommended according to the possible different objectives, namely biomass resource saving or increased power generation. If resource saving is the chosen objective, the action is to shut down the bark boiler and run the recovery boiler with the amount of black liquor that is strictly sufficient to cover the thermal energy demand of the mill. On the other hand, if increased power production is the chosen objective, a condensing steam turbine must be introduced in the CHP system in order to exploit the excess steam generated by the boilers. In addition, it should be noted that the configuration changes introduced in the steam Rankine cycle of the CHP system have also played a significant role in the potential for increased power production.

The results of the scenario analyses about the industrial site featuring a Kraft pulp and paper mill, its supply chain and a wood-pellet plant have also shown large potential for resources saving or increased power production compared to the stand-alone plants. In this case the option of replacing the boiler based CHP system with a bIGCC is also explored. If power production is considered a priority, the bIGCC configuration should be implemented and all the available biomass resources should be gasified. If otherwise resource saving is prioritized, there are two main alternatives complying with this objective. The first alternative saves the BL and 57% of the BFM while the rest of the BFM is burnt in the bark boiler. The second one utilizes the BL in the bIGCC and saves the BFM.

6.2 Biorefinery concepts based on forest industry

The conceptual process for producing high grade lignin and butanol developed in paper III presents an alternative path for the utilization of the underutilized organic portion of woodchips used in chemical pulping process. Extraction of lignin from BL allows for an increased production of wood pulp without introducing major process changes in existing chemical pulp mills. The configuration changes introduced in the MEEV played an important role in reducing the effects of the integration of the conceptual process on the energy balance of the BL evaporation sub-process. This has been confirmed by assessing the impact on the hot utility requirement of the entire mill, especially when only the lignin separation plant is considered. Besides, assigning one of the MEEV units to the new process reduces the capital investment. As regards the lignin separation, it can be concluded that integrating only the lignin separation and BBLH production processes presents an attractive solution resulting in an increase of the pulp production.
The investigations about sawmill-based biorefinery concepts has concluded that biomass utilization is improved when sawmill byproducts are upgraded on-site into final products (SNG and/or electricity) that already have well-established market infrastructures. However, the uncertainties related to the implementation of such large-scale biomass gasification reactors may overshadow such assessments, as the gasification technology used in the CHP system of the integrated biorefinery site plays an essential role towards the realization of these concepts.

6.3 Economic aspects

The general conclusion about the techno-economic analyses performed in this work is that the integrated system configurations clearly have an increased operational profitability, but when the investment costs of the biomass transformation processes are considered the selling prices of the final products should be still higher than the current market prices.

The economic evaluation in paper II shows that the integrated designs perform better than the standalone cases in terms of the IO index. Moreover, the integrated cases with biGCC achieved higher profitability margins due to the large amount of electricity generated by the gas turbine.

The economy of butanol production through the process proposed in paper III has resulted in a rather high butanol selling prices (at least for the main case evaluated). The ABE fermentation yield from BBLH needs to be improved to the levels already reported for pentose substrates in order to have an economically viable process. According to the sensitivity analysis, an improved fermentation yield together with an increased production capacity of the ABE process could result in an economically feasible biorefinery concept based on Kraft pulp mills. The economy of the integrated process has also shown a strong dependence on the selling price of lignin, as lignin is the major product in terms of quantity.

The integrated SNG and/or electricity production processes in a typical Nordic sawmill have resulted in natural gas and electricity selling prices higher than their current market values, stressing the need for biofuel subsidies. The level of subsidies required to achieve a break-even production of SNG and electricity varies depending on sawmill sizes, with larger mills requiring lower subsidies as an effect of the potential benefits from economy of scale.
Conclusions

6.4 Integration with intermittent RES

The integration of CST collectors with a power generation unit fed with gasified woody biomass makes it possible to increase the overall efficiency significantly, according to the results obtained in paper V. It can be concluded that woody biomass, used as non-variable power source, can play a transformative role for the integration of intermittent RES by reducing grid integration problems. In addition, the configuration reported in paper V reduces the issues about the hybrid design of the system, since the only interaction between the two thermal energy sources is limited to the heat exchanger network.

The energy system with a large share of intermittent RES studied in paper VI shows a significant potential for developing a low-carbon power generation sector when power-to-X technologies are implemented on a large scale. The analysis on the impact of carbon policy and fossil fuel prices confirms that a carbon policy support would further incentivize this transformation. In addition, power-to-X technologies with co-electrolysis can facilitate the decarbonization of the entire energy sector, thanks to the reconversion of the CO$_2$ emitted by a wide range of sources that do not belong to the power generation sector.
### Nomenclature

#### Abbreviations

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tbody>
<tr>
<td>ABE</td>
<td>Acetone-Butanol-Ethanol</td>
</tr>
<tr>
<td>ADt</td>
<td>Air Dried ton</td>
</tr>
<tr>
<td>BB</td>
<td>Biomass fuel mix Boiler/ Bark Boiler</td>
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<tr>
<td>BBLH</td>
<td>Birch-wood Black Liquor Hydrolyzate</td>
</tr>
<tr>
<td>BFM</td>
<td>Biomass Fuel Mix</td>
</tr>
<tr>
<td>bIGCC</td>
<td>biomass Integrated Gasification Combined Cycle</td>
</tr>
<tr>
<td>BL</td>
<td>Black Liquor</td>
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<tr>
<td>CC</td>
<td>Composite Curve</td>
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<tr>
<td>CHP</td>
<td>Combined Heat and Power</td>
</tr>
<tr>
<td>CST</td>
<td>Concentrated Solar Thermal</td>
</tr>
<tr>
<td>CSP</td>
<td>Concentrated Solar Power</td>
</tr>
<tr>
<td>DH</td>
<td>District Heating</td>
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<tr>
<td>DM</td>
<td>Dry Matter</td>
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<tr>
<td>DME</td>
<td>DiMethyl-Ether</td>
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<tr>
<td>EA</td>
<td>Evolutionary Algorithm</td>
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<tr>
<td>GAMS</td>
<td>General Algebraic Modeling System</td>
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<tr>
<td>GCC</td>
<td>Grand Composite Curve</td>
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<tr>
<td>GHG</td>
<td>Greenhouse Gases</td>
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<tr>
<td>HHV</td>
<td>Higher Heating Value</td>
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<tr>
<td>HP</td>
<td>High Pressure</td>
</tr>
<tr>
<td>HRSG</td>
<td>Heat Recovery Steam Generator</td>
</tr>
<tr>
<td>I</td>
<td>Integrated</td>
</tr>
<tr>
<td>IGCC</td>
<td>Integrated Grand Composite Curve</td>
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<tr>
<td>IPCC</td>
<td>International Panel on Climate Change</td>
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Nomenclature

<table>
<thead>
<tr>
<th>Acronym</th>
<th>Description</th>
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<tr>
<td>IO</td>
<td>Investment Opportunity</td>
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<tr>
<td>IRR</td>
<td>Internal Rate of Return</td>
</tr>
<tr>
<td>KPP</td>
<td>Kraft Pulp and Paper</td>
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<tr>
<td>LHV</td>
<td>Lower Heating Value</td>
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<tr>
<td>LP</td>
<td>Low Pressure</td>
</tr>
<tr>
<td>MEEV</td>
<td>Multiple-Effect EVaporator</td>
</tr>
<tr>
<td>MER</td>
<td>Minimum Energy Requirement/ Maximum Energy Recovery</td>
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<tr>
<td>MILP</td>
<td>Mixed Integer Linear Programming</td>
</tr>
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<td>MP</td>
<td>Medium Pressure</td>
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<tr>
<td>PA</td>
<td>Pinch Analysis</td>
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<tr>
<td>PI</td>
<td>Process Integration</td>
</tr>
<tr>
<td>ppmv</td>
<td>part per million, volume based</td>
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<tr>
<td>RB</td>
<td>Recovery Boiler</td>
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<tr>
<td>RES</td>
<td>Renewable Energy Sources</td>
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<tr>
<td>SA</td>
<td>Stand-Alone</td>
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<tr>
<td>SNG</td>
<td>Substitute or Synthetic Natural Gas</td>
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<tr>
<td>SQP</td>
<td>Sequential Quadratic Programming</td>
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<tr>
<td>SOEC</td>
<td>Solid Oxide Electrolysis Cell</td>
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<tr>
<td>T-H</td>
<td>Temperature-Enthalpy diagram</td>
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Chemical symbols

<table>
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<tr>
<th>Chemical Symbol</th>
<th>Description</th>
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<tr>
<td>Ca(OH)$_2$</td>
<td>Calcium hydroxide</td>
</tr>
<tr>
<td>CaCO$_3$</td>
<td>Calcium carbonate</td>
</tr>
<tr>
<td>CaO</td>
<td>Calcium oxide (lime)</td>
</tr>
<tr>
<td>CO</td>
<td>Carbon monoxide</td>
</tr>
<tr>
<td>CO$_2$</td>
<td>Carbon dioxide</td>
</tr>
<tr>
<td>H$_2$</td>
<td>Hydrogen</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>Water</td>
</tr>
<tr>
<td>Na$_2$CO$_3$</td>
<td>Sodium carbonate</td>
</tr>
<tr>
<td>Na$_2$S</td>
<td>Sodium sulphide</td>
</tr>
<tr>
<td>Na$_2$SO$_4$</td>
<td>Sodium sulphate</td>
</tr>
<tr>
<td>NaOH</td>
<td>Sodium hydroxide</td>
</tr>
<tr>
<td>O$_2$</td>
<td>Oxygen</td>
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Acknowledgments

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Great thanks are due to my beloved family and friends, both here and abroad. I would like to express my deepest gratitude to my parents, my brothers, and my sister for their love, constant support and encouragement.

Finally, my utmost praise goes to the Almighty God for this step forward.

Sennai Asmelash Mesfun
June 2016
Luleå, Sweden


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Optimization of process integration in a Kraft pulp and paper mill-Evaporation train and CHP system
Optimization of process integration in a Kraft pulp and paper mill – Evaporation train and CHP system

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ABSTRACT

A great interest has been arising about the production of fuels and advanced chemicals from renewable resources such as wooden biomass in the so-called biorefineries. Pulp and paper mills are often seen as the most obvious fundamental module of such industrial sites, because of the common feedstock and the chemical transformations that already occur in the process. In this paper the model of real Kraft pulp and paper mill is developed and optimized from energetic point of view using process integration techniques, in order to assess the potential for energy saving and to establish a starting point for future research on biorefinery sites. Improvements to the configurations of the multi-effect evaporator and of the steam cycle in the CHP system have been introduced, and three different levels of heat integration boundaries have been considered (multi-effect evaporator, mill sub-processes, and total site). Results indicate a significant potential for the decrease in thermal energy requirement and/or the increase in power production for the same pulp and paper production.

1. Introduction

The interest in improving the efficiency of energy conversion and transfer in chemical plants has been growing due to several reasons including rational utilization of resources, profitability and concern for environmental issues. Thus energy intensive industries, such as the pulp and paper sector (which accounts for about 5% of the total energy use in IEA member countries [1]), have turned their focus into maximizing the internal heat recovery among their sub-processes, increasing the efficiencies of components and processes, recycling of waste streams, and expanding their product chain with new branches that can be easily integrated from the material and energetic point of view. In this respect, the transformation of pulp and paper mills into biorefineries can be seen as a natural evolution of the same industrial site, because both types of plants require similar feedstock and part of the chemical transformation steps can readily be shared so that new chains for value added side products can be incorporated generating additional revenues (see e.g., [2–5]). These modifications result in innovative and more complex process configurations, which require advanced design and optimization techniques for material and energy integration analysis (see e.g., [6–11]) to be really effective.

The aim of this paper is to develop a simulation model of a Kraft pulp and paper mill and to investigate the configuration and the design parameters of the complete industrial site with process integration techniques, in order to provide the basic module of a framework in which new biorefinery concepts can be modeled and analyzed.

The benefits of applying process integration techniques to pulp and paper mills have been reported by several authors in the literature (see e.g., [12–14]; see also [15–19] for recent advances in the literature about pulp and paper mills). In the current work, the HEATSEP method [20] has been applied to the flowsheet of the model mill in order to achieve the maximum flexibility about the properties and the matching of the thermal streams of the
system. In fact, the HEATSEP method suggests to enclose the heat transfer section of the system in a 'black box', in which the thermal streams are completely free to interact (i.e. no heat exchanger network is defined, so that stream mass flow rates and temperatures can be varied with fewer constraints) provided that heat transfer feasibility is verified.

The configuration and the process parameters of the model, which is developed in the MATLAB/Simulink environment, are taken from the experimental data on a real pulp and paper mill in northern Sweden. Previous studies performed in [13,21] are based on the same experimental data and use a model developed in MINDE [22]. Three optimization problems are set to explore the potential for thermal integration inside different system boundaries:

1. the multi-effect evaporator is optimized as a standalone unit to minimize the live steam consumption with respect to its design variables (Section 3);
2. the minimization of hot utility requirement of the pulp and paper mill sub-processes, including the multi-effect evaporator with its design variables (Section 4);
3. integration of the thermal streams resulting in the minimum hot utility requirement for mill sub-processes with a CHP system, the design parameters of which are the decision variables of the optimization problem (Section 5).

All the optimizations have pure thermodynamic objectives. The synthesis of the heat exchanger network that realizes the heat transfers among the optimized thermal streams is beyond the scope of the current work. The results indicate the potential for energy saving and additional power generation that can be achieved in the pulp and paper mill with respect to current operation.

2. The Kraft pulp and paper mill model

The model that is developed to investigate the potential energy and resource savings by means of process integration techniques is a stationary model that reproduces the operational flowsheet of a Kraft pulp and paper mill (see Fig. 1). The model is built in the Simulink environment, which is a graphical extension to MATLAB for modeling and simulating generic systems.

The flowsheet of the mill has been modified to allow the application of the HEATSEP method. In fact, all the potential thermal streams of interest were assigned a thermal cut and they were freed from the heat transfer matches dictated by the real plant flowsheet. For instance, if stream A is known to preheat stream B in the real plant, stream A and stream B will be considered instead as a free hot and a free cold stream, respectively, inside the heat transfer black box. Moreover, their mass flow rate and/or initial and final temperatures may be considered as independent variables of the model (and later as decision variables of an optimization problem).

Some of the key design parameters of the model can be modified to alter:

- the separate productions of pulp and paper (the required input in terms of wood chips is then recalculated according to the specifications on final products);
- the mass flow rates and the temperatures of some of the thermal streams involved in the internal heat transfer;
- the mass flow rates and the properties of the steam that is available for electricity production by expansion in the steam turbine.

The model is based on the configuration and the measured data from a real plant, the mill owned by Billerud Karlsborg AB in Kalix (northern Sweden), the main characteristics of which are summarized in Table 1. The same measured data were already reported in [13,21,23] and are critically rearranged here to establish a reference case reflecting real plant operation. In the base case pulp and paper production rates are 5.04 and 4.13 kg/s dry basis respectively, and, accordingly, the input rate of wood chips is 21.24 kg/s dry basis. The optimized cases refer as well to these operating conditions. In the following subsections some details are given about the modeling of the two sub-processes whose parameters are the most involved in the optimization of the configuration and the design parameters of the mill: the multi-effect evaporator and the CHP system.

2.1. Multi-effect evaporator model

Fig. 2 represents a scheme of the streams entering and exiting in generic ith evaporation effect. The vapor generated in the effect is calculated from Eq. (1). Vapor temperature at the exit of the ith effect is estimated by Eq. (2) as the sum of the steam saturation temperature at effect pressure (\(p_i\)) and a boiling point elevation term (\(\Delta T_{boil}\)). Eqs. (3) and (4) provide an expression for estimation of the \(\Delta T_{boil}\), due to the presence of solid substance as a function of BL dry content (DMi) at effect outlet. Eq. (3) applies only to effect 1 (the one in which live steam is used) because of the much higher change in dry content involved [24], whereas Eq. (4) is used for the other effects and contains an additional correction factor...
depending on steam saturation temperature at effect pressure in Kelvin \( (T_{SAT}) \). The exit temperature of the BL is assumed to be equal to the vapor exit temperature \( (T_{VO}) \). Eq. (5) represents the energy balance inside the control volume built around an effect, in which the external heat flow rate \( q^e \) needed to concentrate the BL appears. The enthalpy of the BL is calculated from its specific heat capacity, which is estimated from Eq.(6).

\[
\begin{align*}
\text{Wood chips input (dry basis)} & = 21.24 \text{ kg/s} \\
\text{Pulp production (dry basis)} & = 5.03 \text{ kg/s} \\
\text{Paper production (dry basis)} & = 4.13 \text{ kg/s} \\
\text{Electricity generation} & = 33 \text{ MW} \\
\text{Recovery boiler capacity} & = 221 \text{ MW} \\
\text{Bark boiler capacity} & = 39 \text{ MW} \\
\text{Steam generation in boilers at 60 bar} & = 60 \text{ MW} \\
\text{Steam medium pressure level 10 bar} & = 10 \text{ MW} \\
\text{Steam low pressure level 4 bar} & = 4 \text{ MW} \\
\end{align*}
\]

\[
\begin{align*}
\rho_{BL,i} - \rho_{BL,i-1} & = \Delta \rho_{BL,i} - \Delta \rho_{BL,i-1} \\
T_{BL,i} - T_{BL,i-1} & = \Delta T_{BL,i} - \Delta T_{BL,i-1} \\
\Delta T_{BL,i} - 1300DM_i^2 - 1300DM_i^4 + 46DM_i^8 + 1.1DM_i + 5 & = 0.0036(T_{VO})_i - 10000 \\
\Delta T_{BL,i} & = \left(6.173DM_i - 7.48DM_i^5 + 32.747DM_i^7\right) (1 + 0.006(T_{VO} - 100)) \\
\rho_{BL,i}h_{BL,i} + \rho_{BL,i-1}h_{BL,i-1} - \rho_{BL,i}h_{BL,i-1} & = \Delta h_{BL,i} + \Delta q^e \\
\end{align*}
\]

\[
C_{PB,i} = 4.246(1 - DM_i) + \left(1.675 - \frac{3.11}{10000}DM_i\right)DM_i + 4.87 \frac{20T_{VO,i}}{1000} \left(1 - DM_i\right)DM_i (6)
\]

2.2. CHP system

The HP steam generated by the two boilers (or by the RB alone) has three usages:

- Part is consumed in the process itself.
- Part supplies thermal energy to some process cold streams.
- Part is used for electricity generation by expansion in a steam turbine.

Most of the steam is then recovered to form a steam cycle, which is the core of the CHP system, but of course this is not possible for the fraction which is consumed in the process, since it has mixed with some steam of the fiber line. A mass flow rate of make-up water (at 1 bar, 5 °C) equal to that of consumed steam must therefore be supplied into the steam cycle.

In the real plant, a steam Rankine cycle is operated with one HP steam generation level at 60 bar and 450 °C. The HP steam is then expanded down to 4 bar in a back-pressure turbine with extractions at three levels: a small amount is extracted at 30 bar (just for soot blowing in the RB), MP steam at 10 bar and LP steam 4 bar. Both MP and LP steam are used both for supplying heat to process streams and for direct mixing with process streams. MP steam is required for soot blowing in the BL, preheating the cooking and bleaching chemicals, steaming wood chips and drying in the paper machine. LP steam is used for preheating and impregnating wood chips, drying in the pulp dryer and paper machine, and supplying the live steam demand of the multi-effect evaporator.

In the model a different steam cycle configuration has been considered, because the potential benefits from increasing cycle efficiency are to be investigated and the design parameters of the steam cycles that are currently used in this kind of industry have been constantly improving in the last years. HP steam is produced at 100 bar and superheated up to 450 °C (still below the highest cycle parameters that can be found in the largest real applications, maximum pressure up to 120 bar, maximum temperature up to 540 °C as in case of traditional steam power plants). A reheat has been introduced at 30 bar to the maximum cycle temperature (450 °C). Moreover, the possibility of having an additional condensation level in a condensing turbine at 0.05 bar (which corresponds
to a temperature which is sufficiently above the typical ambient temperature in northern Europe) has also been explored.

### 3. Optimization of the multi-effect evaporator

The multi-effect evaporator is one of the sub-processes in the Kraft pulp and paper mill that require the largest amounts of heat, which is directly supplied by LP steam. This makes it important to optimize its design parameters in order to reduce the overall thermal energy demand, and therefore the resources that are used to satisfy it.

The purpose of the train of evaporation effects is to evaporate water from the thin BL in order to increase BL calorific value before combustion in the RB. In the real plant, LP steam from the CHP system (live steam) is used to concentrate the BL up to 70–75% dry content. An overview of the base case configuration is shown in Fig. 3. To minimize steam consumption, live steam is used only in effect 1 whereas the vapor generated in each effect is used to supply the thermal energy required for evaporation in the subsequent effect. The evaporation train has seven units and an integrated stripper that uses part of the vapor generated in effect 1. The BL flow is mixed in the base case configuration, i.e. partly counter-current (effects 1, 2 and 3) and partly co-current (effects 4, 5, 6 and 7) to the vapor flow. The pressure inside the effects decreases sequentially from effect 1–7 to the vapor flow. The pressure inside the effects decreases sequentially from effect 1–7 ($p_1 = 1.71$ to $p_7 = 0.21$ bar) and so does the temperature of the vapor according to Eq. (2). The temperature of the BL stream follows the same trend as the vapor when it flows from a higher pressure effect to a lower pressure one (i.e. co-current to the vapor flow from effect 4–7). On the contrary, when the BL flows from a lower pressure effect to a higher pressure one (i.e. counter-current to the vapor flow) it may require intermediate heating depending on the pressure difference between the effects, as the corresponding temperature increase can be too high to be compensated by the thermal energy supplied by the vapor of the preceding effect. This is the case when the BL steam leaves effect 7 at a temperature of 64°C ($\theta_w,7$) and flows to effect 3, which is at 0.94 bar (corresponds to $\theta_{w,3} = 102^\circ$C). The evaporation train therefore includes a HEX between effect 7 and effect 3, as shown in Fig. 3, which must be supplied by an external thermal energy source. A technical issue also contributes to complicate the structure of the evaporation train. Intermediate BL (about 40% dry content) is extracted and mixed with the incoming thin BL (14% dry content). This is done to avoid foaming in the effects by increasing the dry content of the BL up to about 21% before it enters the first evaporation unit [25] (which in the base case is effect 4).

A new sub-process configuration has been introduced and optimized in the current work. The potential thermal streams that are enclosed in the black box representation of the heat transfer section as suggested by the HEATSE method are:

- the streams of vapor from effects 2–7 and from effect 1 (after subtracting the mass flow rate required by the stripper). These are all hot streams, which can make available latent heat first, at the evaporation temperature of the effect they come from, and then sensible heat down to 20°C;
- the streams of BL entering the effects, which can be hot or cold streams according to the temperature difference between the incoming BL and the temperature inside the effect;
- the thermal requirements of the effects, which can be represented as cold streams at constant temperature (the evaporation temperature inside the effect).

Accordingly, the potential thermal streams that are identified in the new configuration can be optimized without assuming any predefined match among them, and, as a consequence, without any predefined configuration for the vapor flow. An overview of the new configuration is shown in Fig. 4. As it appears, this configuration cannot be described as neither co- nor counter-current flow and, more importantly, the need for intermediate HEX is avoided. The mixing of the incoming thin BL with the intermediate one coming from effect 2 (in order to have a 21% dry content at the inlet of effect 7) is maintained as a technical constraint.

It is worth noting that the numbering of the effects may make the new configuration seem quite different from the base case one. Actually, the numbering of the effects is almost irrelevant, because the dry content of the BL must always increase along the BL path, while effect temperature (and pressure) can increase or decrease along the BL path according to how vapor flows are connected to the effects. In the case of the new configuration, the vapor generated in each effect is free to exchange with any other sink and the vapor flow connections are dictated by the results of the optimization.

In this section, the new configuration of the multi-effect evaporator (see Fig. 4) is optimized as a standalone subsystem in order to minimize its live steam consumption and to compare it with that of the base case configuration in the real plant. The main design parameters of the multi-effect evaporator are used as decision variables in the optimization problem, in particular the values of effect pressures and BL dry content at the exit of the effects. The temperatures in the effects, which are also the temperatures at
the boundaries of the heat transfer black box, are dependent variables since they are function of the effect pressure and boiling temperature rise according to Eq. (2). The BL dry content at the exit of effect 1 and the pressure of effect 1 are fixed as target values that are equal to those of the base case configuration (71% and 1.71 bar). The BL temperature and dry content at the entry of the evaporation train are also equal to those in the base case configuration (71% and 26°C). The BL temperature and dry content at the exit of the mixing tank, which is a function of the number of evaporation units.

The details of the optimization problem are as follows:

\[ \min \, f_i(x_i) \]

\( f_i \) returns the minimum live steam demand according to the Problem Table algorithm that is run with the multi-effect evaporator thermal streams, which are in turn calculated as a function of the following decision variables \( x_i \):

- DM, BL dry content at the exit of ith effect (\( i \) is integer variable such that \( i \in \{2; 7\} \)).
- DM \( e \in \{35.5; 50\}, \) DM \( \epsilon \in \{32; 35.5\}, \) DM \( \epsilon \in \{28.5; 32\}, \) DM \( \epsilon \in \{26; 28.5\}, \) DM \( \epsilon \in \{24; 26\}, \) DM \( \epsilon \in \{22; 24\}, \) DM \( \epsilon \in \{20; 22\} \).
- \( p_i \) is the pressure of effect \( i \) (\( i \) is an integer variable such that \( i \in \{2; 7\} \)).
- \( p_2 \in \{1.4; 1.71\}, \) \( p_1 \in \{1; 1.71\}, \) \( p_6 \in \{0.2; 1.4\}, \) \( p_7 \in \{0.2; 1.4\}, \) \( p_5 \in \{0.2; 1.4\} \).

A minimum temperature difference \( \Delta T_{\text{MIN}} = 4 \, ^\circ\text{C} \) is considered for the heat transfer among the thermal streams of the multi-effect evaporator, which are presented in Table 2. The subscript \( j \) stands for an integer variable such that \( j \in \{1 ... n\} \), where \( n \approx 7 \), i.e. the number of evaporation units. \( T_{\text{BL}} \) is the temperature of BL at the exit of the mixing tank, which is a function of \( T_{T_{\text{IF}}} \) and \( T_{T_{\text{IF}} \text{MIN}} \) as well as the mass flow rate of both streams (see Fig. 4).

The algorithm chosen to solve the optimization problem is a single-objective evolutionary algorithm based on the Genetic Diversity Evaluation Method (GeDSE) presented in [26]. The motivation for this choice is given by the mathematical nature of the optimization problem itself. In fact, small changes in the decision variables may cause the pinch point(s) of the Problem Table to move from one temperature level to another and this result in discontinuities in the first derivative of the objective function that traditional optimization algorithms are not able to deal with [27]. Moreover, the objective function is expected to have a multi-modal behavior because the different combinations of potential pinch points at the temperature levels of the different effects may result into several local optima. The number of individuals in the population was set at 200, with a mutation probability of 0.05 (due to the discontinuities expected in the objective function), and the algorithm was run for 10,000 generations. Decision variables are represented as real numbers, and intermediate recombination is applied as crossover operator. The mutation operator changes at random one of the decision variables of an individual. Objective function values of infeasible solutions were assigned a heavy penalty factor. The population of the new generation is selected applying a pure elitist criterion according to a hybrid evaluation of the objective function values and the genetic diversity of the individuals in the union set of current and offspring population (see [26] for more details).

In the optimal solution found by the optimization algorithm the live steam demand of the new configuration of the multi-effect evaporator is 19.76 kg/s. In the base case configuration the live steam demand is 21.48 kg/s, plus the 15.97 MW of the HEX load that correspond to 7.06 kg/s of live steam energy equivalent. This means that the overall reduction obtained in terms of thermal energy consumption is about 21%. The grand composite curves of the

---

**Table 2. thermal streams of the evaporator effects.**

<table>
<thead>
<tr>
<th>Subsystem</th>
<th>Hot streams</th>
<th>Cold streams</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( T_i ) (°C)</td>
<td>( T_j ) (°C)</td>
</tr>
<tr>
<td>Evaporator effects</td>
<td>( T_i )</td>
<td>( T_j )</td>
</tr>
<tr>
<td></td>
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</tbody>
</table>

* Actually, this thermal stream can be hot or cold depending on initial and final temperature.
base case and new configuration with the data about effect pressures, dry contents (DM) and the corresponding effect temperatures are presented in Fig. 5 and Fig. 6, respectively. From the GCC of the base case configuration it is apparent that the thermal requirement of effect 1 (the horizontal line at the higher temperature level, which can be covered by the live steam) is lower than the overall hot utility requirement of the evaporation train. Moreover, the heat loads do not appear to be evenly distributed among the effects, and this contributes to increase the distance of the curve from the ordinate axis, that is to increase the hot and cold utility requirements. On the contrary, in the new configuration the thermal requirement of effect 1 corresponds exactly to the overall hot utility requirement of the evaporation train. The heat loads are almost evenly distributed among the effects, so that a pinch point can be found at the temperature level of each effect (as it would happen in the ideal condition, so this is a demonstration of the optimality of the solution found). As regards to vapor flow connections, only some hints can be drawn from the trend of effect pressures and temperatures. In fact, effect 6 is the one having the lowest temperature, so it is likely that its thermal requirement will be mainly covered by the vapor coming from effect 7, the one at the immediately higher temperature level. In turn, effect 7 will be mainly heated by effect 5, and the rest of the temperature trend is descending from effect 1 to effect 5 as in the base case configuration.

4. Hot utility minimization of plant processes

The results obtained in the previous section can be further improved if the multi-effect evaporator is considered not just a standalone sub-process, but as one of the energy demanding sub-processes of the Kraft pulp and paper mill. This means that the boundaries of the heat integration analysis are enlarged to include all the processes of the Kraft pulp and paper mill. The thermal streams from the selected sub-processes are then extracted from the simulation results and used as input to Pinch Analysis techniques. It should be noted that CHP system thermal streams (i.e. those that are excluded in the model for total plant hot utility minimization) account only for the portion of the steam that circulates in the steam cycle, whereas thermal streams required for the generation of the steam that is directly consumed in the process (and must be replaced with make-up water) are taken into account as pulp and paper mill thermal streams.

The hot utility requirement of the process in the base case configuration has been calculated first for comparison. The list of the thermal streams that are involved in the calculation can be obtained by merging the streams listed in Tables 1 and 2. Most of thermal streams have fixed temperatures at the boundary of the heat transfer section black box and their mass flow rates depend only on the pulp and paper production rates, which are set at the measured ones. On the contrary, the thermal streams associated with evaporator effects (Table 2) and steam generation (Table 3) have both boundary temperatures and heat loads that depend on the choice of the design parameters in the multi-effect evaporator and in the CHP system. In base case calculation they are evaluated at the base case conditions derived from the experimental data. The thermal streams of the evaporator effects are evaluated at the effect pressures and dry contents of the real plant. The thermal streams for steam generation are evaluated according to the conditions of the base case CHP system configuration, which is superheated steam at 60 bar and 450 °C without reheating. All the streams from Tables 1 and 2 are then integrated with Pinch Analysis techniques and the ΔT_{min}/2 indicated for each stream is applied in the Problem Table algorithm. The values of ΔT_{min}/2 are selected according to the nature of the stream, so that a reasonable heat exchanger area can be expected for any of the possible matches among the streams.

The hot and cold composite curves and the grand composite curve of the base case are presented in Fig. 7. The main contributions to the shape of the grand composite curve can be identified from the figure according to the temperature intervals. In the upper range (from 140 to 450 °C) the shape of the curve is determined by the streams related to steam generation, the horizontal segment at about 280 °C being the heat of evaporation at 60 bar required by the generation of steam which is directly consumed in the process. In the middle range (from 60 to 140 °C) the curve is dominated by the heat loads of the evaporator effects, which result in large

![Fig. 5. Grand composite curve of the base case multi-effect evaporator configuration.](image-url)
horizontal traits at different temperature levels. Finally, the shape in the lower range (below 60°C) is determined by the cooling of plant condensates and effluents and the preheating of water in the HW and WW subsystem. The overall hot utility requirement for the base case scenario is 78.9 MW. It appears that the largest share of it is due to the thermal requirement of the multi-effect evaporator (roughly 55 MW, which is about the same of the evaporation train alone in the base case, see Fig. 5). This part of the mill thermal demand can be reasonably satisfied with the condensation of LP steam, which occurs at a temperature slightly higher than the temperature of the cold stream representing the thermal requirement of the first effect. The rest of the hot utility requirement (nearly 25 MW) is due to the generation of the steam which is directly consumed in the process. Because of the temperature range, this part of the mill thermal demand can be satisfied only by the high temperature heat released during combustion in the boilers.

The minimization of the hot utility requirement is performed using the modified configurations of the multi-effect evaporator and the CHP system, and by applying the black box representation of the heat transfer section as suggested by the HENESIS method. The details of the optimization problem are given in the following:

$$\min f(x)$$

where $f_i$ returns the minimum hot utility demand according to the Problem Table algorithm that is run with the thermal streams listed in Tables 1 and 2, which are in turn calculated as a function of the following decision variables $x$:

- $DM_i$: dry content at the exit of $i$th effect ($i$ is integer variable such that $i \in \{2, 7\}$).
- $DM_1 \in [35.5: 50]$,
- $DM_2 \in [32, 35.5]$,
- $DM_3 \in [28.5, 32]$,
- $DM_4 \in [26, 28.5]$,
- $DM_5 \in [24, 26]$,
- $DM_6 \in [22, 24]$,
- $p_i$: is the pressure of effect $i$ ($i$ is an integer variable such that $i \in \{2, 7\}$).
- $p_1 \in [1.4, 1.71]$,
- $p_2 \in [1.71]$,
- $p_3 \in [0.2, 1.71]$,
- $p_4 \in [0.2, 1.4]$,
- $DM_6$: dry content at the exit of effect 6.

The algorithm chosen to solve the optimization problem is again the single-objective evolutionary algorithm used in the previous section, for the same motivations mentioned about the behavior of the objective function. The number of individuals in the population was set at 200, with a mutation probability of 0.05 (due to the discontinuities expected in the objective function), and the algorithm was run for 10,000 generations.

The grand composite curve of the optimal design solution found is shown in Fig. 8. The minimum hot utility requirement is lowered to 66.88 MW, with a reduction of 15.3% from that of the base case. From the comparison of the GCCs in Figs. 6 and 7 it is apparent that the main contribution to the reduction of the hot utility requirement in the optimized case comes from the new design parameters of the multi-effect evaporator, while the contribution from the upper temperature range remains substantially unaltered. To be more precise, the rise of the pressure at which steam is generated from 60 to 100 bar and the introduction of the reheating at 30 bar actually increases the hot utility requirement due to the thermal streams in the upper temperature range. In fact, the reduction of the hot utility requirement would have been equal to 18% if steam generation had taken place in the same conditions as in the base case. The GCC in Fig. 8 shows that the solution is very close to the ideal condition in which a pinch point is present at each effect temperature level, but in this optimal solution the design parameters of the effects, in particular the pressure, have quite different values from the solution found in the previous section. The order of effect temperatures (from high to low) is now 1-2-3-7-6-5-4, and this also reflects the most likely sequence of vapor flow connections (from 1 to 2, from 2 to 3, from 3 to 7 etc.). Another aspect that should be noted about the optimal GCC (but it is also present in the base case GCC) is the considerable need of a cold utility (about 43 MW, with a reduction of about 25% compared to the base case). The hot streams that could be coupled to that have unfortunately a quite low temperature (around 50-60°C), so that the energy associated with the rejected heat from mill sub-processes can hardly be recovered for other purposes. The final remark on the results is about the optimal temperature of the thin BL, which is found at the upper limit of the range allowed (120°C, against 87°C in the base case). This result on one hand is quite surprising, because it may seem that the thin BL exiting the cooking digestion step at 169°C cannot be fully exploited as a hot stream down to a lower temperature, but on the other hand the mixing of the thin BL in the multi-effect evaporator with some thicker BL coming from effect 2 is of about 120°C, and this means that the mixing is nearly isothermal in the optimal solution (non-isothermal mixing causes exergy destruction and reduces the vertical distance between the hot and cold composite curves, hence it may...
result in larger hot and cold utility requirements). After the mixing the residual higher thermal energy associated with the thin BL can be still exploited as a hot stream of the multi-effect evaporator.

5. Total site integration

The primary source of the thermal energy supplied to the Kraft pulp and paper mill is the combustion in the boilers where BL and bark are burnt. The thermal energy that is released during combustion is stored in the product gases, which in turn is transferred by radiation and convection (see the thermal streams related to RB and BB in Table 4). This transfer can occur directly to the process streams (as in the generation of the steam which is consumed by the sub-processes even after the portion of thermal energy converted to the electrical energy from the combustion gases (indirectly or partially directly transferred to the process) or it can occur indirectly through the steam cycle of the CHP system. In this respect, it is worth noting that the minimum consumption of fuels (BL and bark) is prioritized, then the result will be a design solution in which the hot utility requirement is satisfied by an equal amount of thermal energy from the combustion gases (indirectly or partially directly transferred to the process streams involved in the hot utility minimization being kept at the optimal conditions determined in Section 5.

However, it is not straightforward to decide how this total site heat integration should be performed, because the objectives to be pursued can differ and lead to different results. If, for instance, the minimum consumption of fuels (BL and bark) is prioritized, then the result will be a design solution in which the hot utility requirement is satisfied by an equal amount of thermal energy from the combustion gases (indirectly or partially directly transferred to the process streams). If, on the other hand, maximum power production were selected as an objective, the optimization procedure would search for a solution in which the objective of minimum utility consumption was satisfied by an equal amount of thermal energy from the combustion gases (indirectly or partially directly transferred to the process streams involved in the hot utility minimization being kept at the optimal conditions determined in Section 5.

The main purpose of the steam cycle is to absorb thermal energy from the combustion gases of the boilers, to supply the thermal energy needed by the pulp and paper mill sub-processes at the most suitable temperature levels (those corresponding to MP and LP condensation levels) and to convert the difference between the absorbed and released thermal energy into electricity by expanding the steam in a turbine. The sub-processes of the pulp and paper mill have therefore to be integrated with the boilers and the CHP system in order to get their thermal requirement satisfied. The boundaries of the heat integration analysis is then further enlarged to include all the thermal streams that are listed in Tables 1–3 in the black box representation suggested by the HEATSEP method, the streams involved in the hot utility minimization being kept at the optimal conditions determined in Section 5.

5. Total site integration

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However, it is not straightforward to decide how this total site heat integration should be performed, because the objectives to be pursued can differ and lead to different results. If, for instance, the minimum consumption of fuels (BL and bark) is prioritized, then the result will be a design solution in which the hot utility requirement is satisfied by an equal amount of thermal energy from the combustion gases (indirectly or partially directly transferred to the process streams) with no electricity production from the CHP system (and the steam cycle would have no reason to be implemented). If, on the other hand, maximum power production were selected as an objective, the optimization procedure would search for a solution in which the maximum possible amount of fuel (the amount of available BL is upper bounded, but that of bark is not) is burnt to increase the thermal energy input to the steam cycle of the CHP system. In this respect, it is worth noting that the amounts of BL and bark that are burnt in the real plant according to the experimental data are far more than sufficient to supply the minimum hot utility requirement for the pulp and paper mill sub-processes even after the portion of thermal energy converted to electricity is considered. Please also note that an economic

Table 3

<table>
<thead>
<tr>
<th>Subsystem</th>
<th>Hot streams</th>
<th>Cold streams</th>
</tr>
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<tbody>
<tr>
<td></td>
<td>Ts (°C)</td>
<td>Tc (°C)</td>
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<td>Digester</td>
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<td>169</td>
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<tr>
<td></td>
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<tr>
<td></td>
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<td></td>
<td>81</td>
<td>2400</td>
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<td></td>
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<td>300</td>
</tr>
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<td>Bleaching plant</td>
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<td></td>
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</tr>
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<td></td>
<td>84</td>
<td>7500</td>
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<td>51</td>
<td>600</td>
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<td>Paper machine</td>
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</tr>
<tr>
<td></td>
<td>84</td>
<td>200</td>
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<td></td>
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<td>Steam generation</td>
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<tr>
<td></td>
<td>4</td>
<td>Tsat</td>
</tr>
<tr>
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</tr>
<tr>
<td></td>
<td>4</td>
<td>Tsat</td>
</tr>
</tbody>
</table>

a According to the configuration used in current work.
optimization of the operational profit based on a given set of prices for BL, bark, electricity and excess heat would anyway have one of the two above mentioned outcomes.

As a consequence, the optimization of the integration among pulp and paper mill sub-processes, boilers and CHP system must include one constraint to avoid uninteresting results. Since the choice of the constraint is completely arbitrary, five different scenarios are considered here according the chosen constraint and steam cycle configuration:

1. All BL is burnt in the RB and the amount of bark taken from the experimental data is burnt in the BB, steam expansion ends in a condensing turbine.
2. All BL is burnt in the RB but no bark is used, steam expansion ends in a condensing turbine.
3. All BL is burnt in the RB and the amount of bark taken from the experimental data is burnt in the BB, steam expansion ends at LP condensation pressure level.
4. All BL is burnt in the RB but no bark is used, steam expansion ends at LP condensation pressure level.
5. No bark is used, the amount of BL burnt in the RB is limited to that required to satisfy the pulp and paper mill minimum hot utility requirement without releasing any thermal energy from the condensing LP steam to the ambient, steam expansion ends at LP condensation pressure level (of course in this case the fraction of BL that is not burnt in the RB will undergo other processes than combustion and the cooking chemicals will be recovered in some other ways).

In all the five scenarios the chosen objective function to be maximized is the net power production from the steam cycle of the CHP system.

The optimization problem is set in slightly different ways for the different scenarios. For scenarios 1 and 2, it is set as follows:

\[ \max \, f_3(x) \]

\[ f_3 \] returns the maximum net power production. It is subject to the constraints set by heat transfer feasibility inside the black box enclosing the heat transfer section and those about the consumed amounts of BL and bark. Decision variables \( x_i \) are:

- Unit P(bar) Fm DM Top (ºC)
- Effect 1 1.710 0.710 129.5
- Effect 2 1.710 0.406 121.9
- Effect 3 1.000 0.355 105.8
- Effect 4 0.212 0.320 63.5
- Effect 5 0.278 0.280 74.2
- Effect 6 0.345 0.247 69.3
- Effect 7 0.790 0.224 98.6

BL temperature (ºC):

\[ \frac{\text{BL temperature (ºC)}}{T_{\text{min}} = 120} \]
– \( m_{10 \text{cylce}} \): the mass flow rate (kg/s) of the condensing MP steam at 10 bar;
– \( m_{04 \text{cylce}} \): the mass flow rate (kg/s) of the condensing LP steam at 4 bar;
– \( m_{00 \text{cylce}} \): the mass flow rate (kg/s) of the condensing steam at 0.05 bar.

For scenarios 3 and 4 the optimization problem set above is the same except for the steam mass flow rate to the condensing turbine, which is set to zero and excluded from the decision variables set (this is because the condensing turbine is not considered in these cases).

For scenario 5 a new decision variable is added to the mass flow rates of the condensing MP and LP steam to express the amount of the BL that is actually burnt in the RB. The new decision variable is a real value in the range between 0 and 1 and multiplies the thick BL mass flow rate coming from the evaporation train.

A traditional optimization algorithm has been chosen to solve the optimization problems in all scenarios, since no discontinuities are expected due to the mathematical nature of the problems. The temperature levels of all the thermal streams are fixed, so the heat transfer feasibility constraint can be expressed by a set of inequalities (one at each temperature level, see [27]) that are linear in the mass flow rates of the thermal streams. The objective function is calculated as an algebraic sum of terms in which specific enthalpy differences, derived from fixed steam conditions, multiply the steam mass flow rates, so it is linear in the decision variables as well. Accordingly, a linear constrained optimization algorithm can be used to find the solutions of the five scenarios.

### Table 4

<table>
<thead>
<tr>
<th>Subsystem</th>
<th>Hot streams</th>
<th>Cold streams</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( T_1 (°C) )</td>
<td>( T_2 (°C) )</td>
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<tr>
<td>CHP system</td>
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Table 4 summary of results for total site integration scenarios.

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<th>Scenario</th>
<th>Steam (MW)</th>
<th>Electric (MW)</th>
</tr>
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<tr>
<td></td>
<td>MP</td>
<td>LP (MW)</td>
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<td>8.3</td>
<td>58.1</td>
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<tr>
<td>2</td>
<td>7.8</td>
<td>55.3</td>
</tr>
<tr>
<td>3</td>
<td>5.2</td>
<td>134.3/80</td>
</tr>
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<td>4</td>
<td>5.1</td>
<td>112.7/62</td>
</tr>
<tr>
<td>5</td>
<td>3.5</td>
<td>46.1</td>
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### Table 5

<table>
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<th>Scenario</th>
<th>Steam (MW)</th>
<th>Electric (MW)</th>
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<tr>
<td></td>
<td>MP (MW)</td>
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<td>2</td>
<td>7.8</td>
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<td>3</td>
<td>5.2</td>
<td>134.3/80</td>
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<tr>
<td>4</td>
<td>5.1</td>
<td>112.7/62</td>
</tr>
<tr>
<td>5</td>
<td>3.5</td>
<td>46.1</td>
</tr>
</tbody>
</table>

### Fig. 9

Scenario 1 integrated grand composite curve (BL: 25.43 kg/s; Bark: 4.28 kg/s).
Fig. 10. Scenario 2 CHP results:
Rankine cycle net power: 51.6 MW
Steam flow:
- $m_{\text{steam}} = 23.931 kg/s$
- $m_{\text{water}} = 22.635 kg/s$

Fig. 11. Scenario 3 CHP results:
Rankine cycle net power: 45.5 MW
Steam flow:
- $m_{\text{steam}} = 2.236 kg/s$
- $m_{\text{water}} = 58.890 kg/s$
- $m_{\text{water}} = 0.000 kg/s$
Excess heat production: 80 MW ($\approx$8bar, 143.8°C)

Fig. 12. Scenario 4 CHP results:
Rankine cycle net power: 39.2 MW
Steam flow:
- $m_{\text{steam}} = 2.283 kg/s$
- $m_{\text{water}} = 48.754 kg/s$
- $m_{\text{water}} = 0.000 kg/s$
Excess heat production: 62 MW ($\approx$4bar, 143.6°C)
The optimal integrated total site solutions are summarized in Table 5 and the corresponding integrated grand composite curves are presented in Figs. 9–13. These curves are used to show in different colors the contribution of separate subsets of thermal streams to the total site heat cascade. The blue curve represents the streams of the CHP system steam cycle, whereas the red curve represents all the other thermal streams (including those associated with the generations of the steam consumed in mill sub-processes). This representation emphasizes how much the streams of the CHP system steam cycle are able to exploit the heat pocket formed by the streams from the boilers and mill sub-processes. In fact, the aim of the CHP system is to avoid the exergy destruction that would be caused by the direct heat transfer from the high temperature hot combustion gases to the low temperature cold process streams, and to convert this exergy gap into electricity.

The shape of the red curve in Figs. 9–13 derives from the composition of the optimized GCC of mill sub-processes (previously shown in Fig. 8) with the radiative and convective heat transferred by the combustion gases of the two boilers. The horizontal segment at 1000 °C represents the radiative heat in the BB and is followed by another oblique segment representing the heat from combustion gases down to 206 °C. In Scenarios 1 and 3 (Figs. 9 and 11) the BB streams are also present: the horizontal segment at 800 °C represents the radiative heat and is followed by another oblique segment representing the heat from combustion gases down to 152 °C. The actual composition with the streams of mill sub-processes occurs at temperatures below 450 °C, and the most evident sign of it is the inversion of the slope of the oblique segments adjacent to the horizontal one at 311 °C (the evaporation of the steam consumed by mill sub-processes).

The shape of the blue curves in those figures is the result of the composition of the latent and sensible heat absorbed and released in the steam cycle. In descending temperature order, the horizontal segments represent: steam evaporation at 100 bar, the condensation of MP steam, the condensation of LP steam, the condensation of HP steam, and the most evident sign of it is the inversion of the slope of the oblique segments adjacent to the horizontal one at 100 °C (the condensation of steam at 1 bar).

Maximum power generation is obtained in Scenario 1 (62.1 MW, +88% than in the real plant), with the same fuel consumption of the real plant and the addition of a condensing turbine. If the BB is shut down (Scenario 2), power production is reduced by 17%, but the thermal energy released by the combustion of BL only is sufficient to supply heat to the process and to a CHP system that generates an amount of power that is still 50% higher than the one experimentally measured in the real plant. In Scenarios 3 and 4 steam expansion ends at LP condensation level, so power production is reduced and a large surplus of heat is available from steam condensation at 4 bar (143.6 °C). When all the available fuel is burnt (Scenario 3), power generation is 45.5 MW, +38% than in the real plant, with 80 MW of surplus heat. If the BB is shut down (Scenario 4), power production is decreased by 14% and the heat surplus by 23%.

Scenario 5 represents a very particular and interesting situation. From the results of the previous scenarios it is apparent that combustion of BL alone produces more heat than the thermal demand of mill sub-processes, but in Scenario 5 the CHP system is requested to supply exactly that demand. The results from the optimization show that only half of the BL is enough to supply the thermal requirement of the mill and to produce 19.2 MW of net power. Provided that the spent pulping chemicals must be recovered and sent back to the chemical loop to form the green liquor, the remaining part of the BL can be used for other purposes. For instance, it can be gasified to produce syngas, which can be used to supply a gas turbine engine (e.g., in a combined cycle configuration) for additional power production or chemically processed to obtain liquid fuels for transportation (see e.g. [22,28,29]). Another process which the BL can undergo is fractionation, which is used to separate the lignin and hemicellulose content from the solution. One example of fractionation is acid precipitation [30], in which BL lignin and hemicellulose are precipitated and subsequently filtered from a supernate which is a solution rich of Na+ and CO32- ions. The precipitate may then be processed to produce green chemicals and fuels via biochemical and/or thermochemical transformations.

6. Conclusions

A detailed model of a Kraft pulp and paper mill has been developed in the MATLAB/Simulink environment to evaluate the potential for resource and energy saving by using process integration techniques. Real plant flowsheet and stream data have been critically revised for the application of the HEATSEP method, in order to obtain the maximum possible heat integration without any predefined match among the hot and cold thermal streams, and some...
improvements have been introduced to the configuration of the evaporation train and the steam cycle of CHP system. Three levels of boundaries for the optimization of heat integration have been considered: the multi-effect evaporator alone, mill sub-processes considered as a whole and total site. The optimized solutions show that a great deal of the potential reduction in thermal energy requirement derives from a different choice of the design parameters of the multi-effect evaporator, while the changes introduced in its configuration play a much less important role. A more even distribution of the thermal loads among the effects is critical in lowering the hot utility requirement of both the evaporation train alone and the mill sub-processes considered as a whole. The use of the HEATSEP method has also allowed redefining of the steam balance between the CHP system and mill sub-processes in the real plant, which is determined by the predefined matches between condensing MP or LP steam and process streams. The reduction of the thermal energy requirement of the mill sub-processes, on one hand, and the improvements introduced in the steam cycle of the CHP system (in particular, the option of having a condensing turbine to expand the steam down to vacuum pressure), on the other hand, make it possible to reformulate a steam balance that is now completely internal to the black box enclosing the heat transfer section of the system. This leads to a substantial increase in power production, since the thermal energy input due to the combustion of biomass fuel in the boilers is much greater than mill requirement, even if the BR is shut down. Another interesting option is to burn as much BI, as requested by mill sub-processes (about only 50%, while the CHP system still produces a considerable amount of power) and use the rest for some other thermochemical process. Future work will see this model and these results being used as the fundamental process streams. The reduction of the thermal energy requirement considered as a whole and total site.

Acknowledgements

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References


Paper II

Integrating the processes of a Kraft pulp and paper mill and its supply chain
Integrating the processes of a Kraft pulp and paper mill and its supply chain
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ABSTRACT
This paper investigates the possibility of combining different forest industries (a pulp and paper mill, its supply chain, and a wood-pellet plant) into an integrated industrial site in which they share a common heat and power utility. Advanced process integration and optimization techniques are used to study the site from both material and energy viewpoints. An existing pulp and paper mill is used as the site core plant and its pulp and paper production rates are kept fixed as they are in reality, while the other material flow links among the plants are based on the current industrial situation in Sweden. Different scenarios are evaluated in order to reflect the two main objectives that can be pursued (increased electricity production or biomass resource saving) and the two technologies that can be considered for the shared CHP system (boilers and product gas fired gas turbines). The corresponding non-integrated (standalone) configurations are compared to these scenarios to quantify the potential benefits of the integration. Investment opportunity is also calculated for the considered scenarios as an indicator of the economic convenience.

1. Introduction
The search for securing reliable, affordable and, more importantly, clean energy supply requires innovative and efficient utilization of sustainable resources. Woody biomass, one of the most important and relatively abundant renewable resources, is expected to play a significant role in easing the dependence on fossil-based resources for energy and motor fuels. The entire productive chain, from woody biomass to end products (such as lumber, pulp, pellets, heat, electricity and biofuels), encompasses different process industries. A holistic evaluation of the different processes involved in the chain is crucial in order to investigate the potential for resources saving or higher profitability.

Increased resources saving, i.e. getting a reliable share of resources without the need to compete with existing industries, would allow margins for innovative conversion technologies such as the production of biofuels through thermochemical (see e.g. [1-11]) and/or biochemical (see e.g. [12-16]) pathways. The potential for this can be investigated by implementing advanced optimization tools (see e.g. [17-19]) aimed at fine tuning the subprocesses of existing forest industries (from a material and energy perspective) and by using efficient process integration techniques to combine them with the subprocesses of other related industrial sectors, so that they all can share the same heat and power utility.

Chemical pulp mills are existing examples of lignocellulosic biomass based biorefineries in which woody biomass is converted into pulp (a cellulose derivative) and other intermediate biomass products. Expanding the boundaries of chemical pulp mill to include other forest industries (such as sawmills and wood-pellet plants) could result in a better profitability and therefore boost the concept of biorefinery further.

Industrial symbiosis in Swedish forest industry is reported in [20] with the objective of reducing the environmental impact and increasing the profit. The issue is further discussed in [21] with the aim of assessing the total cost benefit of integrating forest industries (a sawmill, a wood-pellet plant and a pulp and paper mill). However, both works do not consider the scenario of a real integrated industrial site and the benefits that may arise from an optimal integration among the plants with a shared CHP system. The thermodynamic efficiency gain and the economic benefits of integrating a wood-pellet plant with a pulp mill is investigated in [22]. The focus of that paper is to exploit the excess heat produced in the RB based CHP system of the pulp mill for the drying process in the wood-pellet plant, but no alternative technologies for the pulp mill CHP system were considered. BL gasification is often reported in the literature as an alternative to the RB in the CHP system of pulp mills (see e.g. [23,24]), and the results of these
studies showed potential for reduced resources consumption and increased overall energy efficiency. Several authors have also worked from different points of view on the integration of biofuel synthesis process in pulp mills through the gasification of BL (see e.g. [4,7,9,23]). The economic and energetic perspective of integrating solid biomass gasification process in a pulp and paper mill for the synthesis of biofuels and/or for power generation via a combined cycle has been discussed in [25].

The main objective of this paper is to investigate the outcome of the optimal integration of a chemical pulp mill, its supply chain and a wood-pellet plant with a common CHP system supplied by boilers or product gas fuelled gas turbines. The analysis is based on mathematical models of the industrial processes considered.

The paper is structured as follows. Section 2 presents the description of the considered industrial processes including a list of the thermal and power loads associated with them. In Section 3 the methodology used to perform the integration and the optimization is described and the setups of the optimization problems are presented in detail. The integration scenarios are also described in Section 3 with an explanation of the objective/s in each case. Section 4 presents the main results followed by detailed discussion about the most significant outcomes.

2. Description of the process integration model

The process integration model is developed in MATLAB Simulink environment. The data used in the model are either experimentally measured in existing plants as well as in pilot and demo scale research facilities, or represent the typical Nordic countries forest industries. Each process has been considered in order to identify the potential thermal streams involved and the power required for its operation. In the following subsections brief descriptions of the plants and their processes are presented.

2.1. Pulp and paper mill

The pulp and paper mill model is based on data measured in an existing plant which is representative of the typical technologies employed in integrated chemical pulp and paper mills in Nordic countries. A detailed description of the mill model has been reported in [26]. Only the design parameters which are important for the integration among the plants are highlighted in this section. Pulp and paper production are 5.03 and 4.13 kg/s (dry basis), respectively, and these result in a woodchips consumption of about 21.24 kg/s (dry basis). In fact, a large portion of the woodchips fed to the digester of the pulping process (i.e. the lignin and part of the hemicellulose) becomes the organic content of the BL which has a mass flow rate of about 18.05 kg/s (dry basis). In addition the mill consumes about 50 MW of electricity for internal operations. The thermal demand and part of the power demand of the mill are covered by an internal CHP system based on a BL and a BFM boiler. The BFM is made of 85% bark, 9% sawdust and 6% woodchips in dry wt. The mixing is done because the combustion and/or gasification of bark alone is problematic due its high moisture content, low calorific value and high ash content [27,28]. The composition of the BFM has been maintained as a constraint during the allocation of the resources to integrated industrial sites.

The details of the CHP system model along with its steam cycle configuration have been reported in [26], and this model is simply adopted in this work without any modifications (the thermal streams from the boilers are summarized in Table 1 for quick reference). In addition all the thermal streams involved in the pulp and paper mill subsystems are presented in Table 2.
is carried out by heating outdoor air from a temperature of 2\(^{18}\%\). Therefore, the process includes a lumber drying step which is performed in dedicated rooms called drying kilns. The timber, which is initially at 55–60% moisture content, is dried in lumber with a final moisture content of about 18%. Therefore, the process includes a lumber drying step which is carried out by heating outdoor air from a temperature of 2 \(^{\circ}\)C to 75 \(^{\circ}\)C and circulating it through packages of lumber placed in dedicated rooms called drying kilns. The drying process is assumed to take place at a constant temperature of 75 \(^{\circ}\)C, and is driven by the moisture content difference between the lumber surface and the drying air. The thermal load is satisfied by burning BFM in a dedicated furnace and about 2.3 kg dry of BFM are burnt per dry kg of produced lumber. All the thermal streams in the reference sawmill model are presented in Table 2. The electricity consumption is about 690 kJ per dry kg of produced lumber.

The model of wood chopping does not include any thermal streams, and the electricity consumption is about 81 kJ per dry kg of wood logs.

2.3. Wood-pellet plant

Wood-pellet plant produces densified wood fuels in the form of pellets from the biomass wastes of forest industries as well as from the chopping and grinding of dedicated woody biomass. The raw material is dried and fine grinded prior to the pelletization process. Sawdust, one of sawmill by-products, is a convenient raw material for pellet production as it has already undergone part of the drying process in the sawmill exit is therefore saturated at the operating pressure (the saturation temperature is 181 \(^{\circ}\)C in this case), which means that its latent heat can be used to generate M/P/LP steam in a counter-current condenser [31]. The cooking chemicals are recovered as smelt, mainly composed of Na\(_2\)CO\(_3\) and to a lesser extent Na\(_2\)S, that falls into the bottom of the reactor and gets dissolved in the weak liquor bath to form green liquor [32]. The green liquor exits from the bottom of the reactor as saturated liquid at the operating pressure and is cooled on its way to the chemical recovery process of the pulp and paper mill by exchanging heat with the weak liquor entering the gasifier to replace it.

The thermal energy requirement of the gasification process is supplied by a partial oxidation of the BL which is regulated by the amount of air supplied to the reactor. In this case the equivalence ratio is controlled by the gasification temperature, which in turn is calculated from the overall energy balance of the reactor assuming a heat loss of 2\% on the LHV of the BL input. According to the operating conditions and the chemical equilibrium calculations, the product gas after the cleaning process is predicted to have a molar composition of 55.4 vol.% N\(_2\), 17% H\(_2\), 14% CO, 13% CO\(_2\) and 0.6% as by-products. The chopping of wood logs produces woodchips (64 wt.% of the logs input) and bark (the remaining 36 wt.% of the logs input). The industrial processes that are included in the supply chain to a pulp and paper mill are therefore sawmills and wood logs choppers. A schematic representation of the material flow among the integrated industries is shown in Fig. 1. The model of a typical sawmill in Nordic countries is taken from [29]. The timber, which is initially at 55–60% moisture content, is processed into lumber with a final moisture content of about 18%. Therefore, the process includes a lumber drying step which is carried out by heating outdoor air from a temperature of 2 \(^{\circ}\)C to 75 \(^{\circ}\)C and circulating it through packages of lumber placed in dedicated rooms called drying kilns. The drying process is assumed to take place at a constant temperature of 75 \(^{\circ}\)C, and is driven by the moisture content difference between the lumber surface and the drying air. The thermal load is satisfied by burning BFM in a dedicated furnace and about 2.3 kg dry of BFM are burnt per dry kg of produced lumber. All the thermal streams in the reference sawmill model are presented in Table 2. The electricity consumption is about 690 kJ per dry kg of produced lumber.

The model of wood chopping does not include any thermal stream, and the electricity consumption is about 81 kJ per dry kg of wood logs.

2.4. Biomass gasification and product gas (PG) utilization

Biomass gasification is considered in this work as an alternative to the combustion of BL and BFM in boilers for CHP system. In this case the product gas is used to feed a gas turbine for power production, and the exhaust gases are further used in HRSGs to generate steam that drive the steam cycle of the CHP system.

The modelling of complex gasification reaction kinetics is beyond the scope of this work. However, it is worth to illustrate briefly the gasification techniques considered in order to substantiate the choice of the operating conditions in the gasifiers and the potential thermal streams identified.

2.4.1. BL gasification

BL is a by-product of the pulping process and contains about 50 wt.% organics and 50 wt.% inorganics (mainly the cooking chemicals) on dry basis. The BL, which comes out of the digester at about 14.4% dry matter (DM), is concentrated in a multiple-effect evaporator to about 71% DM prior to its gasification (or combustion). The technology considered for the BL gasifier is similar to that of the pressurized oxygen-blown entrained flow reactor developed by Chemrec AB [30]. In this study, however, the reactor model is air-blown [3] and operated at a temperature of 975 \(^{\circ}\)C and a pressure of 19 bar. The benefits of operating the gasifier at this pressure is twofold: first, the product gas is not to be pressurized before entering the gas turbine combustor and, second, any excess air from the compressor of the gas turbine can be used as oxidizing agent in the gasifier. According to the design and configuration of the Chemrec reactor, the product gas after the reaction zone gets quenched by a spray of water and the weak liquor bath at the bottom of the reactor. The product gas at reactor exit is therefore saturated at the operating pressure (the saturation temperature is 181 \(^{\circ}\)C in this case), which means that its latent heat can be used to generate M/P/LP steam in a counter-current condenser [31]. The cooking chemicals are recovered as smelt, mainly composed of Na\(_2\)CO\(_3\) and to a lesser extent Na\(_2\)S, that falls into the bottom of the reactor and gets dissolved in the weak liquor bath to form green liquor [32]. The green liquor exits from the bottom of the reactor as saturated liquid at the operating pressure and is cooled on its way to the chemical recovery process of the pulp and paper mill by exchanging heat with the weak liquor entering the gasifier to replace it.

The thermal energy requirement of the gasification process is supplied by a partial oxidation of the BL which is regulated by the amount of air supplied to the reactor. In this case the equivalence ratio is controlled by the gasification temperature, which in turn is calculated from the overall energy balance of the reactor assuming a heat loss of 2\% on the LHV of the BL input. According to the operating conditions and the chemical equilibrium calculations, the product gas after the cleaning process is predicted to have a molar composition of 55.4 vol.% N\(_2\), 17% H\(_2\), 14% CO, 13% CO\(_2\) and 0.6% as by-products.

### Table 1

<table>
<thead>
<tr>
<th>Boilers</th>
<th>Thermal streams (hot/cold (*))</th>
<th>Q [kW]</th>
<th>(\Delta T_{\text{min}}/2) [(^{\circ})C]</th>
<th>Radiative heat transfer BB</th>
<th>Convective heat transfer BB</th>
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</table>

\(m_{\text{BL}}\) – BL mass flow rate, kg/s dry.

\(m_{\text{BFM}}\) – BFM mass flow rate, kg/s dry.

### Table 2

<table>
<thead>
<tr>
<th>Boilers</th>
<th>Thermal streams (hot/cold (*))</th>
<th>Q [kW]</th>
<th>(\Delta T_{\text{min}}/2) [(^{\circ})C]</th>
<th>Radiative heat transfer BB</th>
<th>Convective heat transfer BB</th>
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\(m_{\text{BL}}\) – BL mass flow rate, kg/s dry.

\(m_{\text{BFM}}\) – BFM mass flow rate, kg/s dry.
Table 2
Thermal streams of the industrial processes considered.

<table>
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<th>Hot streams</th>
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<td>$T_f$ [°C]</td>
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$m_m$ – Lumber production rate, kg/s dry
$m_m$ – Sawdust mass flow rate, kg/s dry
$m_m$ – Mass flow rate of directly consumed steam, kg/s.

$^a$ Streams associated with the generation of the steam directly consumed in the pulp and paper mill (see the description of the steam cycle in the methodology section of this paper for further details).
CH4. Consequently, about 67% of the total energy that comes with the BL is recovered as chemical energy in the product gas. In the gasifier model, the cooling of the saturated product gas from 181°C to 40°C has been identified as hot stream. In addition, the process consumes electricity to pressurize the BL feed stream from atmospheric to the operating pressure. The power consumption and the thermal streams of the BL gasifier are presented in Table 4.

The expected increase in the energy demand of the lime kiln due to the replacement of the RB by the high-temperature BL gasifier is estimated to be around 16% [33]. In the basecase, i.e. with the Tomlinson boiler, the lime kiln energy demand is supplied by the tall oil recovered from the pulping process of the reference mill. It is assumed that the amount of tall oil produced is enough to balance the expected increment of energy demand in the lime kiln [25].

2.4.2. BFM gasification

The considered BFM gasifier is a pressurized air-blown circulating fluidized bed reactor operated at a pressure of 19 bar and a temperature of 950°C, as the one developed by Sydkraft AB [34] in Värnamo, Sweden. The gasification pressure has been chosen according to the same criterion stated in the description of BL gasification. As a pre-treatment, the BFM needs to be dried and reduced in size in order to make it suitable for feeding and gasification. The BFM, which is available at 55–60 wt.% moisture content, is dried to a moisture content of 8–10 wt.% using hot air as drying medium (the dryer model and its operating conditions are the same as used for the wood pellet plant). The dried BFM is then pulverized in a hammer mill with a final sieve size of 1 mm. Sieving is essential in order to homogenize the size distribution of the biomass particles before the gasification process [5]. The hammer mill power consumption is about 342 kJ per dry kg of milled BFM.

The reactor energy demand is satisfied by the partial oxidation of the BFM and is controlled by the supply of oxidation air to the process. In the model, the equivalence ratio is regulated by the gasification temperature, which in turn is calculated from the overall energy balance of the gasifier assuming a heat loss equal to 5% of the input fuel LHV and correcting this value to take into account char residuals. According to the operating conditions and the chemical equilibrium calculations, the product gas from the reactor is predicted to have a composition of 47 vol.% N2, 8.7% H2, 22.8% CO, 12.5% CO2 and 9% CH4 (on dry basis). Thus, the corresponding chemical energy contained in the product gas accounts for about 73% of the energy in the BFM. The product gas from the reactor is recovered at a temperature of about 800°C [35] and most of its sensible heat is used to generate MP/LP steam in a gas cooler. The power and the thermal streams of the BFM gasification process are summarized in Table 4.

2.5. Gas turbine

Industrial gas turbines are usually designed to be fed with natural gas (NG), which has a much higher calorific value as compared to that of the product gas from biomass gasification (45–50 MJ/kg against 3–6 MJ/kg). This is because the product gas is composed of 50–65 vol.% of inert gases. Apparently, the product gas mass flow rate to the combustor has to be much larger (about 10–17 times larger in this case) in order to achieve the design turbine inlet temperature (TIT). As a consequence, the amount of exhaust gas flow from the gas turbine is much larger than in the case of NG feeding. The mass flow rate is increased to about 10–17 times the NG feeding in order to achieve the desired TIT. The net power output of the gas turbine is calculated as a function of the TIT, the exhaust temperature, and the fuel properties.

Table 3
Gas turbine parameters ISO rated and simulated results.

<table>
<thead>
<tr>
<th></th>
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<th></th>
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<th></th>
<th></th>
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</thead>
<tbody>
<tr>
<td>ISO rated NG</td>
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<td>69.50</td>
<td>1.50</td>
<td>0.00</td>
<td>1250</td>
<td>614</td>
<td>27.82</td>
</tr>
<tr>
<td>Simulated</td>
<td>19</td>
<td>45.82</td>
<td>25.18</td>
<td>22.38</td>
<td>1250</td>
<td>614</td>
<td>26.99</td>
</tr>
<tr>
<td>Simulated</td>
<td>19</td>
<td>58.03</td>
<td>12.97</td>
<td>10.20</td>
<td>1250</td>
<td>606</td>
<td>26.76</td>
</tr>
</tbody>
</table>

![Fig. 1. Material flow connection among the integrated forest industries.](image-url)
Thermal and power streams of the b-IGCC. They and are used as a heat source to generate HP steam in gas turbine model simulations are summarized in Table 3. The exhaust gases of the HRSGs and the net power production of the gas turbines are reported in Table 4 as functions of the biomass fuel fed to the gasifiers.

### Table 4: Thermal and power streams of the b-IGCC

<table>
<thead>
<tr>
<th>Unit/stream description</th>
<th>Thermal streams (hot/cold [°C])</th>
<th>Power [kW]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$T_i$</td>
<td>$T_f$</td>
</tr>
<tr>
<td>BL gasification</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Product gas cooling</td>
<td>181</td>
<td>40</td>
</tr>
<tr>
<td>Gasification air</td>
<td>45</td>
<td>390</td>
</tr>
<tr>
<td>PL pump</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gasification air booster</td>
<td></td>
<td></td>
</tr>
<tr>
<td>BFM gasification</td>
<td>800</td>
<td>30</td>
</tr>
<tr>
<td>Gasification air preheating</td>
<td></td>
<td></td>
</tr>
<tr>
<td>BFM drying air</td>
<td>2</td>
<td>250</td>
</tr>
<tr>
<td>Humid air from dryer</td>
<td>110</td>
<td>45</td>
</tr>
<tr>
<td>Hammer mill</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gasification air booster compressor</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gas turbine</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Preheating $PC_{m}$</td>
<td>40</td>
<td>130</td>
</tr>
<tr>
<td>Air bleed aftercooling</td>
<td>452</td>
<td>40</td>
</tr>
<tr>
<td>$PC_{a}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Preheating $PC_{m}$</td>
<td>40</td>
<td>130</td>
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<tr>
<td>Air bleed aftercooling</td>
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<tr>
<td>$PC_{a}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>HRSG</td>
<td>606</td>
<td>152</td>
</tr>
<tr>
<td>HRSG$_{diff}$</td>
<td>614</td>
<td>152</td>
</tr>
</tbody>
</table>

$m_{BL}$ – BL mass flow rate, kg/s dry.

$m_{BFM}$ – BFM mass flow rate, kg/s dry.

The amount of woodchips required to form the BFM is calculated so that no residual bark is left (i.e. all the bark coming from the sawmill and the wood chopper is used to form the BFM).

### 3. Methodology

#### 3.1. Integration strategy

The main objective of this paper is to investigate the potential for resources and/ or energy savings deriving from considering a large integrated industrial site for biomass processing served by a common CHP system instead of several separated plants having their own utilities. The integrated site comprises a pulp and paper mill, its supply chain, a wood-pellet plant, and one of the two options considered in the previous section for the common CHP system that is either BL and BFM fired boilers or BL and BFM based gasification systems in the so-called Biomass Integrated Gasification Combined Cycle (b-IGCC).

The core of the integrated site is the pulp and paper mill described in the previous section and the strategy for the material stream integration with the other plants is to maintain its production of pulp and paper as it is today. The other material streams of the supply chain are calculated backwards from the woodchip demand of the mill by imposing additional conditions which are needed to eliminate the degrees of freedom due to stream mixing and splitting. These conditions are:

- The amount of woodchips required to form the BFM is calculated so that no residual bark is left (i.e. all the bark coming from the sawmill and the wood chopper is used to form the BFM).
- The total biomass input to the site is calculated to obtain the amount of woodchips required by the mill and by the formation of BFM.
- The proportion between the stream of logs to the wood chopper and that of timber to the sawmill is considered equal in order to reflect the current biomass market situation in Sweden.

Finally, wood-pellet plant production remains fixed by the available amount of sawdust (i.e. the amount of sawdust left after the BFM has been formed). The HEATSEP method [41] is applied to the overall process integration model in such a way that thermal cuts are assigned to the potential hot and cold thermal streams of the processes in the plants (in this case both the initial and final temperatures and the thermal load of the streams are known). The heat transfer interaction among the thermal streams is assumed to occur in a black box without any predefined heat exchanger network. The heat transfer feasibility within the black box is checked by evaluating the composite curves of the hot and cold streams by solving the Problem Table Algorithm according to Pinch Analysis rules [42], that is accumulated heat made available by the hot streams has to be greater than or equal to the accumulated heat demand of the cold streams at all temperature intervals in the thermal cascade.

As a result of the integration with the CHP system shared by all the plants in the site, the heat transfer black box includes:

- the thermal streams listed in Table 2 (pulp and paper mill, sawmill, wood-pellet plant).
- the thermal streams from the combustion of BFM and BL (for a CHP system based on boilers) summarized in Table 1 or the thermal streams from the gasifier(s) and the gas turbine exhaust gases (for a CHP system based on b-IGCC) presented in Table 4, and

Reducing the TIT by reducing the fuel supply. The drawback of this technique is that it lowers the performance of the gas turbine and the power production.

- Accepting the pressure increase in the combustor while maintaining the same mass flow through the expander. The increase in pressure is however limited by the surge limit of the compressor.

- Reducing the air supply to the combustor (by bleeding air from the compressor and/or adjusting the inlet guide vanes (IGV) to minimize the air intake) while maintaining the pressure ratio at the design value.

- Enlarging the expander inlet nozzle size. This is nonetheless a permanent modification, and returning back to natural gas operation mode may not be easy.

For the sake of simplicity the third option, that is reduction of air supply to the combustor by bleeding air from the compressor, has been adopted in the gas turbine model developed (the advantage is that the air bled can be utilized in the gasifiers). The strategy is to have the same reduced mass flow rate through the expander as in the case of the chosen natural gas fired ISO rated aero-derivative gas turbine. The TIT and the compressor pressure ratio (CPR) are also kept at their rated values. The results of the gas turbine model simulations are summarized in Table 3.

The exhaust gases exit the gas turbine at fairly high temperature and are used as a heat source to generate HP steam in HRSGs. The temperature of the exhaust gases at HRSG outlet is set at 152 °C in order to avoid acid gas condensation risks on the surface of the steam generator heat exchangers. The thermal streams of the HRSGs and the net power production of the gas turbines are reported in Table 4 as functions of the biomass fuel fed to the gasifiers.
• the thermal streams of the steam cycle, which are subject to the optimization process.

The amounts of available BL and BFM are the limiting constraint on the size of the heat pocket created by the high temperature hot streams in the CHP system in the GCC representation of the thermal cascade. The steam cycle is designed and optimized to exploit this heat pocket to produce power while supplying the hot utility demand of the industrial site using condensing steam at suitable temperature levels. The Rankine steam cycle is assumed to be operated with a maximum pressure and temperature of 100 bar and 450 °C. The configuration of the steam cycle is as follows;

- steam generation at the maximum cycle pressure,
- steam superheating to the maximum cycle temperature and reheating to the same temperature at 30 bar,
- two or three condensation pressure levels depending on whether a condensing turbine is included in the analysis or not.

It should be noted that the pulp and paper mill subprocesses consume steam at 10 and 4 bar both directly (part of the extracted steam is mixed with the fiber stream and needs to be replaced continuously by a make-up water stream during the steam generation process) and indirectly (part of the extracted steam is condensed to supply process thermal loads and remains in the steam cycle). Therefore, the choice of the condensation pressure levels is predetermined by these technical requirements (see for e.g. [26] for further details). As a result, the decision variables regarding the steam cycle are;

- the mass flow rate of steam condensed at 10 bar that remains in the steam cycle,
- the mass flow rate of steam condensed at 4 bar that remains in the steam cycle,
- the mass flow rate of steam condensed at 0.05 bar (in case a condensing turbine is included in the analysis).

The generation of the steam that is directly consumed in the pulp and paper mill subprocesses is accounted among the process streams and the associated thermal streams are presented in Table 2. Different scenarios are considered in order to explore a wider range of options for the optimal integration of the industrial site in terms of thermodynamic efficiency and operational profitability.

In Scenario 1, the most straight forward case, all the resulting BL and BFM are burnt in the respective boilers to generate HP superheated steam at the maximum cycle pressure and temperature.

In Scenario 2, all the BL and BFM are instead gasified, and the product gas coming from the gasifiers is used to feed a gas turbine. In this case HP superheated steam is generated in HRSGs.

In Scenario 3, all the BL is gasified and the resulting product gas is used to fuel a gas turbine to generate power that would hopefully be sufficient to satisfy the requirement of the industrial site, whereas all the BFM is saved. In this case, the saved BFM can be either simply sold or gasified into high quality syngas (for e.g. using oxygen/steam blown reactors instead of air) and catalytically converted into value added chemicals and biofuels [25].

In Scenario 4, the amount of BFM that is strictly required to supply the hot utility demand of the integrated industrial site is burnt in the BB to generate HP steam, while the surplus BFM and the BL are saved for other processes (e.g. the spared BL can undergo biochemical transformation into biofuels, as in [43], or thermochemical conversion into a high quality syngas suitable for biofuel synthesis).

In Scenario 5, BFM is used to supply the hot utility demand of the integrated industrial site using the b-IGCC pathway, with the intention of saving the BL for other processes. In this case if the BFM were not sufficient to supply the hot utility demand, part of the BL is gasified to supply the deficit.

In all scenarios any deficit in the electricity supply to the integrated industrial site is assumed to be covered by the power grid.

3.2. Set-up of the optimization problems

In all the considered scenarios the objective function is the Rankine cycle net power production, which is to be maximized.

- in scenarios 1, 2 and 3, the steam mass flow rates at the three condensation pressure levels (10, 4 and 0.05 bar) are the chosen decision variables.
- in scenarios 4 and 5 the condensing turbine is excluded from the analysis because the consumption of the available fuels (BFM and BL) is limited just to cover the hot utility requirement of site processes. Thus, the decision variables related to the steam cycle are the mass flow rates of the two higher condensation pressure levels (10 and 4 bar). However, in both scenarios additional decision variables are used to control the BL and the BFM consumption in the CHP system by means of real values in the range between 0 and 1 that multiply their available amounts.

4. Results and discussion

The optimal results of the scenarios described in the previous section assume a greater significance if each of them is compared to a corresponding standalone case in which the plants are considered separately and each served by an own furnace/CHP system to satisfy their respective thermal demands. In order for the comparison to be fair and for the benefits of the integration to be clear, the resource usage must be the same as that of the integrated site. However, since BL or BFM may be differently utilized in the standalone cases, these are either simply saved or burnt/gasified to produce power according to the objectives of that particular scenario. In the following subsections the integrated cases will be labelled with the letter “I” and the stand-alone cases will be labelled with “SA”.

4.1. Optimal integration results

The net power production of the scenarios evaluated for the integrated case are presented in Table 5, and the amounts of BL and BFM saved in each of the integrated cases together with their corresponding standalone cases are summarized in Table 6. Positive net power production shows the surplus power which can be exported from the industrial site whereas negative net power production indicates the amount of power that needs to be purchased from the power grid. Fig. 2 shows a bar chart with the contributions to power generation and consumption in each of the integrated and stand-alone scenarios.

Figs. 3–7 present the corresponding IGCCs of the optimal results of the integrated scenarios described in this section. The red curve represents the GCC of the thermal streams of boiler or gas turbine flue gas (depending on the technology considered) and the thermal streams of the integrated processes. The blue curve represents the GCC of the steam cycle thermal streams and shows how well the steam cycle is exploiting the heat pocket formed by the profile of the red curve. The patches in those figures indicate the magnitude of power generation from the steam and gas turbines (in green color).
increased by 101% compared to its corresponding standalone case. Moreover, the net power production of scenario 2I has presents a better alternative should power production become a prior-
case 1I. This shows that the gasifiers based combined cycle pre-
1I, the net power production further increases by 58% compared to 2I, where the b-IGCC replace the boilers based CHP system of case 3I, the net power production further increases by 58% compared to 2I, where the b-IGCC replace the boiler based CHP system of previous section to produce power. It is apparent from the results of the previous section to produce power. It is apparent from the results of the integrated cases 1I and 2I, respectively. In both of the corres-
Due to the large potential coming from the fuel calorific content. Actually, about 78 MW and 122 MW of excess power are produced in the integrated cases 1I and 2I, respectively. In both of the corre-
heat to the steam cycle of the internal CHP system. In so doing the objective of saving all the BL is maintained in the standalone case as well, and a fair ground for comparison against the integrated one is established. The BFM consumption in case 4SA is calculated by simulating the pulp and paper mill model with zero BL consumption and obtaining the amount of BFM required to produce the same thermal output to the steam cycle. The BFM consumption in the pulp and paper mill is then added to the BFM needed in the sawmill and the wood-pellet plant furnaces. This resulted in an overall BFM demand of 4.48 kg/s dry, which is about 93% of the total BFM available. Compared to case 4I, the BFM consumption of the standalone case is 121% higher, whereas the deficit in net power is about 38% lower. Apparently, the integration among the different plants does not allow to achieve the same benefits as in scenario 3 (i.e. both an increase in power generation and a larger amount of saved biomass fuel). Power production and biomass saving remain conflicting objectives in this scenario and, if power production is not a prior-
2I, the BB is set to burn enough BFM just to provide the hot utility demand of the industrial site, while all the BL is saved for other process. On the other hand, in case 4SA the pulp and paper mill is assumed to shut down its RB and to use only the BFM fueled BB to supply heat to the steam cycle of the internal CHP system. So in doing the objective of saving all the BL is maintained in the standalone case as well, and a fair ground for comparison against the integrated one is established. The BFM and the power consumption of the plants in the integrated site (in brown color).

<table>
<thead>
<tr>
<th>Scenario</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Integrated case (I)</td>
<td>77.76</td>
<td>121.91</td>
<td>2.56</td>
<td>-39.50</td>
<td>60.80</td>
</tr>
<tr>
<td>Standalone case (SA)</td>
<td>36.72</td>
<td>60.60</td>
<td>-24.40</td>
<td>-24.40</td>
<td>-16.54</td>
</tr>
<tr>
<td>CHP plant efficiency (%)</td>
<td>55.0</td>
<td>48.2</td>
<td>36.9</td>
<td>21.9</td>
<td>47.8</td>
</tr>
</tbody>
</table>

Table 5
Net power production of the integrated and standalone cases [MW].

Scenario 1 and 2 (Figs. 3 and 4) present the cases where all the BL and BFM available are consumed to supply the hot utility and power requirements of the integrated industrial site using different pathways. The expectation for positive net power production that can be exported to the power grid is reasonable in both scenarios due to the large potential coming from the fuel calorific content. Actually, about 78 MW and 122 MW of excess power are produced in the integrated cases 1I and 2I, respectively. In both of the corre-

In scenario 3I (Fig. 5) only the BL is consumed in the b-IGCC pathway to supply all the hot utility demand and as much as possible of the power demand of the integrated industrial site. The corresponding standalone case is the same as case 2SA described above, except for the remaining 72% of the BFM is saved instead.

In case 4I (Fig. 6) the BB is set to burn enough BFM just to provide the hot utility demand of the industrial site, while all the BL is saved for other processes. On the other hand, in case 4SA the pulp and paper mill is assumed to shut down its RB and to use only the BFM fueled BB to supply heat to the steam cycle of the internal CHP system. In so doing the objective of saving all the BL is maintained in the standalone case as well, and a fair ground for comparison against the integrated one is established. The BFM consumption in case 4SA is calculated by simulating the pulp and paper mill model with zero BL consumption and obtaining the amount of BFM required to produce the same thermal output to the steam cycle. The BFM consumption in the pulp and paper mill is then added to the BFM needed in the sawmill and the wood-pellet plant furnaces. This resulted in an overall BFM demand of 4.48 kg/s dry, which is about 93% of the total BFM available. Compared to case 4I, the BFM consumption of the standalone case is 121% higher, whereas the deficit in net power is about 38% lower. Apparently, the integration among the different plants does not allow to achieve the same benefits as in scenario 3 (i.e. both an increase in power generation and a larger amount of saved biomass fuel). Power production and biomass saving remain conflicting objectives in this scenario and, if power production is not a prior-

In scenario 3I (Fig. 5) only the BL is consumed in the b-IGCC pathway to supply all the hot utility demand and as much as possible of the power demand of the integrated industrial site. The corresponding standalone case is the same as case 2SA described above, except for the remaining 72% of the BFM is saved instead.

<table>
<thead>
<tr>
<th>Scenario</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Integrated case (I)</td>
<td>BL 0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>18.05</td>
<td>13.16</td>
</tr>
<tr>
<td>BFM 0.00</td>
<td>0.00</td>
<td>15.96</td>
<td>9.21</td>
<td>0.00</td>
<td></td>
</tr>
<tr>
<td>Standalone case (SA)</td>
<td>BL 0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>18.05</td>
<td>18.05</td>
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<tr>
<td>BFM 0.00</td>
<td>0.00</td>
<td>11.48</td>
<td>1.05</td>
<td>0.00</td>
<td></td>
</tr>
</tbody>
</table>

Table 6
The amounts of BL and BFM saved in the integrated and standalone cases [kg/s, dry].

In case 5I (Fig. 7), the BB based CHP system of case 4I is replaced by the BFM gasifier based combined cycle with the same intention of saving all the BL. However, it turned out that in this design solution the utilization of all the available BFM was not sufficient to meet the hot utility demand of the industrial site. This means that part of the BL has to be gasified as well (the optimal solution shows...
that 31% of the available BL is consumed in addition to all the BFM). Case 5SA is the same as case 4SA (which consumes 93% of the BFM available) except that in case 5SA the remaining 7% of the BFM is set to be gasified to produce power in order to establish a fair comparison ground. Accordingly, case 5SA results in a net power deficit of 16 MW, whereas the integrated case produces a surplus of 61 MW. However, case 5SA saves 27% more BL compared to its integrated counterpart.

4.2. Economic evaluation of the optimal cases

The optimal solutions are also evaluated from the economic point of view by using Investment Opportunity (IO) as an indicator of the potential operational profit. IO is an index of the profitability of a design solution based on the difference between the algebraic sum of resource cost and product revenue. The higher is the IO value the wider are the margins for investing more money in a design solution while the cash flow history of the project remains positive at the end of the economic lifetime.

\[
IO = \sum (\text{products revenue}) - \sum (\text{resources expense})
\]

The annual potential IO is evaluated as an economic indicator of the integrated and standalone cases assuming 8000 h of annual operational time and the results are presented in Fig. 8. In all the cases the integrated IO is found to be greater than the standalone IO, showing that the integrated cases present better economic performance, with margins in the range between 155 and 395 MSEK. Cases 2I and 5I, in particular, have resulted in high IO potential due to the large amount of power produced by the gas turbines. If power production is not a priority, cases 3I and 4I present better alternatives at lower IO values while allowing larger margins for saving resources which can be utilized in innovative conversion technologies.

A sensitivity analysis about the IO potential as a function of electricity price (assuming that the selling and purchase price is the same) is further performed for the cases that represent the
extreme situations in regards to the possible objectives pursued by site integration i.e. maximum power production (case 2I, where all the BL and BFM are consumed) and maximum resources saving (case 3I, 0% BL 100% BFM, and case 4I, 100% BL 57% BFM). The results (Fig. 9) show that electricity prices above 516 SEK/MW h (i.e. the abscissa of the point where the lines of cases 2I and 3I intersect) make case 2I more favorable than case 3I, focused on BFM saving, whereas prices above 564 SEK/MW h (i.e. the abscissa of the point where the lines of cases 2I and 4I intersect) make case 2I more favorable than case 4I, mainly focused on BL saving.

In regards to resources saving, case 3I (BB technology) presents better IO potential compared to case 4I (BL based b-IGCC technology) for electricity price greater than 700 SEK/MW h. At the current electricity price (800 SEK/MW h which is close to the abscissa of the intersection point) the difference in the annual IO potential between the two cases is marginal compared to the difference in the resources saving potential evaluated according to the lower heating value of the saved resources (340 MWth (3I) against 416 MWth (4I)). The resources saving potential of case 4I compared to 3I, both evaluated according to the prices of resources in Table 7, is 2.14 times larger.

5. Conclusions

The discussion of the results in the preceding section shows that an integrated industrial site featuring a pulp and paper mill and its supply chain presents a significant potential for resources saving and/or increased power production compared to standalone plants. This potential derives mainly from the integration of the industrial processes (pulp and paper mill, sawmill and wood-pellet plant) with a common CHP utility. The considered different scenarios also help to assess the impact of the technology (boilers or gasifiers) used in the CHP system on the magnitude of the potential for resources saving/increased power production.

Cases 1I and 2I prioritize increased power production over resources saving, and case 2I presents the design solution producing the highest power surplus. On the other hand, cases 3I and 4I focus on resources saving rather than increased power production. Surprisingly, the results from case 3I show that the chemical energy content of the product gas from BL gasification is enough to supply the thermal and power loads of the integrated site without the need of supplementary BFM feeding. Conversely, according to the results of case 4I, about 43% of the available BFM is enough to supply all the thermal and 35% of the power requirements of the integrated industrial site.

From the economic point of view, the integrated cases perform better than the standalone ones in terms of IO potential. Another significant observation is that the annual potential IO tends to be higher in the scenarios that implement b-IGCC technology due to the large amount of power produced by the gas turbines in those cases. Finally, the sensitivity analysis about the IO potential shows that the lower end of the electricity price range in which the objective of increased power production is more convenient than that of resources saving is still much lower than the current electricity price.

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References

Paper III

Black liquor fractionation for biofuels production- A techno-economic assessment
Black liquor fractionation for biofuels production – A techno-economic assessment

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Abstract
The hemicelluloses fraction of black liquor is an underutilized resource in many chemical pulp mills. It is possible to extract and separate the lignin and hemicelluloses from the black liquor and use the hemicelluloses for biochemical conversion into biofuels and chemicals. Precipitation of the lignin from the black liquor would consequently decrease the thermal load on the recovery boiler, which is often referred to as a bottleneck for increased pulp production. The objective of this work is to techno-economically evaluate the production of sodium-free lignin as a solid fuel and butanol to be used as fossil gasoline replacement by fractionating black liquor. The hydrolysis and fermentation processes are modeled in Aspen Plus to analyze energy and material balances as well as to evaluate the plant economics. A mathematical model of an existing pulp and paper mill is used to analyze the effects on the energy performance of the mill subprocesses.

HIGHLIGHTS
- A model for the fractionation of black liquor to produce lignin and butanol is developed.
- A techno-economic assessment is carried out.
- Configuration changes are introduced to the evaporation train.
- Results show potential for a profitable biorefinery concept to chemical pulp mills.

1. Introduction

The sulphate or Kraft process is the dominant production method of chemical pulping and accounted for approximately 75% of the global pulp production in 2012 (FAOSTAT, 2014). Sweden is the largest producer of chemical wood pulp in Europe with a total of 8.1 million tonnes in 2012 (FAOSTAT, 2014). This results in approximately the same amount of black liquor (BL) as a by-product. Currently, the BL is used as a fuel in the recovery boiler (RB). The RB is an important part of chemical pulp mills as it provides the thermal energy and power demand of the mill while also playing a key role in the recovery process of the pulping chemicals. The latter is very crucial for the overall economy of the Kraft process. The BL contains pulping chemicals, lignin and degraded hemicellulose. The hemicellulose fraction of the BL is an underutilized resource in many chemical pulp mills. Extracting that fraction could be one way to generate a sugar feedstock amenable to biochemical conversion into biofuels and chemicals. Precipitation of the lignin from the black liquor would consequently decrease the thermal load on the recovery boiler, which is often referred to as a bottleneck for increased pulp production. The objective of this work is to techno-economically evaluate the production of sodium-free lignin as a solid fuel and butanol to be used as fossil gasoline replacement by fractionating black liquor. The hydrolysis and fermentation processes are modeled in Aspen Plus to analyze energy and material balances as well as to evaluate the plant economics. A mathematical model of an existing pulp and paper mill is used to analyze the effects on the energy performance of the mill subprocesses.
In some cases the RB is referred as a bottleneck towards increased pulp production (Pettersson et al., 2012; Vakkilainen and Valimäki, 2009). The extraction of lignin from the BL can help debottlenecking in those situations. Fractionation of the black liquor therefore represents an interesting business opportunity for chemical pulp mills. The benefits of separating lignin from the BL of pulp and paper mills to increase pulp production have been discussed by several authors in the literature (see e.g. (Vakkilainen and Valimäki, 2009; Olsson et al., 2006; Axelsson et al., 2006)). The advantage is twofold: it opens the opportunity for increased pulp production without altering the ordinary pulping and chemical recovery processes, and lignin can be sold as a fuel by itself thereby increasing the products. The pre-extraction and chemical recovery processes, and lignin can be sold as a fuel by itself thereby increasing the products. The pre-extraction of hemicelluloses from pulp wood prior to pulping for the production of fuels by itself thereby increasing the products. The pre-extraction of hemicelluloses from pulp wood prior to pulping for the production of biofuels and green chemicals have also been previously reported, see e.g. (Huang et al., 2010; Mao et al., 2010).

One subject of this study is to assess the techno-economic feasibility of the recovery of hemicelluloses from the BL and its subsequent conversion into butanol via the conventional ABE process. Butanol can play an important role in alleviating the dependence on fossil derivatives for transportation fuel. N-butanol with molecular formula C4H9OH and boiling point of 118 °C has energy density about three-fourth of gasoline. This makes it superior to ethanol as a biofuel, and more importantly it can be produced from more sustainable feedstock, than bio-diesel for instance (Dürre, 2011; Green, 2011). In addition, butanol can surpass the blending limit of ethanol with gasoline for regular vehicles (which is usually less than 10%) without inducing significant impact on the Otto engine performance. The production of butanol from BL is not mentioned in the literature; however, commercial production of bio-butanol through fermentation of sugars has been used previously, see e.g., (Ni and Sun, 2009).

The production butanol from hemicellulose substrates through the conventional ABE process is hampered by low yield as compared to starchy substrates (rich in sugar content such as corn and molasses) because hydrolyzate derived hemicelluloses contains low sugar which is mostly dominated by xylose. However, the need for efficient solvent recovery remains an issue regardless of the substrate as butanol concentration in the fermentation broth puts a limit on the Clostridia strains (maximum concentrations tolerable 10–12 g/l butanol or 20 g/l ABE (Qureshi et al., 2001, 2007; Ezeji et al., 2004a, 2007; Lu et al., 2012; Xue et al., 2013)). Recent developments regarding solvent recovery techniques can be found in (Qureshi and Blaschek, 2001; Xue et al., 2013; Qureshi et al., 2001, 2005; Kraemer et al., 2011; Ezeji et al., 2004b; Jin et al., 2011; Groot et al., 1992). Gas stripping is reported as the most promising recovery technique both from economic and operability viewpoints (Ezeji et al., 2004a; Jin et al., 2011; Lu et al., 2012; Xue et al., 2013; Qureshi and Blaschek, 2001).

In the current work a conceptual process for the production of high grade lignin and ABE solvents from Birch-wood Black Liquor (BL) is developed. The new process is assumed to be integrated in an existing pulp and paper mill. The model is then used to evaluate the energy and material balances of the system for different process conditions. An economic evaluation of the new process is made based on published price data.

The paper is organized as follows. Section 2 presents a brief description of the experimental data up on which the conceptual process is based and the corresponding Aspen Plus model developed. The methodologies for economic and process evaluations are also presented in Section 2. Section 3 summarizes the main findings and the discussions about the most significant outcomes.

2. Methods

2.1. Process description

The fractionation plant considered in this work produces butanol from BBLH via the conventional ABE fermentation process, while high grade lignin is separated as an intermediate product. A model is developed in Aspen Plus in which the material and energy balances are obtained. The input data to the lignin separation and BBLH producing process are based on the experimental data carried out in an existing pulp and paper mill. The input data to the ABE fermentation is also based on experimental trials conducted on BBLH and the downstream processes (namely product recovery and purification) are based on the literature. The plant is assumed to operate 8000 hours/year and the butanol production capacity is fixed by the amount of BBLH available.

In order to extract lignin carbohydrate complexes (LCC) for the production of biofuels (or green chemicals) via hydrolysis and fermentation pathway, a BBL stream at about 13% DM is diverted from the digester of a pulp and paper mill. The extracted stream contains organic and inorganic (mainly alkali) substances. The organic content in the extracted stream is equivalent to about 10% of the total organic substance present in the pulp wood and the inorganic matter is also about 10% of the total cooking chemicals charged, both on dry matter basis. It is also considered important to minimize the effect of the degradation of hemicelluloses in the digester.

The diverted stream is then sent to one of the multiple effect evaporator units of the pulp and paper mill, to increase its solid content up to 25–30% DM before its pH is lowered by injecting CO2. The decrease in pH causes the LCC in the diverted stream to precipitate. The precipitate, which accounts for about 30% of the total dry solids extracted from the digester, is then filtered to separate it from the alkali rich solution that comes along with the diverted stream from the digester. The alkali rich solution is returned back to the chemical recovery cycle of the pulp and paper mill, whereas the LCC stream with about 50% DM undergoes the hydrolysis and fermentation pathway to produce butanol and high grade lignin as main products, and acetone and ethanol as co-products. The hydrolysis is carried out using diluted sulfuric acid in agitated reactor which is maintained at about 100 °C for one hour. Alkali free lignin, at about 65% DM, is filtered out of the stream exiting the hydrolysis reactor and the rest of stream is let into the downstream processes. The pH of the hydrolysate is increased from about 1, in the hydrolysis reactor, to about 5 prior to its admission into the fermenter by dissolving lime mud (CaCO3) which is freely available in the pulp and paper mill. Overview of the process is shown in Fig. 1.
The BBLH stream is the feed for the ABE fermentation process which was refitted with the BBLH experimental data: ABE fermentation (batch) of BBLH using \textit{Clostridium acetobutylicum} ATCC 824 conducted at a temperature of 37 °C and in agitated reactor set to 200 rpm. The pH was controlled to 5.1 using automatic addition of 45 N\textsubscript{NH\textsubscript{4}}OH during the entire period.

2.2. The Aspen Plus model

The Aspen model is a steady-state flowsheet based on a stoichiometric reactor approach. The total fermentation time considered is 200 h and stripping starts after 20 h. In order to render a continuous process, the overall productivity in the model is maintained using average mass flow rates over the total fermentation time. For the same reason, the substrate intermittently added during the fed-batch fermentation process is simulated as a continuous addition using averaged mass flow rates over the entire fermentation period.

The BBLH stream (pre-fermentation) contains about 38.5 g/l xylose, 3.4 g/l acetic acid, 0.1 g/l furfural, 0.5 g/l hydroxymethylfurfural (HMF), 3.8 g/l phenolic (acid soluble lignin (ASL)) and 2.1 g/l formic acid. In addition about 65 g/l of gypsum is present in the hydrolysate. Moreover, flash evaporators are required to concentrate the BBLH up to 100 g/l sugar concentration for the main feed stream and up to 500 g/l of xylose concentration for the intermittent feed stream. The fractional conversion of the sugars in the BBLH to ABE solvents is based on unpublished experimental data and on the fermentation stoichiometry fitted in the model. The precipitation, hydrolysis and liming processes have also been modeled based on experimental data conducted in a real pulp and paper mill. According to the model 30 vol.% of the carbon in the pentose sugars of the substrate is converted to ABE, 20 vol.% to CO\textsubscript{2} and the rest into acids.

The ABE process solvent recovery system considered in the model is based on gas stripping technique as previously described (Qureshi and Blaschek, 2003) and is accomplished by recirculating fermentation byproduct gases through the broth. According to the model, the fermentation gases produced are more than that of required for the gas stripping process and therefore part of it is bled-off. The selectivity of the stripping process for a specific compound is calculated according to Eq. (1).

\[
\text{Selectivity} = \frac{y/(1 - y)}{x/(1 - x)}
\]

where:
- \(y\) – the concentration of a compound in the product condensate (wt.%)
- \(x\) – the concentration of that compound in the fermentation broth (wt.%)

The ABE are selectively removed during the fermentation process. In the fed-batch fermentation (20–200 h), butanol selectivities were reported in the range of 10–22. Acetone and ethanol selectivities ranged over 6.69–12.72 and 4.45–11.16, respectively (Ezeji et al., 2004a). The solvents are recovered by condensing the gases flowing out of the fermenter to a temperature of 2 °C in a condenser.

Several authors have also published experimental results of bio-butanol production via fermentation of starchy substrates with in situ gas stripping. The composition of the product condensate i.e. concentration fractions of acetone, butanol and ethanol to the total ABE in all the reports fairly agree, see e.g. (Ezeji et al., 2004a, 2007; Lu et al., 2012) based on fed-batch mode and (Ezeji et al., 2004b, 2007; Qureshi et al., 2007) based on batch mode fermentation.

The product separation and purification process is composed of five distillation columns, all based on RadFrac unit operation of Aspen Plus, and a triple phase flash decanter. The final product purity targets are set to 99.9 wt.% for butanol and acetone and 97 wt.% for ethanol. The triple flash decanter and two of the distillation columns are used to separate the heterogeneous butanol–water azeotropic mixture according to the configuration reported in the literature (Luyben, 2008). In addition the final composition of the high grade lignin is set to 65% DM.

2.3. Economic evaluation

The proposed processes are evaluated from an economic point of view by using the cost of butanol production as an economic indicator. The study estimates method together with the Hand
method is used to estimate the capital cost. The pre-requisite for using the study estimates method (that is a flowsheet diagram of the process) is fulfilled. Accordingly, the sets of equipment which are required by the process are sized based on the mass and energy balance flowsheet developed in Aspen Plus. The evaluation of the equipment cost is performed by estimating the purchase cost of the unit operations, based on published data as well as in-house database information, and by multiplying them with their respective Hand factor for the equipment type to account for piping, insurances, installations etc. Moreover, since the fermentation process is assumed to be operated on a fed-batch mode a schedule for the unit operations, based on published data as well as in-house database information, and by multiplying them with their respective Hand factor for the equipment type to account for piping, insurances, installations etc. Moreover, since the fermentation process is assumed and evaluated during the economic assessment. The capital cost is estimated according to Eq. (2).

$$\text{Capital cost} = \sum \left[ \text{Equipment purchase cost} \times h_t \times f_m \right] \times f_i$$

(2)

where:

- $h_t$ = Hand factor,
- $f_m$ = material factor,
- $f_b$ = building factor and
- $f_i$ = place factor.

The cost of equipment for the unit operations involved in the process are initially estimated using correlations and data available in the literature (Brown, 2007). The initial estimates have been corrected to match the pressure and material requirement of the current process using factors reported on the same literature. The estimated costs are based on the chemical engineering plant cost index (CEPCI) 460 (year 2005) and are reported here after being adjusted for inflation for the year 2011 (CEPCI 586).

2.4. Process integration and impact assessment methods

The strategy for the process integration has been to extract 10% of the organic content of the BL and fractionate it according to the processes described in Section 2. In doing so, two integration boundaries are considered during the economic and the hot utility requirement assessments:

- the integration of the lignin separation and BBLH production process (the section of Fig. 1 inside broken line boundary);
- the integration of both the lignin separation and BBLH production process and the conventional ABE process (Fig. 1).

Furthermore, the impact of conceptual processes on the Kraft pulp and paper mill has been investigated using:

- a mathematical model of typical Nordic mill (the impact of the integrated processes on the evaporation unit and the RB);
- the model mill and pinch technology (Kemp, 2007) (the impact of the integrated processes on the hot utility requirement).

During the hot utility assessment the thermal streams of the integrated processes are added to the heat transfer black box of the model mill according to the HEATSEP method (Lazzaretto and Toffolo, 2008) principles. In this case the results present the change in the MER due to the integration of the new processes without considering the synthesis of HEN to realize it.

3. Results and discussion

3.1. Material balance

This section presents the input–output structure of the model developed. The composition of the extracted stream is used as the basis for determining carbon dioxide, sulfuric acid and lime mud demand of the precipitation, hydrolysis and liming processes, respectively. In addition, water is added to dilute the sulfuric acid, initially at 72% H$_2$SO$_4$, to 4% H$_2$SO$_4$ concentration in the hydrolysis reactor.

Alkali free lignin as a biomass fuel and butanol as an alternative transport fuel are the main products considered. However, acetone and ethanol are also produced along with butanol as co-products. The material balance of the integrated process is presented in Table 1.

The limitation on the amount of organics that can be extracted from the BL has resulted in lignin and butanol production capacities which are quite low for an economical sensible biorefinery capacity. This can for instance be improved, if the lignin extraction can be made self-supporting, e.g., by a high price for refined lignin and/or if the extraction can be used to increase pulp production capacity (because of the decreased load on the MEEV and the RB). Another option is to introduce sugar hydrolysate streams from other sources in order to increase the production of butanol thereby bringing the integrated process to an economically sensible biorefinery capacity. The detailed evaluations of these aspects are, however, out of the scope of this work.

3.2. Process utility requirement

The lignin separation and butanol plant utility requirements are classified into heating (using MP) (medium pressure steam), cooling (using water) and electricity. The steam is mainly required by the heat exchangers and flash evaporators and to a lesser extent by the re-boilers of the distillation columns. For the cold utility, water is assumed to be the cooling medium which is mainly required by the coolers as well as the condensers of the distillation columns. The steam, cooling water and electricity demand of the process are evaluated in Aspen Plus and are used as the basis for the evaluation of the cost of utility during the economic assessment. The utility requirements of the conceptual processes are summarized in Table 2.

3.3. Investment and operating costs

The capital cost is estimated according to Eq. (2) and is accounted in the analysis using the annuity method, assuming 15 years plant life time and 13% interest rate. The Hand factors used in the analysis are as follows: for the compressors 2.5, reactors 4, pumps 4, heat exchangers 3.5, pressure vessels 4 and fractionation columns 4. Moreover, the instrumentation factor is set to 1.55 (that is for central control and computerization), and the building factor is set to 1.11 (that is for new solid-fluid processing equipment). The pre-requisite for the equipment type to account for piping, insurances, installations etc. Moreover, since the fermentation process is assumed and evaluated during the economic assessment. The capital cost is estimated according to Eq. (2) and is accounted in the analysis using the annuity method, assuming 15 years plant life time and 13% interest rate. The Hand factors used in the analysis are as follows: for the compressors 2.5, reactors 4, pumps 4, heat exchangers 3.5, pressure vessels 4 and fractionation columns 4. Moreover, the instrumentation factor is set to 1.55 (that is for central control and computerization), and the building factor is set to 1.11 (that is for new solid-fluid processing equipment).
The investment is estimated based on equipment sizes calculated according to the amount of the extracted stream. Besides, configuration changes are introduced in the MEEV so that it supports the organics precipitation process thereby minimizing the capital investment and the operating cost of the integrated process (see Section 3.7). The required number of personnel is estimated using the data available in the literature where fractions are assigned for personnel per unit operation per shift (Brown, 2007). These fractions are multiplied with the number of unit operations of each type and with the number of workers summed up to obtain the total number of persons needed. The fractions used in the analysis are as follows: for the compressors (0.09 persons/unit/shift), for the reactors (0.25 persons/unit/shift), for the distillation towers (0.25 persons/shift/shift/unit), for the heat exchangers (0.05 persons/unit/shift), and for the evaporators (0.15 persons/unit/shift). Assuming 5 shifts, about 15 personnel are required. This has been accounted in the production cost estimation by assigning kUSD 71.5 per person per year. This resulted in an estimated labor cost of 0.5 MUSD/year.

The cost of the extracted stream is excluded from the economic evaluation because its organic content comes from the woodchips and the cost of woodchips has already been accounted in the pulping process. In such case the digester would be providing a pretreated feed stream to the proposed process as one of the mill subprocesses. In fact, the celluloses and part of the hemicelluloses in the wood are converted into pulp and the pulping is not impacted by the extracted stream (Helmerius et al., 2010). The evaporation unit and the RB are the only subprocesses of the pulp and paper mill which are affected by the BBL fractionation plant.

Table 3 summarizes the utilities and raw material prices associated with the lignin separation and the ABE process. It is worth mentioning that part of the cooking chemicals in the extracted stream is lost in the downstream processing. During the economic evaluation a NaOH make-up stream that accounts for the loss is evaluated. The amount of the cooking chemicals lost in the process is estimated to be about 5% of the total amount of cooking chemicals in the extracted stream (Olsson et al., 2006).

Table 3
<table>
<thead>
<tr>
<th>Utility and material usage</th>
<th>Tonne/tonne-lignin*</th>
<th>Price (USD/tonne)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MP steam</td>
<td>1.54</td>
<td>50</td>
</tr>
<tr>
<td>Water (both utility and process)</td>
<td>32.74</td>
<td>0.2</td>
</tr>
<tr>
<td>Carbon dioxide</td>
<td>0.56</td>
<td>52*</td>
</tr>
<tr>
<td>Sulphuric acid</td>
<td>0.32</td>
<td>71*</td>
</tr>
<tr>
<td>Make-up sodium hydroxide</td>
<td>0.16</td>
<td>280*</td>
</tr>
<tr>
<td></td>
<td>kW/tonne-lignin*</td>
<td>(USD/kWh)</td>
</tr>
<tr>
<td>Power consumption</td>
<td>2.10</td>
<td>0.045*</td>
</tr>
</tbody>
</table>

* Lignin on dry matter basis.

3.4. Lignin production cost

One perspective of interest is to consider the option of integrating only the lignin separation and BBLH production plant (see Fig. 1 the process inside the broken line boundary). In this case the BBLH is assumed to be sold as final product boosting the revenues. Considering the fact about 60% of the capital investment is due to the ABE process, exploring this option is deemed interesting. The cost of production accounts mainly for the costs incurred by the investment, the purchase of raw materials, the utilities (MP steam, water, electricity, refrigeration etc.), and the labor. Since there are two products to consider in this case, the reciprocity relationship between the production costs of lignin and BBLH is evaluated. Doing so has resulted in lignin production cost range of USD/MWh 36 to 0 for the corresponding BBLH price in the range of USD/tonne 0–28. Accordingly, the lignin price must be above USD 36/MWh in order the integration of the lignin separation process to be profitable without the incentive from BBLH.

3.5. Butanol production cost

On the other hand, integrating both the lignin separation and the ABE processes results in an increased product portfolio. The cost of butanol production is chosen to be the basis for the economic analysis while the price of lignin is kept fixed, at least for the main case where the lignin price is set to 30 USD/MWh. The reason for this is that, the price of lignin is available in the in-house database as well as in the open literature in a fairly similar range. The prices of solid biomass fuels in general are available in the quarterly report of statistics Sweden (SIA, 2014). That is not the case with the price of butanol. Nonetheless, the selling price of lignin has been a subject of a sensitivity analysis in the next section. For the economic conditions described in the previous section and a lignin selling price of 30 USD/MWh, the butanol production cost is estimated to be 7.12 kUSD/tonne-butanol. The production cost reduces to 5.56 kUSD/tonne-ABE, if acetone and ethanol are included in the product stream. The major contributors to the production cost are the capital cost of the integrated processes (about 40% of the production cost) followed by the labor cost and the utility steam, with about 20% of the production cost each.

The estimated cost of butanol is rather high compared to the current market price 1.03–1.61 USD/tonne-butanol reported in (Mariano et al., 2013). The large difference in butanol prices between the current work and reported values results mainly from the low yield of solvents due to the toxic nature of the BBLH and from the low production quantity of butanol due to the limitations related to the amount of organics that can be extracted of from the pulping process.

In order to assess the impact of these aspects a sensitivity analysis is performed and the results are presented in the next section.

3.6. Sensitivity analysis

Sensitivity analysis of the most influential parameters towards the production cost of n-butanol such as the price of lignin, the solvent yield of the ABE process and economies of scale effects, are considered and the results are presented in this section.

Lignin is the major product with respect to the quantity produced and this makes it an important parameter to the economy of the integrated processes. In fact, a huge part of the earning from the integrated processes is expected to come from the selling of lignin. According to the assumed economic conditions (i.e. 13% interest rate and 15 years economic life time), butanol can be produced in the cost range of kUSD/tonne-butanol 7.12–3.6 for a lignin selling price in the range of USD 30–50/MWh. If acetone and ethanol are included in the product stream, the production cost reduces
the RB toxic nature of the BBL substrate towards the problems.

In order to assess the economies of scale effect a case with 30% extraction of organics from the pulping process is evaluated on the top of production cost sensibility towards lignin price. The results showed about 42% reduction in the production cost both on butanol and on ABE as compared to the case with 10% extraction of organics (Fig. 2b). It should be noted that extraction of organics above 10% would result in local limitations in the subprocesses of the pulp and paper mill, such as combustion problems in the RB. Therefore, if organics extractions above 10% are to be considered they must be supplemented by either increased pulp production or other technical measures in order to avoid the subsequent problems.

The economy of biobutanol production through BBLH fermentation is largely affected by the low yield of solvents as a result of the toxic nature of the BBL substrate towards the C. acetobutylicum. However, there are reports for batch fermentation on pentose substrates where the yield is much higher than achieved from the fermentation of BBLH. A sensitivity analysis has also been performed to emphasize on the effect of the yield of solvents on the production costs (Fig. 3a). A sugar utilization weight ratio of 0.3 (i.e. butanol to sugars in the substrate) has been reported previously (Mariano et al., 2013). A production rate of 0.06 tonne/h butanol corresponds to a conversion ratio of 0.3 g of butanol per gram of xylose in the hydrolyzate during the fermentation. Should future breakthroughs on BBLH fermentation allow the yield to increase up to 0.06 tonne-butanol/h or the corresponding yield on ABE 0.08 tonne-ABE/h (calculated according to the ratio of the solvents yield achieved from the BBLH experiments), the production cost of butanol would reduce from 8.9 to 2.97 kUSD/tonne-butanol and the production cost on ABE would reduce from 6.8 to 2.27 kUSD/tonne-ABE.

Furthermore, in order to emphasize the economies of scale effects the 30% extraction of organics case is imposed upon the production cost sensibility towards the yield of solvents. This has resulted in production cost ranges of 4.17–1.95 kUSD/tonne-butanol and 3.2–1.5 kUSD/tonne-ABE (Fig. 3b).

3.7. Impact of the extraction of organics on the evaporation unit and the RB

The removal of hemicellulose and lignin from the extracted stream will afterward change the organics composition of the BL stream when it comes to the evaporation plant and later to the RB. The inorganics composition on the other hand does not change as much, except for the part that is filtered out with the LCC stream which is then directed to the lignin separation plant and later to the butanol production plant. In addition to the composition, the physical properties of the BL are expected to change. In this work, the physical properties of BL, such as viscosity and boiling point elevation (BPE), after the lignin removal are assumed to remain the same. The viscosity and BPE change are insignificant compared to an ordinary BL (Moosavifar et al., 2006). However, the composition and heating value have been recalculated based on data previously reported in (Vakkilainen and Välimäki, 2009). In fact the same results, as reported, have been achieved by recalculating the composition of BL by assuming the organics removed to be shared according to the ratio of carbon to hydrogen in ordinary BL. Ordinary BL is roughly composed of about one-third of organics, one-third inorganics and the remaining one-third water.

According to the experimental results, up on which the model is based, about 30 wt.% (dry basis) of the extracted stream is separated as filter-cake after the CO₂-precipitation. The rest of the stream is assumed to be recycled and mixed with the BL stream that comes from the digester before entering the evaporation unit. In addition the extracted stream is concentrated to about 30% DM in one the evaporator units after being mixed with a recirculated organics stream with about 60–70% DM content.

The effect of hemicellulose and lignin separation on the pulp and paper mill is performed on a mathematical model of an existing pulp and paper mill with emphasis on the effect of the lignin separation in regard to the energy performance of the evaporation unit and the RB. A full detail of the mathematical model mill is reported in (Mesfun and Toffolo, 2013).

Hereafter, the basecase refers to the ordinary pulp and paper mill process and the new cases refer to the pulp and paper mill process with the integrated lignin separation and butanol production plant. The evaporation unit with the basecase and new configuration is presented in Fig. 4a and b, respectively. The impact of the new evaporation unit configuration on the live steam demand is evaluated over a range of temperatures of the stream entering the evaporator units and for different temperatures of the stream entering effect 3 (E-3, see Fig. 4b). The stream entering E-3 is the mixture of the extracted stream and the organics recirculation, see Fig. 4b. The mass flow rate of the recirculated organic stream is also influenced by the dry content of the recycled alkali stream and the temperature of the mixed organics stream (Tmix_2, see Fig. 5). In the basecase evaporator operation the live steam demand is calculated to be around 6.75 kg/s and is represented in Fig. 5 by the broken line for comparison purpose. It should be noted that the basecase pressure level of the evaporator effects are kept constant during the analysis and no attempt has been made to optimize the
system in anyway. In addition, the BL coming from the digester (BL, thin at 14.4% DM) has to be mixed with a recirculated BL from effect 2 (E-2) to achieve a target DM of 21% before it enters effect 4 (E-4). This is done in order to avoid foaming inside the effects (Olsson, 2009). The target dry content (depicted as DMmix_1 in Fig. 4a) is kept as a technical constraint in the new configuration as well.

The results presented in Fig. 5 show the live steam consumption of the MEEV and the organics recirculation mass flow rate dependence on the dry content of the recycled alkali stream and the temperature of the mixed organics stream (depicted as Tmix_2 in Fig. 4b). The dry content of the recycled alkali stream is directly related to the final quality of the lignin; the cleaner the lignin the more washing it requires, resulting in a more diluted recycled alkali stream. In this case a dry content range between 12% and 23% DM is considered for the recycled alkali stream. Similarly, the evaluations have been made for three mixing temperatures (Tmix_2) of the extracted organics stream and the organics...
which also decreases with increasing $T_{mix_2}$ (Fig. 5).

Evidently, the same holds true for the organics recirculation rate curves.

When the stream enters into an effect with a pressure lower than the lower is the vapor thermal energy needed to concentrate the extracted stream as most of the water content will be vaporized because the amount of water to be evaporated is reduced. On the other hand, the organics recirculation rate decreases with increasing $T_{mix_2}$ because the higher is $T_{mix_2}$ the lower is the vapor thermal energy needed to concentrate the extracted stream as most of the water content will be vaporized when the stream enters into an effect with a pressure lower than its saturation temperature (the saturation temperature of water corresponding to 0.91 bar is 97°C). See Fig. 5 the blue curves. Evidently, the same holds true for the organics recirculation rate which also decreases with increasing $T_{mix_2}$ (Fig. 5 the red curves).

It is obvious that with increasing dry content of the recycled alkali stream the live steam consumption of the MEEV decreases because the amount of water to be evaporated is reduced. On the other hand, the organics recirculation rate decreases with increasing dry content of the recycled alkali stream. This is because at higher dry content of the recycled alkali stream the water content of the BL stream entering the MEEV is reduced which in turn lowers the amount of the vapor generated in the evaporation units. And according to the configuration of the MEEV shown in Fig. 4a and b, the vapor generated in the evaporation units is subsequently used as a heat source in the effect with the next lower pressure level. So if the amount of the vapor generated in the effects is reduced its thermal energy content will not be enough to achieve the target DM of 6–3 (i.e. 30% DM). Consequently more amount of organics recirculation is required to compensate the deficit, see Fig. 5 the red curves.

The MEEV is evaluated for two DM compositions of the recycled alkali stream (60% and 70% DM) to investigate its impact on the mass flowrate of the stream. The results are presented in Fig. 5 (60% DM (a) and 70% DM (b)). Shifting the dry content from 60% to 70% DM has resulted in about 14% reduction on the mass flow rate of the organics recirculation.

The other subsection of the pulp mill which is affected by the extraction organics is the RB. All the thermal energy output of the RB comes from the combustion of the organic part of the BL. Therefore, altering the organic composition of the BL would have a direct consequence on the thermal energy output of the RB. The useful thermal energy output is found to be in the range of 53–30 MWh for an organics extraction in range of 0–30%. However, the fuel balance of the RB puts a limitation on the amount of organics that can be extracted for the integrated process. A 10% extraction of the organics in the woodchips, which is the case in this work, has resulted in about 15% reduction in thermal energy output of the RB. The thermal energy output refers to the useful energy left for steam generation after the subtraction of the energy requirement of the endothermic reduction reaction (the reduction of Na$_2$SO$_4$ to Na$_2$S) that takes place inside the RB. It should also be noted that, in all the cases, the BL is concentrated to 71% DM and fed to the RB at a temperature of 129°C. Moreover, the excess combustion air is kept constant at 5% during the assessment.

3.8. Impact of the extraction of organics on the hot utility requirement of the Kraft pulp and paper mill

The impact on the hot and cold utility demand of the Kraft pulp and paper mill due to the integration of the conceptual organics extraction and ABE processes has been investigated using Pinch Technology. Two integration boundaries are considered:

- the integration of the organic extraction process for producing high grade lignin with the Kraft pulp and paper mill subprocesses (in this case the BBLH is considered as final product);
- the integration of the organic extraction and the conventional ABE process with the Kraft pulp and paper mill subprocesses (in this case the BBLH is used as a feed stream to the ABE process).

As it has been emphasized in the previous subsections, the organics extraction process impacts mainly the operation of the multiple-effect evaporators in relation to its hot utility demand. Depending on the mixed stream temperature ($T_{mix_2}$), the organics recirculation stream and that of the recycled alkali stream DM contents, the live steam demand of the multiple-effect evaporators can be higher or lower than the basecase live steam demand (see Fig. 5). Hence, the hot utility demand of the two extreme cases in which the two evaluated cases refer to is summarized in Table 4. Case I represents the scenario that resulted in highest live steam consumption and case II represents the scenario with the lowest live steam consumption.

Fig. 5. The live steam consumption of the evaporation unit and the mass flow rate of the organics recirculation versus the dry content of the recycled alkali stream for different temperatures of the mixed organics stream ($T_{mix_2}$). Note the ordinate axes scale difference. (a) Organics recirculation at 60% DM and (b) organics recirculation at 70% DM.
The details of the Pinch Analysis of the Kraft pulp and paper mill basecase process is reported in (Mesfun and Toffolo, 2013) and is adopted in this work without further explanation. Results showed that the hot utility demand of case I have increased by about 8% as compared to the basecase and that of case II has shown a marginal reduction (about 2%) compared to the basecase.

Expansion of the integration boundary to include the ABE process has resulted in 18% and 8% increase in hot utility demand for cases I and II, respectively, as compared to the basecase (Fig. 6b). The increase in hot utility demand is mainly due to the flash evaporators and re-boilers of the distillation columns which, according to the Aspen Plus model, take place in the temperature range of 85–105 °C. The profile of the GCCs of the new cases in Fig. 6b confirms this observation. Moreover, the changes in cold utility requirement due to the integration of the new processes remain marginal (Fig. 6).

4. Conclusions

The ABE fermentation yield from BBLH needs to be improved to levels reported for pentose substrates in order the process to be economical feasible. The configuration changes introduced in the MEEV played an important role in keeping the expected change in energy balance of the unit within marginal levels, especially when only the lignin separation plant is considered (Fig. 6a). Besides, dedicating one of the MEEV units to the new process means reduced capital investment. Integrating only the lignin separation and BBLH production plant presents an attractive solution towards increased pulp production and increased revenues.

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References


Integrated SNG production in a typical Nordic sawmill
Article

Integrated SNG Production in a Typical Nordic Sawmill

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Abstract: Advanced biomass-based motor fuels and chemicals are becoming increasingly important to replace fossil energy sources within the coming decades. It is likely that the new biorefineries will evolve mainly from existing forest industry sites, as they already have the required biomass handling infrastructure in place. The main objective of this work is to assess the potential for increasing the profit margin from sawmill byproducts by integrating innovative downstream processes. The focus is on the techno-economic evaluation of an integrated site for biomass-based synthetic natural gas (bio-SNG) production. The option of using the syngas in a biomass-integrated gasification combined cycle (b-IGCC) for the production of electricity (instead of SNG) is also considered for comparison. The process flowsheets that are used to analyze the energy and material balances are modelled in MATLAB and Simulink. A mathematical process integration model of a typical Nordic sawmill is used to analyze the effects on the energy flows in the overall site, as well as to evaluate the site economics. Different plant sizes have been considered in order to assess the economy-of-scale effect. The technical data required as input are collected from the literature and, in some cases, from experiments. The investment cost is evaluated on the basis of conducted studies, third party supplier budget quotations and in-house database information. This paper presents complete material and energy balances of the considered processes and the resulting process economics. Results show that in order for the integrated SNG production to be favored, depending on the sawmill size, a biofuel subsidy in the order of 28–52 €/MWh SNG is required.

Keywords: sawmill; bio-SNG; b-IGCC; process integration; HEATSEP method; techno-economic evaluation

1. Introduction

Sweden has a large sawmill industry with an annual production capacity of about 16.1 million cubic meters of sawn wood in 2013 [1]. The corresponding amount of biomass by-products, in the form of woodchips, bark and sawdust, is about the same in terms of dry mass [2]. Biomass utilization has been gaining interest as a means to achieve the targets related to the reduction of greenhouse gas emissions and to the increase of the share of renewables in the energy mix. Woody-biomass is one of the important renewable energy sources (RES) that are globally expected to play a major role in mitigating the dependence of modern society on fossil-based resources for household, industry and transportation energy use. There are, however, several problems associated with the different forms of RES, such as the uneven distribution of the resources around the globe, the insufficient availability of the resources (as long as the objective is to fulfill a significant share of the global primary energy demand according to the best conversion technology known today) and the difficulties with storing them (in the case of intermittent resources, such as solar and wind) [3].
It is essential that biomass can be converted to energy products that correspond to the standards of existing fossil-based energy carriers, such as electricity, chemicals, gas and transportation fuels. The merits in doing so include the alleviation of the burden on the environment from greenhouse gas emissions (bioenergy is considered as CO2 neutral) and utilization of the existing infrastructure (trade and supply network of the long-established products) and applications [3–8].

A typical case is the production of biomass-based substitute/synthetic natural gas (bio-SNG) from lignocellulosic biomass. The bio-SNG process has been gaining interest as a promising downstream technology via thermal gasification of biomass. Several plausible alternative processes are reported in the literature; see, e.g., [3,6,9–13]. The production of SNG from biomass is generally considered to be made of three main process stages: gasification of the lignocellulosic biomass into raw syngas (mainly composed of H2, CO, CO2 and CH4 on a dry basis), methanation (catalytic conversion of the syngas into crude SNG) and upgrading of the crude SNG to the gas pipeline specifications (which implies CO2 and H2O removal). Others studies (see, e.g., [14–17]) also focus on the production of SNG from various renewable energy resources, such as wind and solar via electrolysis of water to produce hydrogen, which afterwards is used in hydrogenation of CO2 to produce SNG.

In this work, the potential for upgrading the by-products of a typical Nordic sawmill into bio-SNG is investigated from a techno-economic point of view. The main technologies considered for the bio-SNG production are a pressurized bubbling fluidized bed reactor (see, e.g., [18]) for the thermal gasification of biomass, the Imperial Chemical Industry (ICI) process [6] for methanation and amine-based CO2 separation (see, e.g., [19]) for bio-SNG upgrade. In addition, the investigation includes the option of utilizing the product gas in a biomass-integrated gasification combined cycle (b-IGCC) in order to produce heat and electricity. In turn, the b-IGCC configuration has two alternative options: supplying low temperature thermal energy to a district heat (DH) system or condensing the steam below atmospheric pressure in order to maximize the electrical output.

The paper is organized as follows. Section 2 presents a description of the considered processes. The methodologies used for process integration, as well as for energetic and economic evaluations are presented in Section 3. Section 4 summarizes the results and includes a discussion about the most significant findings.

2. Process Description

2.1. Sawmill

The sawmill is a process industry that converts timber (the round beam section of a tree, without branches and tops) into lumber (wooden board used in wood industries and construction). In so doing, sawmills produce a large quantity of by-products (in the form of woodchips, sawdust and bark) that can be utilized by innovative bioenergy technologies.

The model of a typical Nordic sawmill is taken from [2]. Accordingly, about 47% of the timber input is converted into lumber (the main product) on a dry mass basis. The remaining 53% is a by-product with respect to the sawmill process. This by-product, the biomass fuel mix (BFM), is composed of woodchips (about 26% of the timber input on a dry mass basis), bark (19%) and sawdust (8%). If the sawmill is not integrated with other processes/plants that can supply heat, an amount of BFM corresponding to 10.3% of the timber input on a dry mass basis has to be consumed internally in a dedicated furnace mainly to satisfy the lumber drying process [2]. In fact, timber is initially supplied at 55%–60% moisture content, while lumber has a final moisture content of 18%. Therefore, the lumber drying process is quite energy intensive and is carried out by circulating heated outdoor air through packages of lumber that are placed in dedicated rooms called drying kilns. The outdoor air, initially at 2 °C, is heated to about 75 °C and is continuously supplied to the drying kilns to replace the humidified air inside the kilns.
2.2. SNG Process

The SNG process consists of a biomass dryer, an air separation unit (ASU), a bubbling fluidized bed gasifier, gas cleaning units, SNG synthesis units and a CO₂ separation unit; see Figure 1. Oxygen and steam gasification is chosen because it already produces a small amount of methane, and a detailed description of the process model is reported in [20]. The biomass fuel, initially at 55%–60% moisture content, is dried to a final moisture content of about 10% using hot air as the drying medium in a belt conveyor dryer. The drying air and the dried biomass exit the dryer at the same final temperature of about 110 °C. In the gasifier, a mixture of steam and O₂ is used as a gasifying agent. The flow rate of steam is kept constant at the steam to biomass ratio of 0.5 kg/kg-biomass, while the flow rate of O₂ is adjusted to keep the gasification temperature at 850 °C. The ICI process [6] is chosen for the SNG synthesis units to achieve a high conversion to methane. The ICI process consists of three fixed bed catalytic exothermic reactors in series (adiabatic equilibrium reactors), and methanation is assumed to proceed according to reactions R1 and R2.

\[
\begin{align*}
\text{CO}_2 + 4\text{H}_2 & \rightarrow \text{CH}_4 + 2\text{H}_2\text{O} \quad \Delta H^{\ominus}_{\text{GSSK}} = -165 \text{kJ/mol} & (\text{R1}) \\
\text{CO} + 3\text{H}_2 & \rightarrow \text{CH}_4 + \text{H}_2\text{O} \quad \Delta H^{\ominus}_{\text{GSSK}} = -206 \text{kJ/mol} & (\text{R2})
\end{align*}
\]

![Figure 1. Bio-SNG production process flowsheet (circles with H and C represent hot and cold thermal streams, respectively).](image)

The reactors are fitted with intercooling heat exchangers in between in order to compensate the heat from exothermic reactions. In this work, amine-based CO₂ separation is assumed for gas upgrading, as it is a proven technology that is commercially available and used in many industries. The overall SNG process includes also a boiler in which part of the biomass is burnt in order to supply the heat demand of the SNG process, as well as of the sawmill.

2.3. b-IGCC Process

The considered b-IGCC process assumes similar pretreatment, gasification and gas cleaning technologies as in the SNG process except that air is used as a gasifying medium instead of a mixture of oxygen and steam. In this case, the cleaned product gas is first utilized in a stationary gas turbine
engine to generate electricity, then the exhaust gas from the gas turbine is sent to an heat recovery steam generator (HRSG) to generate high pressure steam that is expanded in a steam turbine (backpressure or condensing, depending on the bottom cycle configuration of the b-IGCC) to produce additional electricity. In the alternative with DH, a backpressure steam turbine is used that expands the steam down to 1.9 bar (a pressure level that is suitable to satisfy the sawmill heat demand). The heat derived from steam condensation is first used to satisfy the sawmill thermal deficit, and then, the rest is supplied to the DH system. In the alternative without DH, the steam that is not used to satisfy the thermal demand of the sawmill is expanded down to 0.05 bar in a condensing turbine to generate additional power instead of supplying heat to a DH system.

3. Methodology

Process integration techniques are used to evaluate the profitability of integrating a typical Nordic sawmill with the production of biomass gasification-based bio-SNG and/or power under different process configurations and sawmill sizes. The mass and energy balances of the processes are obtained from plant models developed in MATLAB and Simulink. In all of the cases, the amount of available BFM is limited to the amount of by-products from the considered sawmill size. The mass and energy balances of each process configuration are further constrained by the fulfillment of the heat demand of the sawmill.

Under these conditions, the following three process configurations have been considered:

- Integration of the bio-SNG production process described in Section 2;
- Integration of a b-IGCC system that produces electricity and heat for a DH system;
- Integration of a b-IGCC system with the sole purpose of producing electricity.

3.1. Process Integration

The processes considered in this work involve hot thermal streams at different temperature levels. This surplus of thermal energy can be used to supply the heat demand of the sawmill, the heat and steam demand of the bio-SNG process and, potentially, to cogenerate electricity in a steam Rankine cycle. Accordingly, the HEATSEP method [21] is applied to the flowsheets of the processes (including the sawmill and the steam Rankine cycle) in order to assess the synergy among them. The HEATSEP method allows one to focus on the basic process units of a system configuration by replacing the potential heat transfer devices with the so-called “thermal cuts”. The thermal streams (hot and cold) across the cuts are then grouped into a “black box” where heat exchange interactions are assumed to take place without predefined stream matches. The temperatures at the boundaries of the thermal cuts and the mass flow rates of the streams are included among the decision variables during the search for the optimal configuration and design parameters of the different processes (see, e.g., [22], in which the objective function is to maximize the net power output of a steam Rankine cycle that is shared among different processes in a pulp and paper mill). The feasibility of the heat transfer inside the “black box” is a constraint of the optimization problem and is verified according to the problem table algorithm [23].

In this work, the temperatures (and several of the mass flow rates) across the thermal cuts in the bio-SNG process, in the b-IGCC process and in the sawmill are fixed, so they are not considered among the decision variables of the optimization problem. The fixed values for these quantities are subject to change according to the capacity of the sawmill, mainly because the efficiencies of the components improve when the size is increased. As a result, the corresponding hot and cold thermal loads are not exactly proportional to the capacity of the sawmill, although the configurations of the system remain the same as the sizes are varied. The design parameters of the steam Rankine cycle are therefore those that have to be optimally tuned in order to generate the maximum power from the exploitation of the heat pockets of the grand composite curve of the other processes in the integrated industrial site. These optimal parameters will then depend on the system configuration and on the size of the sawmill.
The bio-SNG process requires steam at 15 and 30 bars during the gasification and methanation steps, respectively. In the process configuration with SNG production only, the surplus heat from the high temperature streams of the bio-SNG process is not enough to generate the amount of steam required, so a boiler that burns a fraction of the available BFM is included in the flowsheet in order to satisfy the deficit. This imposes an additional constraint on the amount of BFM retained for bio-SNG production. The steam cycle is designed to generate steam at pressures that are high enough to meet the bio-SNG process steam demand while cogenerating electricity in a steam turbine. The heat demand of the sawmill is also satisfied using condensing steam at 1.9 bar. It should also be noted that an amount of feed water equal to the amount of steam consumed by the gasification and methanation processes must be continuously supplied to the steam Rankine cycle. As a result, the decision variables are the maximum cycle pressure and temperature of the steam Rankine cycle, the mass flow rate of steam that remains in the steam cycle and the split fraction of the BFM that goes to the boiler (this last one, however, is heavily affected by constraints on the grand composite curve in the middle and high temperature ranges).

On the contrary, the b-IGCC process configurations involve high temperature hot streams resulting in a surplus heat that is larger than the demands of the b-IGCC process and of the sawmill. In this case, the steam Rankine cycle is mainly designed to exploit the excess heat for cogenerating electricity and to add flexibility for the heat transfer interactions among the different processes. In fact, the design of the steam Rankine cycle helps with minimizing the exergy destruction that would have occurred if the high temperature hot streams were used to supply directly low temperature heat demands, such as that of the sawmill. The decision variables in both of the alternative b-IGCC process configurations are the maximum cycle pressure and temperature of the steam Rankine cycle and the mass flow rate of the water/steam that remains in the combined heat and power (CHP) system. The only difference in the settings of the optimization problem between the two process configurations is that an additional constraint is introduced in the alternative without the DH system, so that the amount of steam condensed at 1.9 bar is exactly the one required to satisfy the sawmill heat demand.

3.2. Thermodynamic Indicators

The energy indicators of the SNG and b-IGCC process configurations are evaluated according to Equations (1)–(4). The system boundaries and energy flows used to define the indicators are illustrated in Figure 1. The indicator in Equation (1) compares the energy associated with the BFM input to the gasifier to the chemical energy of the final bio-SNG output, and it is often referred to as the cold gas efficiency. It is worth noting that the denominator of this indicator accounts only for the biomass by-products entering the gasifier. Equation (2) represents the overall energy efficiency of the bio-SNG process by accounting for all of the major flows crossing system boundaries; see Figure 2. Equations (3) and (4) measure the overall energy efficiency of the b-IGCC process with and without the DH option, respectively.

\[
\eta_{\text{cg}} = \frac{m_{\text{SNG}} \cdot \text{LHV}_{\text{SNG}}}{m_{\text{BFM to gasifier}} \cdot \text{LHV}_{\text{BFM}}} \quad (1)
\]

\[
\eta_{\text{en, SNG}} = \frac{m_{\text{SNG}} \cdot \text{LHV}_{\text{SNG}} + Q_{\text{b, sawmill}} + W_{\text{el, net}}}{m_{\text{BFM}} \cdot \text{LHV}_{\text{BFM}}} \quad (2)
\]

\[
\eta_{\text{en, b-IGCC+DH}} = \frac{W_{\text{el, net}} + Q_{\text{b, sawmill}} + Q_{\text{DH}}}{m_{\text{BFM}} \cdot \text{LHV}_{\text{BFM}}} \quad (3)
\]

\[
\eta_{\text{en, b-IGCC}} = \frac{W_{\text{el, net}} + Q_{\text{b, sawmill}}}{m_{\text{BFM}} \cdot \text{LHV}_{\text{BFM}}} \quad (4)
\]
The three process configurations are also evaluated according to their overall exergy efficiencies, using Equations (5)–(7). In the b-IGCC process alternative with the DH system, it is assumed that the thermal energy supplied to the DH system could be converted into electricity with a thermal efficiency equal to 10%.

\[
\eta_{\text{ex, SNG}} = \frac{m_{\text{SNG}} \cdot \epsilon_{\text{SNG}} + W_{\text{el, net}}}{m_{\text{BMF}} \cdot \epsilon_{\text{BMF}}} \quad (5)
\]

\[
\eta_{\text{ex, b-IGCC+DH}} = \frac{W_{\text{el, net}} + \eta_{\text{ex, DH--el}} \cdot Q_{\text{DH}}}{m_{\text{BMF}} \cdot \epsilon_{\text{BMF}}} \quad (6)
\]

\[
\eta_{\text{ex, b-IGCC}} = \frac{W_{\text{el, net}}}{m_{\text{BMF}} \cdot \epsilon_{\text{BMF}}} \quad (7)
\]
3.3. Economic Indicators

The economic evaluation is performed by taking into account the operating costs and the investment costs for the equipment that is involved in the flowsheet of the processes. The capital investment is estimated by evaluating the cost of each component in the major process equipment from literature data according to its size. All cost values are updated to the Euro value of 2013. Commodity prices are those of the last quarter of 2013, and exchange rates used for currency conversions (i.e., 8.86 SEK/€ and 6.43 SEK/$) are those of the same period. Two approaches have been adopted for estimating the total capital investment.

The cost associated with the process units that have been designed in detail in this study (such as the heat exchanger network (HEN), turbomachinery, steam cycle components and methanation reactors) is estimated according to [24]. The installed cost ($C_{in}$) of process equipment can be correlated to the bare module cost by Equation (8):

$$C_{in,i} = (1 + f_1) \cdot C_{BM,i} + f_2 \cdot C_{0BM,i}$$  

(8)

where $C_{BM,i}$ and $C_{0BM,i}$ represent the bare module cost of equipment $i$ evaluated in the actual process operating conditions and in the base case conditions (i.e., ambient pressure and carbon steel construction), respectively. Coefficient $f_1$ accounts for contingency and fee costs depending on the reliability of cost data and completeness of the process flowsheet. A value of 18% is used (15% and 3% of the bare module cost in the actual operating conditions for contingency and fee costs, respectively [24]). Coefficient $f_2$ accounts for the costs related to site development, auxiliary building and utilities. A value of 50% of the bare module cost in the base case conditions is assumed [24]. Further information on the relationships between the equipment bare module cost and purchase cost can be found in Appendix A.

The capital cost of the HEN is estimated from the mass flow rates and the temperatures of the thermal streams in the black box of the HEATSEP method, as they result from the optimization of the process configurations. The loads of the heat exchangers are determined by matching the streams according to the hot and cold composite curves. The area of heat exchangers and then the cost are determined by applying appropriate heat transfer coefficients (depending on the nature of the streams) and minimum temperature differences ($\Delta T_{min}$) that would result in a reasonable heat transfer area for any match among the streams.

The cost associated with the remaining major process units (such as biomass pretreatment, gasification, gas cleaning and bio-SNG upgrading) are estimated by scaling published data using the power law of capacity. The reference costs and sizes used for estimating the purchase cost of these process units are documented in Table A1. The actual equipment purchase cost of a single component is first estimated using the power law of capacity by scaling it from a similar component with known cost and size according to a scaling exponent. The equipment purchase cost is then multiplied by an overall installation factor (also presented in Table A1) to obtain the installed cost of the process unit. In this case, the installed equipment cost is related to the purchase cost according to Equation (9):

$$C_{in,j} = f_j C_{E,j}$$  

(9)

where $f_j$ and $C_{E,j}$ represent the overall installation factor and equipment purchase cost, respectively. The capital investment (CI) is then calculated using Equation (10) as the sum of Equations (8) and (9). The bare module equipment costs are estimated based on sizing parameters and cost factors reported in Table A2.

$$CI = 1.18 \sum_{i=1}^{n} C_{BM,i} + 0.5 \sum_{i=1}^{n} C_{0BM,i} + \sum_{j=1}^{m} f_j C_{E,j}$$  

(10)
The internal rate of return (IRR) on the investment is evaluated as the economic indicator for the considered cases. Table 1 presents a list of the market prices and the other economic parameters that have been assumed.

**Table 1. Market price and other economic parameters used in the evaluation of the IRR.**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Investment interest rate</td>
<td>%</td>
<td>4</td>
</tr>
<tr>
<td>Economic life time</td>
<td>Year</td>
<td>20</td>
</tr>
<tr>
<td>Annual operational time (top)</td>
<td>Hour</td>
<td>8000</td>
</tr>
<tr>
<td>Operators (^a)</td>
<td>p, (^b/)shift</td>
<td>6</td>
</tr>
<tr>
<td>Operator salary (^c)</td>
<td>€/(p. year)</td>
<td>55,000</td>
</tr>
<tr>
<td>Maintenance cost (C(_{cm}))</td>
<td>% of CI</td>
<td>5</td>
</tr>
<tr>
<td>BFM price (C(_{BFM})) (^c)</td>
<td>€/MWh(_{LHV})</td>
<td>22</td>
</tr>
<tr>
<td>Electricity (C(_{el})) (including CO(_2) charge) (^c)</td>
<td>€/MWh(_{el})</td>
<td>68</td>
</tr>
<tr>
<td>Natural gas (including CO(_2) charge) (^c)</td>
<td>€/MWh(_{LHV})</td>
<td>48</td>
</tr>
<tr>
<td>District heating thermal energy (^c)</td>
<td>€/MWh(_{th})</td>
<td>35</td>
</tr>
<tr>
<td>CO(_2) charge (input) (^c)</td>
<td>€/tCO(_2)</td>
<td>36</td>
</tr>
<tr>
<td>Renewable electricity policy support (^c)</td>
<td>€/MWh(_{el})</td>
<td>7</td>
</tr>
<tr>
<td>Tax on profit</td>
<td>%</td>
<td>22</td>
</tr>
</tbody>
</table>

\(^a\) Full time operation requires 4 shifts per day; \(^b\) personnel (p.) for the largest sawmill size; for the other sizes, an exponent equal to 0.7 is used to scale down operator salary costs; \(^c\) price levels represent the Swedish market in 2013 [25].

4. Results and Discussion

4.1. Integrated Grand Composite Curves of the Three Configurations

Figures 3 and 4 present the integrated grand composite curves of the three process configurations for the largest sawmill size. The distinctive features of the bio-SNG configuration (Figure 3, red curve) are the radiative heat from the biomass boiler (the horizontal segment at 850 °C) and the demand for process steam generation at about 310 °C (which is the same as the temperature level of the upper horizontal segment of the blue curve). In order to maximize the BFM input to the bio-SNG process, the amount of steam generated in the steam cycle is just enough to satisfy the heat deficit and steam demand of the integrated industrial site (sawmill plus bio-SNG process); see Section 3.1 for further details.

![Figure 3. Integrated grand composite curves of the bio-SNG configuration for the largest sawmill size.](image-url)
The heat pockets in the red curves of the b-IGCC configuration (Figure 4) are mainly a result of product gas cooling (from 850 °C), the recovery of heat from the exhaust gases of the gas turbine (from 550 °C) and the thermal demand of the sawmill (at 75 °C). In both cases, the steam Rankine cycle exploits these pockets for generating steam (the three upper segments of the blue curves) and satisfies the heat demands of the sawmill (and that of the DH system in Figure 4a) in the lower temperature range using condensing steam (the lower horizontal segment of the blue curves). The blue curve in Figure 4b also shows a further steam condensation level that follows the additional expansion in the condensing turbine. A detailed discussion regarding b-IGCC in the context of integrated forest industries can be found in [26].

4.2. Energetic Perspective

The magnitudes of the energy streams of the bio-SNG process configuration, defined according to the system boundaries shown in Figure 2, are summarized in Table 2 for all of the considered sawmill sizes. In order to reflect on the thermodynamic performance of the bio-SNG process, a generic evaluation of the process flowsheet is presented in Table B1 (description of streams) and Table B2 (thermodynamic state and compositions of streams).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Sawmill Sizes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Annual lumber production</td>
<td>10^9 m^3</td>
<td>400</td>
</tr>
<tr>
<td>BFM available</td>
<td>MWHV</td>
<td>86.85</td>
</tr>
<tr>
<td>BFM to biomass boiler</td>
<td>%</td>
<td>42.99</td>
</tr>
<tr>
<td>SNG production</td>
<td>MWHV</td>
<td>4.48</td>
</tr>
<tr>
<td>Sawmill thermal load</td>
<td>MWth</td>
<td>11.95</td>
</tr>
<tr>
<td>Cold gas efficiency (Equation (1))</td>
<td>%</td>
<td>64.55</td>
</tr>
<tr>
<td>Wobbe index</td>
<td>MJ/Nm^3</td>
<td>44.48</td>
</tr>
<tr>
<td>Power generation</td>
<td>MWd</td>
<td>1.34</td>
</tr>
<tr>
<td>Power consumption</td>
<td>MWd</td>
<td>3.15</td>
</tr>
<tr>
<td>Net power export</td>
<td>MWd</td>
<td>6.88</td>
</tr>
<tr>
<td>Energy efficiency (Equation (2))</td>
<td>%</td>
<td>56.17</td>
</tr>
<tr>
<td>Exergy efficiency (Equation (5))</td>
<td>%</td>
<td>42.40</td>
</tr>
</tbody>
</table>

The magnitude of the energy output streams of the b-IGCC process configurations are presented in Figure 5 for all of the considered sawmill sizes and for both configuration alternatives (with and
without DH systems). It appears that the power production in the b-IGCC configuration without DH is higher than that in the other configuration. This is expected since the steam that is condensed to supply heat to the DH in one alternative is expanded in a condensing steam turbine to generate additional power in the other. In both b-IGCC configuration alternatives, the heat provided to satisfy the sawmill demand is represented as a negative contribution to indicate that it is a constraint that must be satisfied.

![Figure 5](image_url)

**Figure 5.** Energy streams (electricity (El.) from gas turbine (GT) and steam turbine (ST), heat to the DH system, heat to the sawmill) of the b-IGCC process configurations as a function of sawmill size.

The overall energy efficiencies of the considered process configurations (defined by Equations (2)–(4)) are compared in Figure 6a as a function of sawmill size. It appears that the b-IGCC configuration with the DH system results in the highest overall energy efficiency (62%–66%, increasing with sawmill size), followed by the bio-SNG process configuration (about 55%–56% for all of the sawmill sizes considered). The b-IGCC configuration without the DH system has always the lowest overall efficiency (from 43%–55%, increasing with sawmill size).

![Figure 6](image_url)

**Figure 6.** Overall energy (a) and exergy (b) efficiencies of the different process configurations.

Figure 6b compares the exergy efficiencies of the considered process configurations, as defined by Equations (5)–(7). It appears that the ranking of the three process configurations is completely changed:
the highest exergy efficiency is that of the bio-SNG configuration (41%–42%, with increasing efficiency as the sawmill size increases), then the b-IGCC configuration without the DH system (30%–41%, increasing with sawmill size) and, finally, the b-IGCC configuration with the DH system (29%–40%, increasing with sawmill size).

It is worth noting that the b-IGCC configuration with the DH system has the highest energy efficiency and the lowest exergy efficiency because of the low temperature heat output, which is a big amount in terms of energy, but not significant in terms of exergy (the exergy of the electric output from the condensing turbine is higher).

4.3. Economic Perspective

Figure 7 shows how the capital cost shares of the different sections in the bio-SNG process configuration change as the size of the sawmill is varied. For a sawmill annual lumber production greater than 71,000 m³, the investment is largely dominated by the cost of the gasification process (23%–35%, with an increasing share as the sawmill size increases). For sawmill annual lumber production capacities lower than 71,000 m³, the share associated with the HEN dominates the capital cost (10%–30%, with an increasing share as the sawmill size decreases). The shares of the other sections are generally lower: gas conditioning and upgrade 13%–18% each, with an increasing share as sawmill size increases; methanation 3%–9%, with an increasing share as sawmill size increases; steam section and pretreatment, about 5% each.

Bio-SNG and electricity selling prices that would allow one to obtain the imposed IRR values of 4%, 8% and 12% are calculated backwards. Thermal energy for district heating in the b-IGCC + DH case and electricity in the bio-SNG case are considered as by-products and sold at the current market price. The results are presented in Figure 8. The ranges for the market price of natural gas required to meet the imposed IRR values are 77–123 €/MWh (4%), 87–146 €/MWh (8%) and 99–171 €/MWh (12%), the lower values being of course obtained with the larger sawmills. A policy supporting biofuels is therefore required to make the bio-SNG process configuration profitable, but it also appears that in any case, a subsidy would not be reasonable for the sawmill sizes below 106,000 m³ of lumber per year due to the economy-of-scale effects. For example, biofuel subsidy certificates in the range of 28–52 €/MWh (decreasing as sawmill size increases) are required in order to break-even (4% IRR) for the bio-SNG processes integrated with a sawmill having an annual lumber production greater than
106,000 m³. The subsidy levels estimated here are generally higher compared to previous studies. In [27,28], 24–42 €/MWh was reported for similar processes, but with considerably larger biomass input (150–400 MW compared to 20–125 MW in this work). Of course, this difference can be explained by the different economy-of-scale, which plays a role in reducing the levels of required subsidy.

![Figure 8. Market price of NG/electricity in order to meet the imposed IRR values.](image)

Should future policy measures allow the biogenic CO₂ separated during gas upgrading to be included in the emission trading system, additional income can be obtained in the form of emission certificates. For example, additional income in the range of 4–6 €/MWh of bio-SNG produced (increasing with sawmill size) can be achieved by assuming that the benefit from the biogenic CO₂ sequestration certificates is equal to the penalty for fossil CO₂ emission charges and by deducting the CO₂ transportation (13 €/tCO₂ [29]) and storage (9 €/tCO₂ [29]) expenses.

In the b-IGCC configuration alternative with DH, the ranges for the electricity market price required to meet the imposed IRR values are 74–191 €/MWh (4%), 85–234 €/MWh (8%) and 98–282 €/MWh (12%). In the alternative without DH, they become 82–197 €/MWh (4%), 92–235 €/MWh (8%) and 104–278 €/MWh (12%). Apparently, the b-IGCC alternative with DH results in lower values of market prices for electricity compared to the alternative without DH. This is because the amount of heat provided to the DH system is relatively large compared to the size of the sawmill. For example, about 0.13 TWh would be provided annually to a DH system with the largest sawmill size. However, it could be a problem, if not impossible, to find a market for this waste heat if the location of sawmills were inconvenient for DH consumers and in the case of large seasonal fluctuations of the DH demand.

5. Conclusions

The energetic and economic perspectives were investigated regarding the integration of different biomass gasification-based technologies for upgrading sawmill by-products on-site. The main focus was on whether, or under what conditions, the investment in a plant producing bio-SNG from sawmill by-products is economically viable. The option of utilizing sawmill by-products in a b-IGCC
configuration to produce electricity (and heat for a DH system in one alternative) was also explored for comparison.

With a limited availability of relatively low cost BFM, it is essential to choose between the production of bio-SNG and that of electricity/heat. If the production of bio-SNG is prioritized, perhaps in order to support the utilization of biofuels in the transport sector, large-scale plants (i.e., sawmills having an annual lumber production greater than 106,000 m³) with ambitious biofuel subsidy requirements (28–52 €/MWh) are to be favored. In a less ambitious biofuel subsidy scenario, the b-IGCC configuration with the DH system is more attractive compared to the alternative without the DH system, although both cases resulted in a cost for electricity production that is higher than its market value.

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Conflicts of Interest: The authors declare no conflict of interest.

Abbreviations

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Equipment sizing parameter</td>
</tr>
<tr>
<td>BFM</td>
<td>Biomass fuel mix</td>
</tr>
<tr>
<td>b-IGCC</td>
<td>Biomass integrated gasification combined cycle</td>
</tr>
<tr>
<td>bio-SNG</td>
<td>Biomass-based synthetic natural gas</td>
</tr>
<tr>
<td>CEPCI</td>
<td>Chemical engineering plant cost index</td>
</tr>
<tr>
<td>CI</td>
<td>Capital investment</td>
</tr>
<tr>
<td>D</td>
<td>Reactor vessel diameter</td>
</tr>
<tr>
<td>DH</td>
<td>District heat</td>
</tr>
<tr>
<td>HEN</td>
<td>Heat exchanger network</td>
</tr>
<tr>
<td>IRR</td>
<td>Internal rate of return</td>
</tr>
<tr>
<td>LHV</td>
<td>Lower heating value</td>
</tr>
<tr>
<td>P</td>
<td>Operating pressure (bar)</td>
</tr>
<tr>
<td>RES</td>
<td>Renewable energy sources</td>
</tr>
<tr>
<td>SNG</td>
<td>Synthetic/substitute natural gas</td>
</tr>
</tbody>
</table>

Sets

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>i</td>
<td>Set of equipment for which cost estimates are made based on the module costing method.</td>
</tr>
<tr>
<td>j</td>
<td>Set of equipment for which cost estimates are recalculated based on the published cost data or in-house database.</td>
</tr>
</tbody>
</table>

Parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>C_{BM,i}</td>
<td>Bare module cost of equipment i calculated at the actual process conditions</td>
</tr>
<tr>
<td>C_{BM,j}</td>
<td>Bare module cost of equipment i calculated at the base rating</td>
</tr>
<tr>
<td>C_{E,j}</td>
<td>Cost of equipment j recalculated from the data in Table A1</td>
</tr>
<tr>
<td>C_{p,i}</td>
<td>Purchase cost of equipment i calculated according to the module costing method at the base rating</td>
</tr>
<tr>
<td>f_{BM}</td>
<td>Bare module factor</td>
</tr>
<tr>
<td>f_{i}</td>
<td>Overall installation factor of equipment j</td>
</tr>
<tr>
<td>f_{m}</td>
<td>Equipment construction material factor</td>
</tr>
<tr>
<td>f_{p}</td>
<td>Pressure factor</td>
</tr>
<tr>
<td>ΔT_{min}</td>
<td>Minimum temperature difference</td>
</tr>
</tbody>
</table>

Coefficients

<table>
<thead>
<tr>
<th>Coefficient</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>B</td>
<td>Coefficients for evaluating the bare module factor</td>
</tr>
<tr>
<td>C</td>
<td>Coefficients for evaluating the pressure factor</td>
</tr>
<tr>
<td>K</td>
<td>Coefficients for estimating the equipment cost at the base rating</td>
</tr>
</tbody>
</table>
Appendix A

Equipment Purchase Cost Data

Table A1. Reference size parameter used for estimating purchased equipment cost.

<table>
<thead>
<tr>
<th>Unit</th>
<th>Original Unit Cost *(€2013)</th>
<th>Reference Size Parameter</th>
<th>Installation Factor b f</th>
<th>Scaling Exponent c</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Biomass Handling</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Conveyor</td>
<td>0.36</td>
<td>33.5 ton/h</td>
<td>2</td>
<td>0.8</td>
<td>[5,30]</td>
</tr>
<tr>
<td>Belt conveyor dryer d</td>
<td>2.20</td>
<td>959,000 m³/h (air)</td>
<td>2</td>
<td>0.8</td>
<td>[31]</td>
</tr>
<tr>
<td>Grinding</td>
<td>0.35</td>
<td>33.5 ton/h</td>
<td>2</td>
<td>0.6</td>
<td>[5,30]</td>
</tr>
<tr>
<td>Feeding system</td>
<td>0.43</td>
<td>33.5 ton/h</td>
<td>2</td>
<td>1.0</td>
<td>[5,30]</td>
</tr>
<tr>
<td><strong>Gasification</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Direct gasifier (BFB) e</td>
<td>40.8</td>
<td>358 MWth (LHV)</td>
<td>2</td>
<td>0.72</td>
<td>[30]</td>
</tr>
<tr>
<td>ASU</td>
<td>25.7</td>
<td>576 ton O₂/day</td>
<td>1.5</td>
<td>0.75</td>
<td>[30]</td>
</tr>
<tr>
<td><strong>Gas Cleaning</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hot gas cleaning</td>
<td>31.3</td>
<td>74.1 m³/s gas flow</td>
<td>1.86</td>
<td>1.0</td>
<td>[5,30]</td>
</tr>
<tr>
<td>Fabric filter</td>
<td>0.72</td>
<td>19.6 m³/s gas flow</td>
<td>1.86</td>
<td>0.7</td>
<td>[5,30]</td>
</tr>
<tr>
<td>Water scrubber</td>
<td>2.76</td>
<td>12.1 m³/s gas flow</td>
<td>1.86</td>
<td>0.7</td>
<td>[5,30]</td>
</tr>
<tr>
<td>Acid gas removal unit</td>
<td>6.92</td>
<td>1.31 m³/s gas flow</td>
<td>1.86</td>
<td>0.7</td>
<td>[5,30]</td>
</tr>
<tr>
<td><strong>Guard Bed</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Activated carbon bed</td>
<td>0.025</td>
<td>8 Nm³/s gas flow</td>
<td>3</td>
<td>1.0</td>
<td>[30]</td>
</tr>
<tr>
<td><strong>Methanation (ICl)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Reactor 1 f</td>
<td>Cost of reactor vessels, excluding heat exchangers and catalyst material. Prices are estimated according to the module costing method.</td>
<td></td>
<td></td>
<td></td>
<td>[24]</td>
</tr>
<tr>
<td>Reactor 2 f</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Reactor 3 f</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Catalyst material 4</td>
<td>0.12</td>
<td>1 ton</td>
<td>1.0</td>
<td>0.7</td>
<td>[32]</td>
</tr>
<tr>
<td><strong>SNG Upgrade</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CO₂ removal b</td>
<td>5.55</td>
<td>3.9 Nm³/s gas flow, 46.7 vol % CO₂</td>
<td>3.8</td>
<td>0.7</td>
<td>[29]</td>
</tr>
<tr>
<td><strong>Miscellaneous</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Compressors</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Steam turbine</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gas turbine</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fan/blower</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Heat exchangers</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pump</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Electric generator</td>
<td>1.45</td>
<td>11.8 MWel</td>
<td>1.5</td>
<td>0.94</td>
<td>[33]</td>
</tr>
</tbody>
</table>

* Cost data have been updated to €2013 using the composite Chemical Engineering Plant Cost Index (CEPCI). In case the original unit cost has not been given in €, the currency is first converted to € by using the exchange rate of the year in which the money value is reported and then adjusted to the value of €2013 by applying the CEPCI. * Overall installation factor for equipment f. It includes auxiliary equipment and installation labor, engineering and contingencies. Unless values are given by the literature, the overall installation factor is set to 2 for a 70 MWth scale [5]. * Cost scaling is performed using the power law of capacity. * Original equipment cost is estimated using a wet biomass loading of 30 kg/m² of belt area at a loading rate of 10 kg/s and a drying time (τ) of 2500 s, according to the correlations reported in [31]. It is further assumed that the dryer belt is 10 m wide and that the cover is 6 m high. The scaling exponent is adopted from [30]. * Estimated cost of complete gasifier installation according to the National Renewable Energy Laboratory (NREL) report [34]. The scaling exponent is adopted from [30]. * Original cost calculated from module costing in $2001 [24]. Methanation reactors are considered vertical vessels, the diameter being calculated using a mean superficial gas velocity of 0.14 m/s, with maximum vessel diameter not to exceed 4 m. Vessel height is calculated from a regression of the correlation in [12] for fluidized bed methanation reactors. * Catalyst material cost is calculated according to the correlated data in [32], assuming a space velocity of 4.5 Nm³/kg catalyst/h and using an updated specific catalyst cost of 107 €2013/kg. * The original equipment cost of monoethanolamine (MEA) is estimated based on the €2010 cost estimate of the amine-absorption unit that includes the absorber, stripper, condenser, reboiler, HEXs and pumps. Heyne [29] has studied three different CO₂ separation technologies, and amine-absorption is reported to be the best for the overall bio-SNG process economy. A scaling exponent of 0.7 is used [30].
Table A2. Factors used for module costing [24].

<table>
<thead>
<tr>
<th>Process Units</th>
<th>Sizing parameter</th>
<th>Module Cost Factors</th>
<th>Equipment Cost Factors</th>
<th>Pressure Factors</th>
<th>$f_m$</th>
<th>$f_p$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Methanation</td>
<td></td>
<td>B1  B2 K1 K2 K3 C1 C2 C3</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Reactors 1–3</td>
<td>Gas flow</td>
<td>2.86 1.82 3.4974 0.4485 0.1074</td>
<td></td>
<td>-</td>
<td>-</td>
<td>3.5</td>
</tr>
<tr>
<td>Compressors</td>
<td>Shaft power</td>
<td>- 1 2.2867 1.3604 -0.1027</td>
<td></td>
<td>-</td>
<td>-</td>
<td>2.8</td>
</tr>
<tr>
<td>Steam turbine</td>
<td>Shaft power</td>
<td>- 1 2.6299 1.4308 -0.1776</td>
<td></td>
<td>-</td>
<td>-</td>
<td>3.6</td>
</tr>
<tr>
<td>HEN</td>
<td>Area</td>
<td>1.63 1.66 4.3247 -0.0103 0.1634 0.0388 -0.11272</td>
<td>0.08183</td>
<td>1 Equation (A5)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Steam boiler</td>
<td>Thermal load</td>
<td>- 1 0.9617 -1.48 0.3161 2.594072 -4.23476 1.722404</td>
<td>1 Equation (A5)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The capital investment is calculated according to Equations (A1)–(A6):

$$CI = \left[ 1.18 \sum_{i=1}^{n} C_{BM,i} + 0.5 \sum_{i=1}^{n} C_{BBM,i} \right] + \sum_{j=1}^{m} f_j C_{E,j} \quad \text{(A1)}$$

$$C_{BM,i} = f_{BM,i} C_{PB,i} = C_{PB,i} (B_{1,i} + B_{2,i} f_{m,i} f_{p,i}) \quad \text{(A2)}$$

$$C_{BBM,i} = C_{PB,i} (B_{1,i} + B_{2,i}) \quad \text{(A3)}$$

$$\log_{10} C_{p,i} = K_{1,i} + K_{2,i} \log_{10}(A_i) + K_{3,i} \left[ \log_{10}(A_i) \right]^2 \quad \text{(A4)}$$

$$\log_{10} f_{p,i} = C_{1,i} + C_{2,i} \log_{10}(P_i) + C_{3,i} \left[ \log_{10}(P_i) \right]^2 \quad \text{(A5)}$$

$$f_{p,i} = \frac{\left( \frac{P_i+1)^{10}}{10} \right) + 0.00315}{0.0063} \quad \text{(A6)}$$

Appendix B

Description of the Streams in the Process Flow Diagram
Table B1. Stream numbers (as depicted in Figure 1) and description.

<table>
<thead>
<tr>
<th>Stream Number</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>BFM composition as received, <em>i.e.</em>, 55% moisture content</td>
</tr>
<tr>
<td>2</td>
<td>BFM composition prior to gasification, <em>i.e.</em>, dried to 10% moisture content</td>
</tr>
<tr>
<td>3</td>
<td>Air</td>
</tr>
<tr>
<td>4</td>
<td>Air was converted to oxygen based on the mass balance of oxygen. Compression and actual separation units were omitted, and only the power consumption and “pass rate” was defined; 233 g-O₂/kg-air (100% O₂); 353 kJ-el/kg-air</td>
</tr>
<tr>
<td>5</td>
<td>Compressed to 15 bar at an isentropic efficiency of 74%</td>
</tr>
<tr>
<td>6</td>
<td>Preheated to saturated temperature of steam at 15 bar if the gas temperature is lower than that</td>
</tr>
<tr>
<td>7</td>
<td>Saturated steam at 15 bar</td>
</tr>
<tr>
<td>8</td>
<td>Adiabatic mixing of O₂ and steam. The pressure was adjusted to the lower side if there were any pressure difference.</td>
</tr>
<tr>
<td>9</td>
<td>BFM gasification using O₂ steam as the oxidizing agent. The detailed calculation method is stated in our previous publication [20].</td>
</tr>
<tr>
<td>10</td>
<td>Cooked down to 350 °C to recover heat (it is 350 °C in order to avoid tar condensation).</td>
</tr>
<tr>
<td>11</td>
<td>Cooked down to room temperature, hence the water was condensed. We assumed that there was a gas cleaning step here, as well.</td>
</tr>
<tr>
<td>12</td>
<td>Compressed to 30 bar at an isentropic efficiency of 74%</td>
</tr>
<tr>
<td>13</td>
<td>Heated to 398 °C (according to the data from the ICI process)</td>
</tr>
<tr>
<td>14</td>
<td>Saturated steam at 30 bar</td>
</tr>
<tr>
<td>15</td>
<td>Adiabatic mixing of syngas with steam. The pressure was adjusted to the lower side if there were any pressure difference.</td>
</tr>
<tr>
<td>16</td>
<td>Methanation Reactor 1; 10% of pressure loss and 5% of heat loss (adiabatic equilibrium reactor)</td>
</tr>
<tr>
<td>17</td>
<td>Heat exchanger (inter-cooling)</td>
</tr>
<tr>
<td>18</td>
<td>Methanation Reactor 2; 10% of pressure loss and 5% of heat loss (adiabatic equilibrium reactor)</td>
</tr>
<tr>
<td>19</td>
<td>Heat exchanger (inter-cooling)</td>
</tr>
<tr>
<td>20</td>
<td>Methanation Reactor 3; 10% of pressure loss and 5% of heat loss (adiabatic equilibrium reactor)</td>
</tr>
<tr>
<td>21</td>
<td>Condensing heat exchanger (the vapor pressure of the steam remained as saturated pressure after a certain temperature)</td>
</tr>
<tr>
<td>22</td>
<td>Compressed to 30 bar at an isentropic efficiency of 74%</td>
</tr>
<tr>
<td>23</td>
<td>Separation of CO₂ (98% of CO₂ and 1% of CH₄ removed); electricity consumption of 0.576 MJ/Nm³ of CO₂</td>
</tr>
</tbody>
</table>
Table B2. Streams mass flow rates, thermodynamic state and composition.

<table>
<thead>
<tr>
<th>Stream Number</th>
<th>Mass Flow Rate, Thermodynamic State and Energy Content</th>
<th>Solid Biomass Composition wt %</th>
<th>Gas Composition vol %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>m (kg/s)</td>
<td>P (bar)</td>
<td>T (K)</td>
</tr>
<tr>
<td>1</td>
<td>14.09</td>
<td>1.01</td>
<td>275.15</td>
</tr>
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<td>2</td>
<td>7.04</td>
<td>1.01</td>
<td>348.15</td>
</tr>
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<td>3</td>
<td>9.94</td>
<td>1.01</td>
<td>288.15</td>
</tr>
<tr>
<td>4</td>
<td>2.42</td>
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<td>288.15</td>
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<tr>
<td>5</td>
<td>2.42</td>
<td>15.00</td>
<td>658.70</td>
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<tr>
<td>6</td>
<td>2.42</td>
<td>15.00</td>
<td>658.70</td>
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<tr>
<td>7</td>
<td>3.52</td>
<td>15.00</td>
<td>491.45</td>
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<td>8</td>
<td>5.95</td>
<td>15.00</td>
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<td>12.91</td>
<td>15.00</td>
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<td>15.00</td>
<td>623.00</td>
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<td>10.61</td>
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<td>10.81</td>
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<tr>
<td>20</td>
<td>10.81</td>
<td>21.87</td>
<td>671.15</td>
</tr>
<tr>
<td>21</td>
<td>8.85</td>
<td>21.87</td>
<td>313.15</td>
</tr>
<tr>
<td>22</td>
<td>8.85</td>
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<tr>
<td>23</td>
<td>1.75</td>
<td>30.00</td>
<td>347.67</td>
</tr>
</tbody>
</table>

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Thermodynamic performance of a hybrid power generation system using biomass gasification and concentrated solar thermal processes
Thermodynamic performance of a hybrid power generation system using biomass gasification and concentrated solar thermal processes

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Highlights

- A hybrid power generation based on biomass gasification and concentrated solar thermal.
- Investigation of optimal integration by pinch analysis.
- Improvement of system efficiency by co-utilizing surplus heat and solar thermal.
- Efficiency affected by composition and temperature of gasifying agent.

Abstract

This paper describes the investigation of a hybrid power production system from biomass and solar energy. This paper suggests integration through heat exchanger network as a useful approach to obtain the synergy between biomass and solar. Biomass is first gasified in a bubbling fluidized bed (BFB) gasifier, and then syngas is used in a gas turbine. Excess heat exists in this sub-system and concentrated solar thermal process (CSTP) while there is a demand of steam for generating gasifying agent. Steam Rankine cycle exploits the heat created by these thermal streams to generate power while satisfying the steam demands. Thermodynamic performance was analyzed by process modelling with a semi-kinetic model of BFB gasifier and pinch analysers. The composition and temperature of gasifying agent showed some effect on the overall efficiency of the system. Higher overall efficiency of the system was achieved at higher temperature and higher O2 fraction in the O2-steam mixture as gasifying agent. The increase in thermal input from CSTP had positive effect on overall efficiency of the hybrid system until thermal input from CSTP becomes dominant against thermal stream related to the gasifier and the gas turbine.

1. Introduction

Diversifying the portfolio of energy sources is an important issue in order to ensure energy security. Especially, alternative energy of fossil fuel and nuclear energy is required to reduce the risk against fossil fuel depletion and nuclear accident. Power generation with biomass gasification (b-IGCC, biomass-based integrated gasification combined cycle) is one of such alternative technologies, whose output is not affected by the weather conditions like other renewable energies. However, it is still necessary to improve the efficiency to generate electricity from biomass. There is a common strategy to increase the performance of energy plant by integrating two or more processes where heat supplies and demand exist [1]. This paper addresses how the overall performance will be improved when b-IGCC is integrated with concentrated solar thermal processes (CSTP). Gasification is a process that breaks down biomass into smaller molecules to generate syngas, mainly H2, CO, CO2, H2O and CH4, with the help of heat and oxidizer (so-called gasifying agents). Some gasification processes
require heat input to generate steam as gasifying agent or to preheat gasifying agents in order to increase the syngas energy. This means that the integration with other processes that have excess amount of heat may improve the performance of the overall system.

Several concentrated solar power stations (CSP) have started commercial operation in the last decade [2]. Solar radiation is concentrated and stored in a heat transfer fluid (HTF) either with line-focusing (parabolic trough and linear Fresnel) or point-focusing (parabolic dish and heliostat) devices. The temperature of the HTF from a receiver is typically 150–500 °C for line-focusing devices and 500–1500 °C for point-focusing devices, making it attractive for hybrid power generation schemes[3–8].

Dependent on the HTF temperature, concentrated solar energy can be converted into electricity in various power cycles [9,10]: e.g. organic Rankine cycle (ORC) for 0–250 °C, steam Rankine cycle for 250–600 °C, Stirling engines for 600–850 °C, and gas turbines for 850–1500 °C. Nonetheless, steam Rankine cycle is the dominant power generation cycle in commercial-scale CSP plants. Thermal energy storages (TES) are often employed to compensate hourly intermittency of the CSP system. Different forms of TES have been studied, broadly classified as sensible heat storages (such as molten-salt, concrete, ceramics) and latent heat storages (PCM, phase change material) or a combination of both [11–19].

Hybridization is also an important strategy for realizing solar thermal power plant since electricity production cost of stand-alone CSP plants is higher than other sources, typically above 210 USD/MWh [20]. Three major approaches are found in the literature, namely hybrid solar-gas-turbine (HSTG) system, solar-CSP-biomass hybrid system, and solar-assisted biomass gasification. Concentrated solar thermal processes (CSTP) are supplemented to natural gas fired gas turbine in HSTG [3,5,7–9], typically to preheat the compressed air to 850–950 °C prior to combustors. However, the commercial application of this system is limited due to the physical locations of the receiver (top of the tower) and the combustor (on the ground) [7]. Other example of HSTG configuration is to inject steam from CSP system into the gas turbine [5].

Solar CSP-biomass hybrid systems for power production are gaining interest [21] as CO2-neutral power generation. The concept of hybrid biomass-fired boiler and CSP is discussed in [22], where the biomass boiler is used to accommodate the change in the intensity of solar radiation. Similar principle, tri-generation (production shift among electricity, heat and cooling), was also investigated at small-scale (2–10 MWth) hybrid solar-biomass boiler [23]. Several studies [24–30] also focused on using the thermal energy from CSTP to supply energy demand of biomass gasification process. Such configurations boost the yield of syngas and the conversion efficiency of the gasification step. However, the operational instability due to the intermittent solar radiation may overcomplicate process control.

An alternative approach is to hybridize b-IGCC-CSTP through heat exchanger network (HEN), which is the focus of this study. An important aspect of our hybrid b-IGCC-CSTP system is continuous operation of the b-IGCC independent of solar radiation because the hybridization is limited to less-crucial sections, in this case the HEN. Such configurations may enjoy the improvement in energy efficiency while avoiding process control issues. In such systems, electricity production of biomass origin virtually plays the role as base-load, and electricity production of solar origin and efficiency improvement by synergy effect plays the role as peak load. It is reasonable because the peak load of electricity in the area with high solar radiation often coincide with the ambient temperature, thus solar radiation. If it is not the case, CSTP can be still used as temporary energy storage since it can store heat in molten salts at most CSP plants can be operated even during cloudy period or night time. It was reported that commercial CSP plant, Gemesolar, achieved 24 h of uninterrupted electricity production [21].

This study presents the investigation of the optimum way to integrate CSTP with b-IGCC for power production, and its thermodynamic performance by process simulation. We assumed that the new system will utilize a bubbling fluidized bed (BFB) gasifier and CSTP with molten salt heat storage system. An accurate model of the BFB gasifier is important because its behavior significantly affects the performance of the whole system. Therefore, we first
developed a semi-kinetic model of the BFB gasifier in order to accurately estimate gas composition and carbon conversion at various operation conditions. Optimal integration of b-IGCC and CSTP also lies on the proper utilization of heat from CSTP in the b-IGCC. Hence, an optimization of the heat integration among the thermal streams of the processes and those of a water/steam Rankine cycle was carried out in order to maximize the net electric output of the system. Then, parameter sensitivity and the synergy effect of the integration were investigated in detail. It should be noted that detailed modelling of a TES is not considered here as it would not have influence on the synergy of the hybridization studied in this work. In addition, availability of the hybrid system is not among the subjects of the current work. The thermal input for the CSTP is considered as a variable parameter while the b-IGCC unit is operated with a constant biomass feeding under different operating conditions.

2. System configurations

The suggested integrated power generation system mainly consists of the BFB gasifier, a gas turbine, a Rankine cycle, CSTP, and an air separation unit (ASU) as shown in Fig. 1. Mixtures of O₂ and H₂O, as well as enriched air were considered as gasifying agents. O₂ was supplied from the ASU. All the syngas was introduced to the combustor in the gas turbine. In addition to the connection of mass flow, each component was integrated by the heat exchanger network. Heat demands exist at: preheater of gasifying agent; and a heat recovery steam generator (HRSG) of the Rankine cycle. Heat supplies exist at: the syngas cooler before the gas cleaning system; exhaust gas of the gas turbine; and hot molten salt from the CSTP. The following sections show the details of components.

2.1. Bubbling fluidized bed (BFB) gasifier

The fluidized bed gasifiers are suitable for the application in relatively large scale power generation system, and have an advantage for high mass and heat transfer as well as stability in reactions due to the enhanced mixing by fluidization. Hence, the fluidized bed gasifier is a proper option for integration with CSTP considering the scale of CSTP: around 100 MWth. H₂–O₂ mixture or enriched air (O₂–air mixture) was used as gasifying agents. Since the injection of high O₂ concentration gasifying agent has possibility to cause bed agglomeration due to high local temperature, maximum O₂ molar fraction was set at 0.40. The bed temperature was kept at 800 °C by adjusting the flow rate of the gasifying agent. The syngas temperature was assumed to be 720 °C, same as that of freeboard, to be utilized in the heat exchanger network. The lowest temperature of the syngas before gas cleaning was set as 350 °C to prevent tar condensation. Two compressors for gasifying agent were applied. The first one compressed bled air to the gasification pressure, and the other compressed O₂ from ASU to the gasification pressure. H₂O were supplied from the back pressure steam turbine in the Rankin cycle and heated to the supply temperature if necessary.

2.2. Gas turbine

The process conditions of the gas turbine must be modified when syngas is used because it has lower heating value than common gas turbine fuels such as natural gas. The relationship among mass flow, pressure and temperature of turbine inlet is defined by the constant, $m \cdot \frac{T}{p^{0.5}}$, for the same turbine because it is under the choked flow. On the other hand, the pressure of the turbine has upper limit to avoid surging of the compressor. Therefore, we
should modify the configuration of mass flow inside the gas turbine. Among common practices, we applied air bleeding method, in which part of compressed air from the gas turbine is spitted as waste stream to keep the turbine inlet temperature. The technical data of a commercial turbine was analyzed, and used for the calculation of gas turbine. The turbine inlet temperature and the pressure ratio of compressor were at 1183 °C and 14, respectively. We assumed that isentropic efficiencies of the compressor and turbine were 0.76 and 0.9, respectively. We also assumed that 2% of pressure loss in the combustor, and 99.5% of combustion efficiency, 99% of mechanical efficiency and 97.2% of generator efficiency.

2.3. Concentrated solar thermal process (CSTP)

The CSTP generate high quality (i.e. temperature) heat by reflecting solar radiation on mirrors that are set on the field, and by concentrating solar radiation on a receiver. Generally, the heat is used for generating steam for Rankine cycle and is converted to electricity. This study considered heliostats type CSTP, which consists of the receiver at the top of the tower and thousands of the heliostats. It also has a hot and a cold tank to store molten salt and accumulate the heat. The heliostats are mirrors which are set on the field and kept to track sun and to reflect sun light to the solar receiver by adjusting angles. Molten-nitrate salt is used as the heat medium in CSTP. It is supplied from the cold tank and heated in the solar receiver, and then stored in the hot tank. The property of CSTP was referred to Gemasolar [2]. It was reported that the temperature of supplied cold molten salt is 290 °C, and that of hot molten salt heated in the receiver is 565 °C. Hence, we assumed that molten salt can supply heat at 500 °C stably to the heat exchanger network and leave at the temperature of 300 °C.

2.4. Air separation unit (ASU)

The pressure swing adsorption (PSA) was applied as ASU. It was assumed that air is compressed to 0.4 MPa and separated by the adsorber, usually zeolite, and then expanded to the atmospheric pressure. This study considered only electricity consumption in the compressor for the energy consumption in PSA.

3. Calculation methods

3.1. Semi-kinetic model for a bubbling fluidized bed (BFB) gasifier

An accurate model of the BFB gasifier is important because its behavior significantly affects the performance of the whole system. Three targets were set during the development of the BFB gasifier model in this study: (1) to be able to predict methane yield; (2) to be able to predict char conversion; and (3) to make it possible to take into account for the effect of geometry. We selected semi-kinetic modelling approach to achieve these targets while keeping computational time low.

The model consists of three main sections as shown in Fig. 2. Relatively fast reactions such as pyrolysis and volatile combustion were calculated without consideration of kinetic process. Pyrolysis products were used as input to a continuously stirred tank reactor (CSTR) model, which represented the reactions in the bed (water–gas shift reaction, WGSR, and char gasification). Then, the gas outlet from CSTR was introduced to a plug flow reactor (PFR) model taking into account to the WGSR in the freeboard. Similar models can be found that consists of non-kinetic section and kinetic-section [31,32]. The main distinctions of the current model are: (1) PFR was applied for the kinetic model of the freeboard, (2) tar production was neglected for the simplicity, (3) char residence time was calculated by mass balance to keep the bed volume constant, and (4) the effect of pore diffusion was taken into account to the char gasification rate.

Pyrolysis and volatile combustion were assumed to finish instantly (non-kinetic section) when biomass was introduced into the gasifier because their reaction times are much shorter than that of char gasification. Product from this section consists of CO, CO2, CH4, and char. CH4 and char yields in this section were taken from the literature data. CH4 yield in the referred pilot-scale experiments [33] was constant regardless the conditions or feed rate at 7.114 wt.% of raw biomass input. Char yield from pyrolysis was taken from lab-scale devolatilization experiments [34] using same sample as pilot-scale experiments [33].

\[ m_{\text{CH}_4} = \frac{m_{\text{bio}}}{100} \times -15.03 + 50.58 \times \left( \frac{F - 273}{500} \right) - 18.09 \times \left( \frac{F - 273}{500} \right)^2 \]  

The diameter of resulting char was assumed to be 4.2 mm since the wood pellet shrunk around 70% during devolatilization and fragmentation was not observed [33]. We assumed that H2 was first selectively consumed during volatile combustion due to its high combustion rate and the diffusivity. Then, CO, CO2, and H2O yields can be calculated by the C/H/O balances. Char combustion was neglected because it is much slower than volatile combustion.

All the products from non-kinetic section were used as input to the CSTR for bed modelling, except for CH4. CH4 flow was bypassed from the non-kinetic section to the output since the methane stream reforming reactions hardly progresses under the relevant conditions in the BFB gasifier [35]. The balance equations of species \( i \) in CSTR can be written as:

\[ F_{\text{i, out}} = F_{\text{i, in}} + \sum n_{r,i} \dot{V}_{r} \]  

The reaction rate of WGSR (\( \text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2 \)) was calculated as:

\[ r_{\text{WGSR}} = k_{0} \exp \left( \frac{E_{\text{a}}}{RT} \right) \left( \frac{C_{\text{CO}}C_{\text{H}_2\text{O}}}{C_{\text{CO}_2}C_{\text{H}_2}} \right) \]  

where pre-exponential factor, \( k_{0} \), is 2.5 10^11 m^3 mol^{-1} s^{-1}. Activation energy, \( E_{\text{a}} \), is 138 kJ/mol, and equilibrium constant, \( K_{e} \), is 2.65 10^2 exp(32,900/T), respectively [36]. Char gasification with CO2 and H2O were also considered, and their conversion rates can be expressed by:

\[ \frac{dX}{dt} = \frac{M_{c}}{P_{\text{char}}} \left( r_{\text{char}} - r_{\text{char}} - \dot{m}_{\text{char}} \frac{\partial P}{\partial X} \right) \]
Kinetic parameters of char gasification with H₂O or CO₂ are shown in Table 1 [37,38]. For the structural function of conversion, g(X), the overlapped grain model was used [39]. The effectiveness factor, ηŚ, is related to the particle shape, and for a sphere particle is expressed as:

\[ \eta_Ś = \frac{1}{\frac{1}{MT} \left( \frac{1}{\text{PFR}} \frac{1}{M} \right)} \]  

(5)

Here, Thiele module, MT, of the nth-order irreversible reaction can be calculated as:

\[ M_T = \sqrt{n \frac{1}{\sqrt{k_g \frac{1}{C_p} \frac{1}{X}}} \frac{1}{C_n \frac{1}{T}}} \]

(6)

The calculation methods of the effective diffusivity can be found in Supporting material. To simplify the simulation, all the char particles in the bed were represented by the average conversion, \( \bar{X} \), of char extracted from the bed. Hence, the molar flow rate of char, \( \dot{F}_{\text{char, out}} \), can be expressed as:

\[ \dot{F}_{\text{char, out}} = (1 - \bar{X}) \dot{F}_{\text{char, in}} \]

(7)

Char volume in the bed, \( V_{\text{bed}} \), was calculated from the bed volume assuming the bed expansion and void fraction as described in Supporting material.

Mass balance in the freeboard was calculated based on the PFR model with considering WGSR. The same reaction rate was used for the WGSR as shown in Eq. (3). The temperature in bed, \( T_{\text{bed}} \), and that in freeboard, \( T_{\text{free}} \), were assumed to be homogenous, \( T_{\text{bed}} \) was calculated by the energy balance.

\[ \eta_{\text{adiabatic}} = \frac{m_{\text{char}} LHV_{\text{char}} + m_{\text{H₂O}} \int_{T_{\text{char}}}^{T_{\text{in}}} C_p \text{d}T - m_{\text{char}} LHV_{\text{char}}}{m_{\text{char}} LHV_{\text{char}} + m_{\text{H₂O}} \int_{T_{\text{char}}}^{T_{\text{in}}} C_p \text{d}T} \]

(8)

The temperature difference between \( T_{\text{char}} \) and \( T_{\text{in}} \) was calculated by using experimental data.

3.2. Scale-up of the gasifier

For the validation of the BF gasifier model, we used the literature data of the pilot-scale gasifier whose capacity was 28.6–68.3 kW based on LHV. On the other hand, the capacity of the gasifier in the proposed system was assumed to be around 70 MW because it employs the combined cycle. It means that biomass feeding rate would be increased from 10–21.6 kg/h to 1.5 ton/h, and the geometry of the gasifier in the simulation needs to be scaled up using the criteria of the literature [40]. The gasifier pressure was increased from the ambient pressure to 1.5 MPa because the pressure of the syngas should be higher than that of the combustors in the gas turbine. The volume ratio occupied by the bed material in the bed was kept at the same value. The result of the scaling up is shown in Table 2. The scaling up criteria was written in the Supporting materials.

### Table 1

<table>
<thead>
<tr>
<th>Coefficient of the reaction rate of CO₂ and H₂O gasification</th>
<th>H₂O gasification [36]</th>
<th>CO₂ gasification [38]</th>
</tr>
</thead>
<tbody>
<tr>
<td>( h_0 )</td>
<td>( 3.1 \times 10^7 \times 1 \text{bar}^{-0.218} )</td>
<td>( 2.6 \times 10^7 \times 1 \text{bar}^{-0.350} )</td>
</tr>
<tr>
<td>( E_a )</td>
<td>215 kg/mol</td>
<td>217 kg/mol</td>
</tr>
<tr>
<td>( z )</td>
<td>0.38</td>
<td>0.52</td>
</tr>
</tbody>
</table>

### Table 2

<table>
<thead>
<tr>
<th>Result of scaling up gasifier.</th>
<th>Pilot scale</th>
<th>Large scale</th>
</tr>
</thead>
<tbody>
<tr>
<td>Biomass feeding rate</td>
<td>10–21.6 kg/h</td>
<td>1.5 ton/h</td>
</tr>
<tr>
<td>Pressure</td>
<td>ambient</td>
<td>1.5 MPa</td>
</tr>
<tr>
<td>Bed diameter</td>
<td>0.15 m</td>
<td>0.90 m</td>
</tr>
<tr>
<td>Bed height</td>
<td>1.40 m</td>
<td>7.00 m</td>
</tr>
<tr>
<td>Freeboard diameter</td>
<td>0.25 m</td>
<td>1.50 m</td>
</tr>
<tr>
<td>Freeboard height</td>
<td>2.15 m</td>
<td>10.50 m</td>
</tr>
<tr>
<td>Amount of bed material in bed</td>
<td>8 kg</td>
<td>1600 kg</td>
</tr>
</tbody>
</table>

Adiabatic efficiency (\( \eta_{\text{adiabatic}} \)) of the large scale gasifier was estimated by assuming that heat loss from the gasifier is proportional to the surface area of the gasifier. Since the biomass feeding rate was increased around 1000 times by this scaling up and the ratio of the surface area of the large scale to the pilot scale was 10, \( \eta_{\text{adiabatic}} \) of the large scale gasifier was 0.9967.

3.3. Process integration

The processes described in the previous sections (b-IGCC and CSTP) were considered to be operated in an integrated manner, and a water/steam Rankine cycle was also introduced to convert the heat surplus available in the ranges of high and medium temperatures into electricity. The electric output and the thermal efficiency of the system were evaluated after considering the heat integration of the thermal streams from all the processes in the system.

In order to perform the heat integration, the HEATSEP method has been used [31,41,42]. The HEATSEP method makes it possible to optimize the configuration of the system without taking into account the details of its heat transfer section. In fact, all the heat transfers among the thermal streams of the system were assumed to occur in an unspecified “black box”, so that only the conditions at the boundaries of this “black box” (i.e. the initial and final temperatures and the mass flow rates of the thermal streams) were used among the optimization variables.

In this case the configuration of the background processes (i.e. b-IGCC and CSTP) was fixed, as well as the temperatures and the mass flow rates of their thermal streams, as calculated by the system flow sheet. Thus, the decision variables that have to be optimized in order to maximize the net electricity production of the system were:

- the maximum pressure and temperature of the steam in the Rankine cycle (upper limits of the turbine inlet temperature and the pressure are 540 °C and 100 bar, respectively, and the vapor fraction at the outlet of the turbine should not be lower than 0.9); and
- the mass flow rate of the steam that remains in the steam Rankine cycle (it should be noted that part of the generated steam is consumed in the gasifier and an amount of feed water equal to the consumed steam has to be supplied continuously to the Rankine cycle).

The optimizations were performed using a Sequential Quadratic Programming (SQP) algorithm, under the constraint that the heat transfers occurring in the “black box” have to be feasible according to the rules set by Pinch Analysis. This was verified by running a Problem Table Algorithm in order to check that there the cumulative heat surplus is greater or equal to zero in all the temperature intervals of the thermal cascade. The synthesis of the heat exchanger network that realizes the heat transfers within the “black box” is however beyond the limits of the current work.
4. Results and discussion

4.1. Validation of the semi-kinetic model

The semi-kinetic model was validated with pilot-scale experimental data at ambient pressure from a literature [33]. The experiment was conducted under simulated adiabatic and autothermal conditions. Wood pellets were used as sample at 10–21.6 kg/h and air, oxygen and steam were used as gasifying agents at the temperature of 400°C. The bed temperature and the freeboard temperature were 755–840°C and 695–727°C, respectively. In this validation, the bed and freeboard temperatures in the simulation were taken from the experimental data, not from the energy balance. The initial porosity of char, the char pore tortuosity, and the specific surface area of char were used as fitting parameters, and decided as \( e_o = 0.5 \), \( s = 4 \), and \( S_{char} = 2.5 \times 10^5 \text{m}^2/\text{kg} \).

The result of the validation of the semi-kinetic model is shown in Fig. 3 as a parity plot. It is common to occur 20% or more errors in mass balance of pilot-scale experiments due to the difficulties in steady operation or complete analyses [43]. Hence, ±20% of error

![Fig. 3. Parity plots of the semi-kinetic simulation and the experimental data. (a) CO yield; (b) H2 yield; (c) CO2 yield; (d) CH4 yield; (e) cold gas efficiency.](image-url)
was considered as the threshold of the validation. Prediction of CO and H₂ yields and the cold gas efficiency were much closer to the experimental values than those by the equilibrium calculation (see Supplement material), and most of the data were seen within threshold lines. Since the CH₄ yield in the semi-kinetic model was calculated by using that of the experiment, the results were nearly on the y = x line. This result can be thought to be valid after scale up because the decomposition reactions, steam reforming, are virtually frozen under the operating conditions of BFB gasifiers as discussed in the model description. Although the result of the CO₂ yield was dispersed more than other values, we regarded it as valid because there was no systematic deviation. No correlation was found between operation conditions and the ratio of heat loss to biomass energy input or temperature difference between bed and freeboard. Hence we used average values of these values to calculate energy balance in the model (Eq.(8)) in the following sections.

4.2. Effect of gasifier operation on system performance

The integrated b-IGCC/CSTP systems were evaluated by considering a common bottoming steam Rankine cycle as described in Section 3.3. The steam Rankine cycle was designed to exploit the heat pocket created by the high temperature thermal streams of the integrated processes and to generate the steam that is expanded in a steam turbine to produce power while satisfying the thermal demands of the integrated processes at the required temperature levels (in this case steam was extracted at 19 bar to supply the gasification process steam demand). Air or the mixture of O₂ and H₂O are often used as gasifying agents, and it is well known that they have significant impacts on the performance of gasification. In addition, preheating temperatures of the gasifying agents and the compressed air in the gas turbine are adjustable by the heat exchanger network, and they may affect overall system performance. Hence, the effects of these parameters on the net efficiency of the overall system were examined in this section.

Enriched air and the H₂O–O₂ mixture was examined as gasifying agents. Since a high O₂ fraction may result in a local high temperature, molar fraction of O₂ was limited to 0.2–0.4. Temperature and molar fraction of O₂: 0.2 and 0.4) and the heat input from CSTP was 100 MW. In both cases, the increase in the gasifying agent resulted in slightly higher overall efficiencies.

4.3. Synergy effect of integrating CSTP to b-IGCC

This section describes the evaluation of the improvement in the system performance by integrating b-IGCC and CSTP. The efficiency of the stand-alone CSTP power generation system using the Rankine cycle was estimated to be 36.0%. On the other hand, the efficiency of the stand-alone b-IGCC system was 28.1%. To evaluate the improvement of the performance of the hybrid system by the integration, the increase in the electric power output from the stand-alone systems (marginal electricity output) and the ratio of marginal electricity output to the power output of the stand-alone systems (marginal efficiency) were calculated as:

\[ P_{\text{ele, marginal}} = P_{\text{ele, hybrid}} - (P_{\text{ele, CSTP}} + P_{\text{ele, BFG}}) \]  
\[ \text{marginal efficiency} = \frac{P_{\text{ele, marginal}}}{P_{\text{ele, hybrid}}} \]

The dependence of the marginal electricity output and marginal efficiency on the thermal energy input from the CSTP (for the cases 20 and 40 vol.% O₂ in the gasifying agent) was evaluated over three temperatures of gasifying agent as presented in Fig. 6. The marginal electricity output increased with both increasing heat input from the CSTP and temperature of gasifying agent. Moreover, the
increase in marginal electricity output followed two distinct gradients over the considered range of the CSTP thermal input (0–100 MW). The gradient was high over the range 0–10/15 MW of CSTP thermal input and moderate over the rest of the range considered. Evidently, as the CSTP thermal input increase further over 100 MW the gradient of the marginal electricity output is eventually expected to fall to zero (marking absolute dominance of the CSTP) beyond which the contribution of b-IGCC is insignificant.

The benefit of integrating the processes is as well readily perceived from the integrated grand composite curves, Fig. 7. When the CSTP thermal input is low (i.e. 0–10/15 MW), the utility pinch was found to be at a temperature corresponding to the evaporation pressure level of the steam Rankine cycle resulting in higher cold utility requirement (about 5 MW corresponding to a 0 MW CSTP input). When the CSTP thermal input is increased to 15 MW and above, the pinch location shifted to a temperature corresponding to the condensation pressure level of the steam Rankine cycle resulting in a very low cold utility requirement (about 0 MW corresponding to a 50 MW CSTP input). The shift of the utility pinch from the evaporation to the condensation temperature level of the steam Rankine cycle was an indication of efficient exploitation of the heat pocket created by the integrated processes, which resulted in higher net power output as compared to the aggregate of the corresponding standalone cases. As shown in Fig. 6(b), there was no visible change in the efficiency after the shift of the utility pinch at around 10–15 MW of CSTP thermal input.

5. Conclusion

Overall objective of this paper was to investigate an efficient way to employ biomass and solar energy together for electricity production. Two key challenges were addressed during the process integration: accurate assessment of BFB gasifier performance and optimal way to integrate the heat exchanger network.

The semi-kinetic model of BFB gasifiers considers kinetics of char gasification with intra-particle mass diffusion effect and water-gas shift reaction while flaming pyrolysis is assumed to be instant process and steam reforming of methane to be chemically frozen. The developed model described the pilot-scale experimental data from the literature well. In the process integration stage, several scale-up strategies from the literature were applied to determine the dimension of the BFB gasifier. Then, overall performance of the hybrid power generation system using b-IGCC and CSTP was analyzed by using the HEATSEP method. First, effects of the composition and temperature of gasifying agent on the overall efficiency of the system were examined. When the mixture of O2 and steam was used as gasifying agent, higher overall efficiency of the system was achieved at higher temperature and higher O2 fraction in the gasifying agent. Then, the benefit of integrating b-IGCC and CSTP was evaluated by varying the heat input from CSTP at constant biomass energy input. As the thermal input from CSTP increased, the electricity output from
the hybrid system became higher than that of the sum from two stand-alone systems. Significant gain in overall efficiency was seen between 0 and 10/15 MW of thermal input from CSTP. Then, the overall efficiency improved moderately with further increase in thermal input from CSTP. The shift of the utility pinch from the evaporation to the condensation temperature level of the steam Rankine cycle was the major reason of the efficiency improvement.

Acknowledgements

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.apenergy.2015.05.084.

References

Power-to-gas and power-to-liquids for managing renewable electricity intermittency in the Alpine Region
Power-to-gas and power-to-liquids for managing renewable electricity intermittency in the Alpine Region

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Abstract:
Large scale deployment of renewable energy sources (RES) can play central role in reducing CO2 emissions from energy supply systems, but intermittency from solar and wind technologies present grid integration challenges. High temperature co-electrolysis of steam and CO2, in the so-called power-to-gas (PtG) and power-to-liquid (PtL) configuration, could provide a path for utilizing the excess intermittent electricity from a power system by converting it into chemical fuels that can be directly utilized in other sectors, such as transportation and heating. The chemical fuels could also be used in the power sector during periods of deficit in supply. Here, the impact of carbon policy and fossil fuel prices is investigated in relation to the economic and engineering potential of PtG and PtL systems deployment as storage for intermittent renewable electricity and as a source of low-carbon heating and transportation energy among the different energy sectors in the Alpine region. A geographically explicit cost minimization model, BeWhere, is used. Results indicate large-scale deployment of the PtG and PtL technologies for producing chemical fuels from excess intermittent electricity is feasible, particularly when incentivized by carbon prices. In addition, depending on carbon price and fossil fuels market, 0.15–15 Mtons/year of captured CO2 are utilized in the synthesis of the chemical fuels, providing 1–11% of liquid transportation fuel. In this context, it can be concluded that PtG and PtL technologies can enable greater integration of RES into the energy supply chain, with application worldwide.

1. Introduction

The global primary energy demand including industrial, residential and transportation sectors is largely met with carbonaceous fossil fuels. Utilization of carbon-based fossil fuels via conventional conversion technologies (e.g. combustion of natural gas and gasified/powder-coal in utility boilers/ gas turbines or combustion of petroleum derived liquid fuels in motor vehicles) releases CO2 to the atmosphere. CO2 is by far the largest contributor to the accumulation of
greenhouse gas (GHG) in the atmosphere leading to global warming and other detrimental environmental consequences.

In order to mitigate GHG emissions, several technologies are being developed. More notably, carbon capture and sequestration (CCS) is being developed as a post-combustion remedy for fossil fuels based energy processes, e.g. see [1], and for bio-energy processes (in the so-called BECCS configuration) in the context of negative emissions [2–4].

Other mitigation aspects include substitution of fossil fuels with carbon-free or low-carbon energy technologies (such as solar, wind, geothermal, hydro, biomass etc.). Decarbonisation of the energy sector by increasing the share of renewables is an essential step towards the deployment of low-carbon and sustainable energy systems. According to IEA [5], the share of renewables in the global electricity mix is projected to increase over the coming years (from 20% today to 65% in 2050, of which about 2/3 is intermittent in nature). Consequently, the interest for integrating renewables into the energy mix is growing. However, power generated from renewable energy sources (RES), in particular from solar and wind, is largely affected by the intermittency of availability of the resources. In addition to intermittency, the temporal and spatial mismatch between availability of resources (wind and insolation) and energy demand (consumers) imposes further constraint. As a result, utility-scale (megawatts to a few gigawatts in capacity) deployment of solar and wind technologies could impact the reliability of power dispatch systems. Large-scale storage systems, such as batteries, compressed-air, flywheel and pumped-hydro, could help even out this supply-demand mismatch.

Moreover, RES (such as wind, solar and hydro) are typically designed to produce electricity, which means that it can either displace fossil fuels usage in the electricity sector or it can power electrified transportation vehicles. This would limit the role of RES in planning economy scale de-carbonization of energy supply systems, which emit CO₂ from a wide-range of sources outside of the electricity sector.

In this context, the high-temperature co-electrolysis of steam and CO₂, see e.g. [6–15], using solid oxide electrolysis cells (SOECs) can offer an alternative option for converting excess intermittent electricity generated by solar/wind technologies into liquid/gas fuels that can be directly utilized in the transportation, heating or power sectors during periods of deficit in power supply. Co-electrolysis adds flexibility to the energy supply chain by creating links among the different energy sectors. Furthermore, it allows recycling of large volumes of CO₂, which can play significant role together with CCS technologies in curbing CO₂ emissions and de-carbonizing the energy supply system by enabling greater integration of RES into the energy mix.

The main focus of this work is to emphasize on the impacts of temporal and spatial intermittency of RES in power dispatch systems as well as on the utilization of excess intermittent electricity via power-to-gas and power-to-liquids (PtG and PtL) processes into other energy sectors (such as transportation, heating or power). The PtG and PtL processes considered in this work assume co-electrolysis of steam and CO₂ using high-temperature SOECs is mature technology and can be deployed at large scale, see e.g. [16,17]. In this regard, the PtG and PtL technologies can offer benefits that would make it an interesting addition to conventional storage technologies (such as batteries, compressed-air, and flywheel). In this process, the power over-generated from the power dispatch system can be stored in gas/liquid fuels via electrochemical reduction of gas-phase H₂O and CO₂. The reduced gas, essentially similar in composition to synthetic gas (otherwise known as syngas) which is mainly made of H₂ and CO, can then be used for the
synthesis of higher-grade transportation/gas fuels. Here, the impacts of carbon policy and fossil fuel prices are investigated in relation to the economic and engineering potential of PtG/PtL systems deployment as storage for intermittent renewable electricity and as a source of low-carbon heating and transportation energy among the different energy sectors in the Alpine region. The Alps is an interesting study region as it has potential for diverse RES generation, including biomass, solar, wind and hydropower.

Despite several reports examining the role PtG and/or PtL might play in low-carbon energy systems, see e.g. [18–22], to the knowledge of the authors, there is no mention of high-resolution energy planning models that put into perspective the role of PtG and PtL in the literature. In particular, PtG and PtL technologies are integrated in high-resolution decision support model on a regional level. This work has broad significance towards efforts to introduce more renewable energy into the electricity sector, and deep decarbonization of energy systems.

2. Power-to-gas and Power-to-liquid configurations

PtG could play a central role in enabling a greater share of intermittent renewables into energy supply systems. Intermittent renewables are inherently linked with power generation, and PtG and PtL processes can diversify the role of RES into other sectors such as transportation and heating. In this manner, the temporal and spatial power balancing problem characteristic to intermittent renewables is minimized and long term storage of renewable electricity can be realized. Figure 1 illustrates the schematics of the power balancing and long-term storage concept investigated in this paper.

2.1. Solid-oxide electrolysis cell

Electrolysis is central to the functioning PtG and PtL concepts. Electrolysis is an electrochemical process in which direct electric current is passed between two electrodes through ionized media (electrolyte) to deposit positively and negatively charged ions onto their respective electrodes.
The overall conversion efficiency from power to the chemical energy in the final product via electrolysis of water varies between 60–71% (based on HHV of H₂) for alkaline type and between 65–83% for PEM, depending on the operating pressure and cell current density [23,24]. Alkaline electrolyzers use aqueous alkaline electrolyte, operated at temperatures in the range of 70°C to 140°C, pressures between 1 bar to 200 bars [23] and low current densities, 0.2 – 0.4 A/cm²[21]. PEM electrolyzers are limited to a maximum temperature of 80 °C due to the polymeric electrolyte, operated under high current densities 0.5 – 2 A/cm² at pressures between 1 bar to 100 bars [21]. Detailed distinction among the listed electrolysis processes can be found in [21]. The alkaline and PEM type electrolyzers, also categorized as low-temperature electrolysis, are relatively mature technologies and most existing PtG pilot plants employ either of them or both [23].

High-temperature solid oxide electrolysis cell (SOEC) is gaining interest as it can be operated at temperatures in the range of 700–1000°C, meaning that part of the energy required to electrochemically dissociate H₂O (in the case of water electrolysis) or H₂O(g) and CO₂ (in the case of co-electrolysis) is supplied as heat energy minimizing the energy input in form of electricity [6]. Thus, performance of high-temperature SOEC has the advantage of both thermodynamic efficiency and faster reaction rates [25,26]. The heat required can be externally supplied via heat exchanger in the case of low current density operation, or it can be internally generated due to the inevitable ohmic cell resistance when the SOEC is operated at high current densities in order to maintain adequate production rates of H₂ or syngas.

The co-electrolysis characteristic of SOECs is of substantial importance here. This is because co-electrolysis achieves products that can be readily upgraded into liquid/gas fuels with existing market infrastructure in one step process. In principle synthetic gas (syngas) can be produced in a two-step process, electrolysis of H₂O to produced H₂ followed by conversion of H₂-CO₂ into syngas through reverse shift water-gas reaction (RSWG). In subsequent stages the syngas is catalytically upgraded into methane (Sabatier process) or higher grade hydrocarbons [6,21]. In this regard, the co-electrolysis process reduces the process steps by directly depositing high quality syngas (mainly H₂ and CO) on the cathode via simultaneous electrochemical reduction of H₂O and CO₂. In so doing, the gas deposited on the anode is pure O₂, which could also bring additional value to the process. In this study, however, no revenue is considered from O₂. Furthermore, the operation mode of SOECs is flexible towards the syngas produced, for instance by controlling the composition of the feed stream to the SOECs the quality of the syngas can be tailored to enhance catalytic conversion into synthetic fuels at later stages [25].

Recent development and performance improvements have demonstrated efficient co-electrolysis of H₂O(g) and CO₂ in SOEC. The ohmic resistance as well as the cell degradation rates and mechanisms are rather similar as in the electrolysis of steam alone [8,12]. In the light of such developments of SOECs, an overall conversion efficiency of 70% for PtL (the ratio of the calorific value of the liquid fuel produced, such as methanol, to power input) [16,19] and 80% for PtG (the ratio of the calorific value of methane produced to power input) [17] are to be expected. Unless stated otherwise, in this work an overall efficiency of 70% is assumed for both PtG and PtL technologies. This efficiency refers to the calorific value of the final product (liquid methanol in the case of PtL and methane gas in the case of PtG) and the power input to the process.
3. BeWhere Alps model

The BeWhere model, initially developed at IIASA and Luleå University of Technology, is used in this work. BeWhere is a geographic explicit cost optimization model, based on mixed integer linear programming (MILP), written in GAMS and uses CPLEX as solver. Earlier applications of the model are focused on planning and localization of bioenergy systems. So far, several researchers have demonstrated its application under different contexts, for instance methanol via biomass gasification [27–29], second generation biofuels on a EU scale [30,31], cost-effective CO₂ emission reduction through bioenergy [32,33], polygeneration in different locations [34–37].

The BeWhere Alps model is an enhanced version which includes other forms of RES in addition to biomass, namely solar, wind and hydropower. This work particularly focuses on the application of BeWhere to investigate the impact of carbon pricing and fossil fuels market as well as the impact of temporal and spatial intermittency of RES when planning coordinated decarbonization of energy supply system in the Alpine Region.

3.1. Set-up of the optimization model

The overall objective function is to minimize the total cost of the complete energy supply chain including the cost of CO₂ emissions, according to the following expression:

\[
\min f = \sum_c (\text{cost}^{\text{supply chain}}_c + \text{emissions}^{\text{CO}_2}_c \times \text{cost}^{\text{CO}_2}_c)
\]

The model satisfies different sets of constraints in relation to power generation mix: those that ensure the power demand is met at all hours in all the regions, those that ensure the share of fossil based power is generated within the country it is used and those that ensure prioritization of RES based power utilization. The first set of constraints satisfies the power demand using the least expensive options based on generation and existing transmission availability. The second sets of constraints prevent transmission of fossil based power (from baseload coal and dispatchable natural gas plants). The third sets of constraints prioritize the utilization of RES based power generation, i.e. investment on RES is only motivated when it becomes feasible to directly satisfy power demand.

The optimization procedure considers the transmission to be a direct power flow balance. There is no attempt to mimic the voltage phase shift which is highly nonlinear. However, the power flow balance approximation is a reasonable representation for high-voltage direct-current (HVDC) transmission network at a high level [38]. The use of an HVDC transmission instead of high-voltage alternating-current (HVAC) is due to the nonlinear nature of HVAC, which significantly complicates the optimization. However, the HVDC transmission can be thought of as an approximation of HVAC in terms of power flow because it includes electrical losses and it describes the transmission at a high level.

The cost optimization is superior to the load-matching optimization for real world applications for the development of a free market solution to the incorporation of variable generation into an electric power system. The objective function is the total generation, transmission, and storage costs for an electric power system for a selected time frame. It is assumed that there is only one type of dispatchable generator (natural gas combined cycle) as the aim is to consider a high penetration-level and variable-generation system.
Weather data is used for estimating the wind and solar PV power outputs. The natural gas plants are assumed to be back-up generation for when RES based power cannot meet the electrical demand. The basic approach is to take the salient variables (wind speed, solar irradiance, etc.) from a numerical weather prediction model and process them through computer code that mimics the behavior of a wind turbine and solar PV panel. The output will take into account the engineering constraints of the technologies as well as the weather components. To get a normalized value the output from the power model is divided by the capacity of the technology modeled.

3.2. Carbon pricing and fossil fuels market scenarios

The prices of fossil fuels often induce large degree of uncertainty in a long-term planning of energy systems. On the other hand, policies regarding CO₂ emissions vary greatly among countries. Based on the way the objective function is defined in the model, carbon pricing plays central role in enabling greater share of renewables. In this work, the model is used to investigate the influence carbon policy (in terms of price on CO₂ emissions) and fossil fuel prices on the mix of the energy supply chain. With this in mind, the model is run over a range 0–200 €/tCO₂ at an interval of 50 €/ton. In turn, each interval is evaluated for different fossil fuels prices (base case, medium, high). The base case is assumed at 100 €/ton CO₂ and at the current market prices for fossil fuels.

Table 1. Price matrix for carbon and fossil fuels

<table>
<thead>
<tr>
<th>Scenario (FFPs)</th>
<th>Carbon price</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Fossil fuels prices</strong></td>
<td>Base-case</td>
</tr>
<tr>
<td>Base-case</td>
<td>{0, 1}</td>
</tr>
<tr>
<td>Medium</td>
<td>{0, 1.5}</td>
</tr>
<tr>
<td>High</td>
<td>{0, 2}</td>
</tr>
</tbody>
</table>

*S{1, 1}, Scenario{Carbon price, FFPs}
*S{1, 1} represents base case scenario, €100/tCO₂ and market prices for fossil at the reference time.
*S{1, 1.5} and S{1, 2} represent for base case carbon price and FFPs 50 and 100% higher than the current market prices, respectively.
*S{1.5, 1} and S{2, 1} represent for base case FFPs and carbon prices 50 and 100% higher than the base case, respectively.
*S{1.5, 1.5}, S{1.5, 2}, S{2, 1.5} and S{2, 2} represent for scenarios where FFPs and carbon prices are 50 and 100% higher than their base case values, simultaneously and alternately. These price sets illustrate realistic future scenarios as FFPs and carbon price are intrinsically related parameters.
3.3. System boundaries and geographic resolution

The boundaries of the model are limited to the Alpine region, which in turn incorporates part of seven European nations as shown in Figure 2. Liechtenstein is excluded from the analysis on the count of its small size. In the model, the entire Alpine region is divided into about 3000 grid cells with a spatial resolution of 0.1 degree (approximately 10x10 km), see Figure 2.

During the optimization process, each grid cell essentially represents demand area (in terms of heating and transportation), supply area (in terms of resource availability such as biomass, river catchment, insolation and wind) and potential location for new power plant installation.

3.4. Supply chain

The energy supply chain considered in this study is comprised of different technologies and resources. The model includes biomass (for producing electricity, heat and biofuels), hydropower (existing and potential for new installations), solar photo voltaic (PV) and wind. The data collection and processing method for every resource considered is described in detail in the following text. The costs of technologies are documented in Appendix A.

Common to all technologies is the different environmental protections, such as national parks and reserves, regional parks, UNESCO reserves and world heritage, in the Alpine Region. This puts limitation on the availability of resources and constraints the setup of new facilities. The different levels of protection are represented in the model according to their priority order (high, medium, low and no protection). Difficulties related to harvesting of resources (e.g., biomass) and to installation of power plants (e.g., CHP, wind, solar and hydro) due to elevation and landscape profiles put further limitation on amount of energy that can be generated. As a result, locations beyond 2,000 m, in elevation, for a low environmental restriction scenario and 1,200 m for a strict environmental protection scenario are excluded from the analysis.
3.4.1 Bioenergy

Different biomass feedstock (e.g. forest residue, agricultural residue etc.) and conversion technologies (e.g. ST-CHP, bIGCC) can be used for the production of bioenergy. In this work, the biomass feedstock refers to forest residue which is assumed to be converted into heat and power via bIGCC technology. Two bIGCC plant sizes with different heat-to-power output ratios are considered for biomass conversion. The details of the technologies and the associated costs are provided in Appendix A, Table A1.

The potential supply of biomass in each grid cell is estimated based on its forest share net primary production and annual increment of forest biomass. A brief description of the methodology can be found in [33,35,39]. Here the annual biomass increment in each grid cell is explicitly introduced into the model. No distinction is made between different tree species. The available forest biomass is assumed to have a density of 500 kg/m$^3$ (dry weight), with a heating value of 18.5 GJ/ton (lower heating value (LHV) of dry feedstock) and a moisture content of 55%. Hourly energy production estimates are obtained by averaging the annual potential over the total number of hours in a year.

In the model, the transportation of forest residues to production plants can be accomplished by three means truck, train and boat. It could also be a combination depending on cost. Data for the cost of transportation and related emissions used in the model are summarized in Table 2. The transportation cost is composed of fixed (to account for loading and unloading, independent of distance) and variable (to account for distance). A network map of roads, rails and shipping lines is used to estimate the distance between supply and production plants. In fact, shipping lines are not applicable in Alpine region. Details on transport data processing can be found in [40].

Table 2. Biomass (refers to forest residue) transportation cost and related emissions. Energy conversions refer to 18.5 GJ/ton, LHV dry basis, and 55% moisture content. Cost data are adjusted to €2010.

<table>
<thead>
<tr>
<th>Transport type</th>
<th>Transport cost$^a$</th>
<th>Emissions$^b$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>€/TJ/km</td>
<td>tCO$_2$/PJ/km</td>
</tr>
<tr>
<td>Truck</td>
<td>$307 + 6.92 \times d$</td>
<td>5.82</td>
</tr>
<tr>
<td>Train</td>
<td>$648 + 0.96 \times d$</td>
<td>2.97</td>
</tr>
</tbody>
</table>

$^a$Transportation costs are adapted from [41]. $d$ is the transportation distance in km. The transportation cost values are for wet biomass (as received basis, 55% moisture content).

$^b$Emission factors are taken from [31].

3.4.2 Hydropower

Hydropower represents for both existing capacities and potential for new installations. The hydropower potential is estimated based on river catchment areas outside the protected regions of the Alps. Annual power production potentials are estimated based on river flow rates and mean head data acquired from [42]. In the model, hourly generation potential from hydropower is obtained by averaging the annual estimates over the total number of hours in a year. At this stage, seasonal variations in water amount are not considered. The costs assumed for new hydropower plants are documented in Appendix A, Table A2.
3.4.3. Solar energy

The hourly capacity factors and capacity limits for solar energy are derived based on high-resolution global climate reconstruction data. Solar insolation data is collected from an open access database developed at Princeton University [43]. Hourly solar installation estimates from the year 2010 are processed at 3-hourly temporal resolution and 0.25 degree spatial resolution. In order to estimate solar power output from solar insolation, a conversion efficiency of 15% is assumed. Capacity factors for 2010 are taken as the ratio of derived power to maximum power output in the year 2010, for each hour in each grid cell. Capacity limits are taken as the maximum power output in 2010. Data is projected to 0.1 degree spatial resolution in order to match with the resolution used in this work, based on the grid cell with the largest overlap. The solar capacity factors are sampled for the same hours as demand, which is described in a subsequent section. The costs associated with the solar PV technology considered in this work are reported in Appendix A, Table A2.

3.4.4. Wind energy

Like solar, the hourly capacity factors, and capacity limits, for wind energy are derived based on the high-resolution global climate reconstruction data from Princeton University [43]. Hourly wind speed from the year 2010 is processed. Wind speed estimates in areas with high surface roughness, like the Alps, are very uncertain. As such, derived capacity factors should be approached with caution.

The wind energy harvested per unit area-swept by the turbine rotor is derived based on the methodology of the Alpine windharvest Partnership Network [44]. To find the hourly energy output, a specific curve with maximum power of 450 W/m² at a rated cut-out speed is assumed, based on the Austrian Wind Potential Analysis [45]. In order to derive power output in each grid cell, the wind turbines are assumed to be spaced 11 lengths apart. Capacity factor, capacity limits, sampling, and interpolation methods are identical to those used in deriving solar inputs. Likewise, the costs associated with the assumed wind energy conversion technology are reported in Appendix A, Table A2.

3.4.5 Natural gas and coal plants representation

In the model, any deficit in power supply is assumed to be balanced with dispatchable natural gas plants that mimic actual plant operations through a set of regulating ramping constraints. The ramping constraints are implemented such that the aggregated output of dispatchable natural gas plants in a region of the Alps reaches a maximum (90% of the demand in the region) or falls down to zero within 120 minutes. Furthermore, the fossil model includes a coal fired base-load to cover 10% of the demand in each region of the Alps.

Moreover, the costs associated with fossil fuels based energy use are accounted in terms of the market values of the energy carriers, as reported in Table B1. Carbon emission intensities of fossil based energy use represent actual figures of each country belonging the Alps as summarized in Table C1.
3.5. Energy demand in the Alpine Region

3.5.1. Power demand in 2010

The hourly power demand for each country in the Alpine region is derived from the European Network of Transmission System Operators for Electricity (ENTSO-E). ENTSO-E reports historical demand at the country level\(^1\). The year 2010 is chosen, which is consistent with the estimates of wind and solar resources, and future representation of transmission within the Alpine Region.

The hourly demand profiles to the portion of each country within the Alpine Region are scaled based on the fraction of population living in the Alps. This assumes that per capita hourly demand is constant within a country. When data is unavailable in a specific hour, the data from the previous hour is used, or the same hour in the previous day, depending on data availability.

To reduce computational complexity, the demand is sampled every three hours from the peak and median day in each month. This is consistent with sampling methods from previous high-resolution electricity sector planning models [46]. In total, 192 hours are sampled throughout the year 2010 (8 hours/day, 2 days/month and 12 months/year). Figure 3 shows the profile of the power demand of the year 2010 for the sampled hours.

![Figure 3. Aggregated hourly power demand of the Alpine Region in 2010 at the sampled hours](image-url)

To represent the entire year, the sampled days are weighted to represent multiple days by fixing peak days to represent one day of the month and median days to represent the remaining days in the month (i.e. days in a month minus one) [46,47]. Doing so ensures peak conditions are included in the power constraint while economic assessment is dominated by typical demand profile, as peak demand occurrences are rare [47]. Accordingly, all samples (i.e. 8 samples per

\(^1\) Available at https://www.entsoe.eu/data/data-portal/consumption/Pages/default.aspx
selected day) represent 3 hours each, peak days represent a day of the corresponding month and median days represent the remaining days in the month. In total, 8760 hours per year. This procedure is included in the model by means of a time-indexed weighting parameter.

3.5.2. District heating and transportation fuel demand

A distribution system for fossil, biofuels and gas/liquids is assumed to exist or be built within the demand areas. The demand in each area is estimated by introducing per capita-scaled, heating and transportation, fuel consumption parameters. These parameters in turn refer to the fuel consumption data of the country to which the demand area belongs to. The data for carbon emission intensities in relation to fossil fuels use in the district heating and transportation sectors are summarized in Appendix C, Table C1.

Furthermore, the year is divided into three time periods of equal length so as to harmonize with seasonal heating demand variations, see e.g. [31]. Country specific parameters, for heating and transportation fuel demands per capita per unit time, used in this study are summarized in Table 3.

Table 3. Heat and transport fuel demand data used in this study [31]

<table>
<thead>
<tr>
<th>Country</th>
<th>Heat (GJ/capita/season)</th>
<th>Transport (GJ/capita/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( m_1 )</td>
<td>( m_2 )</td>
</tr>
<tr>
<td>Austria</td>
<td>12.1</td>
<td>1.7</td>
</tr>
<tr>
<td>France</td>
<td>49.0</td>
<td>26.0</td>
</tr>
<tr>
<td>Germany</td>
<td>163.3</td>
<td>86.6</td>
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<td>Italy</td>
<td>16.3</td>
<td>2.4</td>
</tr>
<tr>
<td>Slovenia</td>
<td>39.4</td>
<td>5.7</td>
</tr>
<tr>
<td>Switzerland</td>
<td>13.4</td>
<td>7.1</td>
</tr>
</tbody>
</table>

3.6. CO₂ sources in the Alpine Region

Other than water, significant portion of the feed stream to the high-temperature SOEC is CO₂. Preferably, CO₂ must be attained at low cost, high purity and flow rates large enough to match electricity over-generation from the power sector at any given time and location. Different sources can be identified as potential CO₂ providers. Commonly discussed sources include CO₂ from fossil power plants, CO₂ from biomass based CHPs and processes, CO₂ from other industrial processes and CO₂ from air. In this work, all types of power generation technologies that emit CO₂ within the Alpine Region are analyzed. No classification is made on plant type or on how the CO₂ is acquired. Direct air capture is excluded (DAC), which is likely cost-prohibitive [48].

Consequently, it is necessary to identify power plants that emit CO₂ in the Alpine Region. These locations are filtered from the Carbon Monitoring for Action (CARMA) database [49], by overlapping a geographic map of the Alpine Region and a location map of CO₂ emitting industries in ArcGIS. A total of 136 potential CO₂ sources are identified within the region, see Figure 4. The CARMA database includes future projections for CO₂ emissions from the
industrial sites, which is used in this work to constrain production capacities of PtG and PtL plants.

The identified CO₂ sources are also considered as potential locations for PtG and PtL plants. This simplifies the optimization problem in relation to localization of PtG and PtL plants. Transmitting excess electricity from the power grid via existing transmission lines is likely easier than transporting CO₂ to locations along the power grid.

Figure 4. Grid map of the identified CO₂ sources in the Alpine Region

4. Results and discussion

The results and discussions presented herein are reflections of the 192 sampled hours and refer to the sets of prices for carbon and fossil fuels introduced in Table 1. All annual estimates are weighted according to the scheme described in section 3.4. In addition, the impact of high penetration of intermittent renewables in the electricity sector is discussed in relation to reduction in CO₂ emissions.

4.1. Power generation mix

Figures 5–7 present the evolution of the resulting power generation mix at the sampled hours for all the sets of carbon prices and FFPs described in section 3. It should be noted that the contribution of base-load coal (which is set to cover 10% of the demand in each region) and existing hydropower (which covers about 18% of the total demand of the entire region) plants remains constant in all the cases. Consequently, the variations in carbon price and fossil market values mainly affect the contribution of intermittent RES (in this case solar and wind energy) and, to a much lesser extent, the contribution of new hydropower and biomass plants.

For instance, at zero carbon price and base-case FFPs the power generation is dominated by natural gas with minor contributions from new hydropower (about 9%), biomass (1.3%) and wind energy (0.25%), Figure 5–S(0, 1). When the carbon price was increased at an interval of 50 €/ton CO₂, the share of intermittent RES (particularly solar) starts to appear in the mix and increases
progressively to cover about 17% (refers to the power fed to grid) of the power demand at a carbon price of 200 €/ton CO₂, Figure 5. On the other hand, when the FFPs are increased by 50 and 100% of their base case values and at zero carbon price the contribution from solar gradually increases to cover about 11% (Figure 6–S{0, 1.5}) and 16% (Figure 7–S{0, 2}) of the power demand, respectively. Furthermore, at 50% higher FFPs than the base case and zero carbon price the contribution of solar is fully used in the power grid (Figure 6-S{0, 1.5}), whereas at 100% higher FFPs than the base case periods when generation exceeds demand start to appear already at zero carbon price, see Figure 7-S{0, 2}. In all the cases evaluated the contribution of wind energy is found to be insignificant.
Figure 5. Aggregated hourly power dispatch at the sampled hours for carbon prices in the range of 0–200 €/ton and base case FFPs. Over-generation, available power for PtG and PtL, is represented by the area above the demand.
Figure 6. Aggregated hourly power dispatch at the sampled hours for carbon prices in the range of 0–200 €/ton and at medium FFPs. Over-generation, available power for PtG and PtL, is represented by the area above the demand.
Figure 7. Aggregated hourly power dispatch at the sampled hours for carbon prices in the range of 0–200 €/ton and at high FFPs. Over-generation, available power for PtG and PtL, is represented by the area above the demand.

During the sample year, the total power demand was 530 TWh of which, depending on the carbon and fossil prices, about 28–53% is satisfied from RES and the remainder is covered from
fossil fuel, see Figure 8a. The low and high ends of the range correspond to scenarios S\{0, 1\} and S\{2, 2\}, respectively. Figure 8b shows the fraction of RES that is directly fed to the power grid. Accordingly, the fraction decreases with increasing carbon price and FFPs. This behavior can be explained by the fact that at high carbon price and/or high FFPs the share of intermittent RES in the generation mix is high, which in-turn increases occurrences of periods when supply exceeds demand.

Accordingly, the fraction decreases with increasing carbon price and FFPs. This behavior can be explained by the fact that at high carbon price and/or high FFPs the share of intermittent RES in the generation mix is high, which in-turn increases occurrences of periods when supply exceeds demand.

Figure 8. The fraction of power generated from RES that is directly fed to power grid to satisfy demand, a, and its corresponding share of the total RES generation, b.

4.2. Power transmissions

The model also utilizes existing transmission capacities among the regions studied. The transmission capacities are adopted from ENTSO-E. The net annual power transmissions for the investigated sets of carbon and fossil prices are presented in Figure 9. At low carbon price and base case FFPs the contribution of intermittent renewables to the generation mix is insignificant and, therefore, the transmitted power is dominated by hydroelectric which was considered non-variable in the model. On the other hand, at high carbon price and high fossil fuel prices the contribution of intermittent electricity (mainly solar) increases gradually, to about 40% for S\{2, 2\} of which about 60% is directly fed to the grid, reducing the power transmissions, see Figure 9c. The reason for the shift in transmissions trend is due to the increase in solar power generation in net power importing regions, in this case Germany and Italy, as shown in Figure 9. It should be noted that the increase in solar generation reduces the magnitude of power transmissions as a result of the RES prioritization constraints that enforce investments on intermittent generation units are initiated only if there is deficit in power supply.
Figure 9. Power transmissions among the regions over the range of carbon price $0–200\text{€/ton } CO_2$ and different levels of FFPs, base-case, a, medium, b, and high, c.
4.3. Utilization of excess intermittent power in other sectors

The PtG and PtL technologies exploit excess intermittent power during periods when supply exceeds demand. In the sample year an over-generation potential in the range of 0–65 GW is observed, see Figures 5–7, resulting in an annual total in the range of 0–93 TWh.

Figure 10a presents the corresponding amounts of methanol produced from the over-generated power in TWh/year. Accordingly, the model produces mainly methanol and traces of SNG, particularly in the high end of both the carbon price and FFPs ranges considered. This behavior is due to the fact that, in the model, methanol is restricted to replace transportation fuel (gasoline) which generally has higher market value as compared to the gas fuels used in the heating sector. The PtG is linked to SNG production which is restricted to displace fossil fuels in the heating sector.

The production of methanol is found to be rather more sensitive towards variations in FFPs as compared to carbon price. For instance, doubling the FFPs at 0 €/tCO2 increases the share of intermittent renewables in the power supply mix from 0.25 to 19% and the production of methanol from 0 to 6 TWh/year. Whereas, increasing the carbon price from 0 to 100 €/tCO2 at the base case FFPs raises the share of intermittent renewables in the generation mix from 0.25 to 12.5% and methanol production from 0 to 0.6 TWh/year, see Figure 10a.

The potential for displacing gasoline transportation fuel with methanol, produced in PtL technologies, is shown in Figure 10b. Accordingly, depending on the carbon price and FFPs, 1–11% of the gasoline use in transportation sector can be covered with methanol.

4.4. Impact of RES penetration on CO2 utilization and emissions

Another important aspect is that PtG and PtL provide the opportunity to recycle large volumes of captured-CO2 into the fuel supply system. Figure 11 shows the recycle rate of CO2 by assuming a mole of carbon dioxide is consumed to produce a mole methanol or methane. In the range of base case to high carbon price and FFPs considered, 0.15–15 Mtons of captured-CO2 is recycled. Recycling only affects the storage requirements for captured-CO2 [20], which could be crucial in countries where geological carbon storage is not permitted. In principle, by controlling the
recycle rate to be equal to the amount of captured-CO₂, the need for long-term storage can be avoided. However, CO₂ emissions from the industrial sites are not affected by PtG and PtL processes because the CO₂ captured at a previous step is used to produce renewable methanol or methane which is then released upon utilization. As such, CO₂ emissions from the industrial processes are only delayed by one step before they finally are released. But, CO₂ emissions from the transportation and heating sectors are reduced because of displacement of fossil fuels.

In a broader context, this study investigates the potential for decarbonizing the Alpine power sector by increasing the penetration of intermittent RES augmented with PtG and PtL technologies. It worth mentioning that the major benefit in terms of decarbonization is still due to the substitution of conventional fossil based power generation units with renewable ones. As such, it was considered important to highlight on the CO₂ emissions avoided due to the high penetration of RES. As shown in Figure 8b, depending on the scenario, 75 to 99.9% of the RES based power generation is directly transmitted to satisfy demand. Figure 12 shows the amount CO₂ emissions avoided due to direct substitution of fossil based power with RES. As a result, depending on the scenario, 22–103 Mtons CO₂ emissions are avoided annually.
Figure 12. CO$_2$ emissions avoided due to RES penetration in Million tons per year [Mt/year], over the range of variation of carbon price and for the different levels of FFPs.

4.5. Discussion

One impact of the PtG and PtL technologies on electricity dispatch systems is that the need for curtailment is reduced, to a large extent. Here, it is assumed all excess electricity generation can be used for the production of gas and/or liquid should the model finds it feasible to do so. In actual scenario, curtailment may not be totally avoided due to operational constraints of the PtG and PtL processes as well as the corresponding market values of the produced methane or methanol, which in turn influence the size and minimum operational hours of such technologies, see e.g. [20]. It may not be, economically and perhaps even technically, feasible to build PtG and PtL plants that operationally follow the peaks of the power generation profile, shown in Figures 5–7. However, curtailment can be minimized by coupling PtG and PtL with temporary power storages, such as batteries, in order to smooth out periods of peak power supply. Consideration of such operational details of the PtG and PtL are beyond the scope of this work. Figure 13 shows the percentage of curtailment as fraction intermittent RES (solar and wind) for the sets of carbon and fossil fuels prices considered. It should be noted that curtailment in this context refers to the surplus power because the model chose not to build PtG and/or PtL plant purely due to economic infeasibility. The weighting scheme by which the annual estimates are evaluated also adds bias, for instance, an over-generation at a sample hour in a median day would likely end-up being converted into liquid or gas fuel as compared to an equivalent over-generation at a sample hour in a peak day.
Regardless of the share of RES, power dispatch systems are required to have back-up capacity large enough to cover demand at any given time. This leads to increased redundant power generation units, which might result in situations when electricity prices are not reflecting cost of generation. A balance needs to be maintained, meaning the price of electricity should at least allow profit margins, whether fed to the grid, used in PtG and PtL or curtailed. As such, the results presented here assume electricity price of 50 €/MWh for the part that goes to the PtG and PtL.

5. Conclusions

This study focuses on investigating the potential for integrating RES in the energy system of the Alpine region. It mainly emphasizes on the quantification of power over-generation potentials due to large scale integration of RES. The model indicates that over-generation potentials in the range of 0.85 to 65GW are possible for the capacities and economic conditions considered in this work.

The PtG and PtL concepts add flexibility to the energy system by linking power to gas/liquid fuels that can be used in other sectors. This link is important because intermittent RES inherently produce electricity. According to this study, as much as 11% of the gasoline in the transportation sector can be displaced with methanol produced from excess intermittent power.

In addition, PtG and PtL provide the opportunity to recycle large volumes of captured CO2, as much as 15 Mtons/year, into the fuel supply system. Furthermore, PtG and PtL complemented penetration of RES into the power sector allows deep decarbonization. For instance, depending on carbon and fossil prices, 22 to 103 Mtons of CO2 emissions can be avoided due to direct substitution of fossil use in the power sector with RES.

Under the assumed economy and operating conditions of the SOECs (i.e. 70% conversion efficiency and intermittent operations), the results indicate these technologies can enable greater
integration of renewables into the energy system. In particular, under globally enforced policy measures towards fossil CO₂ emissions these technologies could play the role of diversifying electricity into other energy sectors as well as the role of long-term storages.

Nomenclature

BECCS  Bioenergy with carbon capture and sequestration
BC  Base case
bIGCC  biomass integrated gasification combined cycle
CHP  Combined heat and power plant
ENTSO-E  European Network of Transmission System Operators for Electricity
FFP  Fossil fuel price
RES  Renewable energy sources
SOEC  Solid oxide electrolysis cell
SNG  Substitute natural gas
PtG  Power-to-gas
PtL  Power-to-liquids

References

[9] Chen L, Chen F, Xia C. Direct synthesis of methane from CO2–H2O co-electrolysis in


[40] Leduc S. Development of an optimization model for the location of biofuel production plants, PhD Thesis. Luleå University of Technology, 2009.

Appendix A. Cost of technologies

The capital cost of building each type of technology is collected from different sources. Table A1 summarizes the parameters of the reference blGCC technology considered. Costs of other plant capacities are scaled based on the reference plant using the power law of capacity with a scaling exponent of 0.7. Table A2 summarizes the cost of technologies and economic parameters used in relation to PtG, PtL, solar, wind and hydropower technologies. For consistency reason, capital cost estimates are based on future projections for the year 2020, see e.g. [19,24,50]. All cost data refer to Euro value of the first quarter of 2010, assuming currency conversion factor of 1.30 $/€. The investment costs are amortized over the operational life time of the respective technology by assuming 5% interest rate and 25 years of economic life time.
For hydropower and bioenergy systems the investment and O&M costs are pre-calculated based on resources potential in every demand area and supplied to the model as parameters. In the model, coal and natural gas are set to satisfy deficit in energy supply and the associated costs are accounted in terms of the energy carrier market value. Whereas for the rest of the technologies, estimation of capital and O&M costs are internalized in the model based on capacity factors and capacity limits.

**Table A1.** Input data for the reference bioenergy production technologies [51–53]. All costs are adjusted to €2010 using Chemical Engineering Plant Cost Index (CEPCI) 2010. Efficiencies refer to the LHV of biomass on dry basis.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Tech1</th>
<th>Tech2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum size</td>
<td>t biomass/hour</td>
<td>6.35</td>
<td>33.88</td>
</tr>
<tr>
<td>Base plant capacity</td>
<td>MW</td>
<td>6</td>
<td>30</td>
</tr>
<tr>
<td>Base investment cost</td>
<td>M€/year</td>
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<td>11.75</td>
</tr>
<tr>
<td>O&amp;M cost</td>
<td>€/GJ biomass</td>
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</tr>
<tr>
<td>Heat efficiency</td>
<td>%</td>
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<td>40</td>
</tr>
<tr>
<td>Power efficiency</td>
<td>%</td>
<td>35</td>
<td>45</td>
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</table>

**Table A2.** Cost of conversion technologies and economic parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>PtL</th>
<th>PtG</th>
<th>Solar</th>
<th>Wind</th>
<th>Hydropower</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capital cost</td>
<td>1000(^a)</td>
<td>800(^a)</td>
<td>3750(^a)</td>
<td>1980(^a)</td>
<td>4000–5000(^a)</td>
<td>€/kW</td>
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<td>Economic life time</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>years</td>
</tr>
<tr>
<td>O&amp;M fixed</td>
<td>5(^b)</td>
<td>5(^b)</td>
<td>5.14(^d)</td>
<td>6.84(^d)</td>
<td>0.03–0.185(^d)</td>
<td>(%)</td>
</tr>
<tr>
<td>O&amp;M variable</td>
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<td>0</td>
<td>6(^b)</td>
<td></td>
<td></td>
<td>€/MWh</td>
</tr>
<tr>
<td>Electricity</td>
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<td>50</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>CO(_2)</td>
<td>20</td>
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<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>Water</td>
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<td>2</td>
<td></td>
<td></td>
<td></td>
<td>€/ton</td>
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<tr>
<td>Conversion efficiency</td>
<td>70</td>
<td>70</td>
<td></td>
<td></td>
<td></td>
<td>(%)</td>
</tr>
</tbody>
</table>

\(^a\)Capital cost includes both the SOEC assembly as well as the synthesis plant from syngas to methanol in the case of PtL [19] and syngas to methane in the case of PtG [20].

\(^b\)Fixed O&M cost as \(\%\) of the corresponding capital cost [19].

\(^c\)Non-tracking commercial solar PV technology with 4kW (DC) installed capacity is considered for this study. The capital, fixed O&M cost are adopted from [50]. The capital cost estimates are expected to have uncertainties of +25%. 

\(^d\)Fixed O&M cost for solar and wind technologies in €/MWh [50].

\(^e\)Capital cost estimate reported here is for onshore wind turbines, with expected uncertainties of less than +25% [50].
Capital cost of new hydropower systems are averaged ranges. In general, typical capital cost estimates vary between 4000–5000 €/kW depending on plant size. These values are averaged from maximum and minimum estimate ranges of 2500–10000 $/kW for plant sizes less than 1MW, 2000–7500 $/kW for plant sizes 1–10MW and 1750–6250 $/kW for plant sizes greater than 10MW. Capacity levelized capital cost estimate of 3500 $/kW (with uncertainties of +35%) is reported in literature [50], which lays within the above range.

Total O&M cost (in €/GWh) for hydroelectric are averaged ranges. Depending on the size of the plant O&M cost can vary between 0.03–0.185 $/GWh. These values are averaged from maximum and minimum estimate ranges of 55–185 $/MWh for plant sizes less than 1MW, 45–120 $/MWh for plant sizes 1–10MW and 40–110 $/MWh for plant sizes greater than 10MW. Accordingly, the capital and O&M cost for every new hydropower installation is estimated beforehand based on the river catchment potential of each demand area and input to the model as parameters.

Variable O&M for hydropower (€/MWh) [50]. Already included in the total O&M cost.

Appendix B. Energy prices

The prices of energy, by sector and country, used in this study are summarized in Table B1 [31].

Table B1. Energy prices (€/GJ) used in this study [31]

<table>
<thead>
<tr>
<th>Country</th>
<th>Heating</th>
<th>Transport</th>
<th>Power</th>
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</thead>
<tbody>
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<td>France</td>
<td>6.8</td>
<td>12.0</td>
<td>13.6</td>
</tr>
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<td>Germany</td>
<td>7.9</td>
<td>12.3</td>
<td>21.1</td>
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<tr>
<td>Italy</td>
<td>9.5</td>
<td>13.9</td>
<td>22.5</td>
</tr>
<tr>
<td>Slovenia</td>
<td>5.1</td>
<td>12.0</td>
<td>20.0</td>
</tr>
<tr>
<td>Switzerland</td>
<td>6.8</td>
<td>11.3</td>
<td>21.1</td>
</tr>
</tbody>
</table>

Appendix C. CO2 emission factors

Table C1. Emission intensities (kg-CO2/GJ) for displaced fossil energy carriers [31]

<table>
<thead>
<tr>
<th>Country</th>
<th>Heating</th>
<th>Transport</th>
<th>Power</th>
</tr>
</thead>
<tbody>
<tr>
<td>Austria</td>
<td>86.2</td>
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<td>87.3</td>
</tr>
<tr>
<td>France</td>
<td>72.1</td>
<td>78.1</td>
<td>39.3</td>
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<tr>
<td>Germany</td>
<td>88.2</td>
<td>78.1</td>
<td>200.8</td>
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<tr>
<td>Italy</td>
<td>70.6</td>
<td>78.1</td>
<td>200.8</td>
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<td>Slovenia</td>
<td>98.6</td>
<td>78.1</td>
<td>158</td>
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<tr>
<td>Switzerland</td>
<td>76.9</td>
<td>78.1</td>
<td>32</td>
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