



IEA Bioenergy
Technology Collaboration Programme

Gasification applications in existing industrial and agricultural infrastructures for production of sustainable value-added products

Case study 1: Entrained flow biomass gasification in the pulp and paper industry

IEA Bioenergy: Task 33

December 2021





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Case study 1: Entrained flow biomass gasification in the pulp and paper industry

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BACKGROUND

Large volumes of forest biomass are being used for pulp and paper making in the world. The sulphate or Kraft process is the most dominant production method of chemical pulping and accounted for by far the largest share of the global pulp production in 2019¹. Figure 1 shows a generic process scheme of a typical forest-based Kraft pulping process.

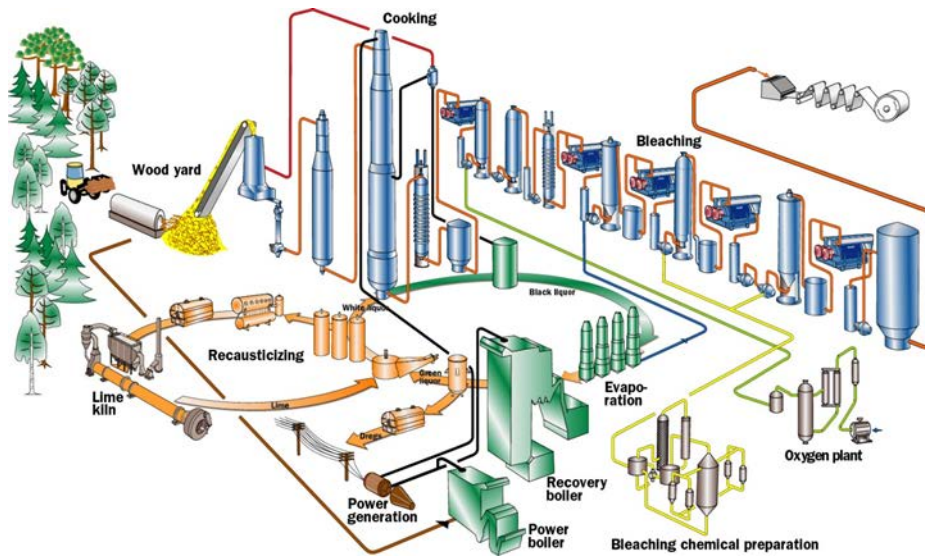


Figure 1. Schematic description of a Kraft pulp and paper mill process (Source: Kvaerner Pulping)

Pulp and paper plants are considered as highly suitable hosts for integration of new biorefinery concepts. Amongst them, thermal gasification-based concepts, where internal by-products or side streams can be converted to energy carriers and products, either for internal use or for export. The product gas from gasification can for example be used as a fuel in boilers and kiln ovens for high temperature heating purposes or upgraded in more advanced applications such as synthesis of chemicals and biofuels². Additionally, the gas can be used for drying purposes, for example in blow-dryers for paper or pulp drying.

Integration of gasification in pulp and paper industries can be beneficial from many perspectives. There are large opportunities for

- an efficient heat integration
- reduced investment costs by co-use of existing processing equipment and logistic synergies
- the experiences of feedstock management and processing and the utilization

There are a handful of pulp mill integrated biomass gasifiers in operation today. These mainly produce a fuel gas for combustion in lime kilns. In Finland, the forest industry company Metsä

¹ FAOSTAT, 2021

² Pio et.al, 2020

Group has a 48 MW_{th} biomass gasification plant at its Metsä Fibre Joutseno pulp mill, where a Circulating Fluidized Bed (CFB) gasifier produces a product gas for combustion in the lime kiln. Metsä Fibre has also a bark gasifier in their Äänekoski bioproduct mill for the same purpose (Valmet, 2021). Additionally, a 12 MW_{th} CFB gasifier is providing fuel gas to Stora Enso's lime kiln at Varkaus³

Other integrated gasification units include a 65 MW_{th} CFB gasification plant at the Zhanjiang Chenming pulp and paper mill in China. Here, the product gas replaces the heavy fuel oil previously used in the lime kiln⁴. The same pulp mill company has installed an 80 MW_{th} CFB gasification plant in its new greenfield pulp mill, Shouguang Meilun also in China. There are also two twin biomass gasifiers in operation at the OKI pulp mill in South Sumatra, Indonesia. The gasifiers are also based on the CFB technology and their capacity is 110 MW_{th} each.⁵

Recently, the technology group ANDRITZ received an order from the largest paper producer in Brazil, Klabin, for a complete biomass gasification plant for their mill in Ortigueira. The scope of supply includes a 51 MW_{th} gasification plant, a belt dryer, a multi-fuel lime kiln burner and biomass handling equipment with auxiliaries. By replacing 100 % of the heavy fuel oil currently burned in one of the mill's lime kilns, the plant will significantly reduce the mill's carbon footprint⁶.

In the Netherlands, Eska Graphic Board converts paper rejects into gas via gasification in a 12 MW_{th} air blown CFB-gasifier. The product gas is combusted in a waste heat recovery boiler to produce saturated steam used in the board-processing machinery.⁷

No integrated biomass gasifier for production of higher value-added products, such as advanced biofuels or chemicals, is however in operation at the time of writing this report. However, a large number of techno-economic studies have been published in the field.

The previously mentioned Kraft pulping method produces after the cooking process (see Figure 1), a residual stream in form of black liquor. The black liquor mainly consists of dissolved lignin, spent pulping chemicals and water and is dried and burned in a Tomlinson-type recovery boiler to simultaneously recover the cooking chemicals and generate process steam and electricity. A literature review found that the most investigated mill-integrated refinery concept is gasification of the black liquor to produce various advanced biofuels⁸. The concept was initially proposed already in the 1960s and several designs have been developed in the intervening decades; however, none of them have seen large-scale commercial implementation.⁹

The drivers behind the development have also undergone a shift, from a desire for increased safety in the 1960s and 70s to power generation in the 1980s and 90s, and more recently, to

³ IEA Bioenergy, 2016

⁴ Andritz, 2021

⁵ Valmet, 2021

⁶ Andritz, 2020

⁷ IEA Bioenergy, 2020

⁸ Malek, 2018

⁹ Furusjö et.al, 2017

the production of advanced biofuels¹⁰. It is for this latter purpose that the Swedish company Chemrec AB developed an oxygen-blown entrained-flow version, which was demonstrated in a 3 MW_{th} pilot facility for more than 28 000 hours in between the years 2005 and 2016¹¹.

Chemrec AB was pursuing a project to scale-up the process to a first commercial plant at the Domsjö sulphite mill in Örnsköldsvik, Sweden. This plant would use the Chemrec front-end gasification technology combined with technology from the petrochemical industry to produce 100 000 tons per year of bio-methanol. The investment was estimated to EUR 300 million, and a support of EUR 50 million was awarded for the Swedish demonstration program after EU state-aid scrutiny. However, after a change in ownership of the mill, the plant Domsjö decided to cancel the project, mainly due to too large market uncertainties.¹²

Objectives and scope

This report gives an overview and summary of recent studies on integrated entrained flow biomass gasification in Kraft pulp mills for production of advanced biofuels or green chemicals. The aim is to illustrate the main techno-economic consequences of integrating such gasification and synthesis concepts. This is done by reporting of the resulting energy balances and economic performances. Table 1 shows an overview of the considered cases divided into type of technology pathway and main product including literature references.

Appendix 1 presents characteristics and chemical compositions of the liquid feedstocks considered in this summary.

Table 1. Overview of considered cases.

Technology/Product	Methanol	Ammonia	Petrol
Gasification of black liquor	Carvalho et.al, 2018		Jafri et.al 2020 Wetterlund et.al 2020
Co-gasification of black liquor & pyrolysis liquid	Carvalho et.al, 2018		Jafri et.al 2020

¹⁰ Whitty, 2009

¹¹ Furusjö et.al, 2017

¹² IEA Bioenergy, 2018

Technology/Product	Methanol	Ammonia	Petrol
Co-gasification of black liquor & raw glycerol	Carvalho et.al, 2018		
Gasification of woody biomass	Andersson et.al, 2014		
Gasification of bark & woody biomass		Andersson et.al, 2015	

INTEGRATED ENTRAINED-FLOW GASIFICATION IN KRAFT PULP MILLS

Two main gasification integration options are considered in this summary report: (i) replacing the bark boiler of a pulp mill with an entrained-flow biomass gasifier, and (ii) full or partial utilization of the black-liquor stream for entrained-flow gasification. Note that the presented cases are not always directly comparable due to different techno-economic assumptions and conditions. However, all relevant background data can be found in the referred publications. Also note that the majority of the pulp mills in the presented cases are state-of-the-art market pulp mills with an electricity surplus.

Replacing the bark boiler with a pressurised entrained-flow biomass gasifier for methanol and ammonia production

Andersson et.al (2014) and Andersson & Lundgren (2015) investigated the techno-economic consequences of replacing the bark boiler in an existing pulp mill with pressurized entrained-flow gasification to produce methanol and ammonia, respectively. The considered pulp mill was an existing, partly integrated, mill with a maximum production capacity of 890 air dried tonnes (ADt) per day of Kraft pulp and 460 tonnes per day of paper.

The most important constraints in these studies were to maintain the pulp production and the process steam balance. Hence, the gasification and synthesis units were dimensioned to supply the same amount of process steam as the bark boiler did. Falling bark originally used for firing the bark boiler was used together with a supplementary amount of biomass as additional fuel to be able to maintain the pulp mill's steam balance. The purge gas from the methanol synthesis was used to fully or partly replace the current fuel used in the lime kiln,

which in these cases liberated tall oil to become available for sale to the market.

Comparisons were made with stand-alone gasifiers with the same thermal inputs operated in parallel to the pulp mill, but with no energy or material exchanges.

Energy performance

Figure 2 shows the resulting normalized energy balances for the methanol production case, normalized to production of 1 MW of product. The annual production for the actual methanol production case was 1.5 TWh per year.

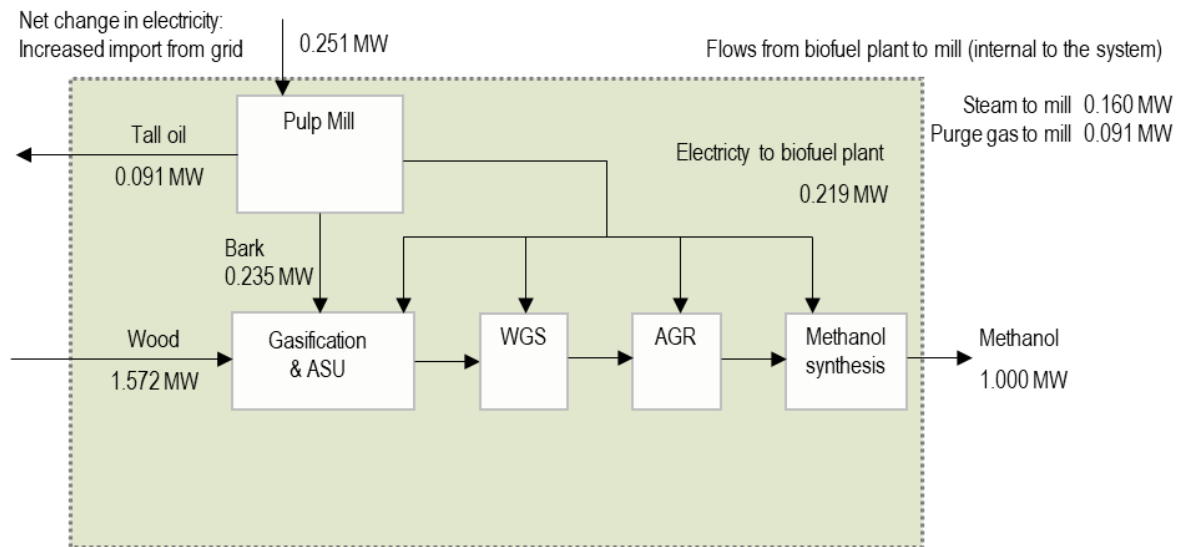


Figure 2. Normalized energy balance for integrated gasification of woody biomass (powder) for methanol production in an existing Kraft pulp mill. ASU stands for air separation unit, WGS stands for water gas shift, while AGR stands for acid gas removal. (Based on Andersson et.al 2014)

The biomass-to-methanol efficiency was calculated to 55 percent on energy basis, while the overall energy system efficiency was 52 percent taken into concern the incoming and outgoing energy flows over the system boundary represented by the dashed box. Re-calculated to electricity equivalents¹³ the system efficiency was 57 percent. The integrated case resulted in seven percent-points higher overall system efficiency than for the stand-alone production unit (calculated to 50 percent, in electricity equivalents).

An important consequence of the integration was however that the imported electricity to the pulp mill increased quite substantially compared to the original mill operation. This was partly due to a decreased internal electricity production and partly due to a higher steam demand following the integration of the new processes.

The main reasons for the higher energy system efficiency of the integrated case were that (i)

¹³ Energy carriers converted to electricity equivalents according to the efficiency of the best-available technologies. This is done to take into concern the exergy of the energy carriers, see for example Malek (2018).

the available surplus of biomass (bark) could be more efficiently converted (e.g., gasified instead of combusted), and that (ii) due to the integration, tall oil could be liberated and exported.

Figure 3 shows the resulting normalized energy balances for the same case, but for ammonia production. The annual ammonia production capacity was approximately 1.2 TWh per year.

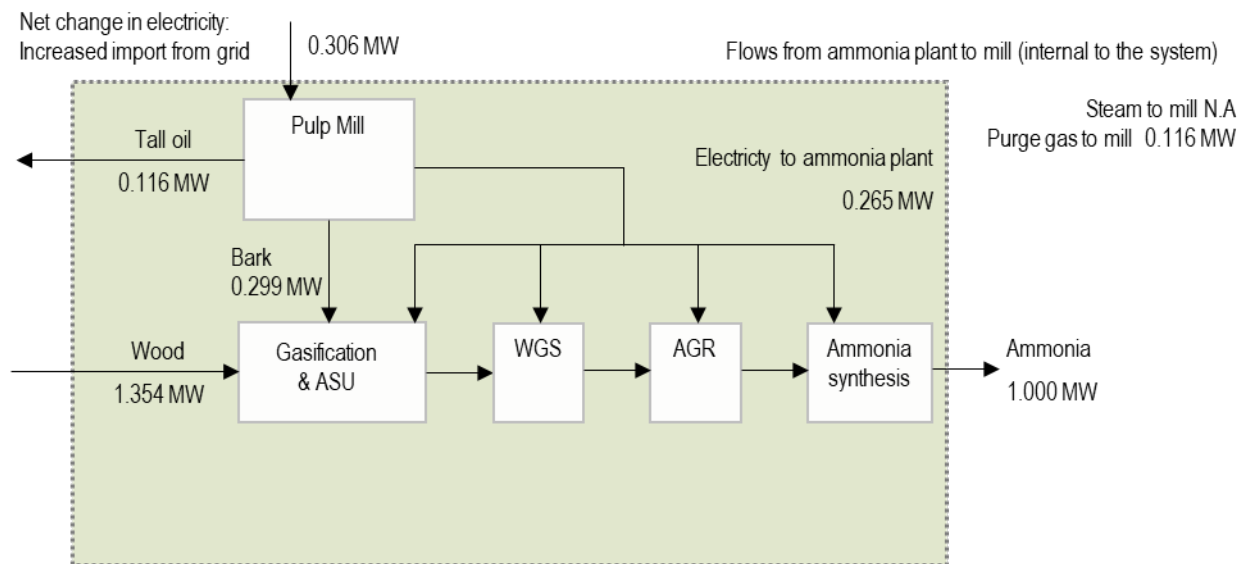


Figure 3. Normalized energy balance for gasification of woody biomass (powder) for ammonia production in an existing Kraft pulp mill. ASU stands for air separation unit, WGS stands for water gas shift, while AGR stands for acid gas removal. (Based on Andersson & Lundgren, 2015)

The process steps required for ammonia production allow a high steam recovery per unit thermal input. Therefore, the thermal input was lower in this case compared to the methanol case (see Figure 2).

The integration led to a 50 percent increase of external biomass supply to the mill as well as a decreased internal electricity production. At the same time, the electricity consumption was almost doubled compared to the original operation of the pulp mill. The calculations showed, however, that the overall energy system efficiency was 10 percent-units higher than for the stand-alone case. This was mainly due to the same reasons as in the methanol case e.g., available internal surplus of biomass (bark) was more effectively converted, and that purge gas could liberate tall oil.

The resulting overall energy system efficiency for the ammonia case was however lower than that of the methanol case, mainly due to the higher specific power requirement per produced unit.

Economic performance

In both cases, the economic performances were better for the integrated concepts, than for the stand-alone production units. The methanol production cost for the integrated case was calculated to approximately 110 EUR per MWh, which was around 15 percent lower than for the stand-alone case.

Generally, ammonia produced via biomass gasification is associated with a high capital and

power intensive process, primarily the synthesis loop and its operating conditions. Approximately 45 percent of the total investment cost was directly connected to the investment of the ammonia synthesis loop. One conclusion of the study was that the production capacities of the studied system configurations (228,000 ton per year of ammonia) were too small to reach economic viability at the current conditions and ammonia price levels. However, by integrating the ammonia production in a pulp mill, the production cost amounted to 458 EUR per ton, which was 12 percent lower compared to the non-integrated, stand-alone alternative (523 EUR per ton).

More details can be found in Andersson et.al (2014) and Andersson & Lundgren (2015).

Entrained-flow black liquor gasification for production of methanol

Black liquor gasification is one of the technologies with the highest level of integration with the pulping process. The integration in between the mill and gasification plant takes the form of materials, namely, black liquor and green liquor, as well as energy exchange. The process can in combination with downstream synthesis for production of value-added fuels and chemicals, potentially replace the recovery boiler of the kraft pulp mill.

Black liquor is an excellent gasification feedstock, mainly due to the catalytic effects of its alkali metals that allows full carbon conversion at relatively low temperatures (1000-1050°C) and at residence times in the order of seconds¹⁴

There are numerous studies on the subject showing that black liquor gasification with downstream biofuels production generally result in good economic and energy performance (see for example Consonni et.al, 2009; Pettersson & Harvey, 2012). Larson et.al (2007) examined in detail the technical and commercial viability as well as the environmental and energy impact of gasification-based biofuel production at kraft and paper mills and concluded that this could result in important economic benefits to the pulp and paper industry.

The examples described in this report consider the production of methanol via pressurized oxygen-blown entrained-flow gasification of kraft black liquor. The modelled capacity was 509 MW fuel input on a HHV basis (including sulphur, see discussion below), but the evaluation of the cases was done using generic balances without considering a specific scale. Hence, balances calculated were assumed to be representative also when scaled up or down. Jafri et.al (2020) evaluated a similar case, but where the methanol was transported to a petroleum refinery for synthesis to petrol (see reference for further information).

Energy performance

Resulting energy balance data for the plants are presented in Figure 4. Readers are referred to Carvalho et.al (2018) (methanol) for additional results, details on system modelling and descriptions of the gasification and syngas upgrading setups.

¹⁴ Sricharoenchaikul et.al, 2002; Öhrman et.al, 2012; Jafri et.al, 2016

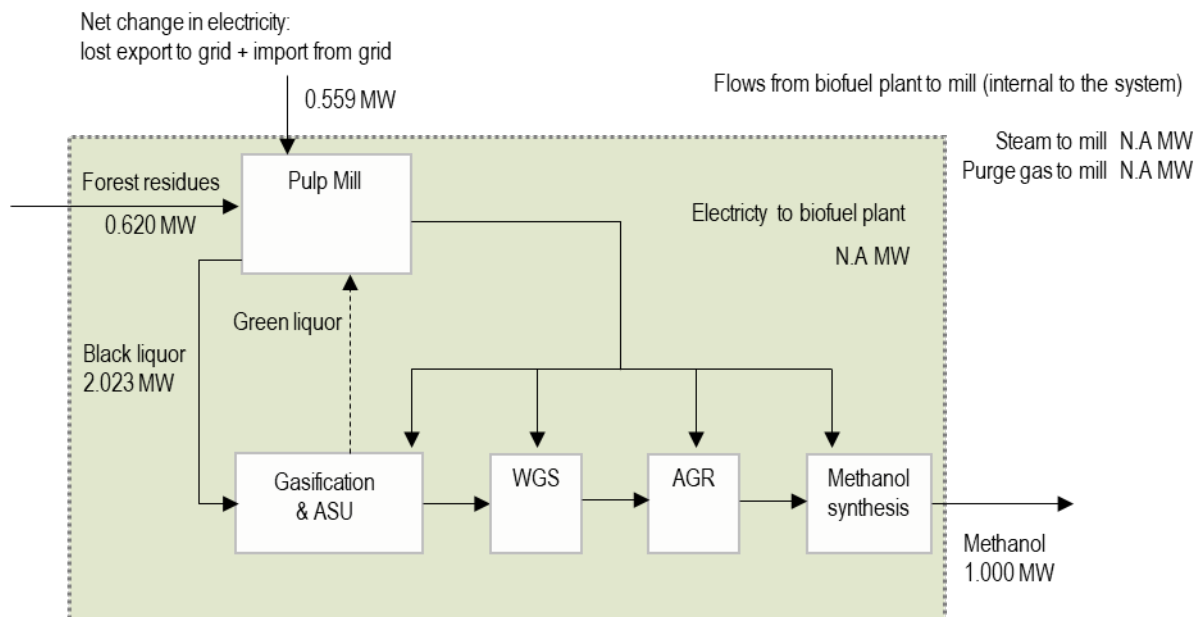


Figure 4. Energy balance is normalized to 1MW HHV of biofuel product. ASU stands for air separation unit, WGS stands for water gas shift, while AGR stands for acid gas removal (Based on Carvalho et. al, 2018)

Here, the recovery boiler was fully replaced by a gasifier that performs the functions of the recovery of pulping chemicals and the production of syngas, used for upgrading to methanol onsite. The gasification and methanol synthesis steps release more heat than is used in the syngas conditioning and cleaning processes and this excess heat, in the form of steam, can be exported to the pulp mill to partly cover the lost steam production in the recovery boiler. Also in this case, purge gas from the methanol plant can be utilized in the mill, in this case for substituting one third of the bark that was used as a fuel in the lime kiln. The freed bark can in turn be used in an expanded power boiler for steam generation. However, the quantities of steam available were not enough, and for every MW of methanol produced, approximately 0.62 MW of forest residues must be added to cover the steam demand of the pulp mill¹⁵.

In the original operation of the pulp mill, electricity could be exported to the grid. After the biofuel integration, however, the mill needed to instead import electricity to satisfy its electricity demand. Hence, for this case, there were two energy supplies to be considered from a system perspective: additional fuel biomass to the pulp mill boiler and the net change in the electricity balance of the pulp mill. Compared to the non-integrated case, additional electricity of 0.56 MW per MW¹⁶ biofuel product was required.

The biomass-to-methanol conversion efficiency was calculated by using black liquor heating

¹⁵ Carvalho et. al, 2018

¹⁶ Note that this figure is the sum of the lost export (the electricity sent to the grid by the mill without biofuel production), and the required power import.

values both including and excluding sulphur oxidation energy¹⁷. Including the sulphur, the efficiency was approximately 49 percent, and without, the conversion efficiency was slightly above 56 percent.

Carvalho et.al (2018) also calculated the marginal overall system efficiency of the production plant to 85 percent. This efficiency is defined as the ratio between the methanol production and the extra fuel biomass to the pulp mill boiler and the net change in the electricity balance of the pulp mill. This efficiency could, for example, be put in relation to the overall energy efficiency of non-integrated, stand-alone gasification-based methanol units, which in general is in the range of 60 to 70 percent¹⁸.

Economic performance

Carvalho et.al (2018) considered a greenfield investment in a new pulp mill. The economic assessment was based on an analysis using the difference between the capital and operating costs of the units required in the integrated and non-integrated pulp mills. Units common to both systems (e.g., debarking, digestion, bleaching, etc.) could therefore be cancelled out and excluded from the analysis. The economic analyses were performed for an Nth plant. The total capital investments were calculated by determining equipment costs and when applicable, installation and indirect costs as well as the balance of the plant (BOP). Cost estimates were based on real estimates from commercial process suppliers as well as on estimations from the literature. For this case, the required methanol selling prices to reach an Internal Rate of Return (IRR) of 15 percent, was calculated to approximately EUR 80 per MWh. More details and a sensitivity analysis are found in Carvalho et.al (2018).

Black liquor gasification with expanded raw material base

The maximum capacity of the black liquor gasifier is determined by the availability of the black liquor, which in turn depends on the pulp production of the mill. This also limits the production of synthesis gas. Additionally, black liquor is an energy-poor fuel and is not transportable. However, it is possible to blend black liquor with a secondary feedstock and co-gasify the blend without reducing the conversion efficiency. The high efficiency of black liquor gasification compared to combustion, is to a large degree due to the strong catalytic effect of the black liquor enhancing the gasification reactions. The reactivity of the black liquor can be maintained even if the black liquor is significantly diluted as illustrated in Figure 5.

Experimental research has confirmed that the reactivity of a blend consisting of black liquor and pyrolysis liquid is very close to that of pure black liquor¹⁹. This means that the thermal input to a black liquor gasification system can be substantially increased by blending. To obtain techno-economic benefits, energy rich and low-cost blend-in feedstocks could be used.

¹⁷ The reason for excluding the sulphur oxidation energy is that the energy is not available in a black liquor gasification process since the sulphur needs to be returned to the pulp mill in reduced state to facilitate pulping chemical recovery.

¹⁸ Börjesson et.al, 2016

¹⁹ Bach et.al, 2015; Jafri et.al, 2017

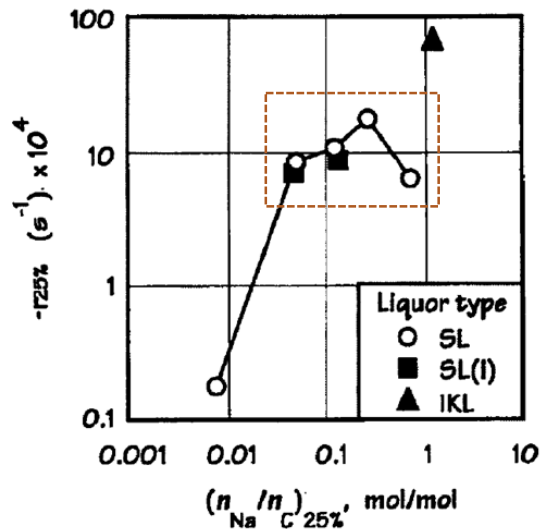


Figure 5. Reactivity of black liquor as a function of the sodium (Na) concentration illustrating that the Na concentration can be decreased substantially without decreasing reactivity (Verril et.al, 1998)

Co-gasification with a blend-in feedstock does not only lead to an increased production volumes and economy-of-scale effects, but also an increased operational flexibility of the plant. The technical feasibility of co-gasification of black liquor and pyrolysis liquid was proven in lab-scale²⁰ and in pilot scale tests in more than 1000 hours of gasifier operation.

Studies have been published regarding the techno-economic performance of co-gasification of black liquor and pyrolysis liquid, mainly for production of grade AA methanol²¹ as well as drop-in fuels in for of petrol²². Carvalho et.al (2018) also evaluated the prospects of producing methanol of different qualities (crude and grade AA) via co-gasification of black liquor and other blend-in feedstocks such as raw glycerol (from biodiesel) and hydrolysis lignin (from cellulosic ethanol). Selections of results and main insights are presented in the following.

Energy performance

Figure 6 shows the resulting normalized energy balances when pyrolysis liquid was co-gasified with black liquor on a fifty-fifty blend ratio for methanol production of grade AA-quality. The total methanol production was 788 MW. Figure 7 shows the normalized balances when 20 percent raw glycerol was added to the black liquor stream for production of grade AA methanol. Here the total methanol production was 451 MW.

²⁰ Bach-Oller et.al, 2015, 2017a, 2017b

²¹ Andersson et.al, 2015, 2016

²² Jafri et.al, 2020

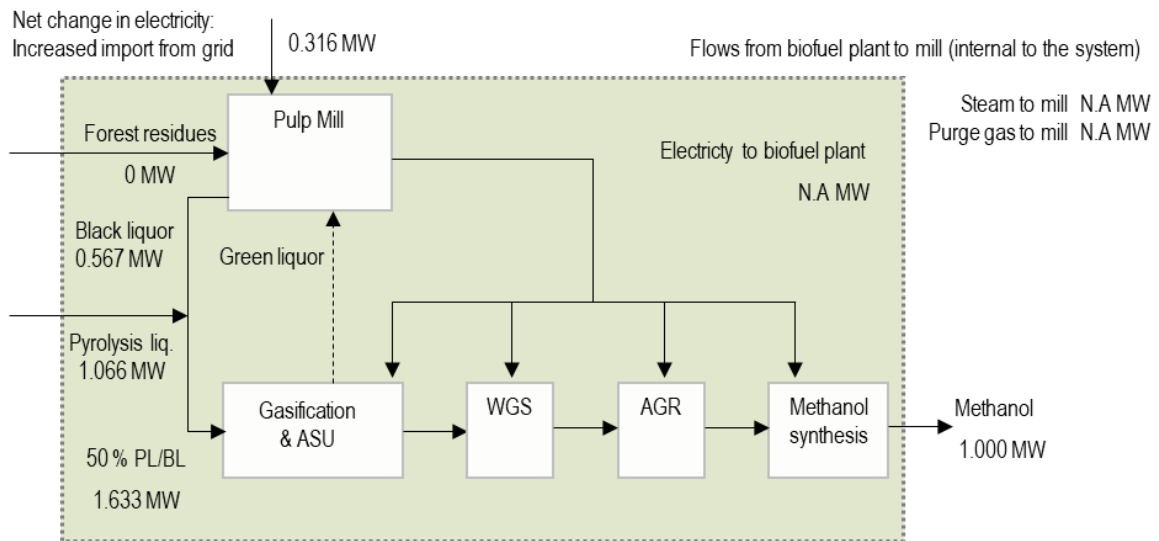


Figure 6. Normalized energy balance when 50 percent pyrolysis liquid on wet mass basis was added to the full black liquor stream for production of grade AA methanol.

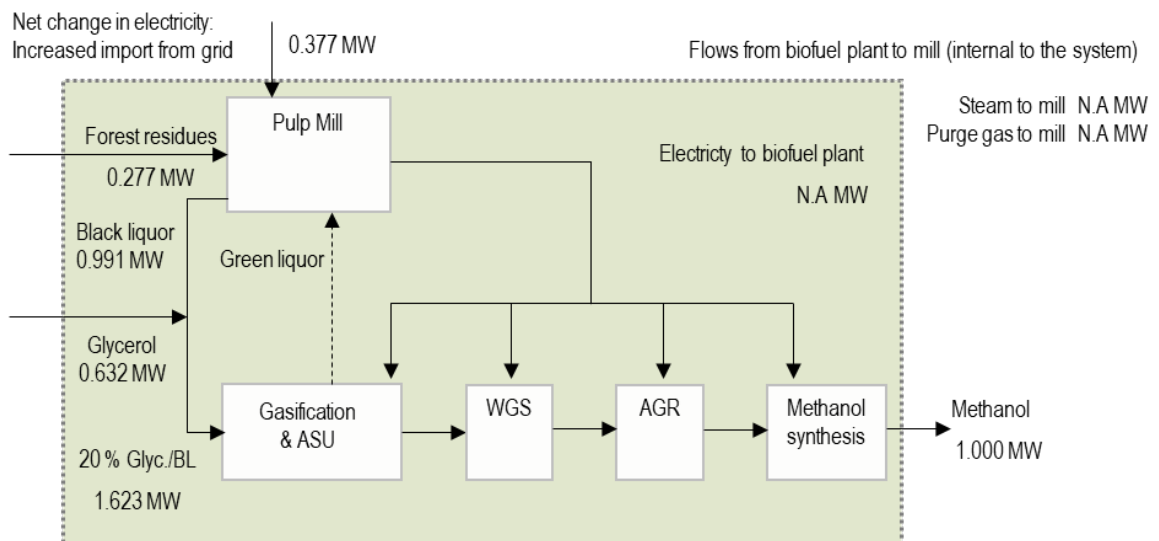


Figure 7. Normalized energy balance when 20 percent raw glycerol on wet mass basis was added to the full black liquor stream for production of grade AA methanol.

By blending a fixed amount of black liquor with a blend-in feedstock with a higher heating value (see Appendix 1), the total thermal input to the plant increases substantially. For a blending ratio of fifty-fifty on wet mass basis, as in the pyrolysis liquid case, the total thermal input was 1 286 MW, or approximately three times higher than if only the pure black liquor stream was gasified. For the glycerol case, the thermal input was 730 MW. Andersson et.al (2016) showed that for mills with pulp production capacities in the range of 200-600 kADt of Kraft pulp per year, the potential methanol production volume via gasification of the full black liquor stream is in the range of 0.6-1.8 TWh per year, while when adding 20-50 percent on wet mass basis of pyrolysis liquid, the corresponding methanol production capacity increases to 0.8-6.5 TWh per year for the same black liquor stream.

Blending secondary feedstocks with black liquor showed positive effects on the cold gas

efficiency as well as on the biomass-to-methanol efficiency. In all co-gasification cases, these efficiencies were higher than that of gasification of pure black liquor.²³

However, the overall system efficiencies²⁴ were 72 percent and 78 percent for the two cases shown above, which were lower than the efficiency for pure black liquor gasification (85 percent). This may seem contradictory, but it emphasizes the strong efficiency improvement of the integration alone. Carvalho et.al (2018) concluded that the largest gain in overall efficiency was caused by the integration itself and not the co-gasification concept. Furthermore, that the efficiency can however increase also with increasing blend-in ratio, but up to a certain level where no excess steam is produced, and no additional biomass fuel supply to the mill is required.

Another important insight of the study was that significant production capacities and consequently economy-of-scale, can be reached by only utilizing a partial stream of the black liquor if blended with a secondary feedstock. This is further discussed later in this report.

Results for other blend-in components and blending ratios can be found in Carvalho et.al (2018).

Economic performance

Co-gasification with an addition of pyrolysis liquid in the blending range of 20-50 percent (on a wet mass basis) applied in pulp mills with a production capacity of about 200-300 kADt per year of kraft pulp, performed better economically than gasification of unblended black liquor. A methanol selling price of in the range of EUR 85 to 100 per MWh would be required to reach a 20 percent IRR depending on pulp mill size. The cost of the pyrolysis liquid is of course critical, but the study showed that co-gasification could be economically beneficial at pyrolysis liquid prices lower than EUR 70 per MWh²⁵.

For larger pulp production capacities than 300 kADt per year of kraft pulp, pure black liquor gasification turned out to be more economically feasible. The main explanation was that the cost for the purchased pyrolysis liquid offsets the positive economy-of-scale-effects reached in the co-gasification case.

It was also found that gasification of pure black liquor as well as black liquor and pyrolysis liquid blends up to 50 percent (on a wet mass basis) resulted in significantly lower production costs compared to gasification of pure pyrolysis liquid²⁶.

Mixing black liquor with raw glycerol could potentially be an economically beneficial option. Carvalho et.al (2018) showed that the required methanol selling price at an IRR of 15 percent could be cost competitive against taxed petrol at a fifty-fifty blend (and if sold as a blend-in component in petrol). Here, the required selling price was around EUR 55 per MWh. At the time of that study, the prices for fossil-derived-methanol in Europe were fluctuating heavily

²³ Carvalho et.al, 2018

²⁴ Defined as the ratio between input and output energy streams crossing the system boundary.

²⁵ Andersson et.al, 2016

²⁶ Andersson et.al, 2016

in the range of EUR 45-85 per MWh.

Gasification of partial black liquor-streams with pyrolysis liquid addition

As previously mentioned, it is possible to extract only a partial stream of the available black liquor and blend with a secondary feedstock to still reach relevant production capacities and economy-of-scale effects. This makes it also possible to keep the recovery boiler. This concept is of particular interest for pulp mills that have reached their capacity ceiling, and where the recovery boiler is a bottleneck for a further production capacity expansion. In such a situation, the mill faces a large investment in expanding the capacity of the recovery boiler, or a very large investment in a completely new recovery boiler. As an alternative, Wetterlund et.al (2020) and Jafri et.al (2020) investigated the opportunities to redirect 10-20 percent of the black liquor to a gasification plant for biofuel production. This means that capacity in the recovery boiler is released, paving the way for an increased pulp production. A prerequisite for this is that any others capacity-limited steps are also remedied. One important benefit compared to biofuel production via gasification of the full stream of black liquor, is that the technical risk is significantly reduced, as, in this case, the recovery boiler can be kept.

Jafri et.al (2020) investigated gasification of a partial black liquor stream blended with pyrolysis liquid for production of methanol for further conversion to petrol at an external petroleum refinery. The syngas was initially upgraded to crude methanol, which was partially distilled to generate a water-containing “stabilized” methanol. This methanol was exported to the refinery, where petrol was synthesized from the stabilized methanol via the methanol-to-gasoline (MTG) process. Here, also small quantities of LPG were obtained as a by-product together with other gaseous by-products from the synthesis loop that were combusted for energy recovery at the refinery. The mill in question was a simulation mill model representing a state-of-the-art market pulp mill with an energy surplus and a pulp production capacity of 2000 tons per day. Total petrol production capacity was 43 MW (based on the higher heating value, HHV).

Energy performance

Figure 8 shows the resulting energy balances when a partial stream of black liquor is blended with 20 percent pyrolysis liquid on wet mass basis for production of methanol, subsequently synthesised to petrol.

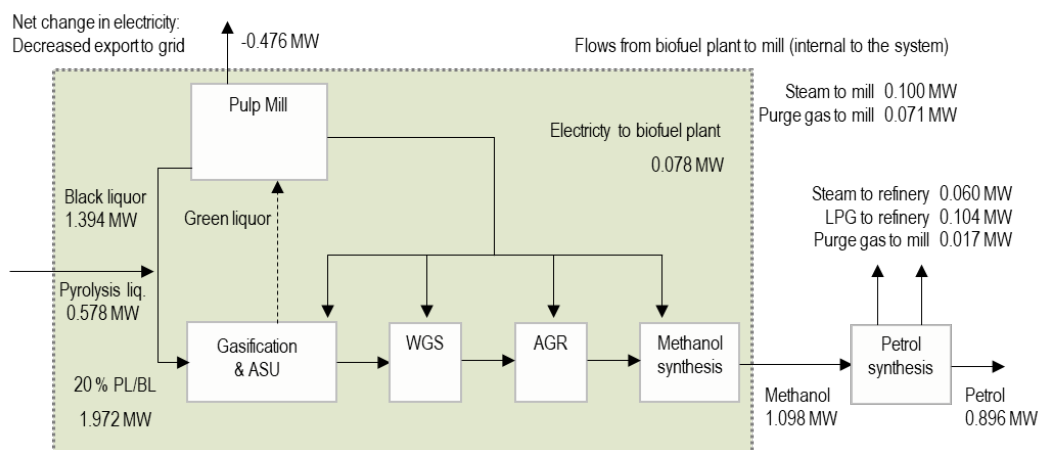


Figure 8. Normalized energy balance when 20 percent pyrolysis liquid on wet mass basis was added to a partial black liquor stream for production petrol.

Jafri et.al (2020) concluded that also when using only a partial black liquor stream for biofuel production, the mill's energy balance was significantly influenced. In this case, the integration of biofuel production led to an increased energy demand at the model mill, which was managed by reduced electricity exports to the grid.

Without considering utilization of side-products and balance changes, the biomass-to-methanol efficiency was calculated to approximately 56 percent, and the biomass-to-petrol efficiency to about 45 percent. If the integration effects at the mill and refinery also were considered, the overall marginal system efficiency was as high as 160 percent. The main reason for this very high value is that low-efficient electricity production from black liquor was replaced by more energy efficient biofuels production.

It should also be mentioned that by decreasing the load of the recovery boiler, an increase in pulp production capacity, estimated to 18.5 percent, was followed²⁷.

Economic performance

In this case, the minimum fuel selling price (MFSP) for the petrol was calculated to EUR 77 per MWh. However, Wetterlund (2020) and Jafri et.al (2020) also studied the impact of pulp mill energy profile on the production costs by examining three different pulp mill configurations as integration sites. The mills had contrasting energy profiles, with one being a state-of-the-art market pulp mill with an energy surplus used to produce electricity (as in the above case), one an integrated pulp and paper mill reliant on energy import for meeting the internal demand, and one a relatively energy-balanced market pulp mill. The results showed that for mills with an existing energy surplus, the production cost was significantly lower than for the other mill types. The import of biomass and electricity due to an increased energy demand has a notably adverse impact on the economic performance and for these mills, the minimum fuel selling price (MFSP) for the petrol was in the range of EUR 91 to 100 per MWh.

CONCLUSIONS

It can be concluded that favorable conditions for integrated biofuels and chemicals production via gasification prevail at Kraft pulp mills, due to, amongst other things, the already available feedstock in form of woody residues and black liquor and the opportunities for efficient heat integration. The overall energy system efficiency is generally higher and the economic performance better if the production is integrated in a pulp mill in comparison to separate stand-alone operations of the mill and the gasification plant.

An important insight was that the pulp mill characteristics such as capacity and type is decisive for what integration concept that is the most favorable from a techno-economic viewpoint. Pulp mills currently operating with an existing energy surplus have a significant advantage as integration sites, while mills that operate their power boilers at, or near maximum capacity may have to invest in extra capacity to meet an increased fuel and

²⁷ Wetterlund et.al, 2020

electricity demand that may follow an integration. Gasification with downstream synthesis to biofuels is for example less appropriate for pulp mills already limited in the lime kiln, due to the increased causticizing load.

Gasification of black liquor with downstream synthesis processes generally result in good economic and energy performance that could result in important economic benefits to the pulp and paper industry. It has also been proven that other biobased raw materials can efficiently "piggy-back" on the black liquor. Blending for example pyrolysis liquid with black liquor for co-gasification is energy efficient and results in potentially large economy-of-scale effects. The production costs were significantly lower compared to gasification of pure pyrolysis liquid. The main finding was that co-gasification is a technically and economically attractive production route for production advanced biofuels.

If only a partial stream is extracted and used for gasification, two primary benefits could be achieved: (i) pulp mills limited by their capacity in the recycling processes have an opportunity for increased pulp production with relatively less investment compared to expansion or replacement of the recovery boiler, while radically reducing the technical risk compared to investment in black liquor gasification on the entire liquor flow, and (ii) some of the obstacles currently existing for investment in a first commercial biofuel production plant based on black liquor gasification can be overcome.

The partial black liquor gasification concept creates improved business benefits for the pulp industry as they get the opportunity to, on one hand, broaden their product portfolio with, in this case, drop-in fuels and on the other, increase their pulp production.

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APPENDIX 1. LIQUID FEEDSTOCK CHARACTERISTICS AND CHEMICAL COMPOSITIONS

	Black liquor ²⁸	Pyrolysis liquid ⁴	Crude glycerol ²⁹	Unit
C	31.9-33.6	61.3	48	[wt.% DS]
H	3.4-3.6	5.6	9.4	[wt.% DS]
N	0.07-0.1	0	0	[wt.% DS]
S	4.6-5.2	0	0	[wt.% DS]
Cl	0.03-0.25	0	0	[wt.% DS]
Na	17.1-19.9	0	0	[wt.% DS]
K	1.23-2.49	0	5.1	[wt.% DS]
O	37.8-39.8	33.1	37.5	[wt.% DS]
HHV	12.2-13.1	23.2	23.7	MJ/kg DS

²⁸ Jafri et,al (2020)

²⁹ Carvalho et.al (2018)

	Black liquor ²⁸	Pyrolysis liquid ⁴	Crude glycerol ²⁹	Unit
LHV	12.5	22.1	21.8	MJ/kg DS
Dry solids	74.8-81.8	75	99.4	[wt.%]



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