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Co-gasification of black liquor and pyrolysis oil: Evaluation of blend ratios and methanol production capacities

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ABSTRACT

The main aim of this study is to investigate integrated methanol production via co-gasification of black liquor (BL) and pyrolysis oil (PO), at Swedish pulp mills. The objectives are to evaluate techno-economically different blends ratios for different pulp mill capacities. Furthermore, the future methanol production potential in Sweden and overall system consequences of large-scale implementation of PO/BL co-gasification are also assessed.

It is concluded that gasification of pure BL and PO/BL blends up to 50% results in significantly lower production costs than what can be achieved by gasification of unblended PO. Co-gasification with 20–50% oil addition would be the most advantageous solution based on IRR for integrated biofuel plants in small pulp mills (200 kADt/y), whilst pure black liquor gasification (BLG) will be the most advantageous alternative for larger pulp mills. For pulp mill sizes between 300 and 600 kADt/y, it is also concluded that a feasible methanol production can be achieved at a methanol market price below 100 €/MW h, for production capacities ranging between 0.9 and 1.6 TW h/y for pure BLG, and between 1.2 and 6.5 TW h/y for PO/BL co-gasification. This study also shows that by introducing PO/BL co-gasification, fewer pulp mills would need to be converted to biofuel plants than with pure BLG, to meet a certain biofuel demand for a region. Due to the technical as well as organizational complexity of the integration this may prove beneficial, and could also potentially lower the total investment requirement to meet the total biofuel demand in the system. The main conclusion is that PO/BL co-gasification is a technically and economically attractive production route for production biomethanol.

1. Introduction

A review of new concepts of biomass gasification concluded that biomass gasification is a key technology that offers high flexibility and efficiency, but also that new knowledge and more efficient and cost-competitive industrial applications are required [1]. The production costs of final products via gasification are strongly dependent on the capacity of the production plants and large capacities are required to obtain beneficial economies-of-scales. Large capacities also increase the required feedstock supply area and put significant demands on the supply chain. Pre-treatment of the feedstock by pyrolysis or torrefaction prior to transportation could potentially improve the economies of the complete biomass logistics chain (i.e. transportation, storage and handling) [2–5].

Heidenreich and Foscolo [1] addressed two different ways to combine gasification and pyrolysis. In the first approach, pyrolysis and gasification are combined directly in a two or three stage gasification process. In the second approach, the pyrolysis and gasification are carried out at different geographical locations. In this concept, pyrolysis is used to concentrate biomass into an oil-char slurry at decentralized small pyrolysis plants for transport of the intermediary products to a centralized large gasification plant. An example of the latter is the bioliq process developed by the Karlsruhe Institute of Technology (KIT) in which low-grade lignocellulosic biomasses, such as straw or forest residues, are utilised [6,7].

A third innovative approach, not addressed in [1], is catalytic co-gasification of a blend of pyrolysis oil (PO) and black liquor (BL). Black liquor gasification (BLG) is a technology which is well proven in pilot scale, with an accumulated operating time exceeding 25,000 h (of which more than 8000 h with downstream biofuel production) in a 3 MW_{th} pilot plant located in Piteå, Sweden [8–10]. The catalytic effect of the alkali metals in the BL (originating in the spent pulping chemicals) allows for a high carbon conversion at lower global temperatures (~1000 °C). Bach-Oller et al. [11] have shown that the BLG process and

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co-gasification of BL and PO can potentially be operated under identical operating conditions. They showed that a similar catalytic effect from BL sodium content can be realized in the co-gasification process, irrespective of the lower alkali content of the mixed feedstock, by experimentally studying single droplet swelling, devolatilization rate and char gasification rate [11]. Preliminary results from initial co-gasification experiments of PO and BL blends in the above-mentioned pilot plant confirm these results. This indicates that co-gasification can achieve a similar catalytic effect from alkali metals and maintain a high carbon conversion at relatively low temperatures (~1000 °C).

It has also previously been shown that a large pulp mill's overall energy efficiency and process economics would be improved by investing in biofuel production via BLG, instead of investing in a new recovery boiler, e.g. [12,13]. The considered mills have a BL availability exceeding 400 MW_{th} and economy-of-scale is one of the reasons for the reported profitability. By blending and co-gasifying PO with BL, the biofuel yield is increased by lower ballast in the system, and the total production volume of biofuels for a given amount of BL is increased. Simultaneously, the operational flexibility is improved and the redundancy requirement is reduced. The co-gasification concept can therefore generate favourable economies-of-scale effects also in smaller pulp mills. Andersson et al. [14] have shown that the profitability of producing methanol in a pulp mill with a BL availability of around 200 MW_{th} would benefit from PO/BL co-gasification, compared to methanol production via pure BLG. The study considered only two fixed blend ratios: 25% and 50% PO addition (on mass basis) to the total wet feed of BL for a specific pulp production capacity (270 kADt/y).

The main objectives of this study are to evaluate biomethanol production via a wider range of PO/BL blends ratios for different pulp mill capacities, in order to identify full-scale co-gasification designs that are economically beneficial when nth plant technology is assumed. The blend cases are compared to three different reference cases for methanol production; (i) via gasification of pure BL only, (ii) via gasification of pure PO in a stand-alone plant, and (iii) via parallel gasification of BL and PO in separate plants. In addition to the techno-economic evaluation of individual mills, the future methanol production potential in Sweden, applying the PO and BL blending concept, is also estimated. Finally, overall system consequences regarding biomass supply requirements are addressed.

2. Methodology and input data

Co-gasification of PO and BL, with different blend ratios, is modelled using simulating thermodynamic models as well as optimising models. Generic Swedish kraft pulp mills with different pulp production capacities are considered as integration sites for the BLG plants. The resulting energy and material balances are used as a basis for the techno-economic evaluation, which includes sensitivity analysis. The overall system efficiency is assessed using electrical equivalents, and the overall system consequences are investigated based on the current Swedish pulp production and recovery boiler capacities and age structure.

2.1. Case description and modelling

The operation of the Swedish Rottneros Vallvik mill was selected as a basis to represent a generic mill. The Vallvik mill produces flash-dried bleached and unbleached kraft pulp, and represents a small to medium sized Swedish pulp mill. Pulp mill steam consumption depends on a number of factors, but in general the specific steam consumption could be expected to decrease with increasing production capacity. Here, however, a conservative approach is applied. Based on the current operation of 200 kADt/y (air dried ton pulp), linear extrapolation is thus used to model five pulp mill sizes in the range from 200 to 600 kADt/y. Key operation data of the pulp mill is summarised in Table 1.

A fifty–fifty PO/BL blend ratio on mass basis is considered as the maximum allowed addition, where the alkali content in the blend can still provide sufficient catalytic effects based on experience of pilot plant sulphite thick liquor gasification [15,16]. Six different co-gasification cases for PO addition are therefore considered, in the range of 10–50% (Cases 10–50) of the available volume of BL on a wet mass basis, corresponding to 2630 kg (73.2% dry matter) BL per ADt. This corresponds to a PO addition in the range of 1250–11,250 kW h/ADt on energy basis to the available BL (5548 kW h_{SF-LHV}/ADt). The considered PO has an LHV of 5.92 MW h/ton dry matter and a density of 1170 kg/m^3. The BL density is 1400 kg/m^3 at the gasifier feed temperature of 140 °C.

The co-gasification cases are compared to three alternative system configurations for methanol production: (i) Case 0, via gasification of the available BL only, (ii) Case 100, via gasification of pure PO in a stand-alone gasification plant, with a PO supply corresponding to the PO input of a fifty–fifty PO/BL blend (i.e. 11,250 kW h/ADt), and (iii) Case REF, which is a combination of Case 0 and Case 100 to quantify potential technical and economic added-values with co-gasification compared to the unblended alternatives.

2.1.1. Gasification plant modelling

The simulations of the gasification process were carried out using a thermodynamic model (SIMGAS) developed for simulating gasification of BL. SIMGAS is implemented in the Matlab platform and based on system Gibbs energy minimization under the assumption of ideal mixtures but with empirical modifications for CH_4 and H_2S as further described in [17]. The tool uses an active-set method to solve the constrained non-linear minimization problem with the chemical composition of gas and smelt phases as independent variables. The model is configured to represent realistic gasifier sizes and heat losses for a commercial implementation of a BLG and a BL/PO co-gasification process. In the simulation, the reactor temperature is fixed at 1000 °C for both BLG and the co-gasification process based on similar reactivity [11]. The specific oxygen consumption is calculated by solving the reactor energy balance at this temperature. Further simulations and model assumptions for Cases 0–50 are described in detail in [14,17].

Due to the absence of catalytic alkali content and a different reactor design, pure PO gasification is simulated using other assumptions regarding reactor temperature and heat losses. An estimated reactor temperature of 1300 °C is used for pure PO gasification, based on information about the Karlsruhe Institute of Technology bioliq plant published before plant start-up [6,7], which also agrees with the pilot scale experiment carried out at the SP Energy Technology Center, in Piteå Sweden [18]. The bioliq

Table 1
Annual average key operation data for the generic pulp mill [kW h/ADt].

	kW h/ADt
BL – LHV dry basis (SF-LHV [a] dry basis)	6473 (5548)
Mill power demand	863
Steam turbine power production	678
Mill steam demand [b]	3084
Lime kiln fuel use	555
Available falling bark (65% DS)[c]	894

[a] Sulphur free – lower heating value.
[b] Mill's demand for medium and low pressure steam.
[c] Prior to the integration of a BLG plant, the falling bark is exported with a 65% moisture content. After the integration of a BLG plant the falling bark is dried to a 35% moisture content and combusted to generate steam.