Energetic and environmental evaluation of two black liquor processing technologies

Miroslav Variny[1,2], Michal Hruška[1], Otto Mierka[1,2]

[1]Faculty of chemical and Food Technology, Slovak University of Technology, Radlinského 9, 812 37 Bratislava 1, Slovak republic

[2]GRUCON s.r.o., Nezábudková 24, 821 01 Bratislava 2, Slovak republic

miroslav.variny@stuba.sk

Abstract

Black liquor (BL) combustion represents a traditional BL processing technology implemented in pulp and papermaking. Alternative ones include BL gasification + combined cycle (=IGCC) among others with literature survey indicating potential of higher electric energy production and lower overall CO_2 emissions compared to BL combustion. To verify this indication we set up material and energy balances for BAT combustion and gasification technology processing BL amounts corresponding to its production in a middle to large size paper mill. The resulting net heat and power productions were compared. Investment costs estimation for both technologies enabled us to calculate the incremental simple payback period of the IGCC. Under current electric energy prices it exceeds 10 years. However the IGCC technology could be economically feasible for pulp and paper industry in case of state support in investment phase or via RES-based produced electric energy bonus. The complex CO_2 emissions evaluation favours the gasification technology.

Key words: *black liquor, combined cycle, gasification, payback period, CO_2 emissions*

Súhrn

Spaľovanie čierneho lúhu (ČL) predstavuje tradičnú technológiu jeho spracovania, uplatňujúcu sa v papierenskom priemysle. Medzi alternatívne technológie patrí jeho splyňovanie s využitím získaného plynu v paroplynovej elektrárni, pri ktorom literárna rešerš vedie k predpokladu vyššej výroby elektrickej energie a následne k nižším globálnym emisiám CO_2 oproti jeho spaľovaniu. Pre overenie tohto predpokladu sme pre BAT technológie spaľovania aj splyňovania ČL v množstve, odpovedajúcom jeho produkcii v stredne veľkej papierni, spracovali materiálové a energetické bilancie a porovnali sme výslednú výrobu elektrickej energie a tepla so zohľadnením vlastných spotrieb. Po odhade investičných nákladov na obe technológie bolo možné vypočítať inkrementálnu jednoduchú návratnosť investície do splyňovania oproti spaľovaniu ČL, ktoré pri súčasných cenách elektrickej energie presahuje 10 rokov. V prípade obdržania štátnej podpory pri investícii alebo formou vyššej výkupnej ceny elektrickej energie z OZE, by implementácia splyňovania ČL v papierenskom priemysle však mohla byť ekonomicky atraktívna. Z hľadiska environmentálneho hodnotenia vyššia netto výroba elektrickej energie pri splyňovaní ČL znamená nižšie emisie CO_2 a iných škodlivín.

Kľúčové slová: *čierny lúh, paroplynový cyklus, splyňovanie, doba návratnosti, emisie CO_2*

Introduction

More than 70 % of total pulp production in European Union stems from chemical pulping [1], where wood chips are cooked in digesters under pressure in an aqueous solution of cooking chemicals that dissolve lignin and hemicellulose, leaving the cellulose fibers for further treatment. Spent chemicals are recovered in the recovery cycle that includes weak spent solution (weak black liquor) evaporation, combustion, quench of chemicals smelt exiting the boiler and lastly the recaustification step. Traditionally the black liquor combustion is carried out in recovery boilers that in turn produce high quality water steam to drive backpressure or condensing-extraction steam turbines. Thus the black liquor combustion provides the majority of steam (and heat) needs of a standard integrated pulp and paper mill with the cogenerated electric energy lowering the necessary power import from outer grid. A paper mill commonly consumes steam at two pressure levels – at a middle pressure level (around 1 MPa) and at a low pressure level (around 0.5 MPa) [1,2].

Starting from early concepts in the 1980's through pilot plant testing and first commercial scale plants in the early 2000's [3] a novel technology for black liquor processing has emerged – its gasification either at atmospheric or elevated pressure, either with air or pure oxygen [2,4,5]. The up to date accumulated experiences with biomass gasifiers operation (including black liquor gasifiers) coupled with combined cycle units – the so called Integrated Gasification Combined Cycle (IGCC) confirm that it has the potential to increase the cogenerated electric energy production compared to that obtained with boilers and steam turbines with some compromise in plant availability [6]. The syngas produced by black liquor gasification contains hydrogen and carbon dioxide among others, offering perspectives for hydrogen separation and purification as well as for carbon dioxide sequestration. Composition of the syngas can vary by changing conditions (operational parameters) in gasifier as demonstrated in papers by Carlsson et al. [4] and Wiinikka et al. [5]. Modelling of black liquor gasifiers and syngas cleaning and utilization gains continuous attention in research projects [7,8] as well as a topic of master and doctoral theses worldwide [9,10]. The aim of all this effort is to analyze most efficient ways of black liquor processing in terms of polygeneration, e.g. simultaneous materials and energies production. Besides the anticipated positive economic effect the polygeneration can also contribute to lowering the global CO_2 emissions.

Our study strives to present an objective independent energy production efficiency analysis of black liquor combustion and gasification process, taking into account latest commercially available development. The system understudy corresponds to a medium to large size integrated pulp and paper mill, producing 1600 t_{DS}/day of 75 % wt. dry solids black liquor. Balances for both systems are set up and their gross and net electric energy production is calculated, while fulfilling the condition of mandatory MP and LP steam export in pre-defined amounts. Difference in electric energy production is then transposed into anticipated national emissions change.

Model system layouts

Black liquor combined steam and power plant (BL CSPP)

The BAT recovery boiler technology includes a modern recovery boiler with enhanced heat recuperation from flue gas, with enhanced regenerative heating of boiler feedwater (BFW) and combustion air, producing very high quality steam with parameters nearly equal to those of power plant live steam [11]. In accordance with this, following boiler parameters were considered:

- Fired black liquor 1600 t_{DS}/day (DS = dry solids), 85 % wt. solids that represents around 7 PJ/year fuel energy based on lower heating value
- Liquor concentration from 75 to 85 % wt. solids performed in an High DS evaporator (HDS) consuming 1,2 MPa (a) steam from steam turbine
- Very high pressure steam (VHP) production 4.25 t/t_{DS}, steam parameters at boiler exit 10 MPa, 510 °C
- Combustion air consumption 4 t/t_{DS}; all air preheated stepwise to final temperature of 210 °C by high-pressure (HP), intermediate-pressure (IP) and low-pressure (LP) steam
- Direct HP steam use for sootblowing in amounts of 5 % of produced VHP steam
- 2 % boiler blowdown rate at 11 MPa; led to blowdown expander and steam utilized in deaerator

Energy system coupled with the boiler encompasses following key components and features:

- Extraction-condensing steam turbine; extractions at 2,3 (a); 1,2 (a); and 0,6 MPa (a); condensing pressure 8 kPa (a); isentropic expansion efficiencies 85, 80 and 70 % considering high pressure, intermediate pressure and low pressure turbine part; mechanic efficiency 95 %
- VHP steam entering the turbine has, due to anticipated heat and pressure losses a by 5 kJ/kg lower enthalpy and a by 0,5 MPa lower pressure compared to state at boiler exit
- Vented deaerator working at 158 °C utilizing LP steam and flash steam with maximum allowed inlet water temperature 138 °C to ensure good deaeration; water heating to 138 °C stepwise by waste heats from paper mill, HDS evaporator condenser and boiler flue gas cooler

- HP, MP and LP steam condensates from BFW and combustion air heating and from HDS condenser are collected in condensates tank, flashed to 0,6 MPa and flash steam is used in deaerator to cover part of its steam need
- BFW pump with overall efficiency of 70 %, delivering deaerated water to the boiler at 12 MPa
- Combustion air and flue gas fans with total Δp of 1.5 kPa and 0,7 kPa respectively and total efficiency of 70 %
- Cooling water pump for steam turbine condenser with overall efficiency of 70 % and total Δp of 300 kPa with maximum allowable cooling water Δt of 15 °C
- Cooling tower fans with total Δp of 250 Pa and total efficiency of 70 %

Schematic process flow diagram is depicted in Figure 1.

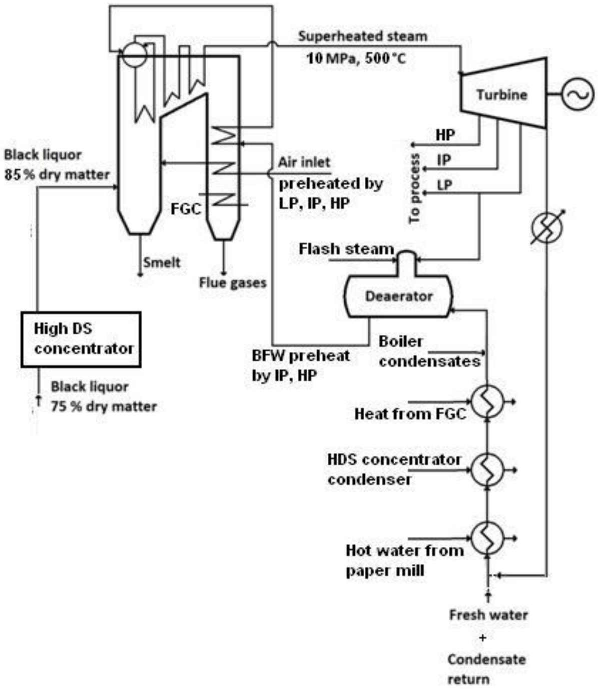

Figure 1 Schematic layout of considered BAT BL CSPP

Black liquor IGCC

General pressurized oxygen-blown gasifier layout and parameters were obtained from study [12] with minor corrections in syngas composition according to [5] and with own equipment changes leading to higher plant efficiency. Syngas exiting the gasifier after flash is available at 3 MPa and 200 °C and is saturated with water steam. It is stepwise cooled down to 40 °C by producing saturated 0,6 MPa (a) low pressure (LP) steam and 0,16 MPa (a) very low pressure (VLP) steam and giving away the rest of its heat content to cooling water. Cold syngas is cleaned in a chemisorber capturing the contained H_2S, with white liquor as absorbent. Cleaned syngas is heated to 100 °C with VLP steam and led to GT combustion chamber. Key features of the combined cycle and auxiliaries are as follows:

- Assumed GT compressor and GT expander isoentropic efficiency is 88 %; its mechanic efficiency is 96 %
- Flue gas leave the combustion chamber at 2,8 MPa (a) and 1300 °C
- Expanded flue gas enters a dual pressure heat recovery steam generator (HRSG) and is contacted with heat exchangers aligned in this order: VHP superheater, VHP evaporator, VHP economizer II, LP superheater, LP evaporator, VHP economizer I, LP economizer, hot water section and exits to stack
- Pinch values of 10 °C assumed generally
- Heat losses of 2 % assumed from each HRSG section
- Produced VHP steam parameters are 6,2 MPa (a) / 450 °C and produced LP steam parameters are 0,6 MPa (a) / 200 °C

- Assumed blowdown rate of 2 % from VHP and LP evaporator as well as from LP and VLP steam generators; blowdown expanded at 0,13 MPa (a) and flash steam used in adjacent deaerator to cover part of its steam needs
- Extraction-condensing steam turbine; extractions at 2,3 (a); 1,2 (a); and 0,6 MPa (a); condensing pressure 8 kPa (a); isentropic expansion efficiencies 85, 80 and 70 % considering high pressure, intermediate pressure and low pressure turbine part; mechanic efficiency 95 %
- VHP steam entering the turbine has, due to anticipated heat and pressure losses a by 5 kJ/kg lower enthalpy and a by 0,2 MPa lower pressure compared to state at HRSG exit
- Vented deaerator working at 105 °C, utilizing VLP steam and flash steam, with maximum allowed inlet water temperature 85 °C to ensure good deaeration; water heating to 85 °C by heat obtained in the HRSG hot water section
- Produced VLP steam is used in deaerator, in absorbent regeneration column reboiler and the rest is compressed in a steam compressor to 0,6 MPa (a) with total compressor efficiency of 60 % and desuperheated to 200 °C with deaerated boiler feedwater
- BFW pump with overall efficiency of 70 %, delivering deaerated water to the HRSG and steam generators at 8 MPa (VHP steam) and 1 MPa (LP and VLP steam)
- Black liquor feed pump to gasifier with total Δp of 3500 kPa and total efficiency of 70 %
- Cooling water pump and cooling tower fans with the same parameters as in the recovery boiler case
- Oxygen production plant with energy consumption 0,5 kWh/Nm^3 oxygen [13] , oxygen consumption 0,3 t/t_{DS} [5].

Schematic process flow diagram is depicted in Figure 2.

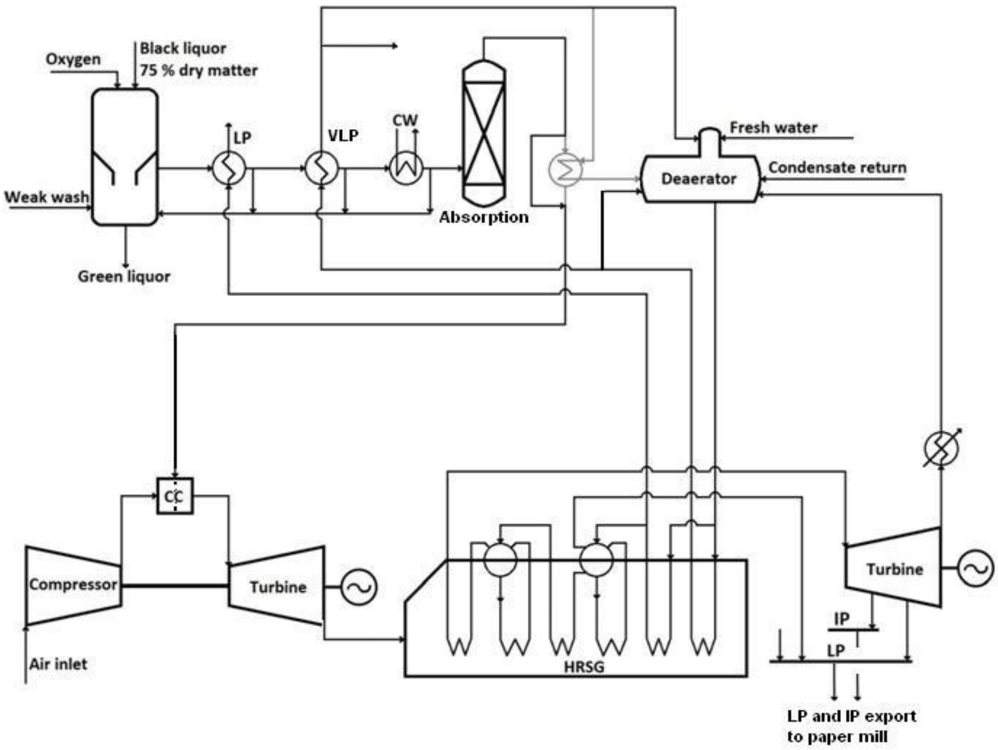

Figure 2 Schematic layout of considered black liquor IGCC

Emission from power production in Slovak republic

Slovak republic produces nearly the same amount of electric energy as it consumes last years. Slovenské Elektrárne, a.s. (SE, a.s.) are traditionally the largest domestic power producer with total market share above 60 %, as documented in Table 1. Sources of electric energy produced by SE, a.s. are the following (rounded up): 80 % nuclear, 10 % water, 10 % fossil fuel (coal).

We decided therefore to implement specific electric energy emission factors of the energy mix of this power producer in our study, obtained from SE, a.s. 2014 to 2016 annual reports [16-18]. An alternative approach considers fossil fuel based marginal power production source that lowers it power production if an increase in power production is achieved by IGCC compared to BL CSPP and applies its specific electric energy emission factors. A substantial difference in emission factors shown in Table 2 emphasizes that the true to be expected emissions change is somewhere within the presented emission factors interval.

Table 1 Key parameters characterizing power production and consumption in Slovakia in the years 2014-2016 [14,15]

Year	Delivered power to grid from SE, GWh	Brutto annual power consumption in Slovakia, GWh	Share of SE on brutto consumption, %
2014	20215	28355	71,3
2015	17892	29579	60,5
2016	17242	30103	57,3

Table 2 Specific emissions from SE, a.s. power production in the years 2014-2016 [16-18]. ** - change in specific emissions in 2016 due to improvement in flue gas cleaning system at brown coal fired power plant Nováky*

Year	Specific emissions				
	CO_2, t/GWh	CO, kg/GWh	SOx, kg/GWh	NOx, kg/GWh	TZL, kg/GWh
2014	121	34,97	1244,2	166,86	15,48
2015	142	39,57	2641,7	217,14	29,79
2016	134	66,35*	370,8*	109,44*	9,80*
Marginal fossil fuel fired source	900	110	500	650	18

Emissions released by BL CSPP and by BL IGCC must also be taken into account. For new modern black liquor boilers following figures serve as good approximation for 8 % vol. oxygen in flue gas: NOx 50 ppmv, SOx 5 ppmv, TZL 37 mg/m^3. For BAT burners for gas turbines NOx emissions are as low as a few ppmv at 15 % vol. oxygen; however 50 ppmv is considered as BAT value in our calculations. SOx emissions are considered zero in the IGCC case as the syngas undergoes cleaning prior combustion in the gas turbine. In both cases the CO_2 emissions are considered the same due to pre-set amount of black liquor to be processed.

Results and discussion

Material and energy balances for both systems have been calculated using the inputs from the preceding chapter. Calculation results are summed up in Table 3, stressing higher power production efficiency of the BL IGCC compared to BL CSPP. On the other hand we see significant amount of steam serving for condensing power production in the CSPP case, meaning that more steam could be exported to the paper mill with a small penalty in electric energy production. With IGCC only small decrease in condensing power production is possible; if more steam is needed in the paper mill it has to be produced in another source a standard paper mill operates – either in a biomass boiler or in a gas boiler, with a consequent economic and ecologic penalty. This aspect is worth further analysis which is a part of our future plans to be done.

Internal electric energy consumption reflects the need for power to drive larger pumps and boiler fans in the CSPP system than in that of IGCC, however more than 80 % of IGCC internal power consumption falls upon the air separation unit that produces oxygen for gasification. Therefore the total internal power consumption is substantially higher in the IGCC system.

What deserves attention as well is the significantly higher steam production in the CSPP system, partly resulting from higher own steam consumption and partly from higher heat efficiency of the system. Higher stack losses are associated with IGCC operation and there is an around 15 % difference between the LHV of the black liquor and the total energy content of the gases exiting the gasifier that indicates quite low energetic efficiency of the gasifier (as mentioned above, the gasifier has not been modelled but the composition and parameters of the exiting gas have been adopted from available literature).

Table 3 Energetic comparison of CSPP and IGCC systems

Parameter/Studied system	**CSPP**	**IGCC**
Total steam production incl. flash steam, t/h	288	144
Internal steam consumption + losses, t/h	92	12
Steam for condensing power production, t/h	83	19
Exported steam LP + MP, t/h	76 + 37	76 + 37
Power produced by gas turbine, MW	-	61,1
Power produced by steam turbine, MW	50,0	9,9
Total gross power production, MW	50,0	71
Internal power consumption, MW	2,6	11,2
Net power production, MW	47,4	59,8 (= +12,2 MW)
Net power production efficiency, %	19,2	24,2 (= + 5 %)

Table 4 Environmental assessment of both systems operation, **assumed 8400 h/year and 70 MW as the total average power consumption of the paper mill, # no applicable data regarding CO and TZL emissions found*

System understudy	**CSPP**	**IGCC**	**Difference**
Power imported to paper mill, GWh/year*	190	87,4	-102,6
Emissions released by SE mix / by marginal source for power production, t/year			
CO_2	25 460 / 171 000	11 712 / 78 660	-13 748 / - 92 340
CO	12,61 / 20,90	5,80 / 9,62	-6,81 / - 11,28
SOx	70,45 / 95,00	32,41 / 43,7	-38,04 / - 51,3
NOx	20,79 / 123,50	9,56 / 56,81	-11,23 / - 66,69
TZL	1,86 / 3,42	0,86 / 1,57	-1,00 / - 1,85
Emissions released by BL CSPP and by BL IGCC#, t/year			
SOx	45,02	Nearly zero	- 45,02
NOx	319,2	356,2	+ 37

Environmental assessment took into account both emissions produced by the systems themselves as well as those produced in order to deliver the remaining needed electric energy to the paper mill. Obtained figures are shown in Table 4. We can conclude that the overall annual emissions decrease in the case of IGCC system compared to the CSPP one is in the order of several tens of thousands tons of CO_2, around 10 tons of CO, nearly 100 tons of SOx, around 1 ton of TZL and the NOx production is almost the same in both cases. The uncertainty in results stems from two possible definitions of the base case for electric energy production – either it is the energy mix of the SE, a.s. or a fossil fuel fired marginal power source.

The difference in net power production of around 100 GWh/year means an increase in the IGCC income compared to Combustion case between 3 to 10 mil. €/year considering electric energy price span of 30 to 100 €/MWh. Obviously the IGCC technology is more costly than the traditional one. Our total investment cost estimate yielded around 80 mil. € for CSPP and 190 mil. € for IGCC system. The difference between those two exceeds 100 mil. € meaning that the marginal simple payback period of the IGCC exceeds 10 years which is a too long period to be economically attractive for industry. This period might be shortened by state support either in form of direct co-financing of the IGCC project or in form of IGCC electric energy bonification. This however deserves further study.

Conclusions

The aim of our study has been to present an objective method for power production potential evaluation for BAT CSPP and IGCC technology applied to black liquor processing. As demonstrated here, for pre-set amount and quality of available black liquor and for pre-set mandatory amounts of steam to be delivered to the paper mill, the IGCC solution offers by around 12 MW (+25 %) higher power production compared to traditional combined steam and power solution. Environmental assessment revealed positive overall effect of IGCC technology in nearly all evaluated items except NOx that seem to be emitted in nearly the same amounts regardless of the technology. A brief economic evaluation showed that the actual difference between the estimated IGCC and traditional technology total investment costs is substantial and the net benefit of +12 MW power production does not ensure attractive IGCC investment pay back period without some additional incentive.

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