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CHP in the pulp industry using black liquor gasification: thermodynamic analysis

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Abstract

Black liquor supplies the major share of the energy used in the production of chemical kraft pulp, the dominant pulping process worldwide. Traditionally, black liquor, which contains dissolved organic and inorganic substances from pulping, is recycled to a recovery boiler that performs the dual function of recovering chemicals and energy. Gasification of the black liquor as an alternative to the conventional recovery system is under intense development, because it offers potential advantages in both functions. The realization of the energy recovery potential of black liquor gasification is strongly connected to changes in the pulp mill's system for cogeneration of heat and electric power. In this work, pinch analysis of the integrated gasification cogeneration system, is used to identify systems which maximize power and heat yield under given process constraints and before considering integration with the mill.

Keywords: Black liquor; Process integration; Gasification; Pulp; Cogeneration

1. Introduction

Black liquor is currently one of the most important sources of energy from biomass (of the order of 3 EJ globally) and supplies the major share of the energy used in the production of chemical kraft pulp, the dominant pulping process worldwide. Traditionally, black liquor, which contains dissolved organic and inorganic substances from pulping, is recycled to a recovery boiler that performs the dual function of recovering chemicals and energy. Gasification of the black liquor as an alternative is under intense development, because it offers potential advantages in both functions, as well as environmental benefits.

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The realization of the energy recovery potential of black liquor gasification is strongly connected to the implementation of changes in the pulp mill's system for cogeneration of heat and electric power. To design an efficient and cost-effective energy system for the mill, it is necessary to be able to predict the effects of these changes ahead of detailed design. It is also desirable to identify possibilities for energy savings that can result from increased integration between the gasification plant and other parts of the mill. The potential impact of a number of combinations of gasification and cogeneration systems on the energy balances of several actual mills has been studied and compared with the conventional technology [1–3]. Mass and energy balance models have been developed for the important parts of the gasification-based cogeneration system, viz. the gasifier, the heat recovery system, the gas turbine and the steam cycle.

The flexibility in meeting the specific demands for heat and electric power of a particular mill is much greater with a gasification and gas turbine based recovery system, with possibilities to design systems with power-to-heat ratios from 0.25 to 1.1, whereas that of the conventional system does not exceed 0.3 [3]. In accordance with other studies [4–6] it has been confirmed that the ultimate potential for power generation is up to 2.5 times that of the conventional system.

In the present study, we have analyzed one of these systems more thoroughly with a view to improving the efficiency through heat integration within the gasification/combined heat and power plant. Although direct integration possibilities with other parts of the pulp mill should be of importance, there will also be situations where it is desirable to limit the integration to exchange of heat via the existing utility (steam) levels. As shown in Fig. 1, in addition to heat integration, the gasification plant will interact closely with the mill in other respects, because the fuel [black liquor (liquor extracted from pulping; mainly lignin, Na and S)] is a product of the process and the dissolved ash [green liquor (the solution containing the inorganic chemicals from the gasifier/recovery boiler; mainly Na_2CO_3 and Na_2S)] is an important intermediate in the preparation of the solution used for pulping [white liquor (the highly caustic solution used for pulping; mainly NaOH, Na_2S)].

2. Black liquor gasification

Several processes for gasification of black liquor are under various stages of development [7–12]. The main dividing line between the different processes is the operation above or below the melting point of the inorganic chemicals which are to be recovered from the gasifier products. Processes proposed to operate at low temperatures ($<700°C$) would employ fluidized-bed gasifiers [10–12]. The potential advantages of low-temperature operation include a higher cold gas efficiency and separation of the inorganics as solids. A disadvantage is the lower reaction rate, which would increase the size of the equipment or may result in incomplete carbon conversion or sulfate reduction. Most of the reduced sulfur is gasified, which calls for equipment to recapture the sulfur, but the resulting "sulfur split" is also anticipated to lead to improvements in the pulping process. In a high-temperature ($>900°C$) process the fraction of gasified sulfur is smaller. At high temperatures the cold gas efficiency is lower, but the reaction rates are higher and the reactor can, therefore, be a smaller, entrained-flow gasifier with a

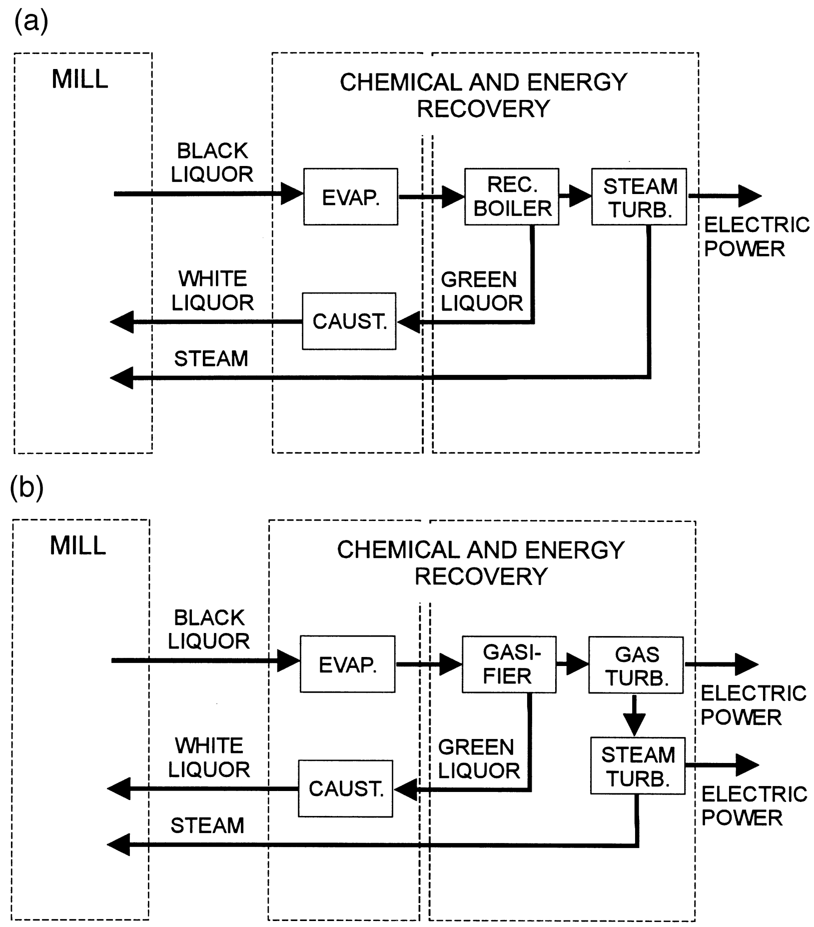

Fig. 1. Simplified schematic representations of (a) a mill with a conventional recovery and CHP system, and (b) a mill with a gasification-based system.

shorter residence time [8, 9]. Care must be taken to avoid corrosion when the inorganics are handled in molten form, much as in the conventional recovery boiler. In the fluidized-bed processes it is important to avoid melting which could cause the bed particles to agglomerate.

Currently, one process is of commercial status. This is a high-temperature, air-blown, entrained-flow gasifier operating at atmospheric pressure. Two plants are in operation, the largest one processing 330 tonne of black liquor solids per day, corresponding to about 60 MW of thermal energy (MW_{th}). For integrated gasification combined cycle (IGCC) applications, the focus is on pressurized systems because the fuel gas must be delivered at a higher pressure than that in the gas turbine combustion chamber. It is more efficient to pressurize the gasification and gas cleaning operations than to compress the flue gas. Of equal or greater importance is the substantial reduction in equipment size that is possible because of the smaller volumetric flow rates. We will limit our discussion here to a system based on a pressurized, high-temperature, entrained-flow gasifier, although the principles can be readily applied to other systems as well. This system includes much heating and cooling in a limited temperature interval, 100–200°C, and pinch technology should therefore be a suitable tool for the analysis.

3. System description

In the present study, the system boundary is drawn to include the gasifier, gas cooling, gas cleaning, gas turbine, steam turbine and heat recovery steam generator (Fig. 2). The remainder of the recovery plant, including black liquor evaporation and white liquor preparation, are not included in the analysis. Thus, the streams crossing the boundaries will be the liquor streams to and from the mill, air, boiler feedwater, steam and return condensate.

Concentrated black liquor (75%DS dry black liquor solids) is injected at the top of the gasifier together with the oxidant (air or oxygen). Final drying, pyrolysis and gasification take place within a few seconds at a temperature of 950°C. The gasifier is nearly adiabatic, so the temperature is controlled by adjusting the oxidant flow rate in order to make the exothermic oxidation reactions balance the endothermic gasification reactions. The products are quenched

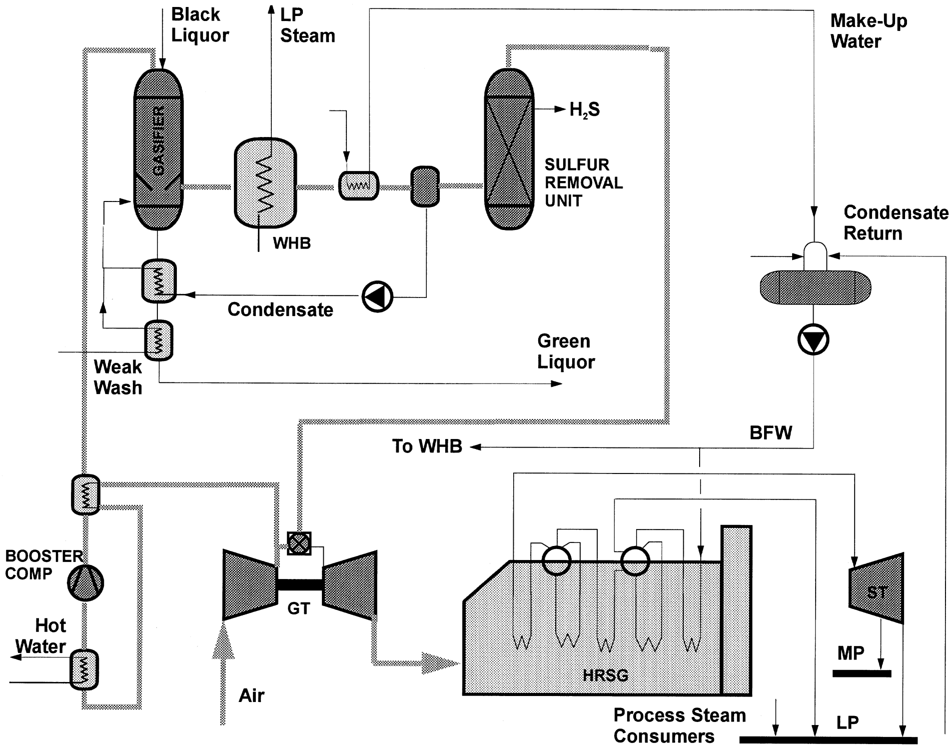

Fig. 2. Example of an IGCC flowsheet with a high-temperature, air-blown gasifier. Abbreviations: waste heat boiler (WHB), gas turbine (GT), steam turbine (ST), boiler feedwater (BFW), low pressure (LP), medium pressure (MP), heat recovery steam generator (HRSG).

with weak wash (dilute liquor recovered from white liquor preparation) and recycled condensate as they exit the gasifier into a gas-liquid separation vessel. Green liquor is drawn from the bottom of the vessel, while the gas (which is saturated at the temperature of the green liquor) exits through a side extraction.

Since some of the sulfur in the black liquor has been gasified, it is necessary to have an efficient system for H_2S removal. Most conventional sulfur removal processes operate at low temperatures (below 100°C) and use absorption in a liquid medium. Low-temperature, wet cleaning also ensures that any gasified alkali is condensed and captured, which is essential for troublefree gas turbine (GT) operation. Cooling of the gas before sulfur removal takes place in a waste heat boiler (WHB), where most of the heat of condensation is recovered along with the sensible heat in the gas. Condensate is separated from the gas and recycled to the quench vessel.

The clean gas from the absorber can be fired in a standard gas turbine. However, since the heating value is considerably lower than that of natural gas, the fuel flow per unit of air to the gas turbine is higher, leading to a larger flow to the expander. The larger flow can be accommodated by: (1) allowing the compressor pressure ratio to increase, (2) reducing the turbine inlet temperature, or (3) modifying the expander inlet nozzles. Two further possibilities are (4) to reduce compressor air flow, or (5) to bleed air from the compressor outlet, in order to minimize the change in mass flow to the expander. The latter measure will be necessary for an air-blown system because the heating value is less than one tenth that of methane, and the fuel flow will thus be more than ten times larger (in effect, this is a minimum because the demand for dilution air is also reduced). For an oxygen-blown gasifier, a combination of 1, 2 and 4 will suffice, provided that there is an adequate margin to avoid compressor surge. Hardware modifications are less likely in the short term, but may be of interest as IGCC technology in general (using other biomass fuels, coal, or oil), becomes a cost-competitive alternative for power generation.

After the gas turbine, hot exhaust flows through a heat recovery steam generator (HRSG) to generate high-pressure steam for a steam cycle; some process steam can also be raised by cooling the flue gas further. Normal steam pressures in the pulp and paper industry are 3–5 bar, with steam extraction at 10–12 bar. Other, intermediate, pressures are sometimes used in paper machines.

4. Method

The gasifier performance was modeled using an in-house global equilibrium model [13], where thermochemical data were taken from a standard reference source [14]. Equilibrium is a justifiable approach for black liquor gasification, more so than for other fuels at corresponding temperatures because the reactions are catalyzed by alkali metals which are abundant in black liquor [15]. A model of the remainder of the gasification combined cycle plant was constructed in HYSYS, a commercial process simulator, using the basic building blocks available (reactors, heat exchangers, etc.). For the pinch analysis we used ProPI, which can be interfaced with HYSYS to extract stream data from a simulated case.

4.1. Simplifications and assumptions

The sulfur removal unit (SRU) and the air separation unit (ASU, for the oxygen-blown system) were not modeled in detail. The SRU is treated as a black box with steam and cooling water consumptions only, thus, accepting the existing internal heat integration as it is. Likewise, the cryogenic oxygen plant is normally contained in a "cold box" where the cold oxygen and nitrogen leaving the plant are used to cool the inflow of ambient air. We modeled only the air compressor that compensates for the pressure drop through the ASU.

The initially assumed target temperatures for the air and gas streams were set by process constraints. An air temperature of 250–500°C is desirable to ensure that the gas heating value is sufficient for stable combustion in the gas turbine. With an oxygen-blown gasifier, the gas heating value is reasonably high and we assumed no preheat. Furthermore, because the oxygen flow is about 80% smaller than the air flow, the amount of heat that may enter the gasifier with the oxidant is much smaller. The oxygen stream has a temperature of about 125°C from the compressor.

Gas cooling, to at least 30°C before the SRU, is necessary for most liquid-absorption processes. Gas reheat is limited to about 450°C, because of fuel valve material constraints [16]. Also, as first assumptions, the target temperatures of the weak wash and the recycled condensate were taken to be 200°C.

The normal boiling point of black liquor at 75%DS is about 115°C, but under pressure the black liquor may be heated above this temperature. Heating reduces the viscosity, allowing the formation of smaller droplets in the burner. We used a target temperature of 150°C.

The minimum stack temperature of the heat recovery steam generator depends on the dew point of sulfuric acid in the flue gas, and hence on the lowest temperature of the cold surfaces in the economizer. For sulfur-containing fuels the minimum allowable metal temperature is about 110°C. However, when firing gasified black liquor, because of the advanced H_2S removal system the flue gas will be nearly sulfur-free from this point of view. For low-sulfur gas, cooling to a metal temperature of abot 70°C is possible with carbon steel economizers [17]. We used a target stack temperature of 110°C for the initial pinch analysis.

All gas turbine calculations were made with a model of a generic "F" generation industrial type turbine, for which component efficiencies, pressure ratio, turbine inlet temperature and cooling air flows have been adjusted to match the manufacturers' data for thermal efficiency, specific work and outlet temperatures. Gas turbines in this class that may match the black liquor flow of an entire mill are, for example, the GE 6001FA and the KWU 64.3, but in the present analysis the gas turbine was not fixed in size. For the oxygen-blown systems the turbine inlet temperature was slightly derated from 1288 to 1250°C and the compressor pressure ratio increased from 15.0 to 15.7, to compensate for the increased mass flow as outlined above. With the air-blown systems it is necessary to bleed air from the compressor outlet and the net increase in mass flow to the turbine is smaller, assumed here to be compensated only by a small rise in pressure ratio.

As a basis for the calculations, the black liquor flow was taken to be 100 tonne DS/h. This number was arbitrarily chosen to simplify the calculations, but it may be noted that this corresponds roughly to the black liquor recovered from a mill producing 500,000 tonne/a of bleached pulp. Table 1 shows the gas and smelt compositions, as obtained from the

Table 1
Black liquor data and output from the gasifier calculations

BLS composition	Mass fraction (%)	Gas composition	Volume. fraction (%)
C	35.2	CO	25.0
H	3.4	CO_2	16.9
S	5.4	H_2O	27.5
O	34.0	CH_4	0.3
Na	19.2	H_2	28.4
K	2.6	H_2S	1.9
Cl	0.2	Gas flow[2] (kg/s)	34.63
HHV (MJ/kgDS)	14.2	Smelt flow[2] (kg/s)	13.06
LHV[1] (MJ/kgDS)	11.7	Oxygen flow[2] (kg/s)	10.66

[1] Corrected for hydrogen and sulfur content.
[2] At a black liquor feed rate of 100 tonne DS/h.

equilibrium calculation for a case with an oxygen-blown gasifier and at a black liquor concentration of 75%DS. With an air-blown gasifier the gas contains more CO_2 and H_2O.

5. Results and discussion

To provide a base level for comparison, the grand composite curve for a case without gas reheat or other regenerative measures is presented in Fig. 3. The grand composite curve includes the HRSG, green liquor cooler, WHB, and some heat from intercooling of the ASU

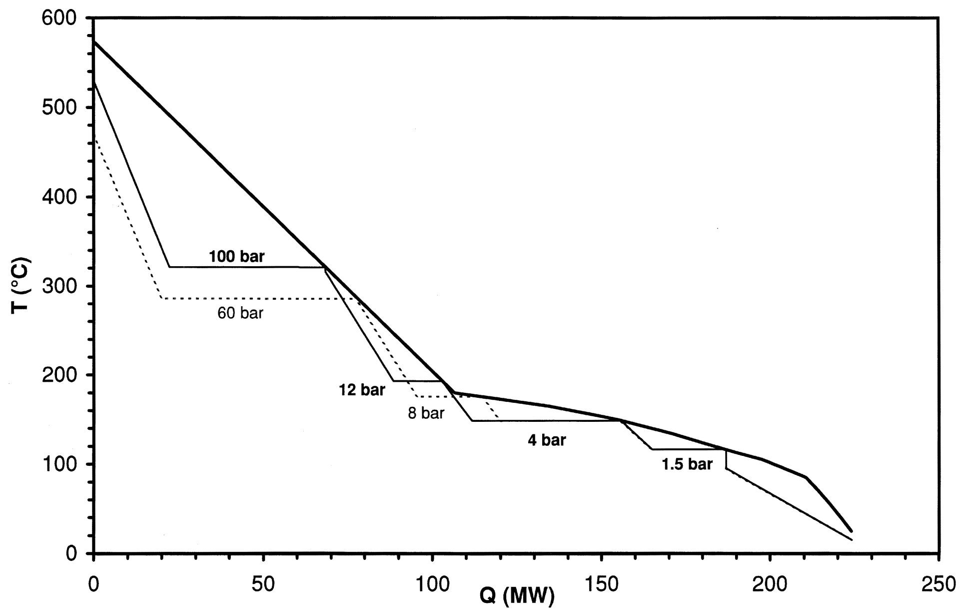

Fig. 3. Grand composite curve for the base level case with examples of two different sets of utility levels.

and oxygen compressors. Roughly 100 MW are available for a steam cycle, and an additional 50 MW to generate low pressure steam. A further 50 MW might be extracted above 100°C, which leaves 25 MW below 100°C. Suitable sinks in the mill for this latter low-level heat may be water for pulp washing and make-up water for the steam cycle. However, mills are moving towards increased water closure, which reduces the need to heat incoming water and may require more process cooling. The gross power produced by the gas turbine is about 85 MW for this case.

Standard utility levels of 12 and 4 bar are shown in Fig. 3, along with a high pressure level of 100 bar for the steam cycle. Also included is a low-low pressure of 1.5 bar which can be used in the deaerator, and may be of possible use in the mill. The steam turbine will generate about 20 MW of electric power. If the steam cycle is laid out for 60 bar instead (corresponding to the normal pressure for recovery boilers), the power produced in the steam turbine will be approximately 17 MW. Also note that other utility levels may provide a better fit with the GCC (e.g. a medium pressure steam level of 8 bar in this case).

5.1. Scope for improvement

5.1.1. Weak wash and condensate heating

In the quench vessel, the gas and smelt are rapidly cooled by weak wash and recycled condensate from 950 to about 200°C. This is a practical way of handling the smelt, but the temperature loss reduces the possibility to convert the sensible heat in the gas into work. Nevertheless, since the aim of the system is to generate both power and heat, the heat from gas and smelt cooling is valuable provided that it is recovered at useful temperatures. The quench cooling relies almost exclusively on the evaporation of water to saturate the gas; the key to heat recovery is therefore that the process is pressurized so that the dew point of the gas is at temperatures high enough for steam generation.

When the very hot gas and smelt are mixed with cold weak wash and condensate, heat will inevitably cross one or several pinches depending on the number of utility levels. By preheating the weak wash and condensate with other streams before they enter the quench vessel, the amount of heat crossing the pinch can be minimized. This will increase the amount of steam that it is possible to generate at each temperature level, as shown in Table 2. For example, by heating weak wash and condensate to 190°C, low pressure steam generation can be raised by 71%, compared with the base case. No more heat is supplied to the system, so the improvement is solely because of the more efficient use of the temperature driving forces within the system. Simultaneously, the amount of heat rejected at temperatures below 100°C decreases

Table 2
Effect on steam production of heating the weak wash and condensate before the quench

Weak wash (°C)	70	100	190	144
Condensate (°C)	30	100	190	144
12 bar steam (MW)	0	0	5	0
4 bar steam (MW)	42	54	72	72
4 bar steam (% incr.)	0%	29%	71%	71%

Table 3
Key results for selected cases

			Oxygen-blown			Air-blown		
Reheat	Gas	°C	30	183	450	183	450	450
	weak wash/condensate	°C	70/30	188	188	188	188	188
	air/oxygen	°C	125	125	125	250	250	500
	total internal heat exchange	MW	0	70	80	105	127	134
Power	GT	MWe	84	86	90	69	77	81
	ST	MWe	17	18	17	14	12	11
	compressors & pumps	MWe	−16	−16	−16	−5	−5	−5
Heat to mill	at 12 bar (to 188°C)	MW	43	43	43	43	43	43
	at 4 bar (to 144°C)	MW	93	125	120	136	127	124
	at 1.5 bar (to 110°C)	MW	30	16	16	25	26	26
	cooling/make-up	MW	34	12	12	16	16	16
Efficiency	electricity	%LHV	26%	27%	28%	24%	26%	27%
	total CHP (to 110°C)	%LHV	77%	83%	83%	87%	86%	86%
Power	net from CHP plant	kWh/tonne DS	852	882	908	781	840	876
Heat	net from CHP (to 110°C)	GJ/tonne DS	6.0	6.6	6.5	7.4	7.0	7.0

(Table 3) when steam production increases, so that a potential demand for cooling water is reduced.

When comparing the GCCs for different levels of weak wash and condensate preheat (Fig. 4) it is evident that the net amount of heat available from the system is the same, while the temperature levels change (note that the GCCs in Fig. 4 correspond to the low-temperature part of the GCC in Fig. 3; for clarity the HRSG has been omitted). To achieve this, however, the amount of indirect heat exchange within the gasification system must increase, as indicated in Table 3. Interestingly, the right part of the curves begin to coincide at the temperature to which the weak wash and condensate have been heated. This leads to the important conclusion that there is no additional gain from preheating these streams beyond the temperature of the desired utility level in the waste heat boiler. This is in accordance with the pinch principles, because the pinch violation in the quench will be eliminated as soon as the entering streams have reached their cold pinch temperatures (about 144°C for 4 bar steam, and 188°C for 12 bar steam).

5.1.2. Gas reheat

The clean gas from the absorber is relatively cold. If the gas is reheated before the combustion chamber, the power output will be augmented because the enthalpy flow to the gas turbine increases. As shown in Fig. 5, the maximum increase in power output is about 6%, but the curve begins to level off at 183°C, since at higher temperatures gas reheat competes with medium and high pressure steam generation. The effect on power output is about double for the air-blown systems (Table 3) because the fuel gas flow is approximately twice that in the oxygen-blown systems. It should be noted that when the gas temperature rises, more air must be delivered by the gas turbine compressor to maintain the same firing temperature. Exhaust

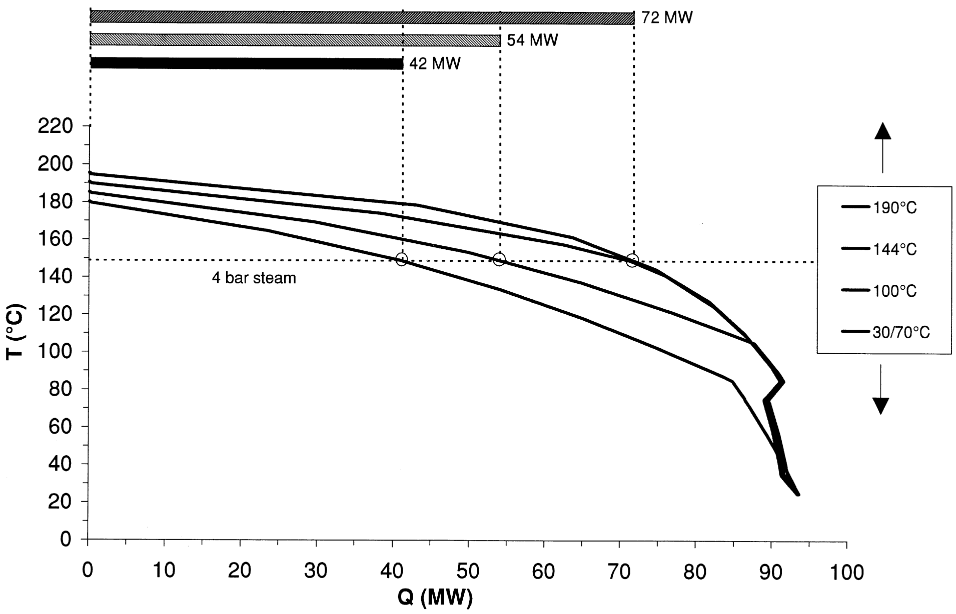

Fig. 4. The shape of the GCC changes as weak wash and condensate are heated before the quench. More steam can thus be raised in the waste heat boiler and green liquor cooler. Numbers to the right indicate the condensate and weak wash temperatures after heating. The bar chart at the top shows the maximum possible LP steam production for each level of preheat.

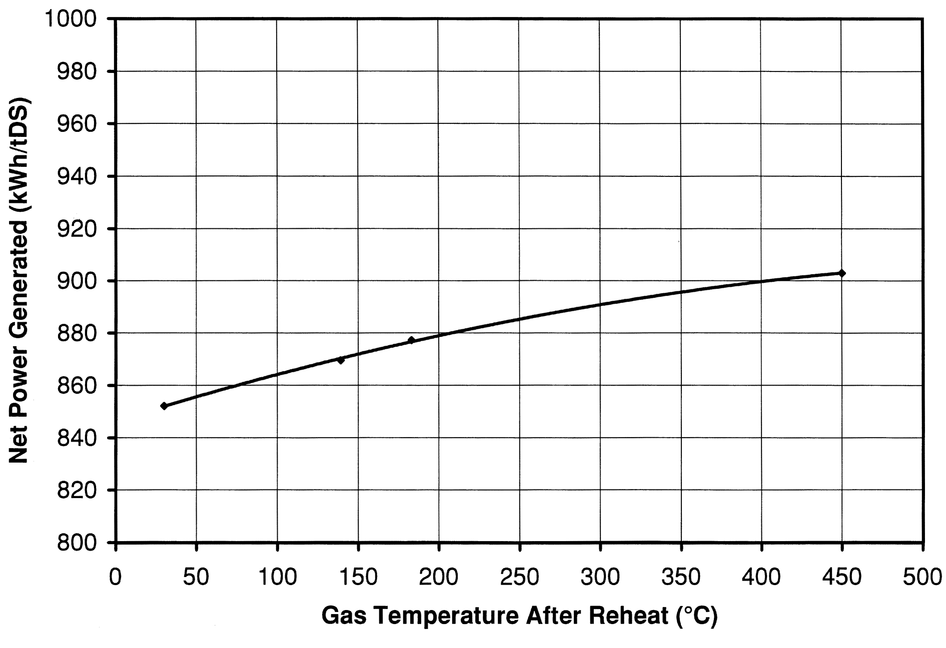

Fig. 5. Gas reheat increases the net power generated. (Case: oxygen-blown gasifier, gas turbine "F", 75%DS.)

gas flow will therefore increase and the flue gas loss will be slightly greater with gas reheat than without, corresponding to 0.5–1% points of the total CHP efficiency.

5.2. Heat exchanger network

Regenerative heating of gas, condensate and weak wash are desirable, and the potential for each has been found from the composite curves. Clearly, much heat must be exchanged within the system to maximize steam and electricity generation. To achieve the target in practice necessitates a well-designed heat exchanger network. In Fig. 6, a maximum energy recovery (MER) network design is shown for the case with the highest power production. There are few hot streams, and several stream splits are therefore necessary to reach the targets for steam production; this leads to a relatively complicated design which may be difficult to control.

It seems that the system would be easier to control if the design is restricted to the matches: (1) weak wash/green liquor, (2) WHB/condensate, and (3) HRSG/gas, and if the black liquor is heated by steam. This network will have some cross-pinch heat transfer as compared with the initial target, which leads to a lower net steam production (MP: −2.5 MW or −17%; LP: −5.7 MW or −8%). The pinch violations can be reduced, however, by adjusting the heat capacity flowrates through the green liquor cooler (Fig. 7); this can be done simply by mixing some of the condensate with the weak wash, since they are both destined for the quench. The improvement is 2.3 MW on low pressure (LP) steam and 1.7 MW on low-low pressure (LLP) steam. About half of the remaining pinch violation is because of the black liquor steam heater.

5.3. Air-blown versus oxygen-blown gasifier

The net power output is up to 12% higher for the oxygen-blown systems. The difference decreases with increasing air preheat and gas reheat, from which the air-blown systems benefit more. Gas and steam turbine outputs are substantially larger for the former systems, but the auxiliary requirements for air separation and oxygen compression balance the greater part of this. Some heat is also removed at temperatures below 110°C from compressors in the ASU. Together with the larger gas flow from the GT this causes the total combined heat and power (CHP) efficiency of the oxygen-blown systems to be slightly lower.

It should be noted that the gas turbine and gasifier are more intimately integrated in the air-blown systems, because the gasification air is bled from the gas turbine compressor. It is possible to have a similar arrangement to integrate the gas turbine with the ASU, but the gain in efficiency must be weighed against the increased complexity. As pointed out before, compressor air-bleed is necessary with the air-blown systems, if a standard gas turbine is to be used.

5.4. Sulfur recovery impact

The SRU requires roughly 6 MW of 30 bar steam to strip H_2S from the absorbent. In most cases, this steam can be raised in the HRSG, but at the expense of MP steam production. Slightly more MP steam needs to be extracted from the steam turbine and the electricity production is, therefore, reduced by approximately 5 kWh/tonne DS. Alternatively, to avoid a

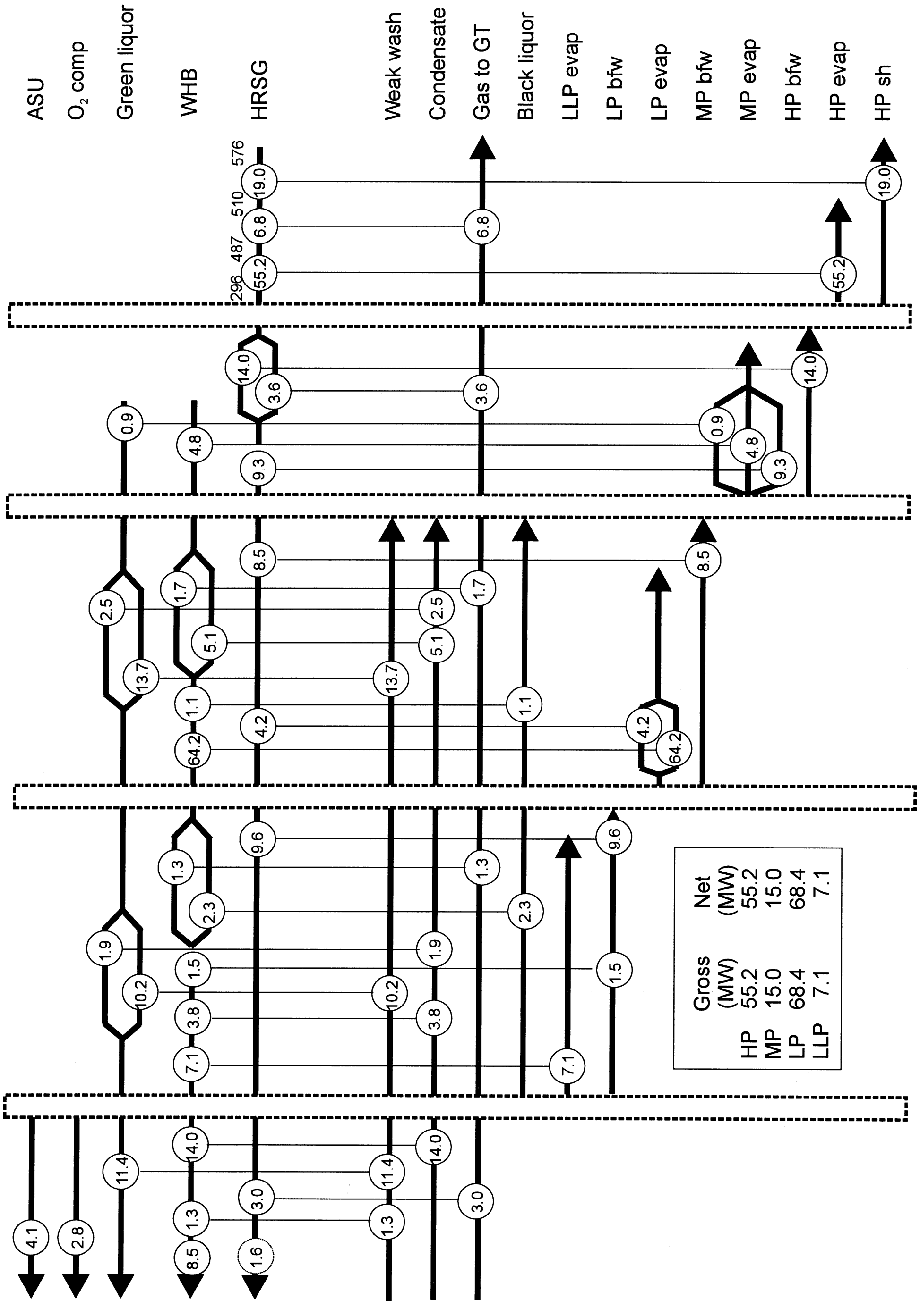

Fig. 6. Maximum energy recovery (MER) network for the case with the highest power production (third column of Table 3).

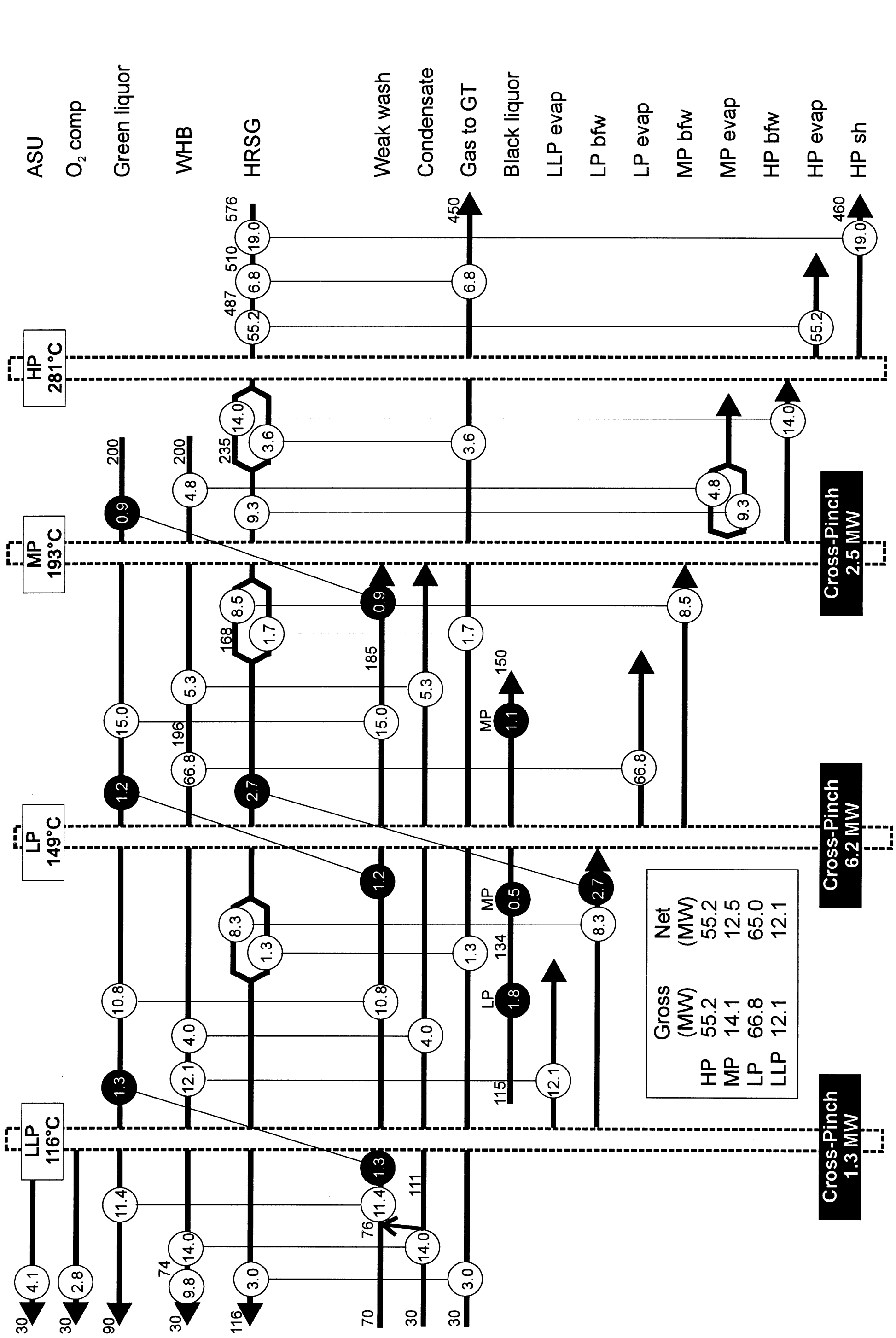

Fig. 7. Simplifed heat exchanger network (same case as in Fig. 6) with better matching of the heat capacity flow-rates through the green liquor cooler, gas reheat in the HRSG, and black liquor heating with steam.

30 bar steam level in the HRSG, the steam for the SRU can be extracted from the turbine at a penalty of about 10 kWh/tonne DS.

Several routes are possible for the conversion of H_2S into useful pulping chemicals; these routes will have different energy balances but the detailed discussion of these lies outside the scope of this paper. It should be noted, however, that some routes will lead to an increase in lime kiln fuel use which must be included when comparing the overall energy efficiency with other recovery processes. In some cases this may correspond to about 0.5 GJ/tonne DS or approximately 4% of the LHV. The cases in Table 3, however, are neutral relative to each other in this respect.

6. Conclusions

- From the pinch analysis and simulations we conclude that the potential for the pressurized black liquor gasification (BLG) cogeneration systems treated here is to generate electricity at 800–900 kWh/tonne DS and steam at 6–7 GJ/tonne DS.
- The pinch analysis has shown that the only pinches in the system are caused by the utilities. If weak wash and condensate are to be heated before the quench, they should be heated to the cold pinch temperature of the desired utility level in the waste heat boiler.
- Gas reheat improves the electric efficiency of the system by up to 6%, but leads to a small decrease in the total CHP efficiency.
- Similarly, oxygen-blown systems will increase power generation. The gain in power output is of the order of 10%.
- Manageable heat exchanger networks can be designed to meet the targets outlined by the pinch analysis.

Acknowledgements

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