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SIMULATION OF COMBINED CYCLES WITH BLACK LIQUOR GASIFICATION

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BREAK

ABSTRACT

Black liquor, a residual of the chemical pulp industry, is a biofuel with great potential for increasing power generation. This may be accomplished by replacing the conventional recovery process using a Tomlinson recovery boiler with black liquor gasification. In addition, there are other considerable advantages.

The gasification of black liquor gives new degrees of freedom since the combustible gas may be used in a multitude of ways. For a full scale plant, a combined cycle may be an efficient way to produce process heat and power. Comparison is made with conventional CHP using a Tomlinson boiler and a steam cycle. It is shown that the generated power could be up to twice that of the conventional plant with an electrical efficiency of 30% and a total efficiency of 77% (HHV). However, the characteristics of a combined cycle in this kind of process integrated application will be quite different than those of a stand-alone power plant. This is clearly shown when compared with conventional natural gas fired combined cycles.

This paper includes thorough analysis of some of the most important parameters of the combined cycle. It is shown that the gas turbine characteristics are the critical factors for providing a high electric efficiency. The effects of supplementary firing are also investigated. Hereby, an optimum for the steam cycle data is established.

NOMENCLATURE

CHP	combined heat and power generation
GT	gas turbine
HHV	higher heating value of fuel
HRSG	heat recovery steam generator
IGCC	Integrated gasification (and) combined cycle (gas turbine and steam turbine)
LPG	liquefied petroleum gas
LP, IP, HP	low pressure, intermediate pressure, high pressure
PFBC	Pressurized fluidized bed combustion

INTRODUCTION

Black liquor or spent cooking liquor is an important residual of the kraft pulping process. It contains valuable substances (mostly Na_2S and Na_2CO_3) that are important to recycle, both economically and environmentally. The contents of black liquor even consists of substances with high fuel value (lignin). Black liquor is therefore an important internal fuel within the pulp industry. With a market share of 65% of the biofuel used on large scale basis in Sweden, during 1991 or a total of 28.4 TWh (Biobränslekommissionen, 1992), black liquor has a leading role in the total energy balance in Sweden, as well as in other countries with large pulp industries.

The traditional way of treating the black liquor is combustion in a so called Tomlinson recovery boiler. There are two main purposes for this type of recovery: chemical recovery of the important cooking chemicals and generation of steam for process demands. Excess steam is expanded in a turbine for power production. The recyclable chemicals form a smelt in the bottom of the boiler and the lignin type of compounds are combusted. The conditions are very alkaline and therefore corrosive. This has been causing severe problems with the water and steam tubes immersed in the boiler. Serious explosions have also occurred when tubes have leaked water into the smelt of sodium sulphide and sodium carbonate in the bottom of the boiler. These drawbacks common to the Tomlinson boiler may be overcome by black liquor gasification, a novel technology recently demonstrated in a pilot plant at the Frövifors pulp and paper mill in Sweden (Boström et al, 1991).

Currently, new techniques for handling the black liquor are being developed in several places around the world. The most promising techniques are based on gasification instead of direct combustion of the black liquor. This has a number of advantages:

(1) A less complicated process unit with no water or steam tubes inside. This leads to less expensive equipment with higher availability.
(2) Eliminated explosion risk, because there are no longer any tubes

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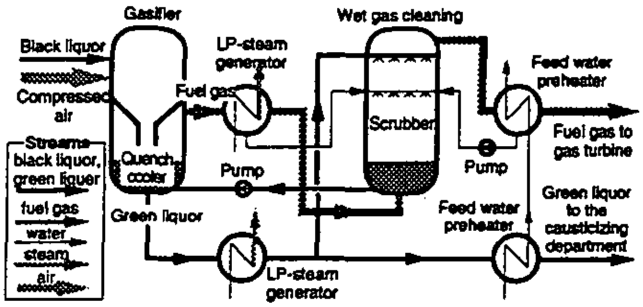


FIGURE 1. THE BLACK LIQUOR GASIFICATION SYSTEM.

inside the vessel.

(3) The recovery boiler is often a "bottle-neck" for production increases. Gasifiers may be built in smaller units and thus modular expansion is possible.

(4) Also the power and process heat generation will be more flexible. The produced gas may be used in a number of ways, including advanced gas turbine cycles or just a simple gas fired boiler.

(5) With a combined cycle, the ratio of generated power to process heat may be increased. This is an option with good potential since most pulp mills generate excess heat that cannot be utilized today.

(6) With fuel gas cleaning after the gasifier, emissions into the environment are reduced since the flow rate of gas is much smaller than for the corresponding flue gas. Calculations indicate that the gas volume to be cleaned decreased by about 95%. (Kohl, 1986) The temperature at which fuel gas cleaning is performed is flexible. Hot fuel gas cleanup is desirable thermodynamically, since keeping a higher temperature increases the possible power production. However, wet gas cleaning at low temperature is an elaborately tested way of reducing pollutants to tolerable levels. The low calorific value of the fuel gas is beneficiary from environmental aspects since the generation of thermal NO_x in the gas turbine thereby is kept at a minimum. (Becker, 1992) Flue gas emissions of NO_x and sulphur are therefore marginal compared with conventional technique. (Andersson, 1990) The increased generation of electricity from black liquor would also directly reduce electric utility CO_2 emissions, since black liquor is a renewable bio fuel. (Larson, 1990)

(7) Depending on the reaction chemistry of the gasification process, there may be new and advantageous alternatives for pulp bleaching sequences. According to Ulmgren et al (1992), sulphide free green liquor is an interesting option.

The processes for black liquor gasification can generally speaking be separated into two types. The smelt phase type operates at temperatures above approximately 900°C. The alkali leaves the gasifier as a smelt consisting of Na_2CO_3 and Na_2S, just as in the conventional recovery boilers. The retention time is short and carbon conversion is near to 100% (Backman et al, 1992). Processes with this concept are being developed in Sweden (Chemrec) and in Finland (Ahlström, Tampella) (Grace, 1991). The solid phase type of gasification operates at a temperature lower than 750-800°C. The alkali content forms a solid powder phase which can be separated in cyclons or likewise. At

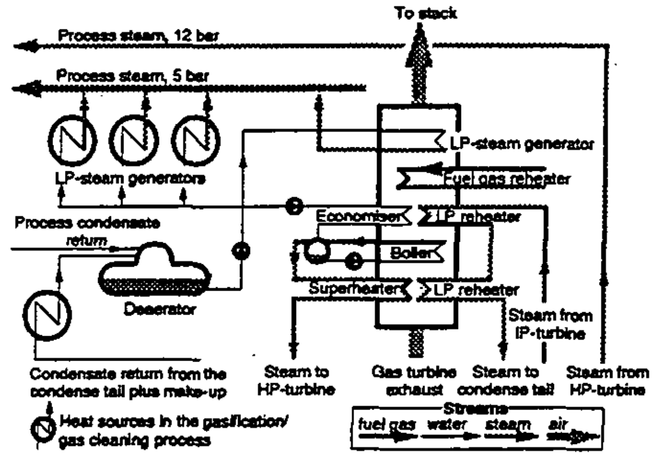


FIGURE 2. THE HRSG AND STEAM SYSTEM.

such low temperatures, the sulphur leaves mainly as $H_2S(g)$ instead of as Na_2S. The green liquor formed can therefore be divided in streams with and without sulphur. The lower temperature also affects the carbon conversion. The retention time needs to be longer in order to get an acceptable level of carbon conversion. Saviharju et al (1992) points out that staged combustion with several feedpoints is an alternate way of improving the carbon conversion. Low temperature processes are being developed in Sweden (ABB) and in Finland (VTT, Tampella) as well as in the USA (MTCI) (Grace, 1991).

If the fuel gas is going to be used in a gas turbine, the gasification process either has to work at elevated pressure or the produced gas will have to be compressed. On the other hand, when incorporating other types of power cycles such as externally fired turbines or just a conventional boiler with a steam turbine it is not necessary to have a pressurized fuel gas, and the gas cleaning requirements become more moderate.

Combined cycles for power and heat generation are well known equipment for fuels such as LPG, natural gas or for integrated coal gasification. The technical development in these areas is rapid. However, the characteristics of a combined cycle applied in black liquor gasification will be quite different than those of conventional power plants.

Because of the high demand for process steam and the special properties of the gasification process, the combined cycle must be optimized using totally different methods. One of the properties of this fuel gas is a low heating value. Gas turbine manufacturers state that a fuel gas of this kind may be run if the purity demands are met (Solantausta et al, 1992; Takano et al 1989). The purity demands are very high since the fuel gas flow rate is much higher compared with natural gas due to the lower heating value (Becker & Schetter, 1992). Comparisons with IGCC or PFBC using other fuels, like coal, peat or biomass indicate that the necessary removal of particulates, tars and alkali-metals could be accomplished by using wet scrubbing technique below the dewpoint of the gas or, preferably, using hot gas clean-up with ceramic candles around 400-500°C (Larson et al, 1989; Odeberg, 1990; Nygren & Svedberg 1990; Lehtovaara & Mojtahedi, 1992; Eriksson et al 1992; Kurkela et al, 1992). It should be noted, though,

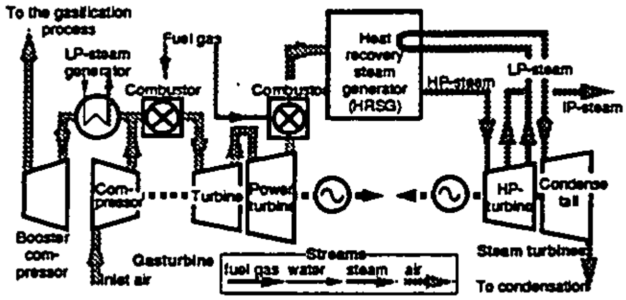


FIGURE 3. THE GAS TURBINE AND STEAM TURBINES.

that long term testing under realistic conditions have nowhere proved this thesis as yet.

PROCESS DESCRIPTION

In this paper, processes including black liquor gasification based on the Chemrec concept (Boström et al, 1991) and different combined cycles have been studied. The systems are based on a theoretical 1000 tpd market pulp mill. The gasification process fundamentals are described in Fig. 1. The Chemrec gasification is performed within the high temperature interval 850°-950°C and air is the oxidant. The produced fuel gas is internally cooled in a quench cooler. The heat generated in the gasification process may therefore only be used at a low temperature. This is done by partly condensing the steam content of the gas in a subsequent LP-steam generator. After cooling, the fuel gas is transferred to a wet scrubber operating at the gas dew point in which the fuel gas is further cleaned. Complete removal of alkali and particulates is thereby assumed. This is followed by another condensation step where the gas is cooled to 100°C. In order to improve the efficiency, the fuel gas is preheated in the last part of the heat recovery boiler before entering the gas turbine. The gasification is running at a pressure of 24 bar and the gas turbine has a pressure ratio of 16. The gas turbine is a simple cycle with no intercooler. The turbine's capacity is matched with the gasification process, so it is not an existing turbine but a theoretical one. The data for the turbine are chosen from existing turbines within the same size range. Part of the air compressed in the gas turbine compressor is lead off to an additional booster compressor. This air is used for the partial oxidation in the gasification process. The steam turbine consists of a high pressure turbine expanding the superheated steam to 5 bar with an extraction point at 12 bar. These two pressure levels are those used in the pulp plant. The surplus of 5 bar steam is superheated in the heat recovery boiler. This is done to prevent the steam from becoming too moist in the final expansion in the condensing tail where the steam expands to a final pressure of 0.04 bar (a maximum of 9.5% liquid phase is allowed). The combustion and turbine system are briefly described in Fig 2.

The gas turbine exhaust gas is used as the heat source in the heat recovery steam generator (HRSG). The major units of the HRSG and the steam system are shown in Fig. 3. The HRSG consists of five sections. The first section is the superheater where the high pressure steam is superheated to its final temperature. For the control case with

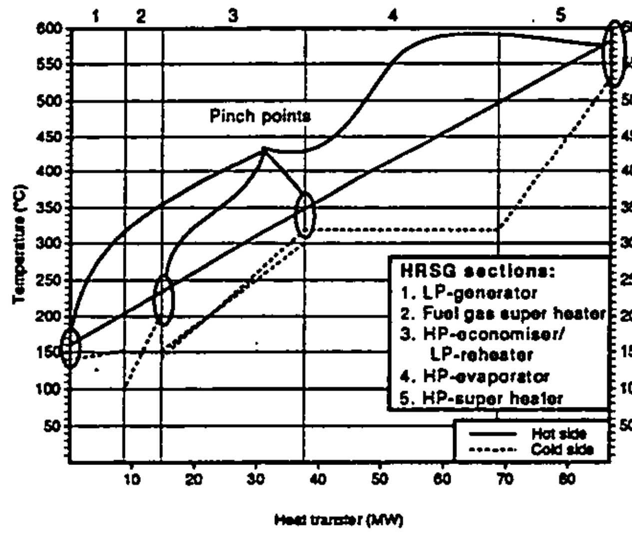


FIGURE 4. EXAMPLE OF TEMPERATURE PROFILES IN THE HRSG.

105 bar steam, the superheating temperature is 520°C. For some extreme low pressure systems, this section of the HRSG is also used to superheat the low pressure steam to 300°C, but for most cases this is done in section three. Section two of the HRSG is the evaporator. This is normally the largest section. Section three is the economiser. It is also used to superheat the low pressure steam before entering the condensing turbine. Section four is the smallest section of the HRSG used to preheat the fuel gas. It is heated to 210°C or lower if there is not enough heat available. The last section of the HRSG is a LP-steam generator, of which there are three more in the gasification process. Due to the excess heat of low temperature in the gasification process and the high return temperature of the condensate from the supplied pulping process steam, the exhaust can not be utilized at temperatures lower than 160°C for power or steam generation. However, it could be used to e. g. produce hot water if such is needed in the pulp/paper process. This lies beyond the scope of this paper.

The HRSG is optimized all the way. It actually has four pinches (i. e. minimum temperature approaches). This is accomplished by varying the steam data and flowrate for the HP-system as well as the flowrate in the LP-steam generator and the temperature in the fuel gas reheater. Also the rate of supplementary firing may be varied in order to change the plant performance. Fig. 4 gives an example of the temperature profiles in the HRSG, showing the different pinch-points. Table 1 summarizes some important admission data.

METHODS

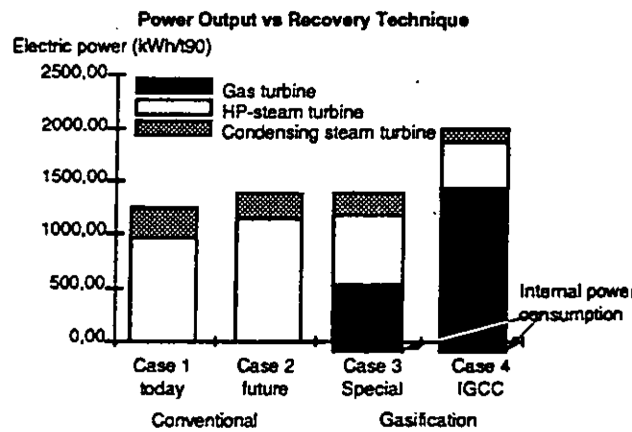
The process has been studied by making computer simulations using the chemical process simulator Aspen Plus™. Although not specifically designed to simulate power cycles, this software turns out to be suitable for this kind of integrated chemical system. The computer models have been constructed for maximum flexibility, so that the total system may be optimized with respect to energy utilization in all parts of the process.

TABLE 1. BASE CASE ASSUMPTIONS AND INPUT DATA.

Description		*Assumptions*
Mill capacity	bleached kraft pulp	1000 tpd
Pulp mill energy demands/return	5 bar steam	7.4 GJ/t90
	12 bar steam	1.8 GJ/t90
	Power	660 kWh/t90
	Condensate return	90%
Black liquor specification	Dry content	80%
	Liquor dry substance	1.64 t/t90
Gasification Process	Type (Chemrec)	Smelt phase
	Temperature	900°C
	Pressure	24 bar
	Cooling	Quench
Fuel gas composition, after gas cleaning (kmol/s)	H2O	7.6148 E-2
	N2	0.8227
	O2	0.0000
	CO2	0.1431
	CO	0.3131
	H2	0.3219
	CH4	4.9266 E-3
	H2S	7.8646 E-3
	O2S	3.9473 E-9
	COS	2.3884 E-4
	LHV	4.33 MJ/kg
	HHV	4.78 MJ/kg
Gas turbine	Pressure ratio	16
	Expander inlet	1100°C
Steam cycle	Live steam data	105 bar/520°C
	Condense tail data	0.04 bar
Temperatures	Ambient conditions	15°C
	Process condensate	140°C
	Stack temperature	160°C
Pressure losses	Gasification	0.5 bar
	Gas cleaning	0.1 bar
	GT air inlet	0.0046 bar
	GT combustor	0.75 bar
	GT fuel inlet	8 bar
	HRSG	0.017 bar
	Other heat exchangers	0.01 bar
Combustion	Gas turbine	99%
	Supplementary firing	99%
Isentropic efficiencies	GT compressor	0.85
	Booster compressor	0.85
	GT expander	0.9
	Steam turbines	0.9
Mechanical efficiencies	Compressors	0.99
	Expanders	0.99
Driver eff.	All pumps	0.9
Pinch points	ΔT gas-gas	30°C/50°C
	ΔT gas-liquid	30°C
	ΔT liquid-liquid	10°C

TABLE 2. BASE CASE PERFORMANCE AND OUTPUT DATA.

Basecase output	
Fuel gas	43.95 kg/s
Fuel gas HHV	210 MW
Power output	86.5 MW
Process steam, 5 bar	39.5 kg/s
Process steam, 12 bar	9.06 kg/s
Power to heat ratio	61%
Electric efficiency (HHV)	28.7%
Total efficiency (HHV)	75%
Gas turbine power/total	77%
Condense tail power/total	7.4%
Gas turbine	70.0 MW (183.4 kg/s)
HP-turbine	12.8 MW (24.8 kg/s)
IP-turbine	2.4 MW (15.7 kg/s)
Condense tail	4.6 MW (6.2 kg/s)
O2 in flue gas	11.0%
Fuel frac to supp. firing	7.8%



Case 3: Gasifcation + Gas Turbine with Gasboiler and Steam Cycle (Warnqvist, 1992)

FIGURE 5. POWER OUTPUT FOR DIFFERENT RECOVERY SYSTEM CONFIGURATIONS

RESULTS

A summary of the base case output is presented in Table 2.

Comparison with conventional recovery boilers

In Fig 5 , the power generation from plants with four different configurations are shown. Case one and two are systems with conventional recovery boilers (Warnqvist, 1992). The difference between them is that the leftmost uses the best available technology today (steam data 80 bar/480°C), while the other uses future technology (steam data 105 bar/520°C). Nygaard (1992) presents a slightly higher output. Cases three and four use black liquor gasification. Case three uses a system layout proposed by Warnqvist (1992), shown in Fig 6. Case four uses IGCC technique as described earlier in this paper. The

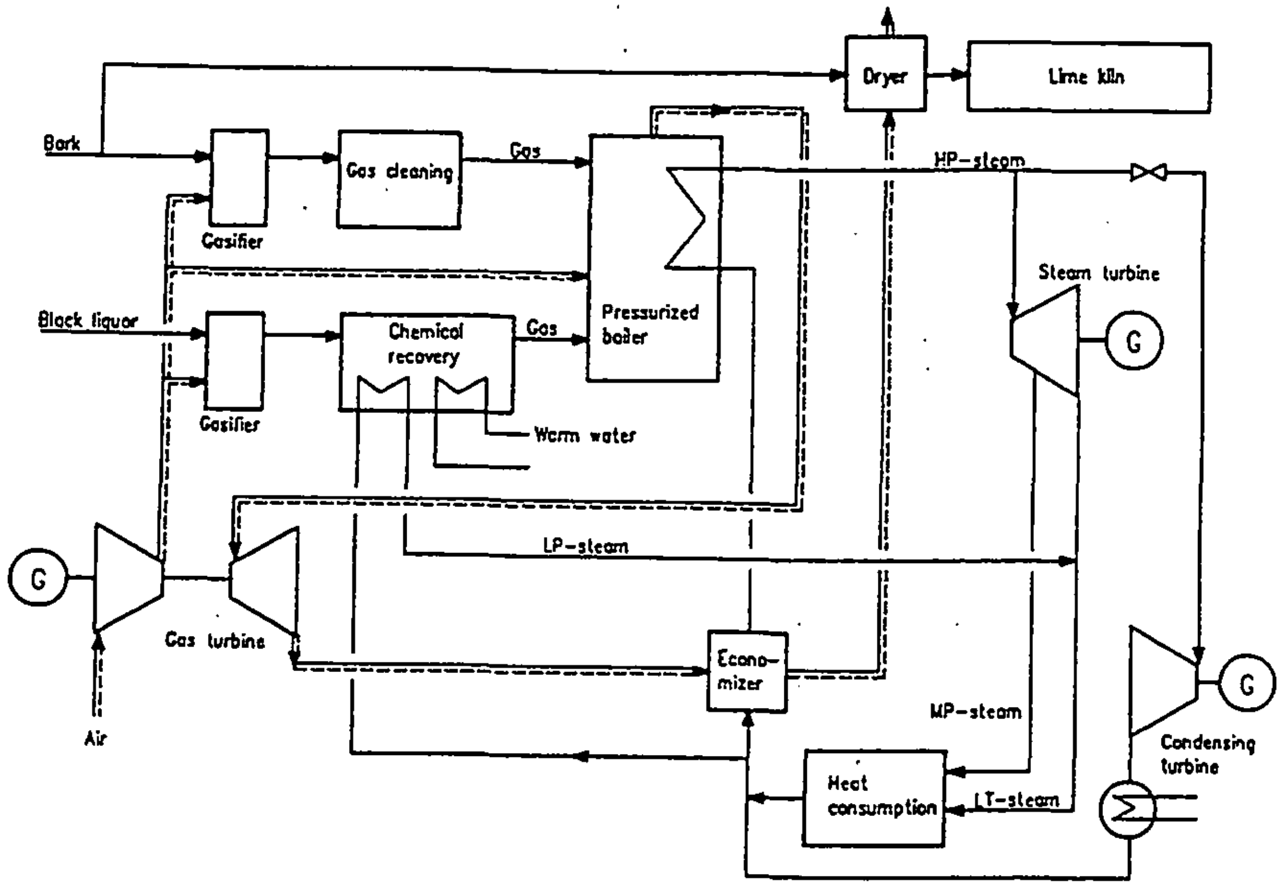


FIGURE 6. THE CASE 3 CONFIGURATION: INTEGRATED BARK AND BLACK LIQUOR GASIFICATION WITH GAS TURBINE, PRESSURIZED GAS BOILER AND A STEAM CYCLE (WARNQVIST, 1992).

diagram illustrates that considerable improvements are reasonable to expect in the conventional technique. However, the potential increase in power output when black liquor gasification is incorporated is shown to be much higher. The importance of thorough analysis of the gasification/combined cycle is verified by the great discrepancy between the two gasification cases. The Warnqvist model appears to suffer from poor gas turbine data, which is a consequence of its layout. Also, the exergy losses are high due to extreme ΔT in the boiler (Ihrén & Svedberg, 1992). The power output of case four is in good accordance with some other sources, e. g. Fogelholm (1991).

Effect of Supplementary firing

Supplementary firing in the heat recovery boiler is a technique used to improve the efficiency when the normal exhaust temperature from the gas turbine is only around 350-550°C, which renders poor data for a steam cycle. The effect of supplementary firing in a normal combined cycle fueled by natural gas is presented in fig 7 (Kehlhofer, 1991). The figure describes cycles with one pressure level for the steam, selected optimally for each case.

The diagram illustrates that there is a maximum efficiency with supplementary firing to a temperature of about 750°C for the gas turbines with lower inlet temperatures and to a slightly lower temperature when the inlet temperature is higher. The explanation is that with an exhaust temperature of around 750°C, the exergetic losses in the economiser is at a minimum because the curves for flue gas and water run parallell (cf. Fig. 4). This means that it is theoretically possible to cool the flue gas to the temperature of the feed-water, thus minimizing sensible heat losses in the flue gas. The reason why the optimum is not exactly the same regardless of the gas turbine is that supplementary firing means that some fuel is not generating gas turbine power output, and the better the gas turbine data, the greater the relative loss of gas turbine power. Therefore, the optimum for the combined-cycle is not the same as for the steam-cycle which is at 750°C for all cases.

The same type of diagram for the case where a combined cycle is coupled to black liquor gasification looks quite different as seen in Fig. 8. There is still a maximum or a terrace point somewhere around 700°C, but it is only a local maximum. The best efficiency for all curves are in cases with no supplementary firing at all. There are basically three reasons explaining the different behaviour of the efficiency curves. First, the exhaust heat can be used for preheating the feed water, but only down to an exhaust temperature of 160°C because the deaerator temperature is quite high due to the high return temperature of the process condensate. Secondly, the exhaust can always be cooled to 160°C, independent totally of the profiles in the high pressure part of the steam cycle since we are using the last part of the HRSG to

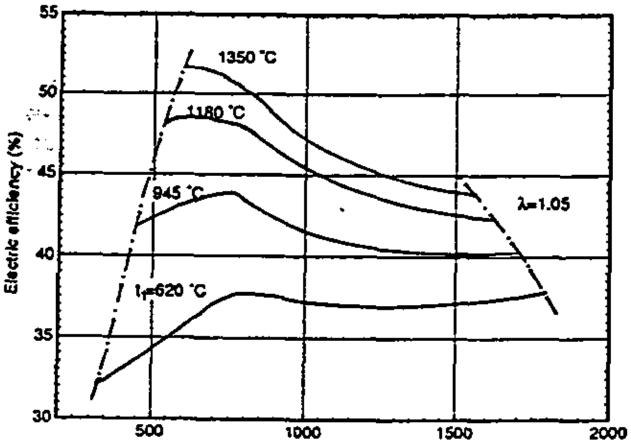


FIGURE 7. ELECTRIC EFFICIENCY AS A FUNCTION OF THE FLUE GAS TEMPERATURE AFTER SUPPLEMENTARY FIRING AND GAS TURBINE INLET TEMPERATURE (KEHLHOFER, 1991). t_i=GAS TURBINE INLET TEMPERATURE, λ=EXCESS AIR RATIO OF THE COMBUSTOR.

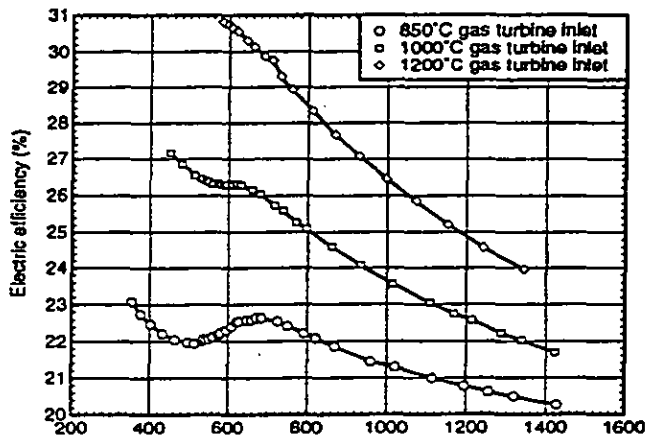


FIGURE 8. ELECTRIC EFFICIENCY VS SUPPLEMENTARY FIRING FOR INTEGRATED BLACK LIQUOR GASIFICATION COMBINED CYCLES. THE EFFICIENCY IS BASED ON THE HIGHER HEATING VALUE OF THE BLACK LIQUOR. THIS DIAGRAM REPRESENTS A COGENERATION SYSTEM WHERE THE PROCESS HEAT GENERATION IS FIXED (CF. TABLE 2).

produce some additional low pressure steam for process needs. The third reason is that a maximum is set for the steam data (140 bar/ 570°C). The first and second reasons lead to the fact that the steam-cycle has an optimum with supp. firing up to 700°C, instead of 750°C as above, and the optimum is only local and less obvious. The effect of steam-cycle improvement with increasing supplementary firing is trivial and therefore it does not dominate the performance of the combined-cycle at low rates of supplementary firing. The third reason, the maximum live steam data, also affects the steam cycle performance, but because steam data is not raised further when the flue gas temperature exceeds 620°C, the improvement of the steam-cycle beyond this point is minor.

Close examination of the bottommost curve gives an indication as to what is actually taking place. If some fuel is taken from the gas turbine and used for supplementary firing, power output from the gas turbine decreases and instead, more power is generated by the steam turbines. However, the steam cycle is so poor with its low temperature and pressure that the result is actually negative. If more fuel from the gas turbine is moved to the steam cycle, eventually a local minimum is reached. When this point is reached, the steam cycle is improved to such a degree that further supplementary firing raises the efficiency. Increasing supplementary firing even more leads to a maximum. At this point, the exergetic losses are minimal at all three high pressure parts of the HRSG, the superheater, the boiler and the economiser. Supplementary firing beyond this point is negative for the steam-cycle, since the Δt in the boiler has to be increased, (cf. Fig. 7) thus increasing the exergetic losses in the boiler and economiser sections. For the gas turbines with higher inlet temperature, the gas turbine power output is the determining element in the performance of the combined-cycle further to the right in the diagam. Therefore, only a terrace-point or an inflection-point at the maximum for the steam-cycle is attained.

A summary of these observations is that the crucial difference between the cases is due to the differences in the last section of the HRSG. With more supplementary firing, a conventional natural gas fired combined cycle can utilize a greater share of the flue gas heat in the recovery boiler because the profile for the hot side is changed. In systems with integrated black liquor gasification combined cycles the profile changes, but it is not thermodynamically possible to utilize any more of the heat in the HRSG.

Effect of Steam Data

Fig 9 graphically illustrates the normalized electric efficiency plotted against the live steam pressure. For each gas turbine, the maximum is 100%. There is an optimum steam pressure for each gas turbine case. This optimum occurs at the maximum pressure that can be achieved without any supplementary firing. To the left of each optimum there is no supplementary firing, but the Δt in the HRSG inlet is higher than minimum. To the right of the optimum the Δt in the HRSG inlet is a minimum, but the fuel fraction to supplementary firing increases gradually. For the low temperature gas turbine, this optimum is as low as 21 bar. It is also shown that the optimum is less apparent the higher the gas turbine inlet temperature.

DISCUSSION

In the simulations, a smelt phase gasifier with a quench cooler is used. Supposing a solid phase gasifier had been used, the major difference regarding the energy system would be that the solid phase gasifier does not cool the fuel gas in a quench cooler. Thereby, the heat generated in the gasification process may be utilized at a higher temperature level, i. e. for a parallell evaporator unit in the steam cycle. This would improve the system slightly if other parameters were kept constant, although the differences would not be qualitative, only

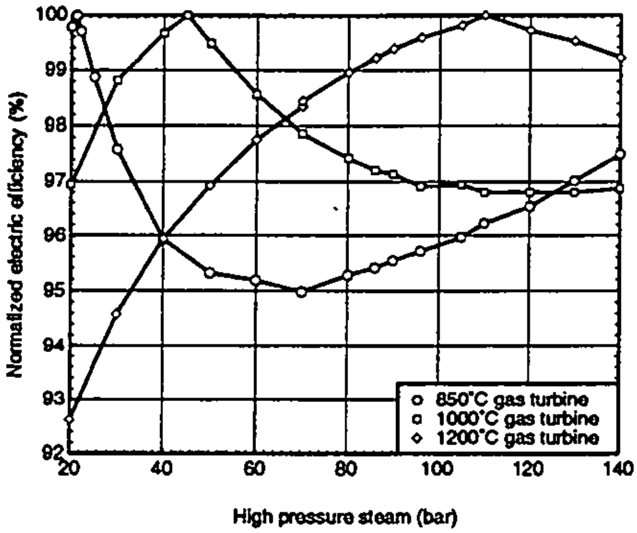


FIGURE 9. NORMALIZED ELECTRIC EFFICIENCY VS LIVE STEAM DATA. THE MAXIMA FOR THE CURVES BECOMES LESS DISTINCT THE HIGHER THE INLET TEMPERATURE OF THE GAS TURBINE.

quantitative. However, reliable data for a solid phase gasifier has not been available.

A theoretical model of a gas turbine is used in the calculations.. The choice of turbine size varies considerably with the fraction of fuel gas being used for supplementary firing. For some designs it may be preferrable to have two parallell gas turbines instead, in order to remain in a moderate size range of gas turbine and for additional control options. There are many (>10) manufacturers of gas turbines in the range 30-80 MW.

The effect of supplementary firing on the electric efficiency for black liquor IGCC is presented in Fig. 8. It must be noted that with a HRSG inlet temperature above 900°C, by using supplementary firing, it becomes necessary to have a water-tube walled construction in the HRSG.

Since a low heating value fuel is used, the combustion efficiency is set at 99% in each combustor. This means that with low supplementary firing, 0.01% of the fuel escapes unburned. With higher fuel fraction to supplementary firing, this figure increases, thus a higher combustion efficiency setting would have been more realistic.

This work deals with combined cycles, but there are other options for the power cycle. Especially interesting are those that maintain a high efficiency, but avoid some of the major complications of the combined cycle. For instance, incorporating externally fired gas turbine cycles would mean that requirements for gas purity are relatively low and that the gasification need not be pressurized. Steam-injected gas turbines or evaporative waste heat regeneration gas turbine systems may also provide good options. Further research is clearly needed in this area.

The potential for increased power generation within the pulp and paper industry will not be the major incentive for commercialization of this new technique. The technique must above all prove to be reliable and more cost effective than current recovery processes.

One reaction to this kind of study might be that this type of advanced combined cycle analysed is not be economically feasible in practical situations. To some extent this may be true, but since the economic outcome is highly dependant on local governmental energy policies it is difficult to define realistic. This study aims at presenting what is theoretically and technically possible and nationally reasonable. In doing so it could be compared with Nygaard (1992) who shows the potential of conventional technique in the same manner.

CONCLUSIONS

This research has demonstrated that black liquor IGCC is capable of substantially raising the electric efficiency for a kraft pulp mill and is in accordance with earlier work presented in the same area. Depending on the particular situation at each pulp mill, the lowered production of steam may cause a deficit that must be compensated for by using some other fuel, such as bark or by heat pumping of lower grade waste heat.

The calculations clearly show that the most important unit in the combined cycle for a high efficiency is the gas turbine. The development of new gas turbines with higher inlet temperatures and higher efficiencies is very rapid. Even techniques for handling low heating value fuel gases are being developed by most gas turbine manufacturers. When designing an IGCC process of this kind it is economically essential that a standard gas turbine be used. This criteria means that the flow rates through the compressor and the expander must have a certain relation. In order to accomodate this and at the same time let the flow rate of black liquor control the process, supplementary firing in some situations can be an important control option.

It has also been concluded from this research that supplementary firing implemented in black liquor IGCC does not improve the electric efficiency, contrary to the results for some other types of combined cycles. With as little supplementary firing as possible, an optimum for the steam data can be established.

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