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EVALUATION OF SURPLUS ELECTRICITY IN BLACK LIQUOR GASIFICATION COMBINED CYCLE

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Abstract. *The pulp and paper sector is intensive in the use of energy, and presents a high participation in the industrial context, specially based in the black liquor, a renewable source generated in the pulp process. Although the black liquor gasification (BLG) is not still completely dominated, it has the potential of becoming an important alternative for the pulp and paper sector, once the burning of BLG in gas turbines should guarantee higher efficiencies, better use of the available renewable resources, also allowing the chemical recovery, as is done nowadays with Tomlinson chemical recovery steam generators. In this article, the traditional steam cycle based on chemical recovery and biomass boilers associated to backpressure/extraction turbine is compared to an innovative black liquor gasification combined cycle (BLGCC) schemes, associated or not to biomass boiler. The objective is to estimate the potential electric surplus that can be produced by the alternative new technologies limited to the technological constraints of black liquor gasification process.*

Keywords: *Cogeneration, black liquor gasification, combined cycle, exergetic analysis.*

1. INTRODUCTION

Currently, the pressing concern related to the environment and reduction of fossil fuels has led researchers to focus on new energy conversion technologies for minimizing environmental impacts and to warrant the human survival on the earth. Renewable energy sources may be a solution to the offering of alternative energy because its dissemination tends to minimize the greenhouse effect and global climate changes.

From the several renewable energy sources available, Brazil stands out for having environmentally-friend resources in abundance, such as hydropower, biomass, solar and wind. However, the water resources are already being affected by climate change, which means that rain dynamics is changing. Vegetal and agricultural biomasses are a source of energy obtained from the sun, a renewable and inexhaustible source in the long term, through the natural photosynthesis process. The use of this fuel is enabled to have a neutral CO_2 emission balance and also there is a great absorption of this gas by the biomass. The great advantage for nature is the reduction of greenhouse effect, as a consequence of CO_2 emissions decrease.

Biomass is available in so many ways, as in the large surpluses of timber forests, agricultural waste and organic fractions of municipal solid wastes. The importance of energy obtained from biomass is dependant of its availability in nature; the conversion of solid biomass into gas is also important to be proposed because some more efficient additional technologies (other than steam thermal power plants, based on steam generators), such as gas turbines, can be employed. One advantage of gasifying biomass is due to the oxygen and water content present in its chemical structure, that are crucial elements in the gasification process. The gasification allows the conversion of solid material into combustible gases, generating energy to be processed to mechanical and/or electrical energy, as well as the production of synthetic fuels and chemicals.

In this paper, an integrated gasification combined cycle scheme was proposed and compared to the conventional scheme of pulp and paper sector, based on chemical recovery boiler and backpressure/extraction steam turbine; in both cases, a biomass boiler was also considered for burning bark and wood residues and wood chips. An energetic and exergetic analysis for the proposed cycle was presented; the black liquor gasification equilibrium was modeled; the mass, energetic and exergetic balance equations were then considered for each component. Energetic and exergetic analysis of each component were detailed, and data relative to the energetic and exergetic analysis of black liquor-integrated gasification combined cycle scheme was then presented.

2. REVIEW OF LITERATURE

The importance of black liquor gasification combined cycle for the pulp sector is due to the need of recovering cooking chemicals with an improvement in the energy production. Traditionally, Tomlinson boiler is the technology used for that, but an improvement in the thermal efficiency of this process and the availability of more electric energy can be obtained by the black liquor gasification and the use of gas/steam combined cycle.

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In a comparative analysis of conventional Tomlinson boiler with integrated gasification combined cycle, Näsholm and Westermark (1997) indicated the potential to double the power output if the conventional system is replaced by this new technology. A hybrid combined cycle was also proposed, in which natural gas is burned in the gas turbine and the fuel gas obtained from black liquor gasification was used as a supplementary fuel for the steam cycle; results indicated that no modifications were needed in the gas turbine for this alternative solution, but the total efficiency was not as high as for the integrated gasification combined cycle.

Berglin and Berntsson (1998) presented a thermodynamic analysis of an integrated black liquor gasification combined cycle cogeneration system. An equilibrium model based on thermo-chemical data taken from a standard reference source was proposed and a commercial process simulator was used for evaluate the combined cycle; an additional pinch analysis was developed to identify potential heat sources to optimize the proposed scheme.

In addition to the thermodynamic and equilibrium studies, it is also important to detail the performance modeling of gasifiers and gas turbine cogeneration systems using different black liquor gasifiers modeled on proposed commercial designs; the paper by Consonni et al. (1998) identified prospective environmental, safety and capital cost benefits for the pulp industry.

A computer program for a thermodynamic comparison analysis of several black liquor gasification combined cycle schemes was structured by Gallego (2004) to consider low and high gasification pressures, low and high temperatures and the use of air or oxygen. Air-blown and pressurized gasifiers were considered in that thermoeconomic analysis. In this work, an exergetic analysis was presented to identify the losses in the integrated gasification combined cycle.

Black liquor gasification combined cycle was also considered by Harvey and Facchini (2004). A Swedish pulp mill state of the art commercially available technology was assumed as the basis for the calculations and an off-design calculation was developed. Klimantos et al. (2009) evaluated an air-blown biomass pressurized gasification integrated with advanced combined cycle aiming at appraising the technical feasibility and the economic viability of such technology for power production.

Naqvi et al. (2010) reviewed several studies related to the black liquor gasification technologies and concluded that, despite they are still under development, they represent prospective environmental, safety, and capital costs benefits compared to the conventional recovery cycle and BLGCC has potential to switch pulp mills from electricity importers to electricity exporters, in the studied cases.

3. PROPOSAL OF A BLGCC SCHEME

Although the black liquor gasification is a process under development, the analysis of BLGCC schemes is of interest to determine the most adequate operational conditions. In the present analysis, a BLGCC scheme is proposed considering the availability of black liquor, bark and wood residues, as well as wood chips, of a conventional Brazilian pulp and paper integrated mill, for which a comparison is done.

The steam distribution and the electric power for an integrated pulp and paper mill that produces 1,200 ton/day of pulp and 1,000 ton/day of paper are presented in Figure 1 (Moraes, 2011). In this case, 220 ton/hour of steam (at 6.0 MPa) is produced by burning 1,740 ton/day of black liquor in a Tomlinson (chemical recovery) boiler and an additional 335 ton/h of steam is produced in the biomass boiler (it was assumed 530 kg/m^3 as the specific mass of wood chips, according to Moraes, 2011). High pressure steam is expanded in a backpressure steam turbine and sent to the processes (at 1.2 MPa and 0.4 MPa). This scheme does not produce electric power to supply the total demand of 60 MW, and 22 MW is then imported from the grid.

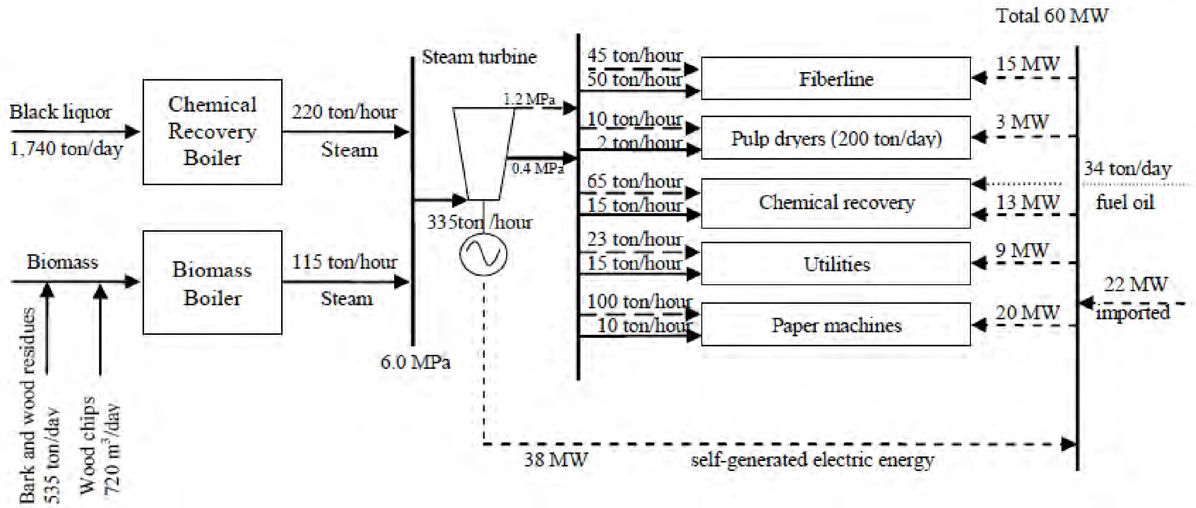

Figure 1 - Integrated pulp and paper mill with backpressure/extraction cogeneration scheme (adapted from Moraes, 2011)

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An alternative to the conventional backpressure/extraction cogeneration scheme is proposed by Moraes (2011) and consists in just change the steam turbine to a condensing/extraction one, according to Figure 2. For this new concept, the biomass boiler needs to burn additional wood chips (augmenting to 2,200 m^3/day) to produce 270 ton/hour of steam, totaling 490 ton/hour and condensing 155 ton/hour. For the same steam consumption of processes and demand of electricity, no surplus nor import of electricity are necessary.

The analysis of a pressurized air-blown black liquor integrated gasification combined cycle and the evaluation of exergetic irreversibility and efficiency are then developed. The proposed BLGCC scheme is based on the gasification unit (gasifier and gasification air booster compressor), gas cleaning system, gas turbine unit, heat recovery steam generator unit, steam turbine unit, condensing unit and the biomass boiler unit and corresponding fuel dryer system, with similarities with the schemes of Näsholm and Westermark (1997) and Larson et al. (2000). Thermodynamic state of equipment were defined according to the technological limits proposed by Larson et al. (2000), Silva (2000) and Santos (2007) relative to the gasification process for the present analysis.

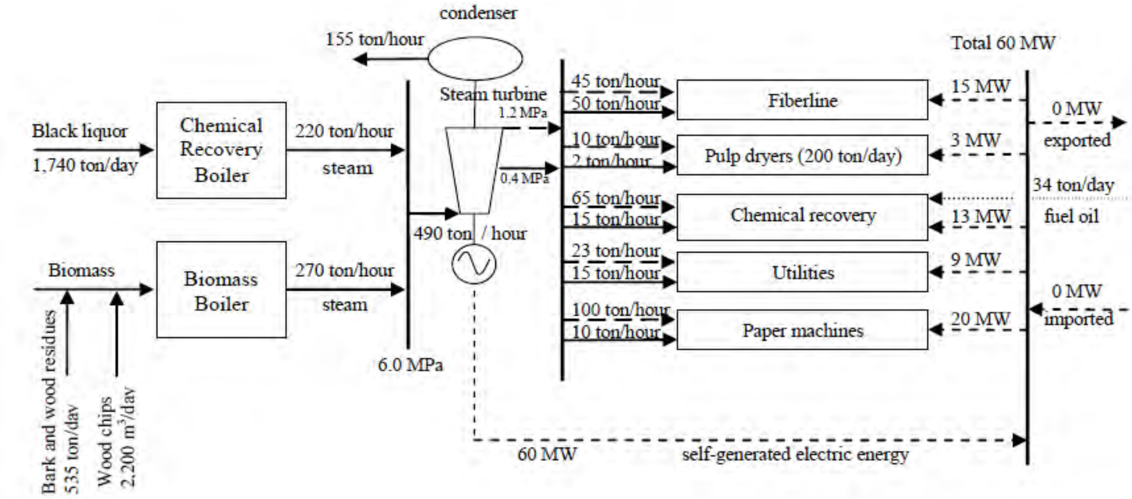

Figure 2 - Integrated pulp and paper mill with extraction/condensing cogeneration scheme (adapted from Moraes, 2011)

Figure 3 illustrates the proposed BLGCC scheme for an integrated pulp and paper mill. In this analysis, steam production in the biomass boiler is maintained in the scheme for a final destination of wood residues generated in the process. The BLGCC cycle was modeled with the commercial software Cycle Tempo (2007) and every main unit is identified by a grey box. A pressurized air-blown gasifier was considered in the modeling due to its low cost in comparison to the other gasification options. The input values of black liquor for the gasifier and of bark and wood residues and wood chips for the biomass boiler are taken from the backpressure/extraction cogeneration scheme of Figure 1. The biomass boiler, in this case, burns the same amount of wood chips of backpressure/extraction scheme, i. e, with the same amount of fuel in both cases. Figs. 4 to 5 illustrate the main blocks of BLGCC scheme. Thermodynamic data is available in the figures according to the convention presented in the legend of Figures 3 and 5.

The analysis of BLGCC scheme revealed that the gross electric power generated is of 157.5 MW, with 97.4 MW in the gas turbine and 60.1 MW in the steam turbine. As total demand of pulp and paper mill is of 60 MW, a surplus of 97.5 MW is available to be commercialized to the grid, and this improves the economic attractiveness of this investment.

As an exergetic analysis is presented for the modeling of such cogeneration unit, energetic and exergetic concepts are briefly addressed for describing the main equations necessary to the analysis. In the energetic analysis, mass conservation equation is expressed by Eq. (1) assuming that steady state for the analysis. The first and second laws of Thermodynamics are defined by Eqs. (2) and (3) for steady state; subscripts "i", "e" and "j" refer respectively to the inlet, exit and heat flows over the control volume (CV) for heat flow ($\dot{Q}$), axis power ($\dot{W}$) and generated entropy ($\dot{S}$) in terms of mass flow ($\dot{m}$), specific enthalpy (h) and specific entropy (s).

$$\sum \dot{m}_e = \sum \dot{m}_i \tag{1}$$

$$\dot{Q}_{cv} = \sum \dot{m}_e h_e - \sum \dot{m}_i h_i + \dot{W}_{vc} \tag{2}$$

$$\dot{S}_{ger_{cv}} + \sum_j \left(\frac{\dot{Q}_{cv_j}}{T_j}\right) + \sum \dot{m}_i s_i - \sum \dot{m}_e s_e = 0 \tag{3}$$

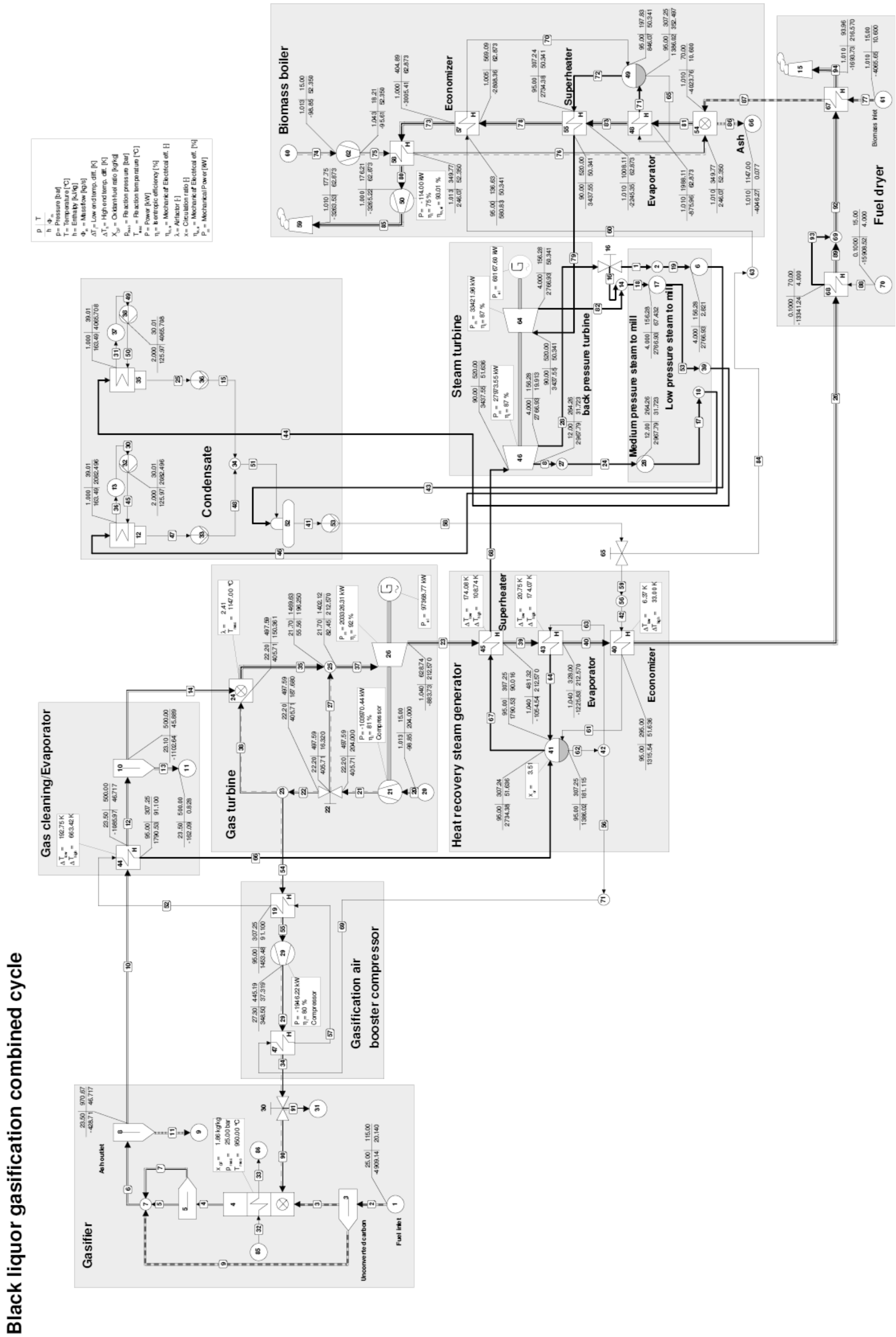

Figure 3 – Air-blown black liquor integrated gasification combined cycle scheme

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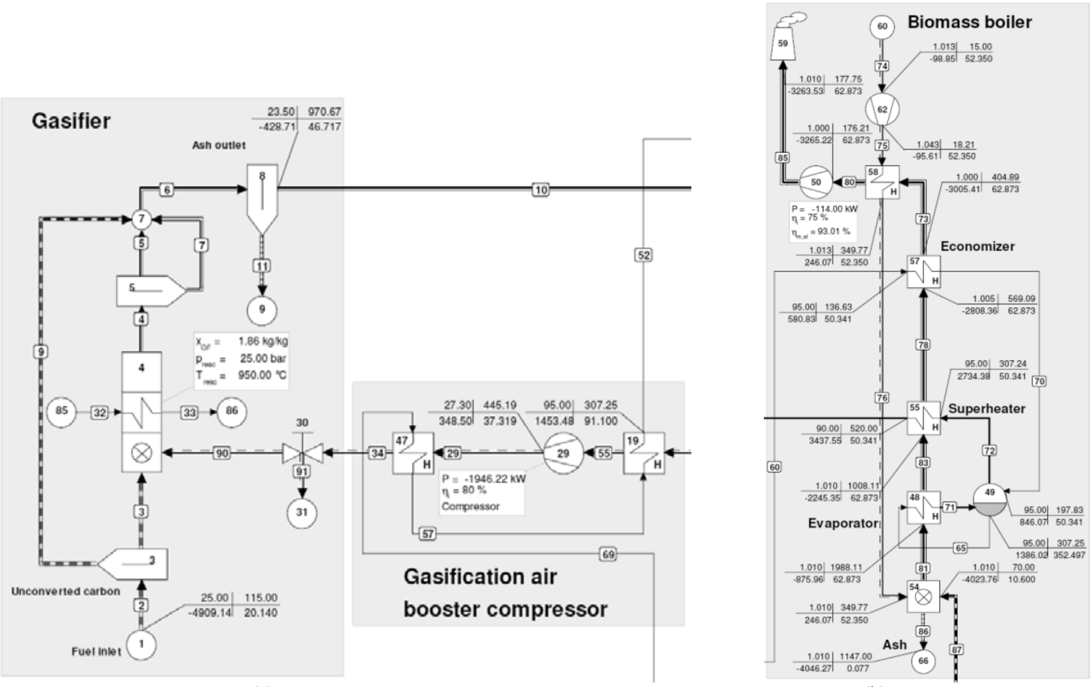

(a) (b)

Figura 4: (a) Gasifier and gasification air booster compressor; (b) Biomass boiler

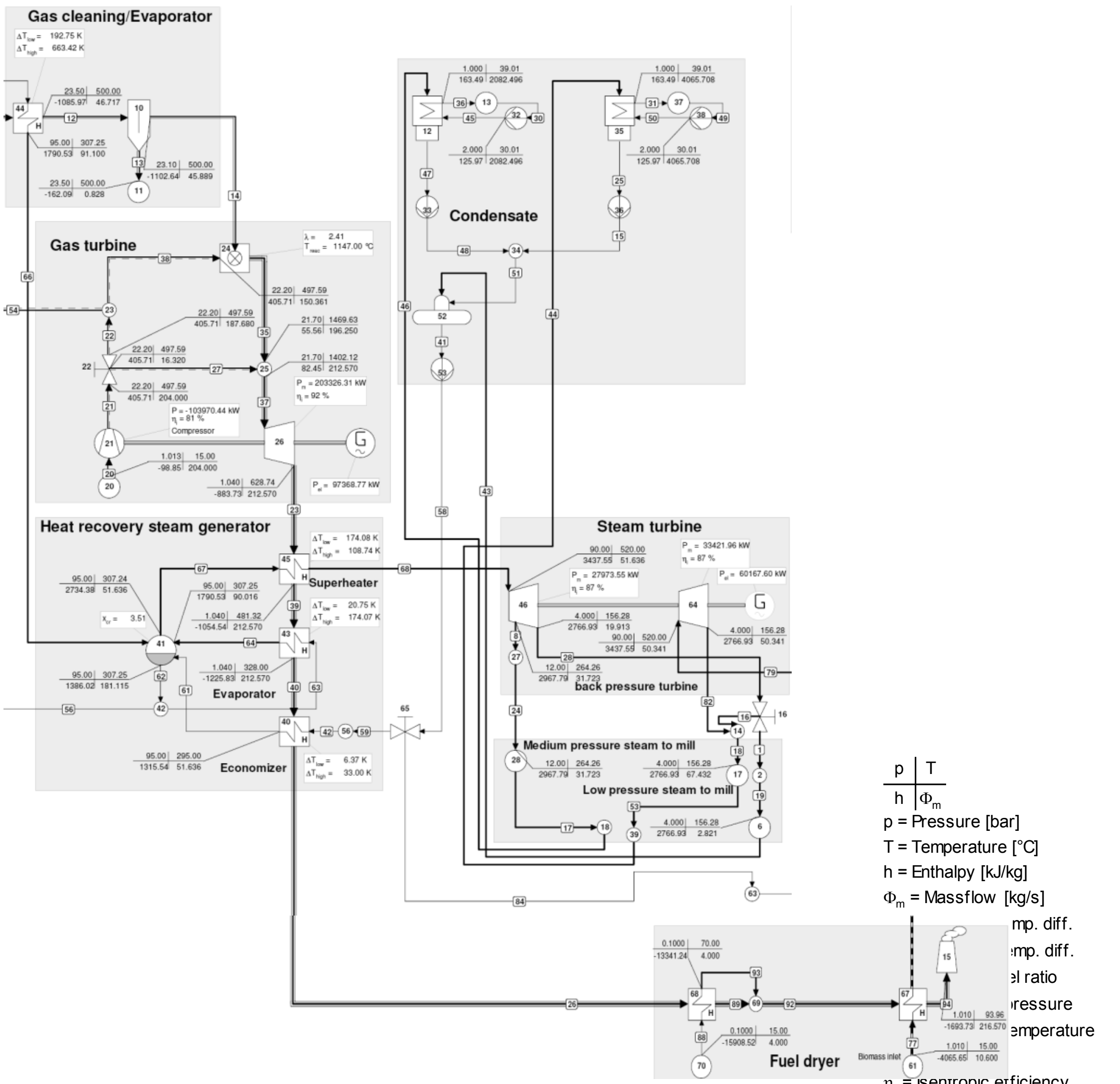

Figura 5: Gas turbine, HRSG and steam turbine.

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In the exergetic analysis, exergy is presented in its separated components (Eq. 4). Kinetic and potential exergy will not be considered in the present analysis; physical and chemical specific exergy are expressed, respectively, by Eqs. (5) and (6). Reference state was defined as P_0=101 kPa and T_0 = 298 K (h_0 = 104.9 kJ/kg, s_0 = 0.3672 kJ/kgK).

$$\dot{E}x = \dot{E}x_c + \dot{E}x_p + \dot{E}x_f + \dot{E}x_q \quad (4)$$

$$ex_f = \frac{\dot{E}x_f}{\dot{m}} = h - h_0 - T_0(s - s_0) \quad (5)$$

$$ex_q = \frac{\dot{E}x_q}{\dot{m}} = \sum_j x_j \bar{e}_j^Q + \bar{R}T_0 \sum_j x_j \ln x_j \quad (6)$$

Exergy analysis is developed considering the irreversibility (destroyed exergy) and exergetic efficiency, as defined respectively by Eqs. (7) and (8). For Eq. (8), the sum of exit exergy represents the products and the sum of inlet exergy represents the “fuel” of each component under analysis.

$$\dot{E}_d = \sum_j (1 - \frac{T_0}{T_j})\dot{Q}_j - \dot{W}_{vc} + \sum_i \dot{m} \cdot ex_i - \sum_e \dot{m} \cdot ex_e \quad (7)$$

$$\eta_{II} = \frac{\sum \dot{E}x_e}{\sum \dot{E}x_i} \quad (8)$$

4. MODELING AND EXERGETIC ANALYSIS OF BLGCC

4.1 Gasifier modeling

For the analysis of the synthesis gas production by the gasification of black liquor, a chemical composition for this residue was defined. The black liquor composition proposed in Table 1 was considered in this analysis and its properties are presented, whose molar composition is $CH_{1.271}O_{0.095}N_{0.018}S_{0.038}$. A previous multi-effect evaporation process was applied for concentration of the black liquor.

Table 1 - Properties of black liquor

Composition	Percentage (mass)
C	34.0
H	3.6
O	4.3
N	0.7
S	3.4
Ashes	44.0
Total	90.0

Source: D’Almeida (1987) apud Carreiro (2009)

The expression by Channiwala and Parikh (2002) in Eq. (9) was considered for estimating the black liquor higher heating value (HHV) for the composition presented in Table 1; the value is expressed in MJ/kg and “A” means “ashes”. The higher heating value of 15,069 kJ/kg was obtained for the considered black liquor composition. This represents a value higher than the 12,000 kJ/kg average black liquor heating value for Brazilian conditions (Carreiro, 2009). Cardoso et al. (2009), Huang and Ramaswamy (2011) consider that the typical higher heating value of black liquor is in the range of 13,400 to 15,500 kJ/kg.

$$HHV = 0.3491C + 1.1783H + 0.1005S - 0.1034O - 0.0151N - 0.0211A \quad (9)$$

For the calculation of black liquor exergy, empirical correlations for solid dry fuels (Kotas, 1985) given by Eq. (10) and given by Eq. (11) (Song et al., 2011) were considered, being e^0 the fuel specific exergy. Equation (11) is valid for (O/C) < 0.5, in which H, C, O and N are the mass fractions presented in Table 1, for which it is resulted β= 1.0584.

$$\beta = \frac{e^0}{LHV} \quad (10)$$

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$$\beta = 1.437 + 0.0140\frac{H}{C} + 0.0968\frac{O}{C} + 0.0467\frac{N}{C} \qquad (11)$$

The lower heating value (LHV) was obtained from Eq. (12) (Good et al., 2006) taking the value of the HHV of 15,069 kJ/kg and 3.6% of hydrogen content, according to Table 1, resulting 15,111 kJ/kg for the specific exergy of black liquor.

$$LHV = HHV - 2442(9.01H) \qquad (12)$$

According to Dinçer and Zamfirescu (2011), the reactor temperature is in the range 900°C to 1100°C. The analysis of power generation of BLGCC cycle is based on the composition of the gasifier generated gas operating with pressurized air at 2.5 MPa and 950 ºC. The molar fractions of the synthesis gas composition exiting the gasifier are presented in Table 2. Simulated results were obtained by the kinetic modeling of Cycle-Tempo commercial software (Cycle Tempo, 2007).

Table 2 – Molar fractions of compositions and operational conditions of gasifier

Compositions (mol)	Simulation
CH_4	0.0074
CO	0.2646
CO_2	0.0165
H_2O	0.0175
H_2	0.1688
N_2	0.5191
H_2S	0.0000
Ar	0.0061
Temperature (ºC)	950
Pressure (MPa)	2.5

According to Dinçer and Zamfirescu (2011), the lower heating value of a gas produced in an air gasification process containing up to 60% N_2 have a typical value of 4 to 6 MJ/Nm^3. Li et al (2004) consider that the high heating value of product gas (HHV_g) can be calculated with Eq. (13), in which the value is expressed in MJ/Nm^3. The higher heating value of 5,789 MJ/Nm^3 was obtained for the considered gas composition of simulation.

$$HHV_g = 12.75\,y_{H2} + 12.63\,y_{CO} + 39.82\,y_{CH4} + 25.105\,y_{H2S} \qquad (13)$$

where y_{H2}, y_{CO}, y_{CH4} and y_{H2S} are the mole fractions of H_2, CO, CH_4 and H_2S, respectively.

The cogeneration cycle was implemented in the simulator with the supply of all the necessary parameters. Figure 6 illustrates the black-box model of gasifier and the corresponding Cycle Tempo model all the equipments of cycle; for the remaining components of BLGCC it is presented just the black-box with corresponding inlet and outlet flows.

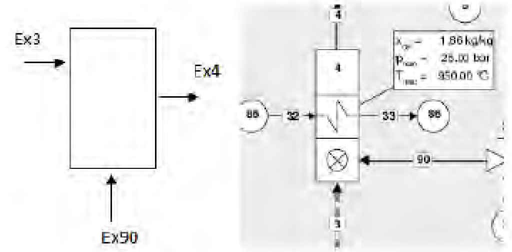

Figure 6 – Gasifier

4.2 Combined cycle modeling

The thermodynamic analysis for the gas turbine (Figure 8) was based on a scheme in which compressor present air extractions for intermediate cooling of stages and part of the air is sent to the gasification process, according to the data synthesis gas composition presented in Table 3.

Due to the change of original fuel by a low heating value fuel as the synthesis gas, modifications in the compressor structure and combustion chamber are expected (Gupta et al., 2010). In this case, as described by Klimantos et al. (2009), the use of one or more control strategies must be adopted in order to avoid compressor instability problems.

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Bleeding air from the compressor is one of these control strategies and a ratio of 1.7 to 2.0 for fuel gas to air bleed flow is considered in the same reference; for the proposed scheme, the fuel to bleeding air ratio is of λ= 1.86. Assuming full load and a mechanical efficiency (η_m) of 0.99, according to Larson et al. (2000), to take into account transmission box and electric generator, a net power of 97,369 kW was obtained for the gas turbine. The mass and energy conservation laws were successfully applied in each component of gas turbine.

Table 3 – Molar composition of the gas that leaves the combustion chamber

Composition of the gas	x_i (%)
CO_2	8.33
H_2O	6.57
N_2	74.81
O_2	9.41
Ar	0.88
Sum	100.00

Based on the synthesis gas composition, the molar composition of the exhaust gas that leaves the gas turbine is presented in Table 4. This gas is send to the heat recovery steam generator, whose exergetic flows are described in the corresponding black-box of Figure 7(other main equipment are also present in the same figure).

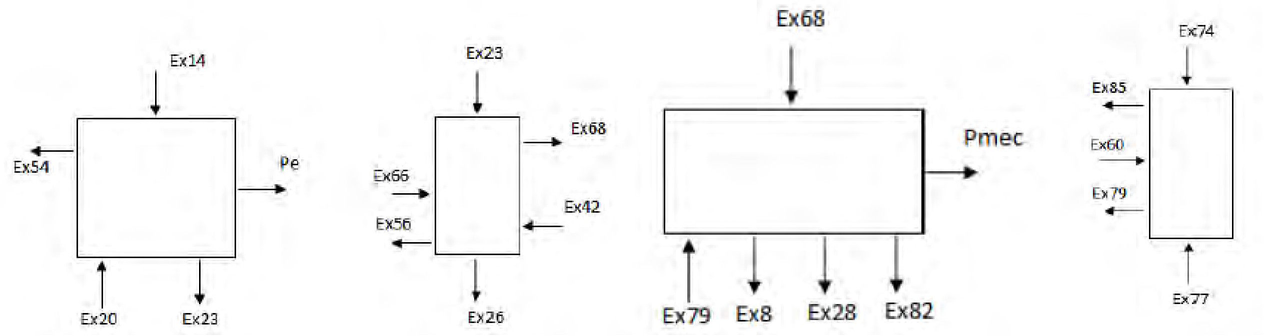

Figure 7 – Black-box for (a) gas turbine, (b) heat recovery steam generator, (c) steam turbine and (d) biomass boiler

Table 4 – Molar composition of the exhaust gas that leaves the gas turbine

Composition of the gas	x_i (%)
CO_2	7.68
H_2O	6.14
N_2	75.00
O_2	10.29
Ar	0.89
Sum	100.00

Table 5 presents the exergetic efficiency calculated according to Eq. (8) for the main components of BLGCC scheme. Total exergy for the main flows of BLGCC scheme are also presented in Table 6.

Table 5 – Exergetic efficiency for the main components of BLGCC scheme proposed in Fig. 3

Description of components	η_{II} (%)
Gasifier	86.40
Gas turbine	75.04
Heat recovery steam generator	86.51
Steam turbine	89.77
Biomass boiler	38.88

Table 6 – Main inlet and outlet exergy flow the BLGCC scheme

Exergy flow	(kW)
Ex_3	342,401.41
Ex_4	323,649.50
Ex_8	31,198.69
Ex_{14}	253,113.41
Ex_{20}	29.66
Ex_{23}	75,183.63
Ex_{26}	12,619.62
Ex_{28}	15,172.49
Ex_{42}	4,853.53
Ex_{54}	17,414.99
Ex_{56}	39,154.80
Ex_{60}	4,731.80
Ex_{66}	57,710.31
Ex_{68}	77,534.11
Ex_{74}	6.54
Ex_{77}	213,770.94
Ex_{79}	75,589.44
Ex_{82}	38,356.80
Ex_{85}	31,521.45
Ex_{90}	32,212.43
P_{mec}	61,395.51
P_e	97,368.77

5. CONCLUSIONS

An air-blown black liquor integrated gasification combined cycle was modeled and an exergetic analysis developed. Black liquor gasification is still a technology under development and this adds difficulties to the comparison of results. Its development is of great importance for the pulp and paper sector because this technology is intended to replace the conventional Tomlinson chemical recovery boiler with a potential increase in the power generation and energy efficiency. The challenges are not just for the black liquor gasification and the synthesis gas cleaning, but also for the gas turbine technology, that must be adequately modified for burning a low energy fuel.

The energetic results of the proposed BLGCC cycle were compared to the conventional steam cycle based on the Tomlinson boiler. For the supplying of steam to the processes of an integrated pulp and paper mill that produces 1,200 ton/day of pulp and 1,000 ton/day of paper and demands 60 MW of electric power, a surplus of 97.4 MW was obtained for the BLGCC cycle. For the same design premises, an amount of 22 MW of electric power must be imported from the grid for the backpressure/extraction steam turbine conventional cycle, and an electric self-sufficiency is obtained for the condensing/extraction steam turbine conventional cycle, the availability of a significant power to be commercialized to the grid certainly improves the economic attractiveness of this investment.

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