

ENCIT-2018-0451

EXPERIMENTAL DETERMINATION OF PYROLYSIS GAS FLOW RATE IN A CHARCOALING KILN

C. B. Schutze
N. G. B. Gomes
C. A. Gasparetto
W. A. Bizzo

State University of Campinas - UNICAMP

cbschutze@fem.unicamp.br, neil.gomes@fem.unicamp.br, gasparetto@fcm.unicamp.br, bizzo@fem.unicamp.br

Abstract. *In order to increase the competitiveness of the industrial charcoal production, several authors suggest the utilization of low heating power emissions from charcoaling ovens for the abatement of toxic components in the gases and for the generation of energy, whether it be thermal or electrical. This work presents a proposed methodology for experimental estimation of the mass flow rate of pyrolysis gases from a charcoaling kiln. Based on experimental data from incoming air mass flow rates and temperature variations in a burner prototype on top of a process kiln, an energy balance equation was proposed. Considering the input data for a complete oven run, an average mass flow rate of 1.22 kg/s was calculated during the process. As part of a larger project for the utilization of these slow pyrolysis gases, further analysis are required in order to generate a more precise mass flow rate curve. These results could support future research of charcoaling gas emissions due to the difficulties in precise mass flow measurements and the design of slow pyrolysis gas burners.*

Keywords: *Mass Flow Rate, Slow Pyrolysis, Charcoal, Low Calorific Values Gases, Energy Balance*

1. INTRODUCTION

Brazil is the world's biggest producer of charcoal from wood pyrolysis with 10.5% of the world production in 2016 (FAO (2016) primarily for the pig iron and steel industries. The slow pyrolysis process currently done in Brazil is very inefficient and produced gas is emitted to atmosphere. These gases represent about 30 to 50% of the initial energy content of the wood (Leme *et al.* (2016) apud de Miranda *et al.* (2013)).

The emissions associated with charcoal production produces greenhouse gases such as CO₂, CH₄ and N₂O (Pennise *et al.* (2001)). Inherently to the slow pyrolysis process among the partial combustion products there are several highly toxic Polycyclic aromatic hydrocarbons (PAHs), according to Barbosa *et al.* (2006) more than 16 different PAHs were found in charcoal production process.

On the other hand the presence of partial combustion products such as CH₄ enables the possibility of exploitation of waste gases from the production process as fuel. The energy utilization of the gases emitted can be utilized for drying the wood to be introduced into the kilns and for generation of electricity for use in the charcoal production unit itself or even distributed across the electricity network (Pereira *et al.* (2017)).

There are several barriers that limit the use of the emitted gases from charcoal production, among them (Vilela *et al.* (2014) cites the low heating value of the gases; variable composition and temperature of the gases in different carbonization stages; variable moisture content of the tar generated during the carbonization process; difficulties in developing a project of an adequate burner for the combustion of the pyrolysis gases with high and variable content of particulates, moisture and condensables, which would require the installation of pre-filters and dilution furthermore cites that occurs partial burning of the gases due to admission of undesired air.

In addition to (Vilela *et al.* (2014) (Pereira *et al.* (2017) argues that fundamental knowledge of the input and output variables is needed such as the temperature, moisture content and gas output flow. There is scarce information regarding the calorific value of the gas and specially the mass flow during the period of carbonization. One of the few studies that presents data from a real operation kiln is (Leme *et al.* (2016) presenting the average mass flow in Table 1.

In the work presented by (Leme *et al.* (2016) the methodology proposed is the AM0041 from UNFCCC which is used under the Carbon Credit market and presents guidelines that, briefly, correlate methane emissions and the gravimetric yield. Although it refers to the composition of the gases "i.e., wood and charcoal weights on dry basis, and collect gas samples throughout the carbonization process" (UNFCCC, 2010, p. 7) it gives no reference how to measure or estimate the mass flow rate during the process. Instead it provides assumptions to calculate a complete mass balance of the whole

process "Based on the experimental results, the mass balance to calculate the mass of methane released for each dry ton of charcoal shall be calculated" (UNFCCC, 2010, p. 34).

Table 1: Average flow and calorific value of the carbonization gases produced in the rectangular kiln Leme *et al.* (2016) sic.

		First 60 hours				Second 60 hours			
		Mean	Max	Min	RSD ¹	Mean	Max	Min	RSD ¹
Flow _{NCG}	kg/h	4498	4808	4364	78	4384	4480	3754	99
Flow _{CG}	kg/h	786	1294	219	261	646	1276	235	242

¹Relative Standard Deviation

With a better description of the emission behavior from wood pyrolysis in a industrial kiln further development can be proposed for a efficient gas burner design and system sizing. The methodology proposed by UNFCCC gives very useful guidelines for green gas emissions but there are no specific guidelines for the charcoal production and specially regarding the mass flow rate.

It is very difficult to measure gas composition throughout the entire process that can last up to 10 days, presence of condensing matter and very complex composition Oliveira *et al.* (1982). Mass flow rate is extremely complex to be done directly due to low pressure and presence of condensible gases Oliveira *et al.* (1982). Normally the exhaust gas is driven by natural convection in a chimney with less than 10m with a rectangular shape in the base into a cylindrical shape as shown in Figure 3 which make difficult for flow stabilization and accurate measurement. Those characteristics for mass flow rate measurements make usual methods hard to be adapted or implying in very costly operating equipment such as non intrusive flow measurements.

1.1 Acquired data from bibliography

Several test performed by ESALQ (Luiz de Queiroz College of Agriculture) for V & M Florestal in accordance with the AM0041 Baseline and Monitoring Methodology where made in charcoaling kilns of similar proportions to the ones where the FLOX burner was to be installed that characterized both the pyro-gas molar composition as well as the generation of condensible vapours throughout the complete pyrolysis process. Figure 1 shows the measured molar composition for the total 120 hours that the process requires in these types of ovens.

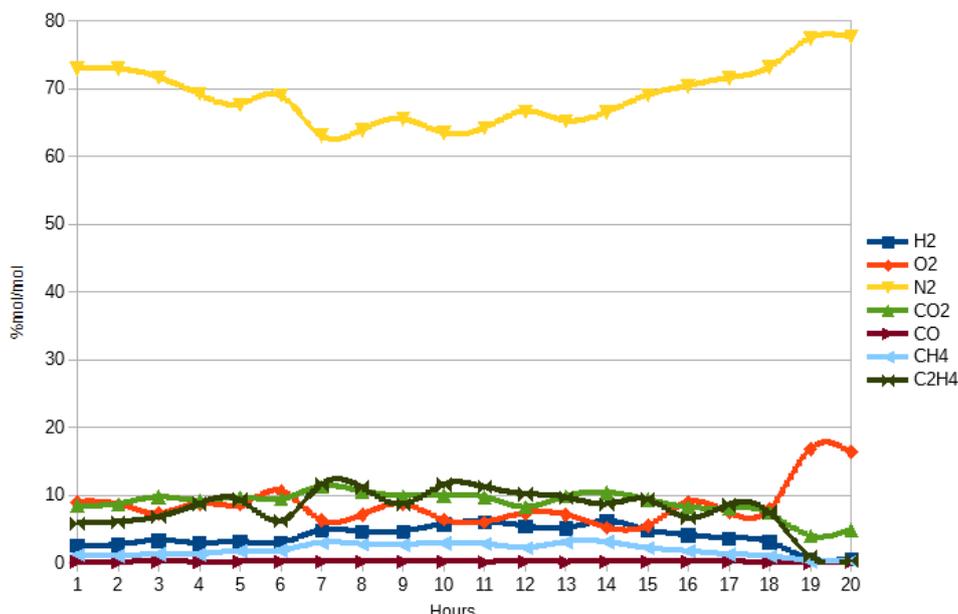


Figure 1: Molar composition of non-condensable pyro-gases as measured by (LQCE, 2011).

Figure 2 on the other hand, presents us with the ratios between the condensible and non-condensable gases collected by the ESALQ team during the characterization of the pyro-gases exiting the charcoaling kiln. Although sadly, the varying composition of these condensates where not characterized through time, this emissions curve can assist in understanding the total mass flow of the complete pyro-gases.

This data, does present some problems however, as can be seen in Fig. 1, the amount of H₂ within the non-condensable gases is higher than amount of CO present. This is an issue considering the literature that presents a greater abundance

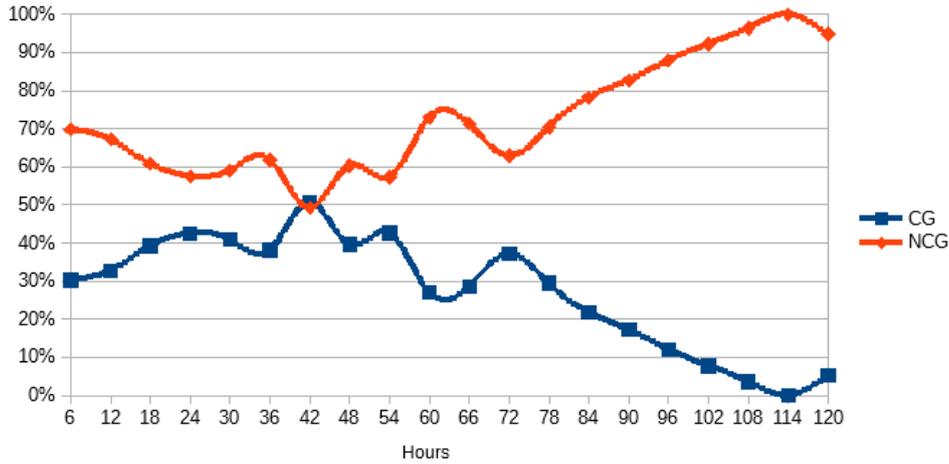


Figure 2: Mass proportions of condensable and non-condensable pyro-gases as measured by (LQCE, 2011).

of CO in relation to H₂ as the works of Kırtay (2011) and Balat and Kırtay (2010) present, signaling a probable error, either in the measurements or in the publication of the results. Either way, these are the most reliable recent emissions compositions published for such a large scale and outdated charcoaling kiln.

2. METHODOLOGY

2.1 Pyro-gas burner: design and flow diagram

The proposed burner design attached to the top of the kiln chimney is a modified FLOX-type burner for gases with a low heating value. The basic requirement for the set-up was to maintain the current pyro-gas flow rate in order to interfere as little as possible with the charcoaling process. Because of this, it was difficult to deal with flame flash-backs due to the pyro-gase's low velocities, high condensate quantities and mass flow variability as well as the ever fluctuating elemental compositions of the exhausts gases. Figure 3 contains a partial view of the installed gas burner Gomes *et al.* (2017).

This prototype contains various temperature and pressure sensors in order to properly control the combustion through the intake of pre-heated air. So, when there is no combustion occurring, the burner becomes a basic mixer with complete instrumentation that allows for the calculation of the incoming and outgoing mass flows.

2.2 Determining total pyro-gas flow

A simple energy and mass balance, considering the installed burner as a black box can assist in calculating the pyro-gas emanating from within the carbonization kiln. Equation 1 is the isolated energy balance for the flows shown in Fig. 4 required to find \dot{M}_{pyro} , or the pyro-gas in kg/s.

$$\dot{M}_{pyro} = \dot{M}_{air} \frac{(Cp_{air}T_3 - Cp_{air}T_2)}{((1 - U_{cg})Cp_{ncg}T_1 + U_{cg}Cp_{cg}T_1) - ((1 - U_{cg})Cp_{ncg}T_3 + U_{cg}Cp_{cg}T_3)} \quad (1)$$

in which:

- \dot{M}_{air} is the measured mass flow of air in *kg/s*.
- Cp_{air} is the specific heat at constant pressure of air in *kJ/kgK* at varying temperatures.
- Cp_{ncg} is the specific heat at constant pressure of the non-condensable gases in *kJ/kgK* at varying temperatures.
- Cp_{cg} is the specific heat at constant pressure of the condensable gases in *kJ/kgK* at varying temperatures.
- U_{cg} is the condensable mass fraction with respect to the total pyro-gas mass flow
- T_1 is the measured temperature of the incoming pyro-gas in *K*.
- T_2 is the measured temperature of the incoming air in *K*.
- T_3 is the measured temperature of the outgoing mixture in *K*.

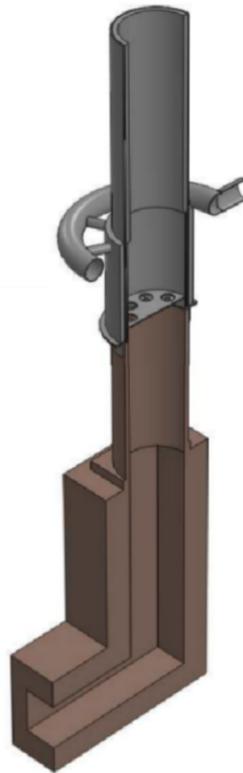


Figure 3: Representation of the Pyro-gas burner prototype, installed on top of the charcoal kiln chimney Gomes *et al.* (2017).

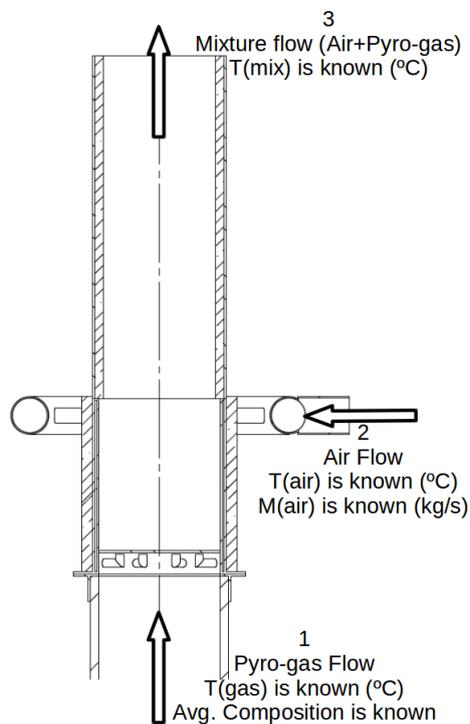


Figure 4: Diagram showing the input and output fluxes considered.

2.3 Specific heat equations

In order to find the specific heats of the various gases that comprise the total condensible pyro-gas, the following equation 2 was implemented to acquire the various specific heats in function of the varying temperatures for the entering

pyro-gas, air and accompanying exiting mixture.

$$Cp_i = (A + BT + CT^2 + DT^3 + ET^4)(R/M_i) \quad (2)$$

in which,

- Cp_i is the specific heat at constant pressure of a compound in $kJ/kmolK$.
- A-E are the constants shown in Tab. 2.
- R is the Universal gas constant at $8.3145 kJ/kmolK$.
- T is the temperature at which the gas is.
- M_i is the molar mass if the compound in $kg/kmol$.

Therefore, with these equation, finding the specific heat of air becomes a simple application of the appropriate constants for the varying temperatures. However, the pyro-gases specific heat requires another simple yet important step in order to properly characterize the specific heat.

Table 2: Specific heat capacity constants from Moran *et al.* (2004).

Element	A	B	C	D	E
H ₂	3.057	2.677	-5.8102	5.521	-1.812
O ₂	3.626	-1.878	7.055	-6.764	2.156
N ₂	3.675	-1.208	2.324	-0.632	-0.226
CO ₂	2.401	8.735	-6.607	2.002	0
CO	3.710	-1.619	3.692	-2.032	0.240
CH ₄	3.826	-3.979	24.558	-22.733	6.963
C ₂ H ₆	1.426	11.383	7.989	-16.254	6.749
Air	3.653	-1.337	3.294	-1.913	0.2763

Knowing the individual specific heats of the gases that make up the non-condensable pyro-gas, eq. 3 can be utilised to find the varying final specific heat or Cp_{ncg} in kJ/kgK .

$$Cp_{ncg} = \sum P_i Cp_i \quad (3)$$

in which,

- Cp_i is the specific heat at constant pressure of a compound in kJ/kgK .
- P_i is the mass composition of the compound in the non-condensable pyro-gas.

Considering that the condensable gases that make up the pyro-gas emanating from the charcoaling kilns are measured and characterized with great difficulty as it varies with time, average specific heats for the remaining condensable vapours where used to calculate Cp_{cg} , or the equivalent specific heats. Table 3 contains the average composition of the pyro-gas condensate of *Eucalyptus Grandis* at age 5.5 years and carbonization temperature of 450°C as gathered from Oliveira *et al.* (1982).

Table 3: Average composition of condensable pyro-gas in volume (dry base) from Oliveira *et al.* (1982).

Methanol	Acetaldehyde	Ethanol	Acetone	Methyl-acetate	Acetic Acid	Soluble Tar
3.15	<0.05	-	0.12	0.16	9.87	11.2

Only, methanol, acetic acid and soluble tars where used to calculate the appropriet specific heat of the consisible pyro-gases, along with the respective water vapour which according to Oliveira *et al.* (1982) would represent approximately 75 % of the volume of total condensates.

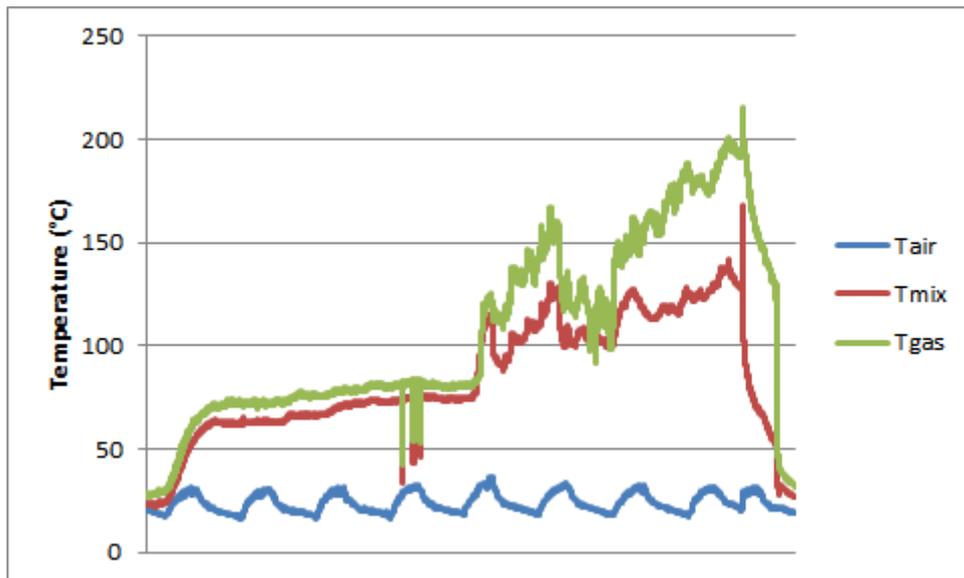


Figure 5: Measured temperatures for air, pyro-gas and mixture in °Celcius.

3. RESULTS AND DISCUSSION

3.1 Temperature and air flow curves

The measured temperatures for the incoming air flow, the pyro-gas exiting from the charcoaling kiln and their resulting mixture can be seen in fig.5.

The temperatures for all three of the flows show a large spike in that is sustained for several hours (approximately 19 hours). This spike was caused by the intended ignition of the pyro-gases as they reach ignitable levels of composition.

Figure 6 on the other hand, shows the injection of air that not only serves to oxygenize the fuel mixture, but to create a suctioning force that would negate the pressure differences applied by the obstruction of the chimney by the actual burner. This is why the air intake is not constant and shows small valleys and peeks. As the pressure difference between the chimney (pre-burner) and the combustion chamber varies with the influx of generated pyro-gas, air is injected to maintain the variations in pressure as low as possible in order to not interfere greatly with the carbonization process.

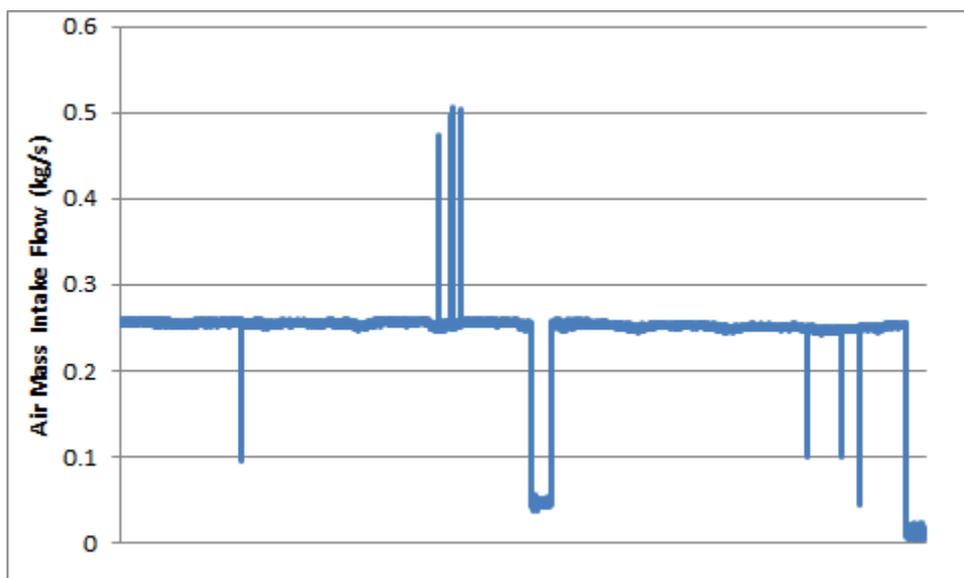


Figure 6: Measured air mass flow rate in kg/s.

3.1.1 Mass flow curve

The resulting pyro-gas flow curve, calculated using the equations from section 2 and both the composition and condensible proportions gathered from the ESALQ team in section 1 can be seen in fig.7.

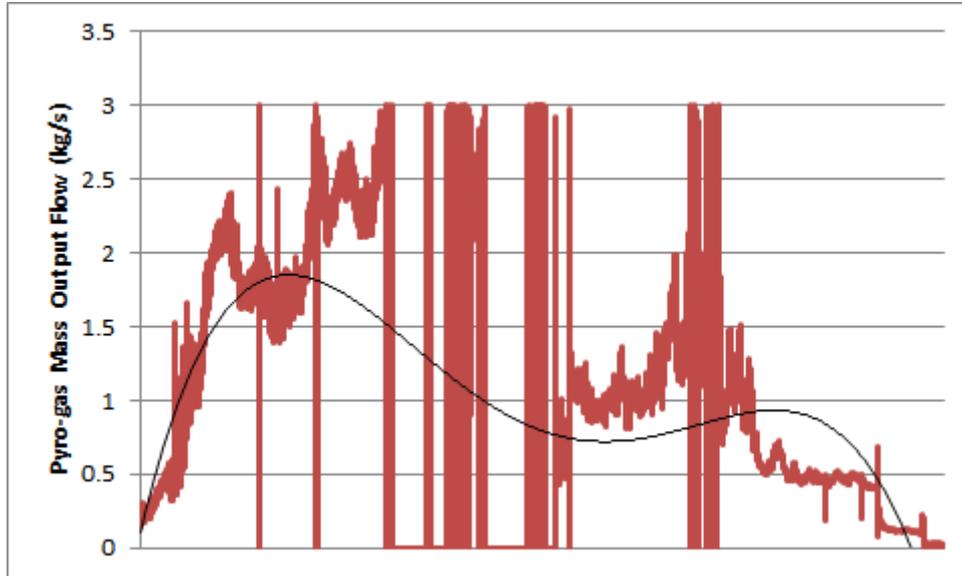


Figure 7: Calculated pyro-gas flow rate in kg/s.

This curve does present some issues, mainly due to the incompatibility of the methodology when the temperature of the mix is less than the temperature of the gas before it enters the burner which actually negates the proposed energy balance. In fig. 7 the peaks greater than 3.0 kg/s per second or less than 0 kg/s were removed.

4. CONCLUSIONS

- The calculated pyro-gas flow rate was an average of 1.22 kg/s which results in a expected emission.
- The gathered information on the composition of the non-condensable gases presented some issues since the beginning.
- The condensable gases were not characterized through time, requiring the use of average compositions to determine the specific heats of the condensable vapours of the pyro-gas.

Proposed solutions are to re-characterize the non-condensates and condensable gases.

5. ACKNOWLEDGEMENTS

This work was financially supported by the research and development program Gerdau/ANEEL. Program in cooperation of Gerdau Florestal, Gerdau Dona Francisca Energética and the R& D program proposed by ANEEL.

6. REFERENCES

- Balat, H. and Kirtay, E., 2010. "Hydrogen from biomass—present scenario and future prospects". *International Journal of Hydrogen Energy*, Vol. 35, No. 14, pp. 7416–7426.
- Barbosa, J.M.d.S., Ré-Poppi, N. and Santiago-Silva, M., 2006. "Polycyclic aromatic hydrocarbons from wood pyrolysis in charcoal production furnaces". *Environmental research*, Vol. 101, No. 3, pp. 304–311.
- de Miranda, R.C., Bailis, R. and de Oliveira Vilela, A., 2013. "Cogenerating electricity from charcoaling: A promising new advanced technology". *Energy for Sustainable Development*, Vol. 17, No. 2, pp. 171–176.
- FAO, 2016. "Forestry production and trade". Technical report, Food and Agriculture Organization of the United Nations.
- Gomes, N.G.B., Chávez, J.V., Medeiros Filho, D., Carvalho, D.J., Santos, R.G. and Bizzo, W.A., 2017. "Methodology for burner design - combustion of pyrolysis gas from charcoal production." *Proceedings of COBEM 2017*.
- Kirtay, E., 2011. "Recent advances in production of hydrogen from biomass". *Energy conversion and management*, Vol. 52, No. 4, pp. 1778–1789.
- Leme, M.M.V., Venturini, O.J., Lora, E.E.S., Rocha, M.H. and Almeida, W.d., 2016. "Estudo técnico, econômico e ambiental da utilização de alternativas tecnológicas para a geração de eletricidade na cadeia produtiva do carvão vegetal no Brasil." *Proceedings of ENCIT 2016*.
- LQCE, 2011. "Projeto carbonização - mitigação de emissões de metano na produção de carvão vegetal da vallourec & mannesmann florestal anexo 1". Relatório, ESALQ, Minas Gerais - Brasil.
- Moran, M.J., Shapiro, H.N. and Boettner, D.D., 2004. *Princípios de termodinâmica para engenharia*. Grupo Gen-LTC.
- Oliveira, J.B., Vivacqua Filho, A., Mendes, M.P. and Gomes, P.A., 1982. "Produção de carvão vegetal - aspectos técnicos". In W.R. Penedo, ed., *Produção e Utilização de Carvão Vegetal*, Fundação Centro Tecnológico de Minas Gerais - CETEC, Belo Horizonte, chapter 2, pp. 59–73.
- Pennise, D.M., Smith, K.R., Kithinji, J.P., Rezende, M.E., Raad, T.J., Zhang, J. and Fan, C., 2001. "Emissions of greenhouse gases and other airborne pollutants from charcoal making in Kenya and Brazil". *Journal of Geophysical Research: Atmospheres*, Vol. 106, No. D20, pp. 24143–24155.
- Pereira, E.G., Martins, M.A., Pecenka, R. and Angélica de Cássia, O.C., 2017. "Pyrolysis gases burners: Sustainability for integrated production of charcoal, heat and electricity". *Renewable and Sustainable Energy Reviews*, Vol. 75, pp. 592–600.
- UNFCCC, 2010. "Am0041 - mitigation of methane emissions in the wood carbonization activity for charcoal production version 1". Technical report, UNFCCC - The United Nations Framework Convention on Climate Change.
- Vilela, A.d.O., Lora, E.S., Quintero, Q.R., Vicintin, R.A. and e Souza, T.P.d.S., 2014. "A new technology for the combined production of charcoal and electricity through cogeneration". *biomass and bioenergy*, Vol. 69, pp. 222–240.

7. RESPONSIBILITY NOTICE

The authors are the only responsible for the printed material included in this paper.