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EXPERIMENTAL AND NUMERICAL STUDY OF COAL AND BIOMASS CO-FIRING IN A DROP TUBE FURNACE

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Abstract. *This study is performed to find optimized kinetic parameters for devolatilization and heterogeneous combustion of char for co-firing of coal and biomass combining a numerical modeling with experimental results. The experimental investigation is carried out in a Drop Tube Furnace for different proportions of coal and biomass. Brazilian coal and rice husk are used and their samples are evaluated to obtain their size distribution and composition. Detailed information for particle burnout are extracted along the axis of the furnace at 1100 ºC, according to the ash-as-tracer method. Using the experimental results obtained in the DTF as input data, the numerical model adopted in this work simulates the combustion evolution considering a particle size distribution of fuels. Therefore, the numerical solution estimates the kinetic parameters that have best fit with the behavior of the fuel obtained during experimental tests. There are still important uncertainties related to the experiments, but the numerical results suggest that the model can be used to predict kinetic parameters of these solid fuels.*

Keywords: *biomass, co-firing, kinetic parameters, numerical modeling, drop tube furnace.*

1. INTRODUCTION

During the last decades, pulverized coal has been widely used for power generation around the world. Nevertheless, concerns about the environmental impact of coal combustion has increased mainly regarding greenhouse-gas emissions. Co-firing of coal and biomass in existing coal-fired units appears as a viable alternative to mitigate part of the CO2 emissions, because biomass has a much lower carbon dioxide footptrint. Additionally, co-combustion has a potential effect in reducing SOx and NOx emissions in comparison with pure coal combustion. However, it is important to understand the effects of co-firing in combustion processes before its application in industrial systems.

Biomass is a primary non-fossil energy source consisting of organic matter from animals and plants. For energetic purposes, the most commonly used biomass mainly includes agricultural residues (such as straw, sugarcane bagasse and animal residue) and woody biomass (such as bark, branch and sawdust). The chemical energy stored in biomass can be converted in different types of energy, as electricity or heat, depending on the thermochemical conversion process applied. Generally, biomass fuels are directly burned in power plants, particularly in co-combustion with coal in a conventional coal-fired facility. However, biomass burning may face some difficulties, for instance, high levels of alkali and chlorine present in the biomass can cause an increase of slagging and corrosion effects (Wang *et al.*, 2014). Therefore, the combustion behavior of biomass has become increasingly critical for development of more efficient conversion technologies.

To evaluate the potential application of biomass to existing pulverized coal units, some work has been done at laboratory scale using drop tube furnaces (DTF). The DTF is a bench scale equipment that can adequately simulate the conditions in combustion process of solid fuels that occurs in industrial scale, in particular the high temperatures, the high heating rates and the short residence times. From assays performed in DTF some important combustion characteristics can be identified such as combustion efficiency (burnout) of the fuels, composition of emitted gases and aspects of

deposits formation. Additionally, the evaluation of kinetic parameters (pre-exponential factors and activation energies) can also be performed using experimental results from tests in DTF as input data for modeling.

Combustion is a very complex process and, depending on the assumptions and the simplifications considered, the model assumed may not describe this process accurately. The usual procedure for determining kinetic parameters is based on fitting the slope of an Arrhenius plot. However, this approach does not take into account the dispersion in particle size of the fuel, instead, a single representative diameter is considered, which reduces the accuracy of the results as shown by Pereira *et al.* (2016). Among other results, the authors demonstrated that a more complex model with less simplification is more realistic, representing better the combustion process.

Ballester and Jiménez (2005) presented a model for deriving kinetic parameters that is based on analyzing the direct simulation of the complete history of the particles, considering the complete particle size distribution. Therefore, the particles are subdivided into particle size classes and the combustion is investigated for each class. In comparison with the traditional Arrhenius procedure, the authors' model was successful to reproduce the burnout curves. In addition, the model proposed by the authors has a better tolerance for noisy data.

This work has been developed to determine optimized kinetic parameters for devolatilization and heterogeneous combustion of char for co-combustion of coal and biomass combining a numerical modeling with experimental results obtained in a drop tube furnace. More specific, the objective is to investigate if variations on the kinetic parameters for coal/biomass blends follow a proportional ratio.

2. METHOD

2.1. EXPERIMENTAL SETUP AND OPERATIONAL CONDITIONS

A Brazilian coal (BC), rice husk (RH) and a mixture of 90% of coal and 10% of biomass (BC90) were studied in this work. Both fuels were sieved and their size distribution was obtained according to ASTM D4749. Table 1 brings their proximate analysis, ultimate analysis, bulk density and particle size distribution. Proximate analysis was carried out following the standard procedures ASTM D3174 and D3175 for ash and volatile matter contents, respectively. Ultimate analysis was performed according with the standard procedures ASTM D5373 for C, H and N, and ASTM D4239 for S.

Table 1. Fuels characterization.

	BC	**RH**	**BC90**
Proximate Analysis, % dry basis			
Fixed Carbon	38.5	16.6	36,3
Ash	32.5	17.6	31
Volatiles	29	65.5	32,6
Ultimate Analysis, % dry basis			
Carbon	52	42.6	51
Hydrogen	3.5	5.7	3.7
Nitrogen	0.8	0.4	0.8
Oxygen	10.4	33.7	12.7
Sulfur	0.8	ND[1]	0.7
Physical characteristics			
Bulk density, g.cm	1,65	0,51	1,54
Particle size, µm			
Under 38	0.5	0	4,1
Under 75	18.8	3.7	40,5
Under 150	71.1	7.3	74,3
Under 300	100	18.8	91,9
Under 600	100	50.8	95,9
Under 1180	100	100	100

[1] not detected

Coal and rice husk present some significant differences in their composition, especially regarding to proximate analysis results, e.g., RH has higher volatiles, and lower fixed carbon and ash content than BC, typical for biomasses (Vassilev, 2010). With respect to elemental composition, nitrogen content, important concerning to NOx emissions is half for RH when compared to BC. In addition, sulfur was not detect for RH sample, which suggests an advantage of using this type of biomass as a possibility in reducing the sulfur content in a mixture with coal and potentially reducing SOx emissions.

Pure coal presents particles concentrated mainly between 75 µm and 150 µm, which is typical for pulverized coal. In contrast, biomasses have a fibrous structure that makes difficult to reduce their size under 100 µm, as can be noticed

for rice husk. Therefore, an increase in the proportion of rice husk increases the presence of coarse particles in the mixture with Brazilian coal.

The experiments were conducted in a drop tube furnace as shown in Figure 1. It is composed of a cylindrical ceramic tube with an inner diameter of 48 mm and a length of 1.6 m surrounded by three concentric tubular furnaces that are electrically heated. The wall temperatures were continuously controlled and monitored by nine type-K thermocouples.

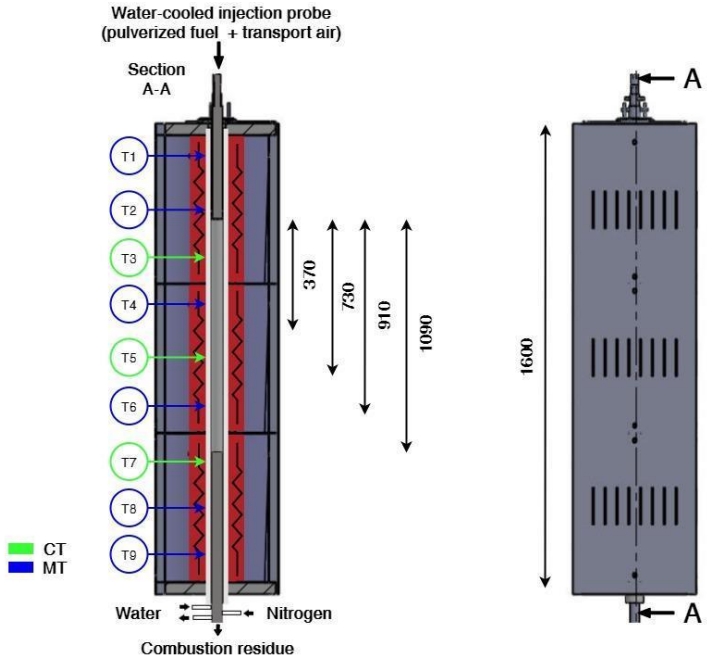

Figure 1. Schematic of the drop tube furnace used in experimental tests. CT = control thermocouples; MT = monitoring furnace temperature thermocouples.

A water-cooled injector, placed at the top of the DTF, is used to feed the solid fuels and the air to the combustion chamber. Before injection, the fuel particles pass through an agitation chamber for sample homogenization. The fuel particles are transported by a twin-screw feeder and enter the tube through the injector with a primary air flow rate that represents 20% of the total air passing through the combustion chamber. This division between primary and secondary air was determined to reach a uniform velocity at the injection point, which approximates the experiments to the one-dimensional flow.

The experiments were performed at a DTF wall temperature of 1373 K and inlet air at room temperature. The solid fuels feed rate was around 28 g/h and the total air flow rate, i.e., the summation of primary and secondary air flow, was 10 L/min. That means that the tests were carried out under high excess oxygen conditions.

Partial burned particles (char) were collected at different distances from the injection point along the combustion chamber axis with a water-cooled, nitrogen-quenched stainless steel probe. On the leaving of the probe, the char samples were retained by a quartz filter. The collected solid samples were placed in an oven at approximately 60 °C to dehydrate to be subsequently analyzed. Each condition was repeated at least three times to guarantee statistical representativeness of the results. The burnout of each char sample was calculated according to the ash-as-tracer method (Eq. 1), which is based on the ash content at the injection point (Ash_{coal}) and the ash content at the distance of sample collection (Ash_{char}).

$$Burnout = 1 - \left(\frac{Ash_{coal}}{100-Ash_{coal}}\right)\left(\frac{Ash_{char}}{100-Ash_{char}}\right), \quad (1)$$

2.2. NUMERICAL MODELING

The numerical model used in the present work is based on the model proposed by Ballester and Jiménez (2005) for deriving kinetic parameters for oxidation from experimental measurements. The model was developed in MATLAB and simulates particle's combustion evolution along the reactor axis estimating parameters as burnout, char characteristics and oxygen partial pressure, besides the optimized kinetic parameters for devolatilization and heterogeneous combustion of char. For optimization process, it is applied the Matlab's *ga* function, which finds the minimum of a function using genetic algorithm. Therefore, ranges of each kinetic parameter are defined in the beginning, based on literature values, and the best value in the range is found by the function. Optimization is completed when the difference between experimental and calculated burnout is as small as possible.

The model is based on a set of equations that describe physical and chemical interactions occurring during combustion through the reactor. Evolution of particle's position and velocity at an instant of time is given by the movement equation,

$$\rho_p \frac{\pi}{6} d_p^3 \frac{du_p}{dt} = (\rho_p - \rho_g)\frac{\pi}{6} d_p^3 g - 3\pi\mu_p d_p (u_p - u_g), \quad (2)$$

where ρ is the density, d is the diameter, u is the velocity, t is the time and μ is the gas viscosity and the subscripts p and g are related to the particle and gas, respectively.

The reaction rate for the devolatilized matter, V, is represented by a single reaction and adopts a one-step devolatilization law,

$$dV/dt = -k_V V, \quad (3)$$

where V is the mass of volatile matter per kilogram of coal and k_V is the chemical reaction rate constant for the devolatilization defined as:

$$k_V = A_V \exp(-E_V/RT_p), \quad (4)$$

where A_V is the pre-exponential factor, E_V is the apparent activation energy for devolatilization, R is the universal gas constant and T_p is the particle temperature.

The char oxidation is modeled based on the outer surface area of the particles,

$$dC/dt = -N\pi d_p^2 k_C, \quad (5)$$

where N is the number of coal particles per kilogram of coal and k_C is the chemical reaction rate constant for the oxidation, defined as:

$$k_C = A_C P_{O2,s}^n \exp(-E_C/RT_p), \quad (6)$$

where A_C is the pre-exponential factor, , P_{O2} is the partial pressure of oxygen, E_C is the apparent activation energy for oxidation and n is considered equal 1.

For a particle, the energy balance determines its temperature,

$$\rho_p \frac{\pi}{6} d_p^3 c_p \frac{dT_p}{dt} = Q_{conv} + Q_{rad} - Q_C + Q_V, \quad (7)$$

where c_p is the specific heat of coal and Q_{conv}, Q_{rad}, Q_C and Q_V are the heat flow toward the particle by convection, the heat flow toward the particle by radiation, the heat of heterogeneous combustion and heat of devolatilization, respectively.

Smith (1971) has proposed a mechanism for the evolution of the diameter and density of the particles along the combustion process, which is expressed as

$$d_p = d_p^0 U^\alpha, \quad (8)$$

$$\rho_p = \rho_p^0 U^\beta, \quad (9)$$

where U is the unburnt fraction and $3\alpha + \beta = 1$. The subscripts *0* refers to the initial condition of the coal. In addition, it is necessary to determine the location of the reaction zone in the particles. For reaction occurring in regime I, the diameter remains substantially constant ($\alpha = 0$) and the density decreases with burnout. For reaction occurring in regime III, the density remains approximately constant and the diameter reaches its maximum variation ($\alpha = 0.33$). An intermediate behavior between the limiting cases is possible, regime II, where both the density and the diameter vary (Pereira *et al.*, 2016).

Finally, the oxygen consumed during combustion enables the determination of the partial pressure of oxygen along the DTF as:

$$\frac{M_{O_2} G}{\rho_g R T_g}\left(P_{O2,g,0} - P_{O2,g}\right) = \frac{1}{\nu_V}\left(V_0 - \sum_j w_j V_j\right) + \frac{1}{\nu_C}\left(C_0 - \sum_j w_j C_j\right), \quad (10)$$

where M_{O2} is the molecular weight of oxygen, G is the mass-flow rate of gas injected into the reactor per mass-flow rate of coal, ν_V and ν_C are related to the mass-based stoichiometric ratio of volatiles and char combustion, and w is the mass fraction of particles for each class j.

A more detailed explanation of the mathematical modeling is shown in a previous work published by the authors in which the model used in this work was validated for coals and a biofuel combustion (Pohlmann *et al*., 2020).

To obtain a response from the simulation, some information is required as input data, which include characteristics of the fuel, boundary conditions, experimental data, initial values for kinetic parameters, physical constants and the step of time (discretization parameter). Among the fuel characteristics, the particle size distribution can be highlighted due to its relevance as a different approach used in this model. Some of the boundary conditions are related to DTF, such as the axial length, the inside diameter, and the wall temperature. The experimental data consists of the sampling points and their associated burnout results, and of the temperature profile of the gas along the furnace. Additionally, two auxiliary variables must be defined in the beginning, one for the selection of the variation mechanism of particle's diameter and density and another one for the selection of the method of discretization of the problem (*Euler* method or fourth-order *Runge Kutta*). The code follows the sequential order shown in Figure 2.

After the input data is entered, some initial calculations are realized at the first position of the furnace for determining gas properties (density and velocity) along the reactor, the mass flow ratio between gas and fuel at the DTF inlet, and a correction for temperature profile from an energy balance on the thermocouple. Then, for each millimeter of the axial distance of the DTF, the main calculations (movement equation, kinetic equations, energy balance and variation of particle's diameter and density) are done for all particle's position and class until convergence. The oxygen is consumed as the fuel burns, decreasing the partial pressure of oxygen available in the furnace for combustion. Thus, after convergence for all classes of particles is achieved, the partial pressure of oxygen at each millimeter is recalculated. The simulation ends when the simulated axial distance is entirely covered by the code.

The results of the simulation with this model provide valuable information about the solid fuel combustion, mainly because of the assumption of particle size distribution. Therefore, the temperature profile of particle and the burnout curves can be accessed for each class of particle besides to the total values. In addition, this model provides the profile of the consumption of oxygen along the DTF.

3. RESULTS

Figure 3 shows the measured particle burnout profiles along the axis of the DTF for the two solid fuels studied. For BC, the burnout profile are in a good agreement with experimental data obtained by Moço *et al.* (2017) in tests with a similar Brazilian coal for similar operational conditions using a similar drop tube furnace. In their experiment, authors have found burnouts varying approximately from 75% to 85% along the furnace axis between 300 and 800 mm from the burner outlet.

For the mixture BC90, it was expected to be observed a positive impact on the burnout curve due to the increase in net volatile content of coal-rice husk mixture, as noticed by Akhtar *et al.* (2018) and Pereira *et al.* (2010). Nevertheless, the opposite effect is observed, with much lower burnout values for the mixture in comparison with burnouts of the pure coal sample. On one hand, the size distribution of rice husk particles, with 80% or particles larger than 300 μm, which is much larger when compared to coal particles, impose a difficulty for heating and burning a fraction of the fuel. On the other hand, biomass is only 10% of the mixture and the effect seems to be much larger than expected.

A possible explanation for this result is the difficulty to get a stable feed rate with the twin-screw feeder recently implemented in the drop tube furnace feeding system. This new feeder was adopted in order to approximate the experiment to a one-dimensional reactive flow, however, a solid fuel agglomeration was sometimes observed in the feeder outlet during tests, which might result in discontinuous feeding rates. Moreover, particle agglomeration results in effective sizes larger than the ones reported in Table 1, which may explain reduced burnouts. Such events seems to be more severe with the addition of biomass, probably because of the much larger size of rice husk particles when compared to the coal particles. Many attempts were made to solve this problem, including changes in the solid feeding rate, changes in the air primary flow rate and its distribution within the solid feeding system, use of screens at the screw end, among others. Clear improvements were obtained for pure coal but results for blends are still unreliable. Thus, this is a work in progress and additional/corrected results will be included in the congress presentation and/or in a new version of this manuscript.

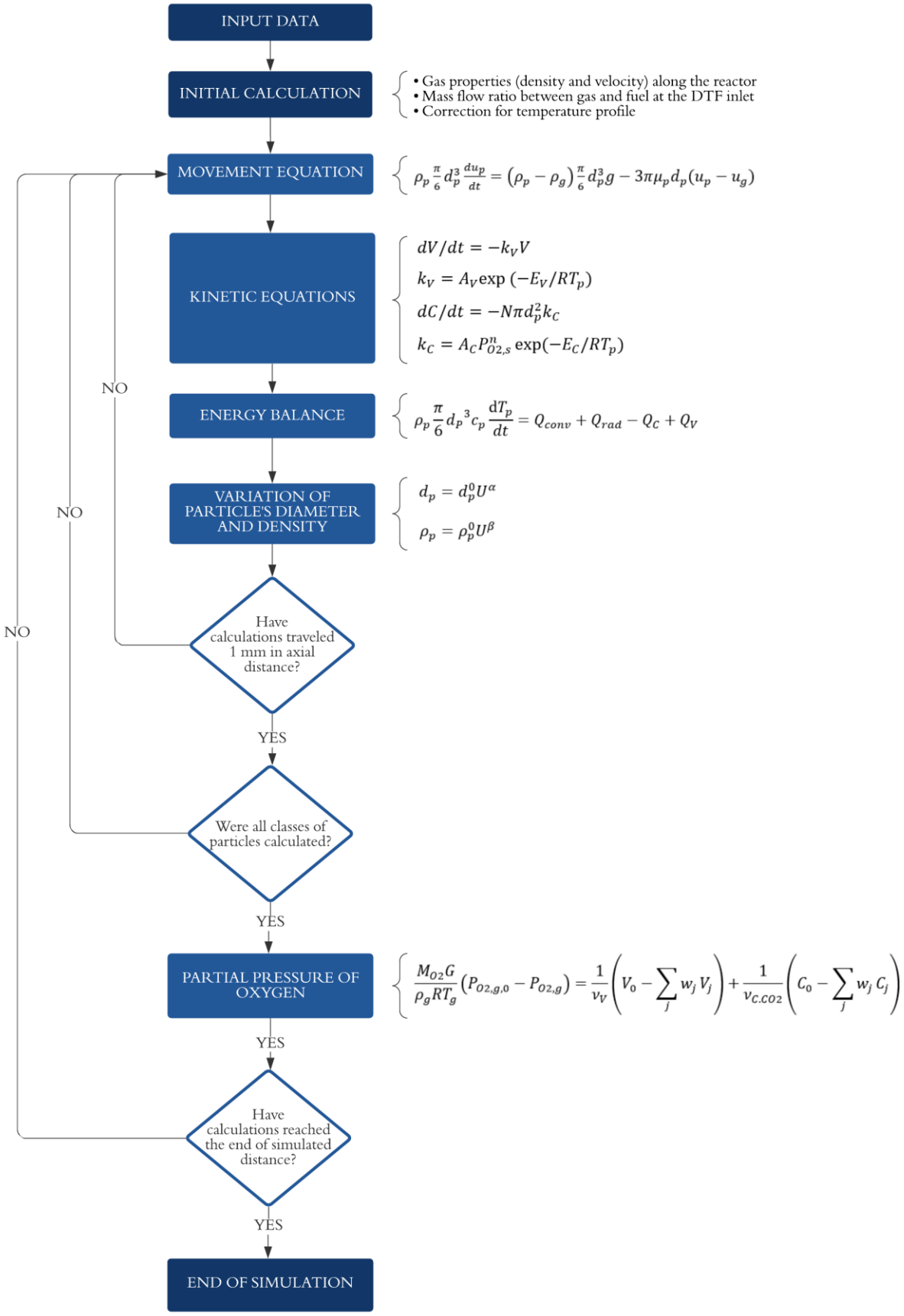

Figure 2. Diagram of sequential order of the numerical solution.

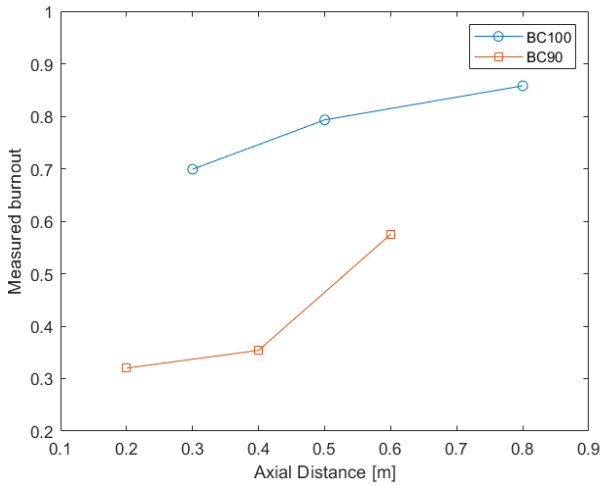

Figure 3. Measured burnout of particles along the DTF for fuels.

Because of the inaccurate experimental results for BC90, optimized kinetic parameters were determined only for BC, using the numerical model previously described in Section 2.2, and they are shown in Table 2. Since there is no information about how diameter varies during combustion, it was assumed in this work reactions occurring entirely on the outer surface, that is, $\alpha = 0.33$. Although there are uncertainties related to experiments, the activation values seem to be fairly close to kinetic parameters reported on the literature for similar coals (Smith, 1982).

Table 2. Optimized kinetic parameters for fuels.

	BC
$\mathbf{A_v}$, s^{-1}	3 x 10^5
$\mathbf{E_v}$, $kJ.mol^{-1}$	143.8
$\mathbf{A_c}$, $kg.m^{-2}.s^{-1}.Pa^{-n}$	4.5 x 10^{-3}
$\mathbf{E_c}$, $kJ.mol^{-1}$	107.0

Figure 4 shows the evolution of burnouts along drop tube furnace axial distance. Four classes were considered in particle size distribution, each one corresponding to an initial mean diameter based on the initial PSD. The predicted profile burnouts were determined using the optimal kinetic parameters referred to above. As expected, finer particles tend to burn faster with associated curves showing a higher slope and ending at short residence times (Ballester and Jiménez, 2005). Larger particles may take all the DTF length to complete combustion (Fig. 4, left). Even with a small amount of experimental points, the comparison of calculated and experimental burnouts indicated good fit of numerical and experimental data, with less than 1.5% of error in the prediction (Fig. 4, right).

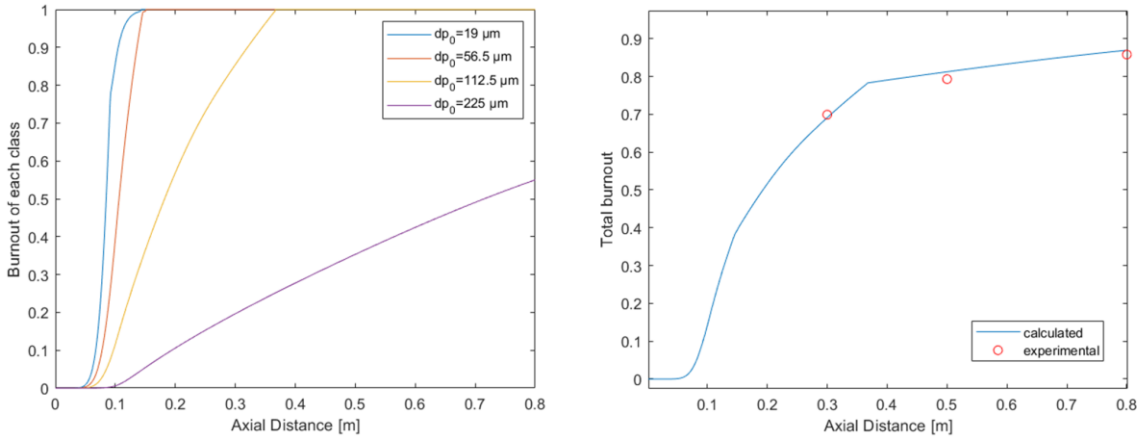

Figure 4. Evolution of burnout along DTF axial distance at different particle size classes at 1373 K (left) and of global burnout compared to experimental values (right) for Brazilian coal.

For comparison purposes, the numerical solution of total burnout for BC90 was obtained assuming the same optimal kinetic parameters found for BC, although this assumption is not correct. Figure 5 shows higher burnouts near to fuel injection point, which is consistent with the increase of volatiles in the mixture (Table 1) and supports the belief that addition of biomass should increase burnout. Larger particles sizes of rice husk do not suggest any negative effects on burnout, reinforcing the problems observed in experimental results of BC90.

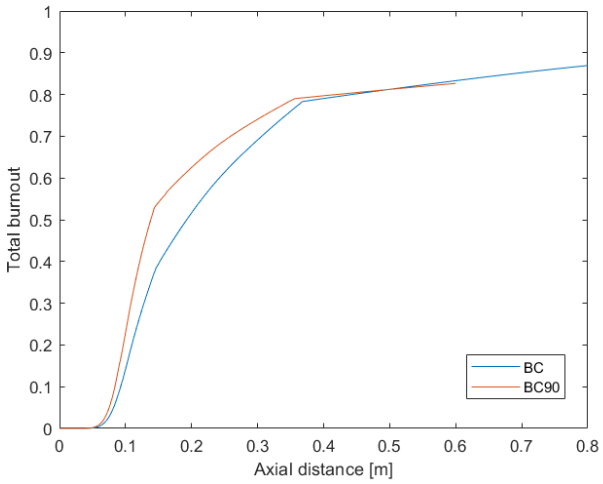

Figure 5. Predicted burnout curves assuming same values of kinetic parameters for BC and BC90.

4. CONCLUSIONS

Brazilian coal and a mixture of this coal and rice husk were burned in a DTF in order to assess their combustion behavior, which was used as input for the numerical model adopted to find optimal kinetic parameters of these solid fuels. However, tests showed a limitation in the feeding system for the mixture, which made not possible to find its kinetic parameters, but additional results will be included in a new version of this manuscript.

Experimental and numerical data presented a good fit for coal burnout along DTF and provided satisfactory kinetic parameters in this work, indicating that this approach has good potential to be employed with typical solid fuels used in industrial processes.

The continuation of this work includes the improvement of feeding system to make experimental results more reliable and the inclusion of different proportions of coal and biomass in the investigation.

5. ACKNOWLEDGMENTS

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