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VALIDATION OF A ONE-DIMENSIONAL NUMERICAL MODEL TO PREDICT SOLID FUEL COMBUSTION IN A DROP TUBE FURNACE

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Abstract. *This work is part of a validation of a one-dimensional numerical model based on the literature and implemented in order to complement experimental studies on coal and biomass combustion in a Drop Tube Furnace recently constructed at the Combustion Laboratory of the Federal University of Rio Grande do Sul. Experimental tests were conducted for pulverized fuels with known particle size distribution and burnout was calculated using the ash-tracer method. The numerical model used as basis of this work simulates the evolution of combustion for size-ranged particle classes and estimates, besides typical combustion parameters, as burnout, partial pressure of oxygen, and chars characteristics, optimized kinetic parameters for devolatilization and heterogeneous combustion of char. Particle size distribution of fuels and of their chars were employed as input data and revealed the best fit related to the combustion regime, which was used for burnout data fitting. A good fit of the experimental and numerical data was observed for burnout at different wall temperature and axial distance along DTF and satisfactory kinetic parameters were found in this work, indicating that this approach could be suitable for application to typical fuels currently used in industrial processes.*

Keywords: *coal combustion, biomass combustion, kinetic parameters, drop tube furnace, numerical modeling*

1. INTRODUCTION

The combustion of solid fuels is still the basis of the world electrical matrix. Despite coal burning is being progressively reduced, the combustion of biomass and waste materials has grown as a sustainable alternative of relatively easy partial or total substitution of coal. Bench scale experiments as well as numerical modeling are valuable tools to contribute in the understanding of the main phenomena that govern particle reactions in industrial systems and to obtain the oxidation parameters of a solid fuel for design and improvement of combustion processes. The use of these tools has even more significance as new fuels emerge.

Drop tube furnaces (DTF) are typical bench scale devices, which suitable reproduce some aspects of industrial solid fuel combustion as the high heating rates and short residence times experienced by fuel particles. Such reactor give valuable information about combustion efficiency of the fuels in addition to gas species composition and deposits formation. The one-dimensional flow and the fact that particles are able to burn without interference from each other in a relatively uniform temperature, allows the different phenomena occurring in DTF (heat transfer between the particle and the medium, mass transport by diffusion in the gas film in addition to the reactions in the gas phase) to be modeled with adequate precision. Thus, it is possible to extract the kinetic parameters (pre-exponential factors and activation

energies in reaction rates in the form of Arrhenius) from the DTF modeling through optimization techniques, having the experimental data as an objective function.

In the combustion of solid fuels, small particles tend to heat rapidly, whereas thicker particles heat more slowly. Thus, it is difficult to predict the reaction rates of a pulverized fuel a priori from its composition, once it depends on the particle size, heating rates and, in the case of char combustion, the final temperature to which it is exposed. Although the approach of Arrhenius plots is a simplified way to access the kinetic parameters from experimental tests, the assumption of a single representative diameter (among other simplifications) reduce the accuracy of the results, thus the particle size distribution has been claimed to be considered since the first studies on gas-solid reactions (Szekely and Propster, 1975).

The determination of the kinetic parameters for the devolatilization and heterogeneous char combustion stages separately have been extensively studied since the 1960s (Field 1969, Smith 1971, Smith et al. 1981). More recently, the growing interest in the study of biofuels focused the determination of kinetic parameters on biomass and derived biomass or residue materials (Costa et al. 2015, Pereira et al. 2016). Costa et al. (2015) determined the kinetic parameters by using first-order models of the Arrhenius equation for the stages of devolatilization and combustion of biomass under different temperature conditions and residence times in a DTF. A global approach was used, in which the determination of kinetic parameters was performed through combustion in one-step (oxidizing atmosphere only). This can be an interesting approach with a smaller number of experiments, with the assumption that volatile combustion do not exert significant influence of the radiative heat transfer on the char particles temperature, as suggested by Tolvanen et al. (2016).

Ballester and Jiménez (2005) developed a numerical model based on the calculation of the particle combustion history to estimate the kinetic parameters of devolatilization and combustion of an anthracite in an EFR (entrained flow) reactor. In contrast to the traditional procedure based on Arrhenius plot, Ballester and Jimenez proposed a more complex model, which considers the particle size distribution of the fuel and subdivides it into particle size classes, investigating combustion for each class.

In this way, the study of combustion by numerical modeling associated with experimental verification has been implemented for a Drop Tube Furnace recently constructed at the Combustion Laboratory of the Federal University of Rio Grande do Sul. This work used the Ballester and Jimenz model as basis to simulate the combustion in this reactor and evaluate the kinetic parameters of coals and a biofuel combustion.

2. EXPERIMENTAL

The fuels tested are a Colombian coal (C), a Brazilian coal (B) and a charcoal prepared through *pinus elliotti* chips carbonization (P). Chemical, physical and petrographic characteristics of fuels are showed in Tab. 1. Both coals are classified as high volatile bituminous according to ASTM D388, with similar vitrinite random reflectance and quite similar volatile matter content in a dry and ash-free basis, but contain substantial difference in ash content, whereas carbonized pine showed a very low ash content, typical of woody biomasses.

Table 1. Fuels characterization.

	Colombian Coal	Brazilian Coal	Carbonized pine
Proximate analysis (%, dry basis)			
Ash	8.7	32.0	1.6
Volatile matter, (% daf)	40.8 (44.7)	27.66 (40.7)	28.7 (29.2)
Fixed carbon	50.5	40.3	69.7
Petrographic analysis (%)			
Vitrinite randon reflectance	0.47	0.48	-
Physical characteristics			
Bulk density ($g.cm^{-3}$)	1.35	1.65	1.50

daf= dry and ash-free basis.

Particle size distribution of fuels was evaluated using the laser diffractometer Cilas 1190 Particle Size Analyzer and is showed in Fig. 1. The fuels were prepared in the particle size typical of pulverized systems, so that about more than 90% passed 75 μm sieve. Since particle size strongly affects the conversion of solid porous, the use of the particle size distribution instead of a single mean particle size gives a better approach in procedures used for deducing kinetic parameters from experimental measurements. Therefore, samples were size-graded in classes and used as a differential input in this model.

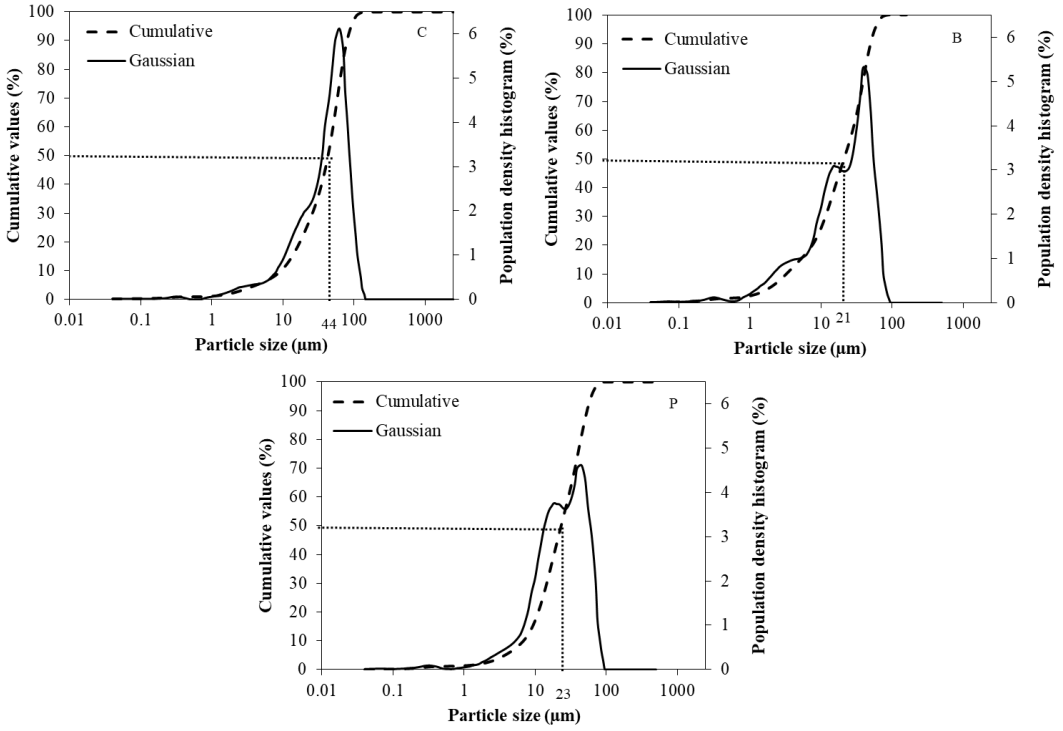

Figure 1. Particle size distribution of fuels.

The combustion tests were performed in a drop tube furnace (DTF, Fig. 2) with a 1.6 m long ceramic tube surrounded by three concentric tubular furnaces, each one with three thermocouples, for control and monitoring furnace temperature. In each test 1 g.min^{-1} of sample was introduced through a water cooled injector placed at the top of the furnace with an air flow rate of 22 L·min^{-1}. That means that the tests were carried out under high excess oxygen conditions.

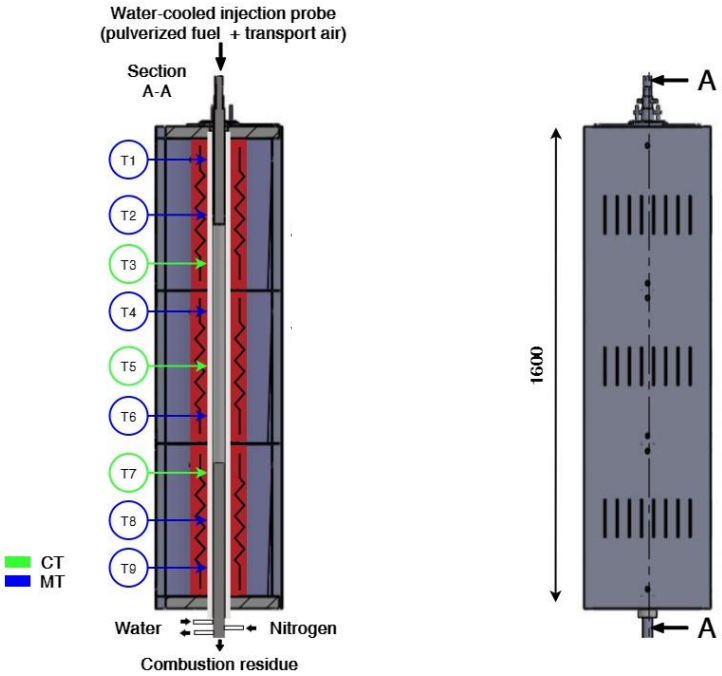

Figure 2. Drop Tube Furnace used in combustion tests. T3, T5 and T7 = control thermocouples; T1, T2, T4, T6, T8 and T9 = monitoring furnace temperature thermocouples.

The fuels experienced different wall temperatures conditions in the DTF and partial burned particles (chars) were collected at varied distances from the injector outlet point along the combustion chamber axis (Table 2) using a water-cooled, nitrogen-quenched stainless steel probe and were retained by a quartz filter to be subsequently analyzed. The axial average temperature along the furnace was measured using a K-type thermocouple and corrected for radiative and convective heat transfer effects. Each condition (wall temperature, axial distance) was repeated at least three times to guarantee the statistical representativeness of the results. Combustion efficiency (*Burnout*) was calculated using the ash-tracer method as in Eq. (1), which is based on the ash mass fraction entering the reactor (Ash_{coal}) and the ash mass fraction at the sampling point ($Ash_{char\text{-}comb}$).

Table 2. Experimental conditions of DTF tests.

Experimental parameters	Colombian coal	Brazilian coal and Carbonized pine
Wall temperature	1173, 1273 and 1373 K	1273 K
Axial distances	0.30, 0.40, 0.50, 0.60 and 0.70 m	0.37, 0.55, 0.73 and 0.91 m

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

3. COMBUSTION MODEL

The model proposed by Ballester and Jimenez (2005) to estimate the kinetic parameters is the basis of this work. The modeling was developed in MATLAB. The input data of the model comprehend the physical constants required for the set of equations, the fuel properties, among which the particle size distribution is a differential approach of this model, boundary conditions (DTF characteristics and feeding rates of fuels and air), experimental data (fuel burnouts and gas temperature measured along the furnace), the initial estimation of kinetic parameters (literature-based data) and the time step (discretization parameter). Additionally, an auxiliary variable selects as the particle's diameter and density vary as it burns according two models (Smith, 1982) and another auxiliary variable selects either the *Euler* method or the fourth-order *Runge Kutta* for the discretization of the problem. The kinetic parameters as a response of the simulation are obtained with an optimization process (Matlab's *fminsearch* function).

The model is basically based on: the equation of motion, to determine the velocity and position of the particle in an instant of time; the kinetic equations, to model the burning of fuel along the axial distance; the energy balance in the particle, to determine its temperature; the mechanism that describes the variation of the particle's diameter and density and finally, the determination of the partial pressure of oxygen along the DTF. The sequential order of each step in the code occurs as shown below.

Considering that gravity, buoyant and viscous forces act on the particle surface, the velocity and position of the particles are evaluated for each class i in an instant of time from the momentum equation (Eq. 2). In Eq. 2 ρ is the density, d is the diameter, u is the velocity, t is the time, μ is the gas viscosity and the subscripts p and g are related to the particle and gas, respectively. The positions of the particles can be associated to their residence times in the furnace.

$$\rho_{p,i}\,\frac{1}{6}\,\pi\,d_{p,i}^{3}\,\frac{du_{p,i}}{dt} = \left(\rho_{p,i} - \rho_g\right)\frac{1}{6}\pi\,d_{p,i}^{3}\,g - 3\,\pi\,\mu_g\,d_{p,i}\,(u_{p,i} - u_g) \quad (2)$$

The combustion along the DTF is modeled according to the devolatilization law (Eq. 3 and 4) followed by the heterogeneous oxidation of the fixed carbon (or char) (Eq. 5 and 6).

$$\frac{dV}{dt} = -k_V V \quad (3)$$

$$k_V = A_V \exp\left(-\frac{E_V}{RT_p}\right) \quad (4)$$

$$\dot{m}_c = \frac{dC}{dt} = -N\pi {d_p}^2 k_C \quad (5)$$

$$k_C = A_C {P^n}_{O2,s} \quad \exp\left(-\frac{E_C}{RT_p}\right) \quad (6)$$

where V and C are the mass of volatile and char per kilogram of coal, k is the reaction rate, A is the pre exponential factor, E is the activation energy, R is the universal gas constant, T is the temperature, N is the number of coal particles per kilogram of coal (1/initial mass) and the subscripts v and c are related to the devolatilization and char combustion steps, respectively. n is assumed to be equal 1 and the partial pressure of oxygen at the particle surface (P_{O2s}) can be

obtained from the Fick's Law rewritten as Eq. 7, where D_{O2} is the mass diffusivity of oxygen, M_{O2} is the molecular weight of oxygen and v_c is the mass-based stoichiometric ratio of volatile combustion.

$$2\pi\, N\, d_p\, D_{O2}\, \frac{M_{O2}}{RT_p}\left(P_{O2,g} - P_{O2,s}\right) = \frac{1}{v_C}\pi\, N\, {d_p}^2\, A_C\, P^n_{O2,s}\, exp\left(\frac{-E_C}{R \cdot T_p}\right) \tag{7}$$

It is necessary to solve an energy balance (Eq. 8) on the particle to obtain its temperature. In Eq. 8, C_p is the specific heat of coal and Q_c, Q_v, Q_{conv} and Q_{rad} are the heat of heterogeneous combustion, heat of devolatilization, heat flow toward the particle by convection and heat flow toward the particle by radiation, respectively and are evaluated by Eq. 9-12

$$\rho_p\, \frac{1}{6}\, \pi\, {d_p}^3 c_p \frac{dT_p}{dt} = -Q_C + Q_V + Q_{conv} + Q_{rad} \tag{8}$$

$$Q_V = \frac{1}{N}\frac{dV}{dt} H_V \tag{9}$$

$$Q_C = \frac{1}{N}\frac{dC}{dt} H_C \tag{10}$$

$$Q_{conv} = \pi\, d_p\, Nu\, k_g\, \left(T_g - T_p\right) \tag{11}$$

$$Q_{rad} = \pi\, {d_p}^2\, \varepsilon\, \sigma\, \left(T_w^4 - T_p^4\right) \tag{12}$$

where *Hv* and *Hc* represent heat of devolatilization and heat of heterogeneous combustion, respectively. *Nusselt* number (*Nu*) is taken equal to 2 and the particles are assumed to be gray bodies with constant emissivity.

Particle´s diameter and density variation (Eq. 13 and 14) depends on the unburnt fraction (Eq. 15) and on the parameters α and β, where 3α+β=1 and the superscripts *0* refers to the initial conditions of coal.

$$d_p = {d^0}_p U^{\alpha} \tag{13}$$

$$\rho_p = {\rho^0}_p U^{\beta} \tag{14}$$

$$U = \frac{C + V + A}{C^0 + V^0 + A^0} \tag{15}$$

Oxygen is consumed by combustion of volatiles and char along the furnace and its content available on the gas flow for each class *i* may be obtained from Eq. 16. *G* is the mass-flow rate of gas injected into the reactor per mass-flow rate of coal, v_v and v_c are related to the mass-based stoichiometric ratio of volatiles and char combustion, and ω is the mass fraction of particles for each class.

$$\frac{M_{O2}\, G}{\rho_g RT_g}\left(P^0_{O2,g} - P_{O2,g}\right) = \frac{1}{v_v}\left(V^0 - \sum_i w_i V_i\right) + \frac{1}{v_c}\left(C^0 - \sum_i w_i C_i\right) \tag{16}$$

4. RESULTS

Particle size distribution was evaluated for each char collected in the different condition in DTF (wall temperature, axial distance) for the Colombian coal. The mean diameter was plotted against burnout in Fig. 3, and compared to the plots of Eq. (13) calculated for alpha varying from zero to 0.33, as also did Ballester and Jimenez (2005) to find an optimal alpha value which fits with experimental data. The mean diameters of 1173 K chars fitted fairly well closer to the alpha = 0.16 curve, whereas as temperature increased the mean diameter fitted closer to the alpha = 0.33 curve. Similar trend was observed when *D[4,3]* of chars was plotted against burnout. Since in a general way all experimental data were, in a sense, homogeneously dispersed between alpha = 0.16 and 0.33, an intermediate alpha value of 0.24 was adopted for simulations. The decay of the mean particle diameter with burnout between a range of intermediate alpha values indicates that there is a probable density variation at the same time as the diameter varies, typical of regime II of

combustion (Smith 1971). However, the bulk density of chars was not was not evaluated in the scope of this work. The alpha value estimated for the Colombian coal was also used for simulation with the other samples of this work.

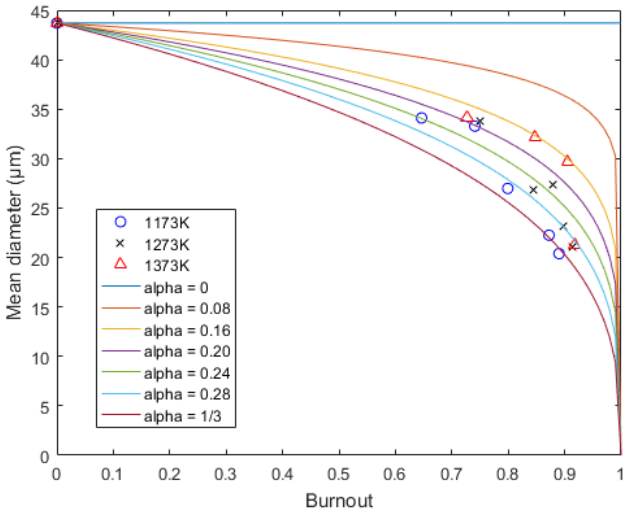

Figure 3. Mean diameter of Colombian coal chars in function of Burnout and curves according Eq. (13) for alpha varying from zero to 0.33.

Figure 4 shows the evolution of burnouts along the axial distance for the fuels. As expected, the burnouts calculated for each particle size class along the furnace shows that conversion is faster as finer the particles, and larger particles may take the entire length of the furnace to complete combustion, depending on the temperature of the test (Fig. 4, left). The comparison of modeled and experimental burnouts for the three temperatures along the axial distance of the furnace for the Colombian coal (Fig. 4, right) indicated good fit of numerical and experimental data, for all conditions of wall temperature and axial distance, with standard errors less than 1.3 %. Even with a smaller amount of experimental points, the Brazilian coal and the carbonized pine showed also a good fit with numerical simulation, with errors less than 1.3 % for the coal and 0.6 % for the biofuel.

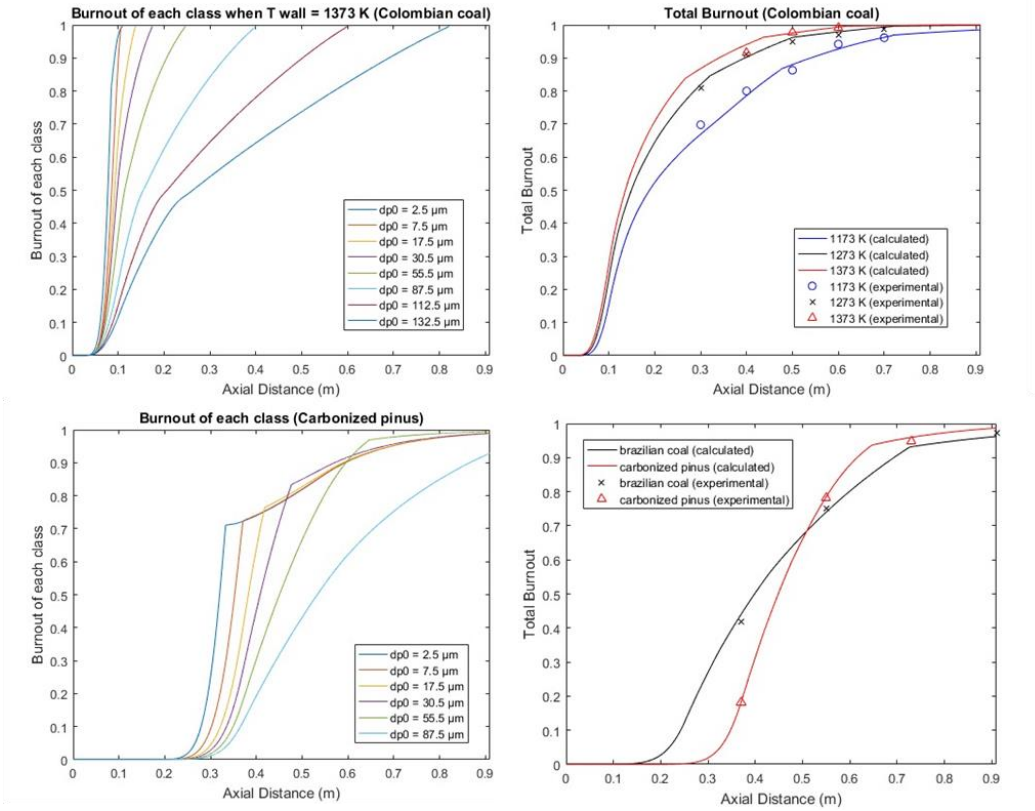

Figure 3. Evolution of burnout along DTF axial distance at different particle size classes at 1173 K (right) and of global burnouts compared to experimental values (left).

The optimized kinetic parameters obtained in simulated conditions are presented in Table 3. The activation values seems to be according to the main properties of the fuels. Despite both coals are of the same rank, Colombian coal is probably to devolatilize and burn easier than Brazilian coal due to its higher volatile matter content, therefore presenting lower values for activation energy, especially during devolatilization stage. In the same way, the carbonized pine could be expected to be more reactive than coals and present a lower activation energy, especially during char oxidation stage, due to its typical less aromatic and highly porous carbon structure (Hillig et al. 2020). The model predictions showed a quantitative agreement with kinetic parameters reported on the literature for coals of similar rank (Smith 1982), but no data (obtained under similar conditions of heating rate) was found for a carbonized biomass. This indicates the potential of using this technique to study alternative fuels.

Table 3. Optimized kinetic parameters of fuels.

	Colombian coal	Brazilian coal	Carbonized pine
A_v (s^{-1})	5.85×10^5	1.07×10^5	1.03×10^5
E_v $(kJ.mol^{-1})$	82.1	117.0	110.1
A_c $(kg.m^{-2}.s^{-1}.Pa^{-n})$	3.88×10^{-3}	1.44×10^{-3}	1.50×10^{-3}
E_c $(kJ.mol^{-1})$	73.0	83.9	67.8

The burnout profiles are a result of both devolatilization and char combustion reaction, once fuels had not pass through previous pyrolysis. The model considers fuel devolatilization and char oxidation as independent and concomitant reactions, so one pair of kinetic parameters describe each one of these steps. The good fit of the experimental and numerical data and the satisfactory kinetic parameters found in this work indicate that this approach could be suitable for application to typical fuels currently used for industrial processes. The continuation of this work involves a better understanding about the behavior of physical properties of chars (like particles diameter and density) with combustion in order to improve the reproduction of experimental data with this one-dimensional model.

5. CONCLUSIONS

An one-dimensional numerical model based on the literature was implemented to experimental data in order to evaluate the kinetic parameters that describe the pyrolysis and heterogeneous char oxidation rates of solid fuels. Particle size distribution of fuels and of their chars were employed as input data and revealed the best fit related to the combustion regime, which was used for burnout data fitting.

A good fit of the experimental and numerical data was observed for burnout at different wall temperature and axial distance along DTF and satisfactory kinetic parameters were found in this work, indicating that this approach could be suitable for application to typical fuels currently used in industrial processes.

6. ACKNOWLEDGEMENTS

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