# SENSITIVE ANALYSIS OF A COAL COMBUSTION MODEL ON A DROP TUBE FURNACE

Leonardo Zimmer, leo.zimmer@mecanica.ufrgs.br
Fernando Marcelo Pereira, fernando@mecancia.ufrgs.br
Paulo Smith Schneider, pss@mecanica.ufrgs.br
Universidade Federal do Rio Grande do Sul – Departmento de Engenharia Mecânica
Rua Sarmento Leite 425, Porto Alegre, Rio Grande do Sul, Brazil, 90050-170

Abstract. In the present work one-dimensional model coal combustion on a Drop Tube Furnace (DTF) is developed. The modeling is developed from literature, and the discrete equations that characterize the flow, heat transfer and coal combustion reactions are programmed in FORTRAN 90 language. The results are compared with a reference model and experimental data, and showed good agreement. A sensitivity study is performed to understand the behavior of coal combustion due to changes of some working parameters of the DTF. The sensitivity analysis showed the versatility of the model and thereby the experiment. From the variation of the oxygen concentration, operating temperature and input flow rates a range of results can be obtained. Using the model presented here together with experimental results leads to a detailed study of the kinetic parameters of coal combustion.

Keywords: Drop Tube Furnace, coal combustion, kinetic parameters, numerical model

## 1. INTRODUCTION

Coal remains the most abundant fossil fuel on earth and accounts for most of the production of electricity of several countries, according to CGEE (2012), and will continue to play an important role in world energy supply for many decades. According to BP (2012) coal consumption grew 5.4% in 2011 and was the only fossil fuel to register above average growth. It also was the form of energy that has been rapidly growing, despite renewable energy. In addition, the coal in 2011 was responsible for 30.3% of global energy consumption, the highest share since 1969.

Considering the global energy participation, the petroleum had greater participation in global primary energy supply, with 33.2%, by the year 2008, according to EPE, 2011, followed by coal with 27%. For the global supply of electricity, coal had the greatest participation, with 41%, followed natural gas, with 21.3%, for the same period.

However, in Brazil the primary energy supply, by the year of 2010, was distributed among the non-renewable sources, with 53%, which coals is included with 0.9%, and renewable sources, with approximately 47%, according to EPE (2011). In terms of electricity supply, the hydropower generation accounted for 74%, while coal accounted for only 1%, for the same period. Nonetheless coal use in Brazil for power generation grew 28.3% in 2011, in respect to the previous year. National coal is produced in the states of Parana, Santa Catarina and Rio Grande do Sul, and it is essentially, 80%, used to power generation, CGEE, 2012.

To understand more about the combustion of national coal, its efficiency and how to optimize it, is necessarily an extensive study of coal combustion, in addition to bench tests and facility tests. One useful experiment is the Drop Tube Furnace (DTF), which is able to create an environment that simulates conditions similar to those encounters in industrial burner, such as short residence time, high temperatures and high heating rates, Ulloa *et al.* (2005). Therefore, DTF may be used to produce pyrolysed char or for analysis of the coal reactivity. From the knowledge of the phenomena, which govern the combustion within a DTF, is possible, through numerical modeling and experimental data, to obtain the chemical kinetics parameters, Ballester and Jimenez (2005).

The main objective of this work is the study of burning coal in a DTF. From the literature, one-dimensional numerical model is built for a DTF focusing on chemical reaction processes of burning coal. The developed model is compared to a model adopted as the reference and to experimental data. Also, a sensitivity study of the model is done to understand the behavior of the system due to changes of some working parameters.

# 2. DROP TUBE FURNACE

The problem to be addressed in this work is the combustion of coal along a reactor DTF. The formulation of the problem is detailed for the specific case simulated, followed by models of heat transfer of the burning coal. The modeling used in this work aims to reproduce the experiments conducted in a DTF described by Ballester Jimenez (2005). The simulation developed uses the geometry and experimental data of the reactor for a specific sample of coal. However, the model can be adapted for other types of coals. The reactor used in the present work is shown in Fig. 1.

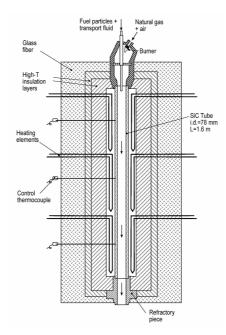


Figure 1. DTF scheme used in present work, source Ballester and Jimenez (2005)

As shown in the Fig. 1, this DTF is heated by heating elements (electrical resistances) and by flue gas from natural gas burner. The furnace reactor is a SiC tube, with 78mm of internal diameter and 1.6 m long, and it can be operated up to 1773 K. The pulverised fuel is injected through the top of the tube with the transport fluid (air) together with the co-flowing gases. The co-flowing gases consist of combustion products of natural gas. The oxygen concentration at the entrance of the furnace can be controlled either by adjusting the flow rate of combustion air and transport fluid or by injecting pure oxygen. The operational conditions used in the simulation of the present works are presented in Table 1.

Table 1 – Working condition of the DTF, adapted from Ballester and Jimenez (2005)

Working temperature of the reactor (K)	1313	1448	1573	1723	1573
CH <sub>4</sub> flow (l/h)	70	70	70	70	70
Air flow for burn CH4 (l/h)	800	800	800	800	770
Transport fluid (air) flow (l/h)	250	250	250	250	250
O2 flow added (l/h)	0	0	0	0	43

The present work uses the same coal as the reference work, anthracite coal, characterized by proximate analysis (by weight) as: 69.09% of Fixed Carbon,  $C^0$ ; 10.28% of Volatiles,  $V^0$ ; 19.17% of Ash,  $A^0$  and 1.46% of Moisture,  $H^0$ ; by ultimate analysis (by weight) as: 70.3% of C; 3.03% of H; 1.63% of N; 2.28% of S; and its HHV is 27.59 (MJ kg-1). The particle diameters for all simulations were 52.2  $\mu$ m and were feeding at the rate of 30 g/h.

Ballester e Jimenez, 2005, found different kinetic parameters for different modes of shrinking core of the particle,  $\alpha$ . The shrinking core model is presented in the next section. For  $\alpha$ =0 Ac is  $4.00 \times 10^{-4}$  (kg/m².s.Pa) and  $E_c$  is  $8.30 \times 10^{4}$  (J/mol); for  $\alpha$ =0.08 Ac is  $5.60 \times 10^{-4}$  (kg/m².s.Pa) and  $E_c$  is  $8.60 \times 10^{4}$  (J/mol); and for  $\alpha$ =0.33 Ac is  $1.49 \times 10^{-3}$  (kg/m².s.Pa) and  $E_c$  is  $9.42 \times 10^{4}$  (J/mol). The value of  $\alpha$ =0.08 is the best fit found by the reference to represent the reduction of particle size and density for a multiple size coal population. These kinetic parameters of coal burning are used in the present work.

The combustion monitoring is based on the unburned fraction, U, and it is calculated from the proximate analyses of fuel and char samples, according to the ash-as-tracer method from Ballester and Jimenez (2005). The relation to the model parameters is presented in the next section.

# 3. COMBUSTION MODEL

Thereby, for the present work it is assumed these simplification hypotheses: One-dimensional uniform flow and Steady-state regime; Are neglected: thermal and mass diffusion in gas phase, gravity effect, particle interaction, viscous dissipation, and gas phase radiation; Coal particles are composed by fixed carbon, volatile, moisture and ash; Ash is an inert component and remains in the particle; The only product of devolatilisation is methane,  $CH_4$ , which is instantaneously burnt; Coal particles have sphere format; Char reaction is a one-step reaction CO formation only, and is represented as  $C + \frac{1}{2}O_2 \rightarrow CO$ , which is instantaneously burnt; Initial particle diameter is just one; Atmospheric pressure in all domain and is defined as dry-air; All fluids are modelled as perfect gases; Temperature particle is

assumed be uniform within the particle; Particles are dragged by the flow and take its velocity; The drying process of the coal particle occurs immediate, due to the small quantity of moisture in the coal and the high heating rate of the reactor ( $\sim 10^4$ K/s); The particle is global modelled, without internal gradient of temperatures and concentration.

#### 3.1. Governing Equations

According to Smoot and Smith (1985), Williams et al. (2000) and Glassman (2008), the devolatilisation process can be represented by a first order single step reaction, following an Arrhenius rate. The rate of mass loss of devolatilisable matter dV/dt can be described as:

$$\frac{dV}{dt} = -k_{\nu}V\tag{1}$$

$$k_{\nu} = A_{\nu} \exp\left(E_{\nu}/RT_{\mu}\right) \tag{2}$$

where  $k_{\nu}$  is the devolatilisation rate (s<sup>-1</sup>),  $A_{\nu}$  is the pre-exponential factor (s<sup>-1</sup>),  $E_{\nu}$  is the activation energy for devolatilisation (J mol<sup>-1</sup>),  $T_p$  is the temperature of the particle (K), and V is the volatiles content (kg of devolatilisable matter/kg of particles). Based on the work of Ballester e Jimenez (2005) the pre-exponential factor used is  $6.0 \times 10^5$  (s<sup>-1</sup>) and the activation energy for devolatilisation is  $144 \times 10^3$  (J/mol).

The carbon reaction is modeled by an apparent kinetics, applied to the outer surface of the particle, which can be found in Turns (2000) and Coelho e Costa (2007). This model was then adapted to the main kinetic parameters used by Ballester and Jimenez (2005), and the rate of consumption of carbon can be showed as:

$$\frac{dC}{dx} = -\left(\frac{y_{O_2}}{R_{cin} + R_{dif}}\right) \frac{N_p}{u_p} \tag{3}$$

$$R_{cin} = \frac{M_{O_2}}{4\pi r_s^2 k_C M_{mixt} p} \tag{4}$$

$$k_C = A_C \exp\left(\frac{-E_C}{RuT_p}\right) \tag{5}$$

$$R_{dif} = \frac{S_{O_2,CO} + y_{O_2,s}}{4\pi r_s \rho D_{O_s}^M} \tag{6}$$

$$N_{p} = \frac{1}{\frac{1}{6}\pi \left(d_{p}^{0}\right)^{3}\rho_{p}^{0}} \tag{7}$$

where, C is the char content (kg of char/kg of particles),  $y_{Oz}$  the mass fraction of  $O_2$  in the ambient,  $N_p$  the number of coal particles/kg of coal (kg<sup>-1</sup>),  $u_p$  the velocity of the particle (m/s),  $R_{cin}$  and  $R_{dif}$  the kinetic and diffusion resistances (s/kg), respectively,  $M_{O2}$  the molar mass of  $O_2$  (kg/kmol),  $r_s$  the particle radius (m),  $k_C$  the constant rate of char oxidation (kg/m<sup>2</sup>s),  $M_{mixt}$  is the molar mass of the mixture (kg/kmol), p is the pressure (Pa),  $A_C$  is the pre-exponential factor of char oxidation (kg/m<sup>2</sup>sPa),  $E_C$  is the activation energy (J/mol), Ru is the universal gas constant (J/molK), Tp is the particle temperature (K),  $S_{O_2,CO}$  the ratio of  $O_2$  mass per C mass to form CO,  $y_{Oz,s}$  the mass fraction of  $O_2$  on the surface of the coal particle,  $\rho$  the gas density (kg/m<sup>3</sup>),  $D_{O2}^M$  the diffusivity coefficient of  $O_2$  into the mixture (m<sup>2</sup>/s),  $d_p^0$  and  $d_p^0$  the initial particle diameter (m) and density (kg/m<sup>3</sup>), respectively.

Analyzing the change in mass fraction of carbon Eq. (3), together with the variation of mass fraction of volatile, Eq. (1), and knowing the mass of a particle is composed of mass fractions of carbon, volatile, ash and moisture, it can be obtain an expression for the mass flow rate of particles,  $m_p$ , such as:

$$m_p = \frac{(m_A + m_H)}{1 - (C + V)} \tag{8}$$

where,  $m_p$ ,  $m_A$  and  $m_H$  are mass of particles, ashes, and moisture, respectively, in kg.

From Eq. (8) and definitions of mass fractions of carbon and volatile is possible to obtain the reaction rates in weight basis (kg/s) according to Eq. (9) and (10):

$$\dot{w}_C = \frac{dm_C}{dt} \tag{9}$$

$$\dot{w}_{V} = \frac{dm_{V}}{dt} \tag{10}$$

Auxiliary equations:

The unburnt fraction, U, is a dimensionless measure of the quantity of material which can react, and is calculated as:

$$U = \frac{C + V}{C^0 + V^0} \tag{11}$$

where C and V are mass fractions of fixed carbon and volatile matter, respectively. The superscript  $\theta$  means that the variable's value is taken at the point of injection.

The model of reduced particle size is based on studies of Smith, 1971a and 1971b, which proposed a relation to the evolution of the diameter and density of the carbon particle over time in relation to the burnt mass fraction of the particle. The relation can be written as function of unburnt fraction, U, as:

$$(d_p)^3 \rho_p = (d_p^0)^3 \rho_p^0(U) \tag{12}$$

Introducing the relation  $3\alpha + \beta = I$ , the diameter and the particle density at any point can be found as:

$$d_p = d_p^0(U)^\alpha \tag{13}$$

$$\rho_n = \rho_n^0(U)^\beta \tag{14}$$

Depending on the particle shrinking mode, it is possible to use the parameters  $\alpha$  and  $\beta$  for classifying the reduction model in a model of the particle constant diameter ( $\alpha = 0$ ), of constant density ( $\alpha = 1/3$ ) or intermediate ( $0 < \alpha < 1/3$ ).

Gas phase equations:

The reactions taking place in homogeneous gaseous phase occur instantaneously as CO and CH<sub>4</sub> are produced and are represented by Eq. (15) and (16):

$$CO + \frac{1}{2}O_2 \to CO_2 \tag{15}$$

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \tag{16}$$

Conservation of mass species:

$$\frac{d\left(\rho u y_{j}\right)}{d x} = \sum_{i,k} \dot{W}_{j,k} \tag{17}$$

where u is the flow velocity (m/s),  $y_j$  mass fraction of the specie j,  $\dot{w}_{j,k}$  the source term (kg/sm<sup>3</sup>) which subscripts j and k represent the specie and the reaction, respectively.

Energy Equation:

The energy equation for a particle can be written as:

$$\rho_{p} \frac{1}{6} \pi d_{p}^{3} c_{p} \frac{dT_{p}}{dx} = \frac{1}{u} (\dot{Q}_{V} + \dot{Q}_{C} + \dot{Q}_{conv} + \dot{Q}_{rad})$$
(18)

where:  $c_p$  is the specific heat of the particle at constant pressure (kJ/kgK),  $\dot{Q}_v$ ,  $\dot{Q}_c$ ,  $\dot{Q}_{conv}$ ,  $\dot{Q}_{rad}$ , represent devolatilisation heat, the heat of combustion of fixed carbon, the heat exchange by convection and radiate heat exchange, respectively, in W. The density of the particles of coal is obtained from literature data, the value used in this study was 1500 kg/m³, obtained from Basu (2006). The specific heat at constant pressure of the particle adopted is 1000 kJ / kg K, and was obtained from Tomeczek and Palugniok (1996).

The heat transferred between the gas and particle through convection can be described as:

$$\dot{Q}_{conv} = \pi d_p N u \lambda_g \left( T_g - T_p \right) \tag{19}$$

where Nu is Nusselt number based on particle diameter,  $T_g$  is gas temperature (K), and  $\lambda_g$  is the thermal conductivity of the gas (W/m K). As stated earlier, the relative velocity of the particle in relation to the gas flow velocity is zero, so Re is zero and Nu is considered equal to 2.0 as Basu (2006).

The heat exchange by radiation only occurs between the particles and the walls of the reactor. Whereas the size of each particle is much less than the diameter and tube length, it can be said that the form factor of each particle in

relation to the reactor is equal to one. Thus, the heat transfer between the wall of the reactor and the particle through radiation can be described as:

$$\dot{Q}_{rad} = \pi d_p^2 \varepsilon \sigma \left( T_W^4 - T_P^4 \right) \tag{20}$$

where  $\varepsilon$  is the emissivity of the particle,  $\sigma$  is the Stefan–Boltzmann constant, and  $T_w$  is the wall temperature (K). The heat release in the combustion and the heat to the endothermic process of devolatization can be written as:

$$\dot{Q}_C = \frac{1}{N_P} \frac{dC}{dt} H_C \tag{21}$$

$$\dot{Q}_V = \frac{1}{N_p} \frac{dV}{dt} H_V \tag{22}$$

where  $H_C$  is the combustion enthalpy (J/kg) and  $H_V$  is the devolatilisation enthalpy (J/kg).

All properties are calculated from the literature. The thermal conductivity of the gas according to Poling *et al.* (2000), the oxygen diffusivity into the gases and viscosity are calculated through Chapman-Enskog equation.

## 4. NUMERICAL METHOD

It is applied the finite volume numerical method for solving partial differential equations. The problem is parabolic, is treated as convective-dominant, and the interpolation scheme for convective terms is upwind scheme. The solution of the problem takes place in a marching procedure, where the next volume is only achieved when all variables converged. The convergence criteria used was 10<sup>-6</sup> for all variables, and to achieve this criteria the mesh was refined till 70 thousand volumes. The program was written in Fortran 90 programming language using the Gfortran compiler (gcc-4.6.0) with all values expressed in double precision. The duration of each simulation was approximately 70 seconds in a MacBook Pro with Core 2 Duo 2.4 GHz processor and 8 GB of RAM.

#### 5. RESULTS

The experiment from Ballester and Jimenez (2005) is chosen to compare the results, which from now on is called the reference experiment, for simplicity. Table 1 shows the operation data of the experiment of reference, and from these data, it is calculated the mass flow rates of the species of the gases of the process necessary to feed the numerical model.

Comparison with the reference work:

This section will compare the results of this model with those presented by the reference work for the same operating conditions of the reactor DTF. The main differences are: The reference model assumes a relative velocity of the particle with respect to gas flow, while in the present work the particle velocity is the same flow; The present model has a more detailed model of the diffusion of oxygen; The thermo-physical properties in the current model are calculated for each point in the reactor, while in the reference model are considered constant; For some situations, the reference work employs a sample with variable particle size, while in this study it is considered as fixed size of  $52.2 \,\mu m$  diameter; Additionally, the reference work obtained the kinetic parameters of the coal reaction by setting their model with experimental data. Therefore, these parameters can be very sensitive to the adopted model and its assumptions.

The first comparisons between models were at temperatures of 1573 K and 1723 K. Also it was considered three values of  $\alpha$  for the model to reduce the particle size, reported in Table 3. The model showed the same trends in unburnt fraction U for the three models of reduction of particle size. However, the values found had a certain deviation from the reference model, as the reference model had a higher conversion of char than the present model for all values of  $\alpha$ . The differences were due to the models adopted, the simplifying assumptions and also to the fact that the kinetic parameters used in this work were obtained particularly for the reference model. The best fit to experimental data was using  $\alpha$  equal to 1/3, meaning that the constant density-shrinking model reduced the differences between the present model and the experimental data. From this statement, the second analysis is made comparing the present model, taking account  $\alpha$  equal to 1/3, over different working temperatures, as shown in the Fig. 2.

From Fig. 2 it can be noticed that while the present model uses a different shrinking model ( $\alpha$ =0,33) and mono size particles the results are similar of those encountered by the work reference ( $\alpha$ =0.08) and experimental data, which considered multiple size particles. It is also interesting to note that the results of this work and from the reference model for the case of 1573 K with the addition of  $O_2$  are similar. One reason for this similarity is that the differences between the models are eventually suppressed by increasing the reaction rate, caused by the higher concentration of  $O_2$  at the entrance of DTF.

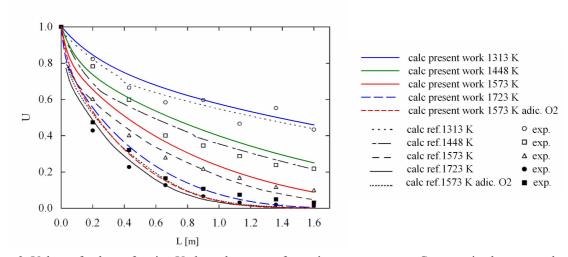


Figure 2. Values of unburnt fraction U along the reactor for various temperatures; Comparative between values of Ballester e Jimenez, 2005, model in black and white,  $\alpha$ =0.08 and with multiple size particle; and present model, in colors,  $\alpha$ =0.33 and  $d_p$  = 52.2  $\mu$ m.

#### Sensitivity analysis

Sensitivity analysis seeks to determine the effect on the system when you change one of its parameters, while keeping the others constant. Here it is presented the main parameters which had most effect on the system. The nominal values are based on the values of the experiment running at 1723 K.

#### $O_2$ concentration:

The nominal value of 8% of the  $O_2$  molar fraction on a dry basis is taken as reference, and is obtained with the  $CH_4$  flow rate of 70.69 l/h and with air flow rate of 799.61 l/h. As a result, the velocity of the gases is 0.41 m/s. From these values  $O_2$  molar fraction is ranged from 6 to 10 %. As a condition for these variations the total mass flow of the gases at the entrance was maintained constant at 3.89 x  $10^{-4}$  kg/s. The behavior of unburned fraction U in terms of these new molar fractions is shown in Fig. 3.

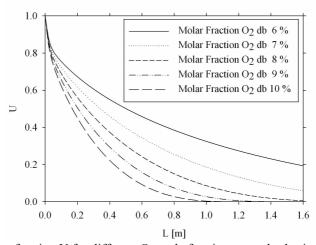


Figure 3. Behavior of unburnt fraction U for different  $O_2$  mole fractions on a dry basis at the entrance of DTF to the temperature of 1723K and  $\alpha = 1/3$ .

Figure 3 shows the variation of oxygen concentration directly affects reaction rates, to the point that with 8% of  $O_2$  molar fraction the unburnt fraction U approaches zero. As the oxygen concentration increases, the reaction rate becomes increasingly intense, and this information is represented by the intensity of the decrease slope of the curves of U.

## Total flow mass:

For this set the concentration of oxygen was maintained at 8% and the total flow mass was changed from 80 till 120% of the reference value. The behavior of unburnt fraction U due to change the mass flow of gases is shown in Fig. 4a. Looking at the results in Fig. 4a it can be seen that even increasing the mass flow, and the velocity of the gases directly, there were no major changes in the behavior of the reactor. The amount of oxygen required for complete combustion

does not change with increasing mass flow of gas. However, when reducing the mass flow it also affects the amount of  $O_2$ , and will eventually reduce the reaction rate. Fig. 4b shows the mass fractions of  $O_2$  at the ambient,  $y_{O_2}$ , and at the surface of the particles,  $y_{O_2,s}$ , under the same above conditions.

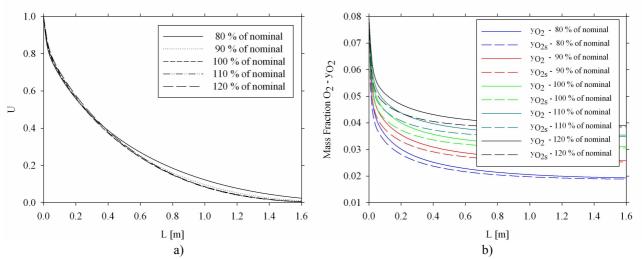


Figure 4. a) Behavior of unburnt fraction U due to change the mass flow of gases at the entrance of DTF to the temperature of 1723K in the system,  $\alpha = 1/3$ ; b) Variation of the mass fraction of  $O_2$ ,  $y_{O_2}$ , and mass fraction of  $O_2$  on the particle surface,  $y_{O_2,s}$ , along the reactor for varying the mass flow of gases to a temperature of 1723 K,  $\alpha = 1/3$ 

Figure 4b shows the variation of mass fraction of oxygen in the reactor,  $y_{O2}$ , and the particle surface,  $y_{O2,S}$ , along the reactor. It is possible to note that although the mass fraction and the initial  $O_2$  concentration are the same for all cases, the amount of oxygen available for reaction is different for each case, changing  $O_2$  diffusion to the particle surface, and thus the reaction rate is changed. So, for this particular case, the reduction in mass flow reduces the overall heterogeneous reaction rates even with flow velocity is reduced.

# Particle diameter:

The nominal particle diameter was set at 50  $\mu$ m, and different values of particles diameter were simulated around the nominal. The feed rate was maintained at 30 g/h. The behavior of unburnt fraction U as a function of variation of particle diameter is shown in Fig. 5a.

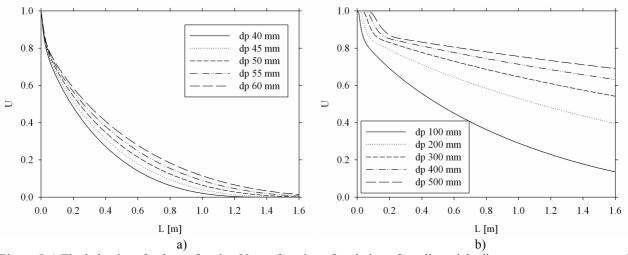


Figure 5.a) The behavior of unburnt fraction U as a function of variation of small particle diameters on temperature of 1723 K,  $\alpha = 1/3$ ; b) The behavior of unburnt fraction U as a function of variation of big particle diameters on temperature of 1723 K,  $\alpha = 1/3$ 

Figure 5a shows an expected relationship that has the diameter with the unburned fraction U. As it is maintained the coal feed rate, the quantity of carbon is the same but the diameter was changed, and consequently the reaction rate is altered. As the diameter decreases, the rate of conversion increases, i.e., the slope of the curve U becomes more pronounced. To investigate the sensitivity of the system to larger particle, bigger than 100 µm, particles of 100 to 500

 $\mu$ m were then tested and are shown in Fig. 5b. As a result of the increased on the particle size the difficulty of conversion increases. The intense conversion rate at the entrance of the reactor can be explained by the intense mass loss of the particle due to devolatilisation process. However, the  $O_2$  diffusion is inversely proportional of the particle diameter causing a decrease in the combustion rate, showed as less intense U curves.

## 6. CONCLUSIONS

In this work, a one-dimensional numerical model of a drop tube furnace (DTF) was developed from the literature. The results were compared with a reference model of Ballester and Jimenez, 2005, and with experimental data. Then a sensitivity study was conducted to understand the behavior of DTF and coal burning front to change some parameters of the system.

It was used a  $\alpha$  equal to 0.33, which represent that all reactions occurs only on the particle surface, for a population of particles with a diameter of 52.2  $\mu m$  and the kinetic parameters obtained by reference. From these parameters, the results showed good agreement with the reference model and also the experimental values. Sensitivity analysis demonstrated the versatility of the model and consequently the experiment. Using the model presented here concurrent with experimental results outcome a detailed study of the kinetic parameters of coal combustion. The application of a numerical model to simulate the coal combustion in a DTF allowed a better understanding of each stage of this complex process. Moreover, this kind of tool has a greater versatility and ability to explore models of coal combustion than commercial software.

With the experience gained on this work it is possible to suggest further studies: add a particles size distribution model, add a model of relative velocity of the particles, compare other models of burning coal, and build a DTF based on the knowledge reported here.

#### 7. ACKNOWLEDGEMENTS

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