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EXPERIMENTAL AND NUMERICAL INVESTIGATION OF COAL DEVOLATILIZATION IN A DTF

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Abstract. *This study investigates experimentally and numerically the devolatilization process of a low-rank Brazilian coal. Coal devolatilization is the first and faster reaction during the convencional air combustion. With the purpose of reducing the reaction velocity, the experiments will be investigated at different temperatures, varying from 600 to1100°C and different residence times, varying from 160 ms to 315 ms under* N_2 *atmosphere. The physicochemical characterization of the coal was performed by means of high heating values, activation energy determination, ultimate and proximate analysis. Preliminary DTF experiments have been carried out within the investigation of the coal combustion evolution and the results confirmed that the burnout increases as the residence time increases. However, the different temperatures and the devolatilization step have not been exploited. The goal of this study is to obtain a devolatilization response surface, scanning the completely accessible reactive area in the DTF. In parallel, a model of devolatilization and combustion of solid fuels in the DTF has been developed. The model includes the energy, momentum and mass balances, and allows for the prediction of burnout as well as the residence time and particle size variation within the DTF. It has been associated in a routine developed for the estimation of the kinetic parameters of the reaction steps of mixtures of particles of variable granulometry of single fuel. This work is in an early stage of development. Therefore, the main conclusions will be obtained in the next stages of the research.*

Keywords: *devolatilization, response surface, coal, DTF, Kinetics.*

1. INTRODUCTION

The combustion of low-grade fuels has received more and more attention due to the growing energy demand and scarcity of resources (Yu *et al*., 2018, Engin and Atakül, 2018). Low-grade coal is very abundant in several regions of the world, contributing with 45% of the total global coal reserves (Yu *et al*., 2018). Brazil has large reserves of coal of varying quality, around 106 tons, mainly located in the southern region (Balanço Energético Nacional – BEN 2016). However, most is low grade coal, which presents low calorific values, high ash and sulfur content (Restrepo *et al*., 2015), hence, the use of these fuels in an efficient and clean way is the focus of many researches (Valdés *et al*., 2016, Valdés and Chejne 2017, Rathman *et al*., 2009).

During coal combustion, the first and faster reaction under convencional air condition is the devolatilization (Valdés *et al*., 2016). This step involves multiple chemical reactions coupled with transport phenomena (Howard, Fongand and Peters, 1987) and is very important to determine subsequent combustion mechanisms (Han *et al*., 2018; Valdés *et al*., 2016, Valdés and Chejne 2017). According to Valdés et al. 2016, parameters such as temperature, reaction time, pressure and type of coal highly affect the distribution of products during coal devolatilization, making it difficult to isolate the individual phenomena (Valdés and Chejne 2017).Thermogravimetric analysis (TGA) is a widely used technique to investigate the release of the volatile matter. Despite plotting a behavioral trend, these studies cannot be extrapolated to practical applications. In order to enhance the devolatilization investigation in a larger scale, a DTF will be employed to evaluate the release of volatiles during coal combustion. The study will be performed under an inert atmosphere (N_2), different temperature and residence time conditions, established according to an experimental design. Furthermore, a numerical investigation will be performed to estimate the kinetic parameters during devolatilization.

2. METHODOLOGY

2.1 Experimental approach

A low grade coal from the South region of Brazil was investigated. The coal characterization was performed based on the higher heating value (HHV), ultimate and proximate analysis. Thermogravimetric experiments will be performed under 10, 20 and 30°Cmin^{-1}, with N_2 gas flow rate of 100 mLmin^{-1}, to determine the coal thermal decomposition profile and the activation energy related to the devolatilization step. The Kissinger–Akahira–Sunose (KAS) model will be used to determine the activation energy according to Eq. (1) (Vyazovkin *et al*., 2011):

$$\ln\left(\frac{\beta}{T_\alpha^2}\right) = \ln\left[\frac{RA}{E_\alpha g(\alpha)}\right] - \left(\frac{E_\alpha}{R}\right)\frac{1}{T_\alpha} \quad (1)$$

where E_α [kJmol^{-1}] is the activation energy related to each conversion value (α), β is the heating rate [K min^{-1}], T_α is temperature [K], related to each α value, and $g(\alpha)$ is the integral form of the inverse function $f(\alpha)$, that is the model reaction.

The devolatilization process can be influenced by several factors such as temperature, residence time, atmosphere, soon. Thus, to obtain a response surface of coal devolatilization, to evaluate the interaction between the factors, reduce the number of experiments and to optimize measurable parameters, a Central Composite Rotatable Design (CCRD) experimental design will be used. The CCRD is a statistical tool that employs multiple regression analysis and provides a second-order model for the production of a response surface. Furthermore, this technique is advantageous because it describes the interactive, cumulative, and individual effects of the test variables on the process yield (Colla *et al*., 2010). The experiments are designed with 5 replicates at the central point, four axial points, and 2^2 plus star configuration, in which are performed randomly. In this study, temperature (T) and residence time (t) are the variables evaluated at five different levels, which were selected on the basis of preliminary experiments. The coded and real values of the variables are shown in Tab. 1.

Table 1: CCRD matrix design of the coded and real values of temperature and residence time.

Run	Temperature Coded and real values (°C)	Residence time Coded and real values (ms)	Predicted Response
1	-1 (670)	-1 (183)	Y
2	1 (1030)	-1 (183)	Y
3	-1 (670)	1 (294)	Y
4	1 (1030)	1 (294)	Y
5	$-\sqrt{2}$ (600)	0 (238)	Y
6	$\sqrt{2}$ (1100)	0 (238)	Y
7	0 (850)	$-\sqrt{2}$ (160)	Y
8	0 (850)	$\sqrt{2}$ (315)	Y
9	0 (850)	0 (238)	Y
10	0 (850)	0 (238)	Y
11	0 (850)	0 (238)	Y
12	0 (850)	0 (238)	Y
13	0 (850)	0 (238)	Y

The quadratic polynomial model related to the response obtained in Table 1 is represented by Eq. 2.

$$Y = \beta_0 + \sum_{j=1}^{k} \beta_j x_j + \sum \sum_{i<j} \beta_{ij} x_i x_j + \sum_{j+1}^{k} \beta_{jj} \, x_j^2 + \varepsilon \quad (2)$$

where Y is th predicted response, β_0, β_j, β_{ij}, and β_{jj} are constant coefficients, i and j take values from 1 to the number of variables, *xi* and *xj* are the coded independent variables or factors, and ε is a random error.

The analysis of variance (ANOVA) will be employed to perform the statistical significance as well as the Student's test (t).

The predicted response (Y) is the percentage of devolatilization obtained in the DTF experiments and, subsequently determined by means of Proximate Analysis.

The DTF experiments will be conducted applying the conditions established in Tab. 1. The combustion chamber of the reactor is constituted by a ceramic tube of 40 mm of inner diameter and 1.6 m in length. The temperature of the reactor's wall is continuously measured by nine Type K thermocouples uniformly distributed along the combustion chamber. A water-cooled injector, placed at the top of the reactor is used to feed solid fuel and fluid transport into the combustion chamber. In each experiment, the mass flow rate of solid fuel is approximately 50 g h^{-1}. The samples are feed into the reactor at the required conditions continuously for 15 minutes. After passing through the tube, the particles are collected with the support equipped with a quartz filter. The proximate analysis of the collected material will provide the devolatilization yield. The proximate analysis will be performed according to D7582-15 Standard Test Methods for Proximate Analysis of Coal and Coke, using TGA 701 Thermogravimetric Analysis - LECO Corporation.

2.2 Numerical modelling

In order to describe the evolution of the thermal conversion processes taking place within the DTF, a one-dimensional transient model of flow and heat transfer of solid fuel particles has been developed.

The model is based on the works of Wang, 2014 and Ballester and Jiménez, 2005, and allows for the prediction of burnout, residence time and particle size variation along the heated length of the furnace as functions of the DTF operating conditions and particle size distribution. Coupling of the model with a proper objective function in the scope of an optimization problem allows for the estimation of kinetic parameters of the combustion of solid fuels in the DTF.

3. PRELIMINARY RESULTS

The high heating rate, the proximate and ultimate analyses of the Brazilian coal are shown in Tab. 2.

Table 2: Proximate and ultimate analysis of theBrazilian coal (dry basis).

Proximate Analysis	
Moisture (%)	4.06
Volatilematter (%)	27.66
Fixedcarbon (%)	40.28
Ash (%)	32.06
UltimateAnalysis	
Carbon (%)	53.80
Hydrogen (%)	3.52
Oxygen (%)*	8.92
Nitrogen (%)	0.87
Sulfur (%)	0.83
HHV (kJ g^{-1})	20.568

* by difference.

The preliminary experiments were performed under air atmosphere condition. Fig. 1 shows the burnout obtained at 1100 °C and different residence times (axial positions).

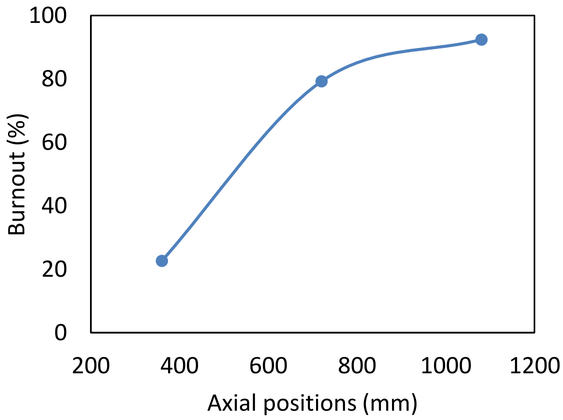

Figure 1. Coal burnout under air atmosphere in the DTF.

The coal reactivity under conventional air conditions is evaluated according to the burnout. The burnout indicateshow the combustion evolves as the temperature and the residence time (measured as axial positions in the DTF) change. Fig. 1 shows that after 720 mm (residence time of 210 ms), the burnout is higher than 80% because the temperature and residence time are high enough to provide the complete volatile releasing and the consumption of the remained char. The results are in agreement with those found in the literature (Steer *et al*., 2015; Wang *et al*., 2014).

To evaluate the devolatilization it is expected that the inert atmosphere and lower temperatures proposed in this study will affect the release of volatile and displace the event to longer residence times.

Some preliminary calculations have been performed in order to estimate the effect of the DTF temperature on the devolatilization of the coal in conditions of inert atmosphere. The results are presented in Fig. 2.

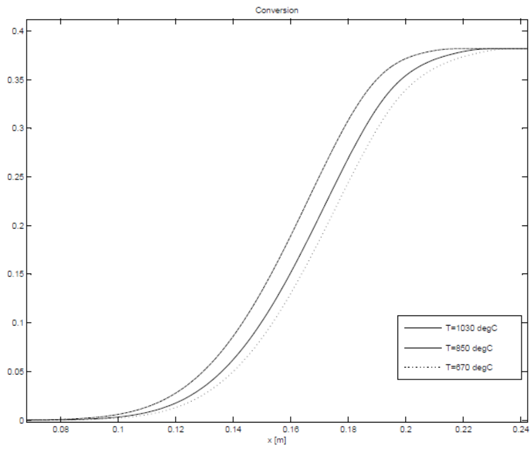

Figure 2. Predicted devolatilization of coal under inert atmosphere in the DTF.

In these calculations, single sized coal particles with 60µm in the diameter are fed at room temperature in the DTF. Inert atmosphere is obtained by setting oxygen partial pressure equal to zero. Since the model requires that the measured temperature profile be known, results of previous studies of combustion in air have been used in these preliminary calculations.

It can be observed that the increase in the temperature leads to shorter devolatilization lengths. This is expected as a result of the increase of the devolatilization rates for higher temperatures.

The present work is in the initial stages of development; hence, no conclusions are available.

4. ACKNOWLEDGEMENTS

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6. RESPONSIBILITY NOTICE

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