Mathematical modeling of the coal activation process in rotary cylindrical kiln

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***Abstract*—**The activation of coal applying physical or thermal method is carried out under an atmosphere in the presence of air, dioxide carbon or water vapor, at temperatures between 800 °C and 900 °C. In this research, it get the mathematical modeling of the coal activation process in order to predict the behavior of the gas and coal temperature distribution inside a rotary cylindrical kiln. The proposed model aims for acquiring useful information to select operating conditions and design parameters. The model groups a non-linear differential equations system, the equations to determine the temperature of the cylinder inner wall and the heat transfer coefficients. The Runge-Kutta fourth order numerical method was used. The comparison of the results obtained from the modeling of the gas temperature inside the cylinder with the experimental data showed that the variation is negligible, with an error less than 5 %.

***Keywords*—**mathematical modeling, activation, vegetal coal, rotating cylindrical furnace.

I. INTRODUCTION

ACTIVATED coal is a product with a reticular crystalline structure similar to graphite. It is extremely porous and can develop surface areas of the order of 1 500 m^2/g of coal, which by means of adsorption, capture on its surface a high range of molecules [1]. This product is used in the extraction of metals, water purification and treatment, medicine, clarification and elimination of odors, the purification of glycerin, the control of gas emissions in automobiles, in purification filters. These applications development numerous investigations related to its preparation and characterization, example: biomass residues from agricultural activities [2].

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Although rotary kilns are widely used by the chemical industry, remain among the pieces of equipment most difficult to be properly analyzed due to the complexity of internal phenomena (heat, mass, and momentum transfer) along with the eventual chemical reaction phenomena [3]. Rotary kilns are used by almost all the most important world manufacturers; however, few studies about the modeling of processes to activate coal with steam in rotary kilns have not been published.

Due to the specifications of each material and the difficulties of associate those to the activation reactions kinetics, the mathematical models for the activation of coal are obtained experimentally, this allows predict the stationary and dynamic behavior in rotary kilns [4]. Ortiz et al [3] have presented a model related to the coal activation with steam rotary cylindrical ovens, which is used by Kim [5] to study the process of pyrolysis in a rotary cylindrical furnace, for the recovery of the volatile organic compounds contained in the process.

The coal activation is carried out by the thermal or physical method, through the action of oxidizing gases such as water vapor, carbon dioxide or air. The Oxygen is responsible for burning the most reactive parts, giving the carbon absorptive properties [6].

In this method the reactions kinetic parameters of the oxygen with the coal molecules are very sensitive to energy variations. When the reaction is exothermic, the coal reacts at about 700 K, and more precipitously at 1600 K. That is, the magnitude of the reaction has to be limited with temperature, for this reason is necessary the rigorous control of the temperature [7].

Other works use the thermogravimetric analysis (TGA), which is an extensive research method for studying thermal behavior. Thermal characteristics of ignition temperature, burnout temperature, maximum combustion rate, and combustion kinetic parameters can be obtained to analyze the thermal process of fuel [8]-[9]. In the research, developed by Qiang [9], pyrolysis and combustion behaviors of three coals were investigated and their combustion kinetics were calculated by the Freeman–Carroll method to obtain quantitative insight into their combustion behaviors. Moreover, the effects of coal size, airflow, oxygen content, and heating rate on coal combustion behaviors were analyzed. Results showed that the three coals have a similar trend of pyrolysis that occurs at about 670 K and this process continuously proceeds along with their combustion. Combustion characteristics and kinetic parameters can be applied to analyze coal combustion behaviors. However, the thermal reaction process of coal in a rotary kiln has not been clearly understood.

Other researchers developed a theoretical-experimental models take in consideration the flowed and dense phases. With the employment of the pattern it is simulated the dependence of the pressure losses, the solid flow and the concentration of the mixture in function of the gas flow of pneumatic transport systems. The authors predict the areas of lower energy consumption and make the correct selection of the systems [10]-[11].

In previous works, phenomenological models are presented to describe the behavior of the process variables throughout the cylinder length. However, the rotary cylindrical ovens are still empirically designed due to the lack of an appropriate model of heat transfer that characterizes the behavior of the variables in the process [12]-[13].

The objective of this research was to establish a model to predict the temperatures behavior of the coal and combustion gases during the coal activation process in a rotary cylindrical furnace.

II. METHOD DEVELOPMENT

For the modeling of the coal activation process in a rotary cylindrical kiln, it starts from the energy and mass balance, through which is obtained a differential equations system, that includes the chemical reactions between coal and water vapor. In addition, the equations that characterize the heat and mass transfer coefficients in outside and inside of the kiln are presented. This method of modeling has been used successfully by various researchers for the analysis of similar equipment such as rotary kilns and cylindrical dryers [14]-[15]. For the elaboration of the model are made the following considerations:

- The changes in velocities for coal and gas, as well as coal particles dragged by the gas, are negligible.
- All the variables are uniform in the radial direction.
- Steam flow and pressure in the kiln are constant.
- The chemical reaction obeys to the Arrhenius Law and the secondary reactions are not taken into account.

There are three phases inside a rotary kiln: gas, solid, and wall [3]. Beside the activation chemical reaction, the phenomena occurring are: mass transfer (solid's moisture and gaseous reaction products are transferred from the solid to the gas phase) and heat transfer (convection and radiation between gas and solid bed, gas and wall, wall and solid bed, and wall and surroundings, and conduction inside of the wall).

In the solution of the model was used the numerical method Runge Kutta fourth order, with the help of the MATLAB professional software, for the design conditions of the kiln.

A. System features

The rotary cylindrical kiln is 1.6 m in diameter and 12 m in length; it has an inclination angle of 15 sexagesimal degrees, with respect to the horizontal plane, to facilitate the mixing and transfer of the material. It works in co-current (Figure 1) and with the operating conditions shown in Table 1.

To guarantee the energy in the process are used the gases generated in the combustion chamber (1), which are supplied to the inside of the cylinder (2) together with the saturated steam (4). The coal for the activation is supplied through conveyor belts that discharge to a hopper (3) with dosers that guarantees the flow of coal for the process. The activated coal leaves the kiln by means of conveyor (5).

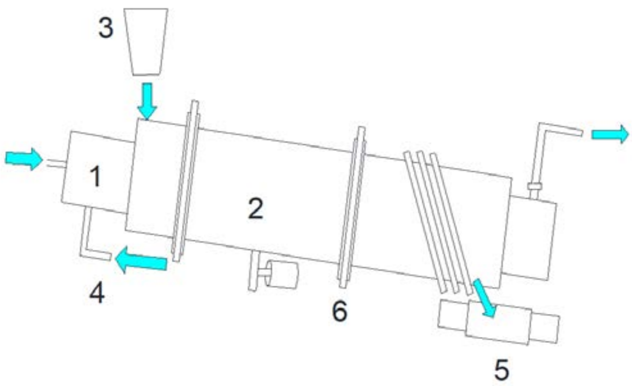

Figure 1. Coal activation kiln.

In the internal part of the kiln are arranged a fins blades that favor the lifting of the material, while the cylinder rotates supported on rollers (6), allowing a better contact between the solid and the gas flow. The fins are inclined with respect to the axis of the cylinder to facilitate the material movement along the same.

Table 1. Operating conditions in the rotary cylindrical kiln used for coal activation

Process parameters	**Values**	**Unity**
Coal flow to the kiln	150-200	kg/h
Steam flow	300-350	kg/h
Rotation speed	6	rpm
Material permanence time in the kiln	3-4	h
Steam temperature	123-140	^{o}C
Steam pressure in the kiln	2	kgf/cm^2
Combustion chamber temperature	1100-1400	ºC
Gases exit temperature	750-800	ºC

B. Identification of heat transfer processes

Figure 2 shows the heat transfer processes in the rotary cylindrical furnace used in the kiln activation. The gases and saturated water steam transfer heat by convection and radiation to coal and to the inner wall of the cylinder not covered by the solid (1). The heat absorbed by the wall is transferred by conduction: a part, to the solid in contact with the wall (2) and, the other part, by conduction from the inner wall to the outside and of this by convection and radiation to the surroundings (3).

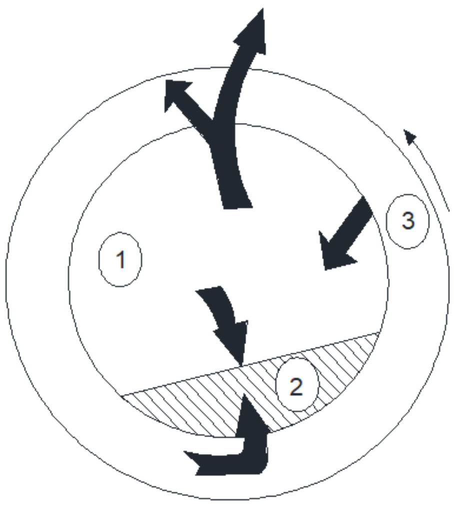

Figure 2. Cross section of the rotary cylindrical kiln.

The term convection is used to describe the energy transfer between a surface and a fluid. Although the diffusion mechanism contributes to this transfer, generally the dominant contribution is the overall movement of the fluid particles. Convection appears only in fluids, where this movement of matter can take place, characterized by a thermal flow transmitted, given by the empirical relationship known as Newton's Law of Cooling [16].

Thermal radiation is the propagation of electromagnetic waves, in certain wavelengths, emitted by a body because of its temperature. The transfer of heat by radiation does not require the presence of matter since heat can be transmitted through absolute vacuum unlike heat transfer by conduction and convection [17].

Some researchers suggest that the coal inside the reactor is behaves like a fluid and propose to estimate the heat transfer between the solid and wall using the convection heat transfer [18].

C. Mathematical modeling for the solid loss by chemical reaction

In order to model the transfer phenomena the differential mass and energy balances are posed, the following assumptions were adopted:

- Both, solid and gas axial linear velocity changes are negligible.
- All variables are supposed uniform in the radial direction due to the small size of the kiln.
- There is neither solid nor gas axial mixing. Then, the two phases are considered as a plug flow model. Solid drag by gas is negligible.
- Variations in rotary kiln wall temperature in the angular direction are neglected.
- Constant steam flow rate is assumed.
- Pressure in the kiln is constant.
- Axial heat transfer by conduction and radiation is negligible.

According to [19], during the activation process part of the coal reacts with water vapor, obtaining carbon monoxide and dihydrogen, releasing 28.5 kcal/mol (equation 1).

$$C + H_2O = CO + H_2 \tag{1}$$

The amount of solid mass lost is determined by experiments (equation 2), in similar conditions to rotary kiln for the coal activation. The kinetic equation has account to the Arrhenius law and considers the steam flow [4].

$$r_s = -\frac{\partial \dot{m}_s}{\partial z} = \frac{6{,}005 \cdot \left[exp^{\left(\frac{-8033}{T_s}\right)}\right] \cdot \dot{m}_v \cdot \dot{m}_s}{V_s} \tag{2}$$

Mass balance

To determine the variation of moisture in the solid, they are taken into account the two phases of drying a substance. The first one through evaporation and is considered that the solid surface remains saturated. In the second, the drying process is obtained by the moisture diffusion from inside the particle to its surface [20]. For critical humidity of 10 % and assuming continuity of the two phases, the variation of moisture in the solid in the axial direction is expressed by equation 3.

$$\frac{\partial \dot{m}_h}{\partial z} = \frac{h_t \cdot A \cdot (T_g - T_s) \cdot \dot{m}_h}{H_v \cdot (0{,}1 \cdot \dot{m}_s)} \tag{3}$$

Taking into account the analysis of solid loss by chemistry reaction, and the variation of moisture (equations 2 and 3) it get the expression 4, for the mass balance of the coal along the cylinder. It allows determine the variation of the coal flow along the kiln, considering the movements of the solid through the kiln in the positive direction of the "z" coordinate.

$$\frac{\partial \dot{m}_s}{\partial z} = -K_e \cdot e^{-\left(\frac{8033}{T_s}\right)} \cdot \dot{m}_v \cdot \frac{\dot{m}_s}{V_s} - \frac{h_t \cdot A \cdot (T_g - T_s) \cdot \dot{m}_h}{H_v \cdot (0{,}1 \cdot \dot{m}_s)} \tag{4}$$

The mass balance for gas, which considers the increase in the gases flow due to the evaporation of moisture from the solid bed and the gases product of the chemical reaction, is done by equation 5.

$$\frac{\partial \dot{m}_g}{\partial z} = K_e \cdot e^{-\left(\frac{8033}{T_s}\right)} \cdot \dot{m}_a \cdot \frac{\dot{m}_s}{V_s} \cdot \frac{30}{12} + \frac{h_t \cdot A \cdot (T_g - T_s) \cdot \dot{m}_h}{H_v \cdot (0{,}1 \cdot \dot{m}_s)} \tag{5}$$

Energy balance

The gas heat flux variation along the "z" coordinate depends on the convection and radiation heat transfer and the gas-solid, gas-wall combination, plus the sensible heat added by the water vapor to the coal activation. The variables K_1, K_2, K_3, K_4, K_5 and K_6 are the convection and radiation heat transfer coefficients from the gas to the solid (equations 7-12).

$$\frac{\partial(\dot{m}_g \cdot C_g \cdot T_g)}{\partial z} = -K_1 \cdot (T_g - T_s) - K_2 \cdot ({T_g}^4 \cdot e_g - {T_s}^4 \cdot A_v) - K_3 \cdot (T_g - T_w) - K_4 \cdot ({T_g}^4 \cdot e_g - {T_w}^4 \cdot A_v) + \frac{\partial \dot{m}_h}{\partial z} \cdot C_v \cdot (T_s - 373) \tag{6}$$

Where

$$K_1 = h_{gs} \cdot L_{ss} \tag{7}$$

$$K_2 = \frac{\sigma \cdot L_{ss} \cdot e_w}{[1-(1-e_s)\cdot(1-A_v)]} \tag{8}$$

$$K_3 = h_{gw} \cdot L_{pnc} \tag{9}$$

$$K_4 = \frac{\sigma \cdot L_{pnc} \cdot e_w}{[1-(1-e_w)\cdot(1-A_v)]} \tag{10}$$

$$K_5 = h_{sw} \cdot L_{pcs} \tag{11}$$

$$K_6 = \sigma \cdot L_{ss} \cdot \phi_{sw} \cdot e_w \cdot e_s \tag{12}$$

The variation of the solid heat flux along the "z" coordinate depends on the heat transfer by: convection and radiation of the gas to the solid; convection and radiation from the wall to the solid; the heat of vaporization and the heat of the solid chemical reaction for activation. To consider all the heats mentioned above the energy balance equation for coal is the following.

$$\frac{\partial(\dot{m}_s \cdot C_s \cdot T_s)}{\partial z} = K_1 \cdot (T_g - T_s) + K_2 \cdot ({T_g}^4 \cdot e_g - {T_s}^4 \cdot A_v) + K_5 \cdot (T_w - T_s) + k_6 \cdot ({T_w}^4 \cdot e_w - {T_s}^4 \cdot A_v) - \frac{\partial \dot{m}_h}{\partial z} \cdot H_v - K_e \cdot e^{-\left(\frac{8033}{T_s}\right)} \cdot \dot{m}_v \cdot \frac{\dot{m}_s}{V_s} \cdot \Delta H$$

(13)

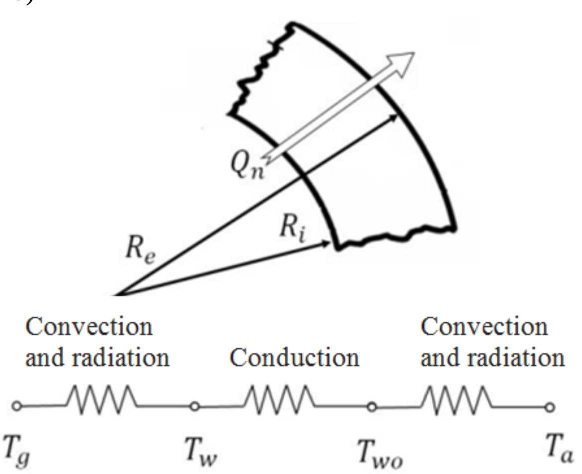

Figure 3. Resistance circuit for the heat flow from the inside to the outside of the cylinder.

To solve the above equations (equation 12 and 13) is necessary to know the temperature of the wall inside the cylinder, for this purpose an energy balance is made considering the net heat flux transferred from inside to outside the cylinder (Figure 3). In this case the heat transferred by conduction and convection between the covered wall and the solid is not considered because is assumed that both have the same temperature (equation 14). The wall temperature inside of the cylinder is considered equal to the gas temperature since the equilibrium state between the gas and the inner wall is reached.

$$T_w = \frac{\left\{\frac{T_a}{h_i \cdot R_i} + T_g \cdot \left[\frac{1}{\left[\left[1+\frac{T_a}{T_{w0}}+\left(\frac{T_a}{T_{w0}}\right)^2+\left(\frac{T_a}{T_{w0}}\right)^3\right] \cdot e_{w0} \cdot \sigma \cdot (T_{w0})^3 + h_0\right] \cdot R_e}\right] - \frac{\ln \frac{R_i}{R_e}}{K_w}\right\}}{\frac{1}{h_i \cdot R_i} + \frac{1}{\left[\left[1+\frac{T_a}{T_{w0}}+\left(\frac{T_a}{T_{w0}}\right)^2+\left(\frac{T_a}{T_{w0}}\right)^3\right] \cdot e_{w0} \cdot \sigma \cdot (T_{w0})^3 + h_0\right] \cdot R_e} - \frac{\ln \frac{R_i}{R_e}}{K_w}} \tag{14}$$

In the model identification is necessary to use an iterative procedure starting from the reference state, and the Runge - Kutta fourth order method that takes into account the behavior of the derivative in four points of each interval. This method, like part of the iterative process is used to solve the theoretical model and to find the values of the characteristic parameters in the coal activation.

The number of experiments was determined from the application of a multifactorial design. In agreement with the determined levels of each variable, at least eight levels of each parameters. The confirmation of the validity of the experimental results with the theoretical model is developed through the relative error, that is, the difference between the experimental value "X_{exp}" of the temperature and the theoretical value "X_{teo}" obtained by the model for the same conditions of the experiment. The relative error is calculated by the following expression:

$$E_p = \left| \frac{X_{exp} - X_{teo}}{X_{exp}} \right| \cdot 100 \tag{15}$$

The average relative error is expressed for:

$$E = \sum_{i=1}^{n} \left| \frac{X_{exp} - X_{teo}}{X_{exp}} \right| \cdot \frac{100}{n} \tag{16}$$

The methodology used during the realization of the experiments is the following:

- The instruments used to measure the values of the variables were calibrated.
- The instruments connection of the data acquisition system was verified for the registration and monitoring of the variables.
- The material flow is constant during the process, according to the experiments design without taking into account the thermal profile of the kiln.
- The measurements of the process parameters were made

III. RESULTS AND DISCUSSION

Figure 4, shows the behavior of the gases temperature as a function of the cylinder length using the experimental results and those obtained by the model. La temperatura de los gases varía entre 1350 K y 1100 K para el proceso de activación del carbón, estos valores se corresponden con los establecidos por la literatura.

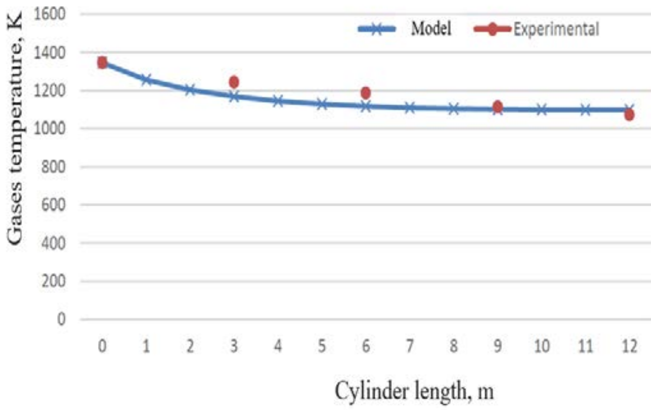

Figure 4 behavior of the gases temperature as a function of the kiln length.

The magnitude of the relative error is less than 5% and the average relative error is 1.19 % (Figure 5). These results confirm the validity of the proposed model to predict the temperature behavior of the gas inside the rotary cylindrical kiln used for the activation of said material.

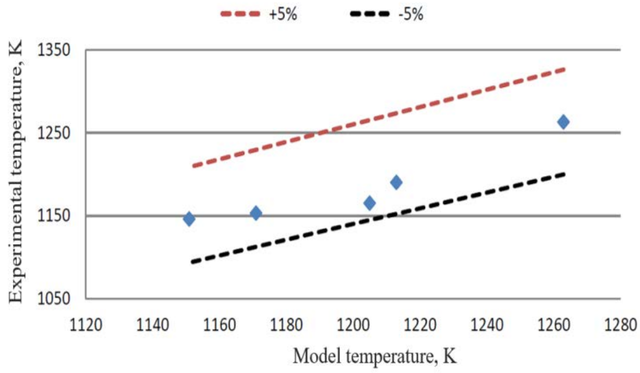

Figure 5 behaviors of the relative errors for the experimental and model temperatures values.

The behavior of the gas and the coal temperature as a function of the furnace length was obtained through the proposed model (Figure 6). The solid temperature (T_s) increases rapidly in the first three meters of the kiln, obtaining values between 750 K and 900 K. Then, it remains practically constant in values slightly higher than 1 000 K, which constitutes the temperature profile required for activation. In the case of the gases temperature, decrease between 1 350 K and 1 150 K is logical and consistent with the results obtained in the system.

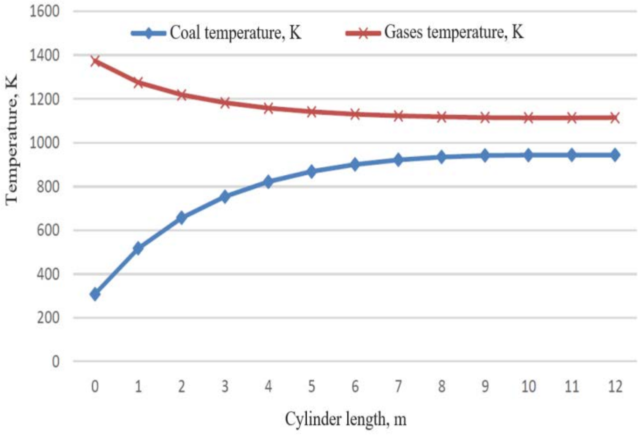

Figure 6 behavior of the gas and coal temperature as a function of the kiln length.

Another behavior obtained through modeling was the variation of carbon and gas mass flows inside the cylinder (Figure 7).

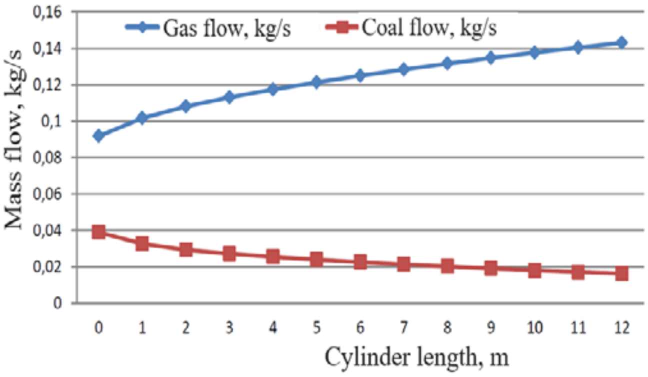

Figure 7 behavior of the gas and coal mass flow as a function of the kiln length.

The gas mass flow increases from 0.09 kg/s to 0.143 kg/s for 12 m oven length. The coal mass flow varies between 0.04 kg/s and 0.02 kg/s for the same conditions. The practical application of the proposed mathematical model lies in the possibility of predicting the temperature behavior of the activated carbon at the outlet of the horizontal rotary kiln, under different operating regimes, in order to guarantee the adequate temperature of the coal activation process, contributing to the saving of energy carriers.

IV. CONCLUSIONS

- The mathematical model that characterizes the heat and mass transfer during the charcoal activation process in a rotary cylindrical furnace was established. The model consists of a system of ordinary differential equations of the first order, obtained from the energy and mass balance. The accuracy of the model is greater than 95%, satisfactory for the energy evaluation of the investigated process.
- The gas mass flow increases from 0.09 kg/s to 0.143 kg/s for 12 m oven length. The coal mass flow varies between 0.04 kg/s and 0.02 kg/s for the same conditions. The practical application of the proposed mathematical model lies in the possibility of predicting the temperature behavior of the activated carbon at the outlet of the horizontal rotary kiln, under different operating regimes, in order to guarantee the adequate temperature of the coal activation process, contributing to the saving of energy carriers.

Nomenclature:

V_s - Coal flow speed, m/s
$\dot{m}_s$ - Coal mass flow, kg/s
Z - Cylinder length, m
K_e - Reaction kinetics constant, s^{-1}
H_v – Heat of vaporization, kJ/kg
$\dot{m}_v$ - Vapor mass flow, kg/s
$\dot{m}_h$ - Water content in the coal, kg/s
A - Gas-solid contact surfaces, m^2
T_s – Coal temperature, K
T_v – Gas temperature, K
h_t – Inside heat transfer coefficient, $W/m^2.K$
$\dot{m}_g$ - Combustion gases flow, kg/s
C_g - Gas specific heat, J/kg.K
C_v - Vapor specific heat, J/kg.K
T_w – Wall temperature, K
K_1 - Convection heat transfer coefficient per unit length, gas - solid, W/m.K
K_2 - Radiation heat transfer coefficient per unit length, gas - solid, W/m.K
K_3 - Convection heat transfer coefficient per unit length, gas - wall, W/m.K
K_4 - Radiation heat transfer coefficient per unit length, gas - wall, W/m.K
K_5 - Convection heat transfer coefficient per unit length, solid - wall, W/m.K
K_6 - Radiation heat transfer coefficient per unit length, solid - wall, W/m.K
e_g - Gas emissivity, dimensionless
A_v - Adsorption, dimensionless
h_{gs} - Convection heat transfer coefficient, gas - solid, $W/m^2.K$
h_{gw} - Convection heat transfer coefficient, gas - wall, $W/m^2.K$
L_{ss} - Cylinder arc length that is in contact with the gas, m
L_{pnc} - Cylinder wall arc length not covered by the solid, m
L_{pcs} - Arc Length that forms the wall covered by the solid, m
e_w - Wall emissivity, dimensionless
e_{wo} - Outer wall emissivity, dimensionless
e_s - Coal emissivity, dimensionless
σ - Stefan-Boltzmann constant, $W/m^2.K^4$
ϕ_{sw} – Radiation constant, dimensionless
ΔH - Heat due to the chemical reaction between the gas flow and the coal, kJ/mol.K
R_e - kiln outside radius, m
R_i - kiln inside radius, m
K_w -Wall thermal conductivity, W/m.K
h_i - Inside heat transfer coefficient, $W/m^2.K$
h_o - Outside heat transfer coefficient, $W/m^2.K$
T_a - Outside temperature, K
T_{wo} - Outside wall temperature, K
r_s - Coal mass flow that reacts with water vapor, kg/h

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