ANALYSIS OF THE DYNAMIC BEHAVIOUR OF A BURNING POROUS CHAR PARTICLE

by

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The mathematical model developed describes the dynamic behavior of a porous char particle during combustion. The model of the char particle involves two exothermic reactions: $C + O_2 = x \cdot CO + y \cdot CO_2$ *– heterogeneous reaction on the internal and external char particle surfaces, and* $CO + O = CO_2$ *– homogeneous reaction inside the char particle. The temperature and gas concentration fields inside the char particle are defined by partial differential equations of heat and mass conservation. Changes of internal surface area and porosity during combustion are included in the mathematical model. The mathematical model results were compared with experiments done in a laboratory fluidized bed reactor. The comparison showed that the model predictions are in a very good acccordance with experiments. The model succesively followed combustion regime changes with bed temperature and coal type.*

Introduction

The overall rate of char particle combustion or gasification depends not only on external conditions, but also on internal reactions either heterogeneous or homogeneous and on intraparticle heat and mass transfer. Which of these processes plays the most significant role in the overall reaction rate depends on the conditions surrounding the particle and the particle structure. In a case of relatively low mass transfer rate toward the char particle, the overall reaction rate is under external diffusion or external mass transfer control. In that case the char particle structure and internal heat and mass transfer are not of importance. But if the external mass transfer rate is sufficiently high to enable an intensive reaction rate, the limit for the overall reaction rate could be internal mass transfer or chemical kinetics. Then the structure of the porous char particle becomes an important parameter, and besides initial internal surface area and porosity, their evolutions are also needed. Using initial parameters of char particle combustion, one could predict some combustion regime, but it does not mean that the combustion regime defined would remain the same in later stages of char particle conversion. Usually, at not so high reaction temperature, the char particle passes through several

combustion regimes. In the first period of combustion, it could be a combustion regime under kinetic control. Along with char particle conversion, the kinetic combustion regime could transfer over a transition regime to a diffusion combustion regime. Whether char particle combustion is under kinetic or under diffusion control depends on many factors. In order to simultaneously comprise all of these factors one needs a complex mathematical model of porous char particle combustion, which can spatially and temporally consider processes within the char particle. One of the first models of char particle reaction, regarding the char particle as a porous medium, was done by Srinivas and Amundson [1]. They analyzed gasification of a porous char particle applying a microscopic approach. Analysis was made for steady state conditions. They obtained gas concentrations profiles within the char particle, but they considered only the initial stage of the particle gasification process, therefore they did not take into account the evolution of the porous structure of the char particle. After that several models followed, but the greatest contribution to the char particle reaction analysis was given by Sotirchos and Amundson [2] or Sotirchos and co-authors (Sotirchos and Burganos [3]; Hsuen and Sotirchos [4]) In their latest paper [4] they took into account three chemical reactions within the char particle and in the boundary layer around the char particle, and considered steady state conditions. In the earlier article Sotirchos and Burganos [3] treated the dynamic behaviour of a char particle during combustion, taking into account only one chemical reaction. In all models of the authors mentioned the heat transfer within the char particle was not microscopically analyzed, that is heat transfer was not affected by local char particle properties.

Mathematical model of char particle combustion

The mathematical model developed describes the dynamic behaviour of porous char particle combustion. The model includes external and intra-particle heat and mass transfer and chemical reactions inside the particle and on the external surface of the particle. Only two chemical reactions are considered: heterogeneous reaction on the external and internal surface of the particle (Reaction 1) and homogeneous reaction within the pore volume of the particle (Reaction 2). The particle is composed of one solid reactant (carbon), mineral matter and voidage. The mineral matter has no catalytic effect on combustion. The environment surrounding the char particle consists of O_2, CO_2, CO, H_2O and N_2. The role of H_2O is only as a catalyst for the homogeneous reaction of CO oxidation.

Reaction 1: $C + O_2 \rightarrow x \cdot CO + y \cdot CO_2$ $\qquad R_1 = A_1 C_{O_2} \exp\left(\frac{-E_1}{R_g T}\right)$

Reaction 2: $CO + O \rightarrow CO_2$ $\qquad R_2 = A_2 C_{O_2}^{0.5} C_{CO} C_{H_2O}^{0.5} \exp\left(\frac{-E_2}{R_g T}\right)$

The molar ratio CO/CO_2 in reaction 1 is defined with the following relation [5]:

$$\frac{CO}{CO_2} = 2512 \exp\left(\frac{51880}{RgT}\right)$$

A microscopic approach to the char combustion model was adopted. Temporal and spatial evolution of char particle physical properties, heat and mass transfer characteristics, as well as temperature and gas concentrations are defined. The pore structure locally evolves proportrional to the local particle conversion and affects the heat and mass transfer properties of the particle.

Char particle shrinkage is allowed. The criterion for particle shrinking in the model is that local degree of conversion of the external surface layer is over 98%. It is known that there are different shrinking characteristics for different coal types, and shrinking depends not only on the degree of burn-off but also on the strength of the particle solid matrix.

The temperature and gas concentration fields within the char particle are defined by the partial differential equations of heat and mass conservation:

$$C_p \frac{\partial T}{\partial \tau} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \lambda_{eff} \frac{\partial T}{\partial r}\right) + \Sigma(C_{p,j} N_j)\frac{\partial T}{\partial r} - \Delta H_1 R_1 S - \Delta H_2 R_2 \varepsilon$$

$$\frac{\partial C_j}{\partial \tau} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 D_{eff} \frac{\partial C_j}{\partial r}\right) + \nu_{1,j} R_1 S + \nu_{2,j} R_2 \varepsilon \qquad j = O_2, CO_2, CO, H_2O$$

The boundary conditions are defined as follows:

$$r = 0 \Rightarrow \frac{\partial C_j}{\partial r} = 0, \quad \frac{\partial T}{\partial r} = 0$$

$$r = R_c \Rightarrow -\lambda \frac{\partial T}{\partial r}\bigg|^{-} = \alpha(T_s - T_b) + (1-\varepsilon)\Delta H_1 R_1 + \sigma \varepsilon_{rad}(T_s^4 - T_b^4)$$

$$-D_{eff,j} \frac{\partial C_j}{\partial r}\bigg|^{-} = k_m (C_{j,s} - C_{j,b}) + R_1(1-\varepsilon)$$

Heat and mass transfer between burning char particle and fluidized bed are defined with relations of Baskakov *et al.* [6] and La Nauze *et al.* [7]. The numerical method of control volume was applied [8]. The char particle is divided in numerous spherical shells. The change of physical properties of the solid material, the heat and mass transfer characteristics, and temperature and gas concentrations are defined for each spherical shell. The evolution of the porosity of the char particle and the internal surface area of each spherical shell are described by the following relations (Bhatia & Perlmutter) [9]:

$$S = S_0(1 - X_{con})\sqrt{1 - \psi \ln(1 - X_{con})}$$

$$\varepsilon = \varepsilon_0 + X_{con}(1 - \Pi - \varepsilon_0)$$

It was assumed that porosity and internal surface area are function of the char particle conversion only. Additional effects on their values are expressed with two constants: the arbitrarily taken constant ψ and the ash content Π. The constant ψ defines the manner of internal surface change. If the value of ψ is greater than 2, the internal surface area increases in the initial period of conversion. As the value is greater, the increase of the internal surface area is greater. The parameter ψ in the model had values in the range of 5–20, depending on coal type.

Heat and mass transfer within porous char particle are very important processes and very sensitive on the characteristics of the porous medium. It was assumed that mass transfer by convection is negligible, since the process of char combustion or gasification is a low generating process, which could not produce remarkable pressure drop within the char particle. For that reason diffusion is the only significant mechanism of mass transfer within the porous char particle during combustion. In addition it was assumed that great pores (macropores) act as the main transport channels for reactant and reaction products, whereas their contribution to the heterogeneous reactions is negligible. On the other hand micropores and mesopores have no effect on macroscopic transfer, and they only act as a source of reaction products, that is, the active surface area of porous char is concentrated only in the micro- and mesopores. Knudsen diffusion, what is characteristic for mass transfer through the micropores, was also ignored. Therefore effective diffusion coefficient was defined by the corrected binary diffusion coefficient [10]:

$$D_{eff} = \zeta \cdot \varepsilon^2 \cdot D_{ij} \qquad \zeta = 1/3$$

Heat transfer through the porous burning char particle was assumed to be carried out by conduction and radiation. Thermal convection can be neglected for the same reason as the mass transfer by convection was ignored. With the analysis previously made [11] it was assumed that heat resistance of solid and gas phases in the porous medium of the char particle are in serial mode. The serial mode of heat resistances was drawn out from comparisons of mathematical model results obtained by using different relations for λ_{eff} with measurements of particle temperature and burning rate. An important role in total heat transfer through the char particle at temperatures over 973 K has radiation between pore walls. The radiation contribution to the total heat transfer within the porous particle is added to the conduction:

$$\lambda_{eff} = \frac{1}{(1-\varepsilon)/\lambda_s + \varepsilon/\lambda_g} + \lambda_{\mathrm{rad}}$$

Radiation conductivity is described by the following relation:

$$\lambda_{\mathrm{rad}} = 4 \cdot F \cdot \sigma \cdot L \cdot T^3$$

where F is a constant comprising porosity and emisivity, and L is related to the photon free path. For the model it was adopted that [12]:

$$F = \frac{\varepsilon}{1-\varepsilon} \quad \text{and} \quad L = d_p$$

Comparison of the model results with experimental ones

Experiments in a laboratory fluidized bed reactor were made (Fig. 1) to test the mathematical model. Three Serbian coals were chosen: anthracite Vrška Čuka, brown coal Aleksinac and lignite Kolubara. Prior to the experiments in the fluidized bed, chars were prepared by heating up at 900 °C in a laboratory oven in inert atmosphere. Then the char particles were cooled and preserved from getting wet. Proximate and ultimate analyses of the investigated chars as well as some their physical properties are given in Appendix. The dry and cold char particles having room temperature were introduced in to the hot fluidized bed reactor. With a thermocouple in the bed and a gas sample probe above the bed, the bed temperature and gas concentrations of O_2, CO, and CO_2 were continuously measured during the experiments. Since the mathematical model considered the fluidized bed without chemical reactions, experiments had to be planed in such

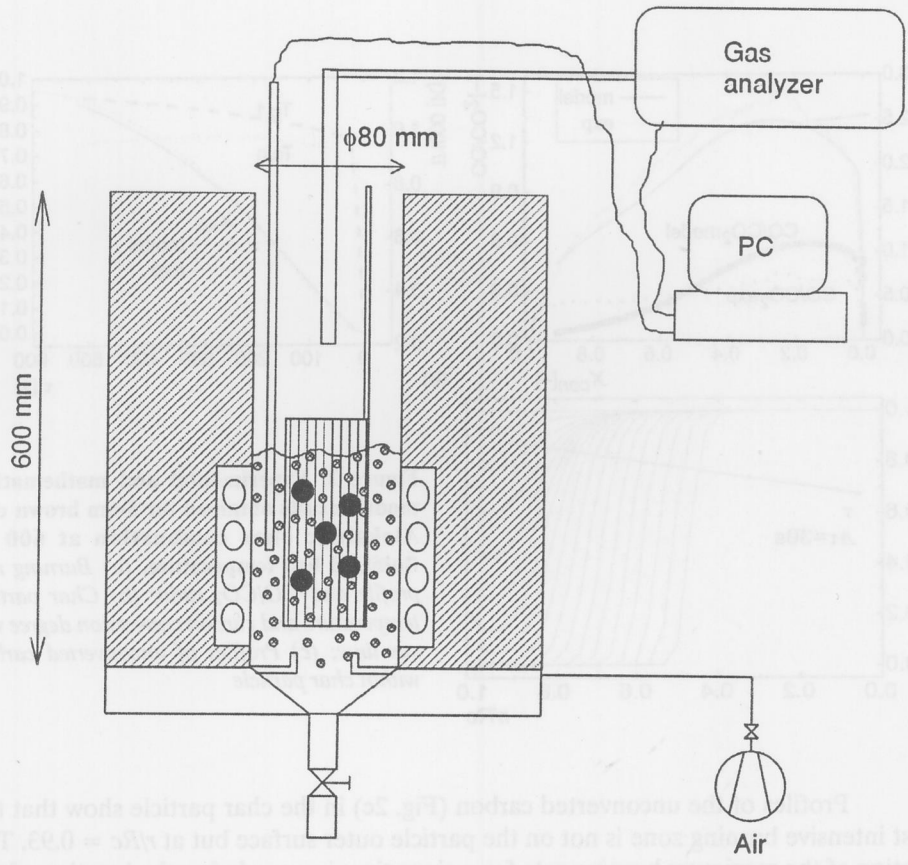

Figure 1. Experimental setup for char combustion investigation in fluidized bed

a way that the experimental results obtained could be used for testing the mathematical model of char particle combustion. Therefore the bed temperatures were chosen to avoid significant oxidation of CO, that is the bed temperature was in the range between 500 and 700 °C.

The bed material was silica sand with particles of 0.5 mm in diameter, while the bed height was 80 mm. Fluidizing velocity was in the range 0.45–0.55 m/s. For validation of the mathematical model with experimental results several tests were made: (1) burning rate versus conversion degree, (2) change of conversion degree versus time (3) CO/CO_2 ratio during combustion.

The burning rate profile of char particles which originated from Aleksinac brown coal implies that combustion was in transient regime, but mostly under kinetic control at 600 °C fluidized bed temperature (Fig. 2a). The maximum burning rate of the char particle was achieved at the overall conversion degree of 0.25 . The combustion rate of the char particle was so intensive that mass transfer within the char particle could not provide enough oxygen for burning in the whole particle volume.

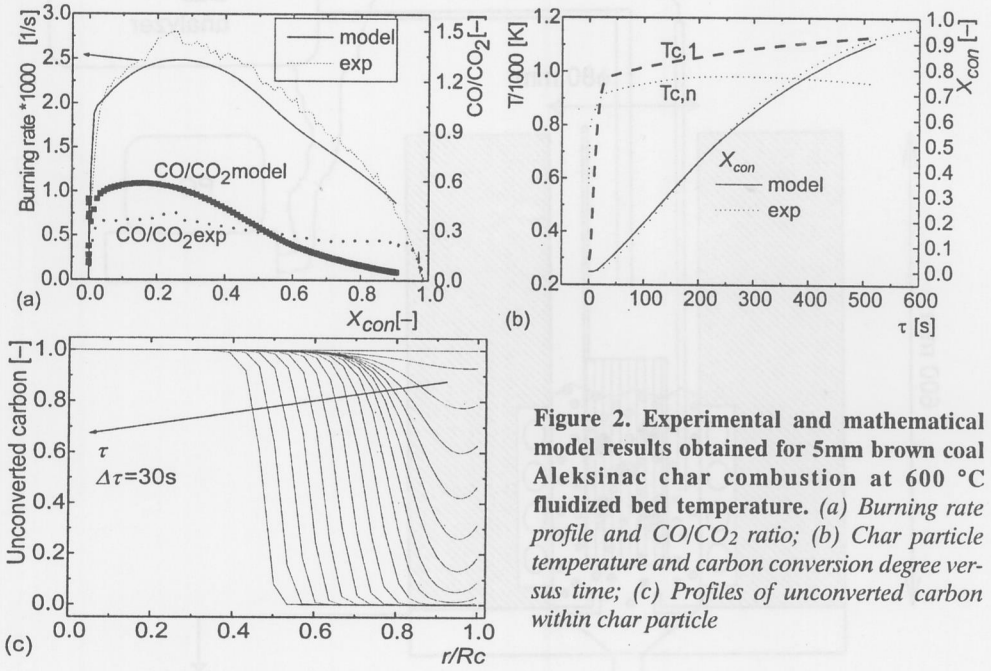

Figure 2. Experimental and mathematical model results obtained for 5mm brown coal Aleksinac char combustion at 600 °C fluidized bed temperature. *(a) Burning rate profile and CO/CO_2 ratio; (b) Char particle temperature and carbon conversion degree versus time; (c) Profiles of unconverted carbon within char particle*

Profiles of the unconverted carbon (Fig. 2c) in the char particle show that the most intensive burning zone is not on the particle outer surface but at $r/Rc = 0.93$. The location of the maximum burning rate for a given time is regarded as the location of the greatest change of the local carbon conversion degree, which can be recognized in figure

of unconverted carbon profiles (Fig. 2c). Besides, the profiles show that the particle surface reached conversion degree of 0.8 after 180s, when the overall conversion degree of the char particle was greater than 0.4. After initial burning period, in which the particle was burning up to $r/R_C = 0.6$ (Fig. 2c), the particle temperature was increased, so the pore diffusion rate limited the further increase of the particle burning rate. During a great part of the burning time (up to $X_{con} = 0.5$) the char particle was burning up to $r/Rc = 0.7$, which means that almost 65% of the volume of the char particle was simultaneously burning.

The temperature at the particle outer surface was increasing during the first period of combustion, up to the overall conversion degree of 0.5 (Fig. 2b). This conversion degree corresponds to the moment when all carbon at the particle surface was totally converted. After that conversion degree the temperature of the particle outer surface was decreasing. Temperature in the center of the particle was constantly increasing. For the conversion degree of 0.5 the difference between the temperatures of the center and the outer surface of the particle was around 80 K. As the combustion process proceeded, the temperature difference increased, reaching the maximum value of more than 150K at the end of the combustion process (Fig. 2b).

Comparison of the experimental burning rate with burning rate profiles obtained by the model and the change of conversion degree in time, gives that there are no significant discrepancies among them. There is some discrepancy in the final CO/CO_2 molar ratio in gaseous products. In the initial burning period up to $X_{con} = 0.55$, it seems that oxidation rate of CO predicted by the model was not enough high, or the primary CO/CO_2 ratio, the ratio formed in the heterogeneous reaction (reaction 1) was overestimated. While for the period of burning after reaching the conversion degree of $X_{con} = 0.55$, it seems as the predicted oxidation rate of CO was too high.

Using burning rate profiles one can detect the combustion regime of char particles. Figure 3. shows the change of burning rate profiles with reaction temperature and burning particle size changes of the Aleksinac brown coal. As the reaction temperature increases the maximum of burning rate shifts toward the initial period of combustion. That characteristic was detected by measurement during the experiments and pre-

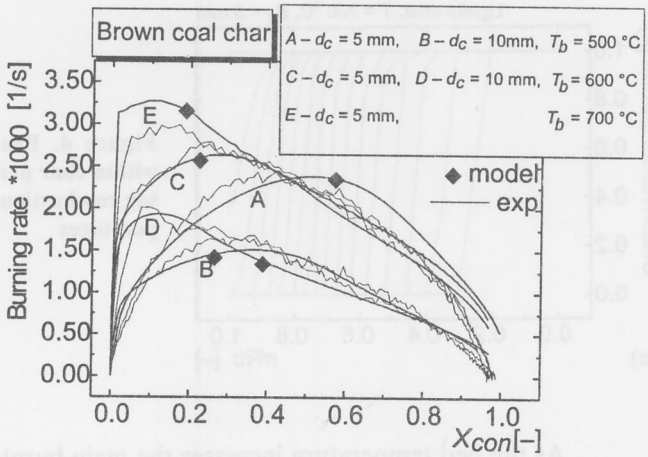

Figure 3. Burning rate profiles for the Aleksinac brown coal for different bed temperatures and char particle sizes (experimental and model results)

dicted with the mathematical model also. Besides, the increase of the char particle size leads to the shifting the maximum burning rate towards the initial period of combustion. Figure 3 shows that the model generally predicts the burning rate profiles as they were experimentally obtained.

During the initial stage of combustion at 500 °C bed temperature, the char particle of the Kolubara lignite was burning over its whole volume (Fig. 4a). That means that the combustion of the char particle was in the kinetic regime. As the char particle burnt, its temperature increases, the consumption of oxygen also increases in parts of the char particle close to the outer surface, and the internal mass transfer rate was not intensive enough to provide oxygen at the center of the char particle.

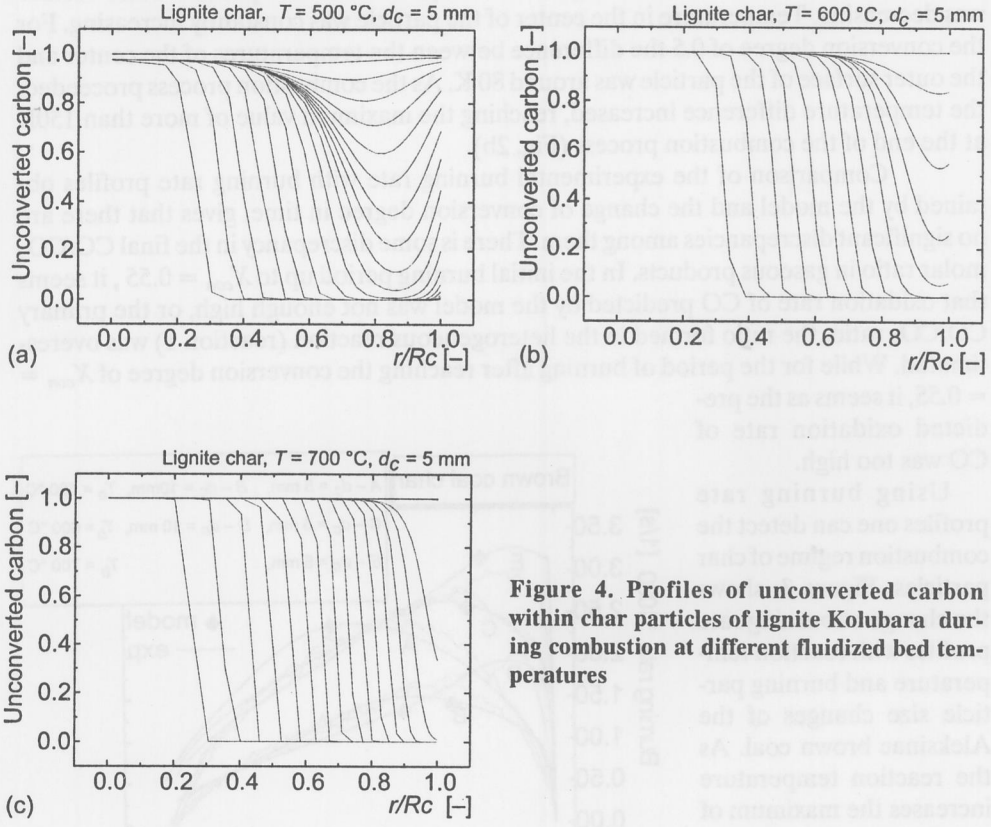

Figure 4. Profiles of unconverted carbon within char particles of lignite Kolubara during combustion at different fluidized bed temperatures

As the bed temperature increases the main burning zone of the char particle shifts toward the outer surface. The profiles of the unconverted carbon shows that combustion can occur in a relatively narrow or a wider zone, depending on temperature. The burning rate along the radius of a char particle was not uniform.

At 500 °C, the burning zone was from $r/Rc = 0.5$ up to $r/Rc = 1$ (Fig. 4a), which means that almost 90% of the volume of the char particle was simultaneously burning. This implies that the combustion process was in kinetic regime. In the first 30 seconds of burning, the char particle was burning throughout its volume. In that period the combustion zone was the whole particle but with a low burning rate. After that initial period, the diffusion rate was not sufficiently high, comparing to the burning rate, to enable presence of oxygen in the particle center, hence the combustion zone was shorted.

As the bed temperature increases, the burning zone of the char particle was shifted, toward its outer surface, and the burning zones became narrower. At 600 °C bed temperature, the burning zone of the char particle was from $r/Rc = 0.7$ till to $r/Rc = 1$, with the maximum burning rate at $r/Rc = 0.97$. Obviously, pore diffusion had a significant influence on the combustion process, although almost 65% of the volume of the char particle was burning (Fig. 4b).

At 700 °C (Fig. 4c), the maximum burning rate was just at the outer surface of the char particle and the burning zone was very narrow. In this case the combustion process of the char particle was under diffusion control. At bed temperatures 500 °C, 600 °C and 700 °C the locations of the maximum burning rate were at $r/Rc = 0.85$, 0.97. and 1.0 respectively.

Conclusions

The mathematical model developed for prediction of the behaviour of char particles during combustion in a fluidized bed. The mathematical model was tested using experimental results of char combustion in the fluidized bed reactor. Burning rate profiles obtained by the model were very alike or almost equivalent to the experimental ones for different sizes of char particles and different bed temperatures.

Under which combustion regime the char particle would burn depends on three main mechanisms. It depends on mass transfer, which defines the oxygen concentration in the pores, on heat transfer, which defines the temperature distribution within char particle, and on kinetics of chemical reactions. All these three mechanisms play an important role in defining the burning rate of a char particle. Their importance in defining the burning rate of a char particle depends on ambient conditions: temperature and oxygen content and on pore structure of the char particle. At low bed temperature (500 °C) the char particle was mostly under kinetic control, whereas at 700 °C the combustion regime was under internal diffusion control. For this temperature range the model predicted, in accordance with the experiment, changes of combustion regimes. It implies that all three main mechanisms, namely mass and heat transfer and reaction kinetics, are very well balanced.

The mathematical model estimates solid carbon and gas concentration profiles within char particles, which are in accordance with the predicted combustion regimes. Well defined burning rates and overall behavior of burning particles give good starting point for prediction of combustion process in real furnace.

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Nomenclature

A_1, A_2 – preexponential factors for chemical reactions 1 and 2
E_1, E_2 – activation energies for chemical reactions 1 and 2
C_j [mol$_j$/m^3] – concentration of j-th species
C_p [J/m^{3}K] – specific heat capacity of porous char particle
C_{pj} [J/mol$_j$K] – specific heat capacity of gas species
d [m] – diameter
D_{ij}, D_{eff} [m^2/s] – binary and effective diffusion coefficients
$\Delta H_{1,2}$ [J/mol] – heat of reactions 1 and 2
k_m [m/s] – mass transfer coefficient between char particle and fluidized bed
N_j [mol$_j$/m^{2}s] – molar flux
R_g [J/molK] – universal gas constant
R_1 [molC/m^{2}s] – reaction rate for 1st chemical reaction
R_2 [molC/m^{3}s] – reaction rate for 2nd chemical reaction
r, R [m] – radius
S, S_m [m^2/m^3] – specific surface area of porous char particle
T [K] – temperature
$T_{c,1}$ [K] – char temperature at the particle center
$T_{c,n}$ [K] – char temperature at the particle surface
X_{con} [–] – char particle conversion degree

Greek symbols

τ [s] – time
α [W/m2K] – convective heat transfer between char particle and fluidized bed
λ [W/mK] – heat conductivity
ε [–], [%] – porosity
ε_{rad} [–] – emisivity
$\nu_{k,j}$ [–] – stoichiometric coefficient in reaction k of species j
ρ [kg/m^3] – density of solids
ρ_ε [kg/m^3] – density of porous particle
σ [W/m^{2}K^4] – Stefan-Boltzman constant
ψ – model parameter for porosity evolution
Π [–] – ash content
ζ – coefficient of pore torousity

Index

b	– bed	o	– initial status	H_2O	– water
c	– char particle	p	– pore	j	– gas species, O_2, CO_2, CO, H_2O
eff	– effective	rad	– radiation		
g	– gas	O_2	– oxigen		
s	– surface, solid	CO	– carbonmonoxide		

Appendix

Table 1. Proximate and ultimate analysis of the investigated chars

	Kolubara lignite	Aleksinac brown coal	Vrška Čuka anthracite
Proximate analysis			
Moisture [%]	–	–	–
Ash [%]	25.7	21.8	7.4
Cfix [%]	74.3	78.2	92.6
Volatiles [%]	–	–	–
Heating value [MJ/kg]	23.7	24.3	30.5
Ultimate analysis			
C [%]	68.4	68.2	89.0
H [%]	1.0	1.2	1.4
S [%]	–	–	–
N [%]	0.3	0.5	0.4
O (%)	4.6	8.3	1.8

Table 2. Measured and adopted char particle parameters for the model

	Kolubara lignite	Aleksinac brown coal	Vrška Čuka anthracite
Measured values			
S_m (BET, N_2) [m²/g]	120	63	2
ρ [kg/m³]	1850	2280	1780
ε (Hg) [%]	49.4	39.8	14.7
ρ_ε [kg/m³]	850	1420	1540
Adopted values			
S_m [m²/g]	150–175	78–95	5.6–6.7
ε [%]	51–58	41–48	10–12

Table 3. paraheat values of the reactions 1 and 2

Reaction 1 ($C + O_2 \rightarrow x \cdot CO + y \cdot CO_2$)	Reaction 2 ($CO + O \rightarrow CO_2$)
$\Delta H_1 = x \cdot \Delta H_{CO} + y \cdot \Delta H_{CO_2}$	$\Delta H_2 = \Delta H_{CO_2} - \Delta H_{CO} = -282965$ [J / molC]
$E_1 = 179400$ [J/mol] (ref. [13])	$E_2 = 55695$ [J/mol] (ref. [14])
$A_1 = 254160$ [molC/m²s] (ref. [13])	A_2 [m³/mol·s]
	– anthracite Vrška Čuka: $1.43 \cdot 10^4$ (ref. [14])
	– brown coal Aleksinac: $7.15 \cdot 10^5$ (estimated)
	– lignite Kolubara: $7.43 \cdot 10^5$ (estimated)

$\Delta H_{CO_2} = -393505$ [J / molC] $\Delta H_{CO} = -110540$[Jmol / C]

Some physical properties of chars and modeling parameters

$\lambda_s = 1.89$ [W/mK] $C_{p,s} = 1800$ [J/kg]

$\varepsilon_{rad} = 0.8$ $C_{H_2O} = 1\%$

$C_p = (1 - \varepsilon)\rho C_{p,s} + \varepsilon C_{p,g}$

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