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Structural evolution of biomass char and its effect on the gasification rate

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HIGHLIGHTS

- A comprehensive model was developed to describe the evolution of biomass char structure.
- An effectiveness factor was used to account for the intra-particle chemical and physical processes.
- The effect of the structural evolution of the multi-pore structure on biomass char reactivity was analyzed.
- The multi-pore model yields results in satisfactory agreement with experiments.

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ABSTRACT

The evolution of char porous structure can affect the conversion rate of the char by affecting the intra-particle transport, especially in the zone II conversion regime. A multi-pore model based on the capillary pore theory is developed to take into account different conversion rates for pores with different radii. The model is valid for biomass chars produced under relatively low heating rates, when the original beehive structure of the biomass is not destroyed during the pyrolysis stage. The contribution of different pores with different radius is taken into account using an effectiveness factor presented for each pore radius with respect to different reactions. As the char conversion proceeds, the pore enlargement increases the contribution of micro-pores; consequently the effective surface area will increase. The increase in the effective surface area leads to an increased reactivity of char during the entire conversion process. This model is used to analyze the steam gasification process of biomass char of centimeter sizes. The results from the present multi-pore model are in better agreement with experimental data than those from a corresponding single pore model. Since the multi-pore model accommodates the detailed intra-particle transport, it is a useful basis toward developing a more predictive model for biomass char gasification.

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1. Introduction

The concern on sustainable development and environment protection calls for fossil fuel independent energy systems in the future [1]. Gasification of biomass and production of high quality biogas [2] and bio-oil [3,4] from renewable sources are viable options to achieve such a goal. In biomass thermochemical conversion processes, the heterogeneous reactions of char are often the rate controlling processes in the overall conversion of biomass, in particular under low heating rate and/or large particle conditions. The heterogeneous char reactions occur at the surface of porous

structure of the char. For the regime where the intra-particle mass transfer and chemical kinetic rates have comparable time scales (which is known as zone II reaction regime), the evolution of porous structure is proven to be important. There are different theories developed for representing a porous medium, such as discrete models and capillary models. Models based on the capillary theory are by far the most commonly used models in this field [5]. One of the well-known models in capillary models is the *random pore model* (RPM) developed by Bhatia et al. [6,7] and Gavalas [8]. The RPM assumes cylindrical capillaries randomly located and oriented with any distribution of pore radii. The surface area of the structure may increase due to the surface reactions or decrease due to different capillary overlapping. The specific surface area (S_c) is related to the conversion degree of carbon structure based on the following equation,

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Nomenclature

A	pre-exponential factor, 1/s or m/s K	ε	void fraction
C_p	specific heat capacity, J/kg K	η	effectiveness factor
D_{eff}	effective diffusion coefficient, m ² /s	ρ	density, kg/m ³
d_{ch}	characteristic particle length, m	ν_i	stoichiometric coefficient
E	activation energy, kJ/mole	Φ	Thiele module
h_c	convective heat transfer coefficient, W/m ² K	ψ	structural parameter in random pore model
h_m	mass transfer coefficient, m/s		
k	kinetic rate constant, 1/s		
k_{eff}	effective thermal conductivity, W/mK		
M	molecular weight, kg/mole		
Nu	Nusselt number		
r	radial position, m		
r_p	pore radius, m		
\mathfrak{R}_c	char reaction rate, kg/m ³ s		
\mathfrak{R}_{Heat}	source term in energy equation, J/m ³ s		
Sh	Sherwood number		
S_c	char specific surface area, m ² /m ³		
T	temperature, K		
u_g	gas velocity, m/s		
Y_i	gaseous species mass fraction		
X_c	conversion		

Subscripts

c	char
$cond$	conduction
$conv$	convection
i	pore size group
int	intrinsic
j	species
k	reaction index
Kn	Knudsen diffusion coefficient
n	reaction order
0	initial state
∞	ambient state far from the particle surface

$$S_c = S_{c,0}(1 - X_c)\sqrt{1 - \psi \ln(1 - X_c)} \quad (1)$$

where $S_{c,0}$ is the initial specific surface area, X_c is the carbon conversion ($X_c = 0$ for zero conversion and $X_c = 1$ for a complete conversion) and ψ is a structural parameter depending on the carbon structure. The structural parameter can be determined using the original pore size distribution; however, it is usually estimated by fitting to experimental data.

For coal, RPM predicts a maximum in the specific surface area and hence in the char reactivity as the conversion proceeds. The specific surface area first increases as the pore radius is enlarged due to the ongoing reactions and at a certain point, starts to decrease as the pores merge and coalesce. For biomass, on the other hand, some experimental evidence shows a constant or a continuously increasing trend for the specific area during the conversion. Dasappa et al. [9] measured the specific surface area of different biomass chars at various conversion degrees, up to 80% conversion, using the BET technique. The specific surface area varied weakly as the conversion proceeded. In another experiment, Lussier et al. [10] reported a constant adsorbed hydrogen concentration between 0.5% and 40% conversion of Saran char. A continuous increase in the “reactive” surface area was reported in Ref. [11] for a beech wood char.

The reason for these different behaviors can be the different origin of the coal and the biomass char. Coal is formed under pressure and heat, which causes the structural order of the original source to be destroyed during the process of coal formation. Biomass char usually has two orders of magnitude greater specific surface areas compared with coal. Hence, wood chars present a remarkably regular structure similar to a beehive network, whereas the coal chars are characterized by highly random structures due to their origin [9]. Because of these differences, the theories originally developed for evolution of a pore structure of coal during the conversion may fail for biomass [12].

Because of the insufficient experimental data and dependency of char structure on the condition that the char is produced, it is difficult to draw a general conclusion on the evolution of the surface area. In addition, other complexities such as the formation of cracks and the presence of ash bring additional difficulty to the problem. Some simple models have been used in the literature,

one of which assumes a constant specific surface area during the conversion [13,14]. The model produces reasonable results for high-temperature oxidation where the boundary diffusion plays a more important role and the evolution of the internal surface area is of less importance. Mermoud et al. [12] developed a simple model based on the assumption that the surface area increases in such a way that the conversion rate of a gasifying char remains constant. This model showed good agreement with the few experimental data that were obtained for up to 60% conversion, c.f. Fig. 1. In another study, Dasappa et al. [9] argued that assuming a monotonous increasing pore size without any merging and coalescing is more relevant for the ordered structure of wood than using the RPM that was originally developed for coal. This assumption implies that the specific surface area increases continuously. This model is based on the classical capillary tubes model and relates the specific surface area to the porosity and pore radius r_p by the following equation,

$$S_c = \frac{2\varepsilon}{r_p} \quad (2)$$

where ε is the void fraction of the char particle, also known as the porosity.

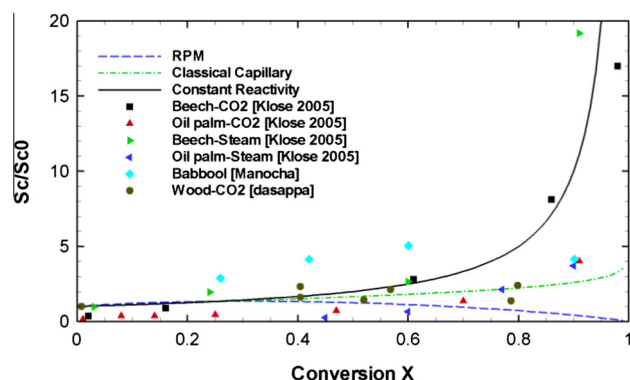


Fig. 1. Evolution of specific surface area of some biomass chars at different conversion degree.

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