FORMULATION OF A SINGLE CHAR PARTICLE MODEL FOR NAPHTHALENE REMOVAL IN BIOMASS GASIFICATION

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ABSTRACT

Tar removal in biomass gasification systems is one of the key challenges to overcome for the successful commercialization of this technology. This study focused on tar conversion over the active char particle besides the simultaneous carbon conversion of the char particle. In the presented model, the char particle was assumed a sphere surrounded by a gas film in a bulk gas composition of naphthalene, nitrogen, steam, hydrogen, carbon monoxide, carbon dioxide and methane. The model treated the heterogeneous gasification reaction kinetics, external and internal mass transfer and changing particle properties during gasification. In addition, the effect of different parameters on naphthalene and carbon conversion will be investigated. The investigated parameters were particle size, temperature and time on stream. The gas and solid mass balance equations besides the energy balance equation are to be solved in the radial direction using a set of initial and boundary conditions.

Keywords: gasification; biomass char; particle model; catalyst; naphthalene; tar.

1. INTRODUCTION

There is a global trend to reduce the dependence on fossil fuels by creating a diversified energy portfolio. This portfolio includes a certain share of renewable energies that varies according to the available renewable sources. Biomass is considered one of these promising renewable energy sources because of its abundance as it provides 14% of the global energy consumption [1], and its potential to reduce global CO_2 emissions [2]. There are many technologies available to utilize the energy content in biomass. Among these technologies, biomass gasification is considered one of the preferable options due to its higher efficiency in power production than the combustion technology. In addition, it can be used in the production of chemicals and fuels. Nevertheless, this technology faces certain technical obstacles that need to be resolved in order to be successfully commercialized. Tar removal is considered the most challenging technical obstacle.

Tars is defined as a generic term comprising all organic compounds present in the producer gas excluding gaseous hydrocarbons (C_1-C_6) and benzene [3]. Tar can be removed by various methods among which hot gas cleaning is considered the most economical one. This method applies a catalyst under a temperature that essentially matches that of the gasifier [4]. There has been extensive research in literature on catalysts for tar removal. However, there is a limited research that focuses on biomass char despite its promising potential as a catalyst for tar removal.

Because of the growing interest in biomass gasification, several models have been developed in order to explain and optimize the gasification process. In addition, models for tar reduction over different catalysts have been proposed. However, limited modeling has been done on tar reduction over biomass char as a catalyst downstream the biomass gasifier. Several char particle gasification models are available in literature. Most of these models are concerned with the rate of gasification of a single particle [3, 4] and the evolution of the particle pore structure during gasification [5]. Raza et. al. [5] developed a simplified particle model to simulate the gasification of char particle in the gasifier. They used the carbon dioxide as the gasification agent that reacts with the char. Wang et. al. [6] developed a generalized model for the prediction of single char particle gasification dynamics, accounting for a chemical reaction with multicomponent mass and heat transfer, structure evolution and peripheral fragmentation. The importance of the model presented in this paper is the focus on tar conversion over active char besides the simultaneous carbon conversion in the char particle.

In a previous work done by Abu El-Rub et. al. [7, 8], biomass char showed high activity for tar removal under fixed bed conditions. It was found that the pore structure and mineral content of the char particle are the key elements for the char activity. In order to get a better understanding for the effect heat and mass transfer on tar reduction with char, a single char particle model was developed. The model was developed for naphthalene reduction using a porous char particle in an environment of N_2, H_2O, H_2, CO, CO_2 and CH_4. The investigated parameters were particle size, temperature, and time on stream. This paper presents only the single char particle model formulation. In the next stage, the results of the single char particle model will be produced and further extended to a fixed bed reactor model that will be validated with experiments.

2. MODEL DEVELOPMENT

In this model, the char particle was considered as a sphere surrounded by a gas film in a bulk producer gas with a constant composition. The model treats the intrinsic gasification reaction kinetics, external and internal mass transfer, and changing particle properties during gasification. The model reference conditions are given in Table 1.

Assumption

The following assumptions were incorporated in the model:

1) The char particle is spherically symmetric. This allows a simplified one-dimensional solution to the species conservation equations.
2) The char particle has a constant external diameter. This assumption approximates the findings presented in a previous work conducted by Abu El-Rub et. al. [9]. It was found that the char conversion reaction does not proceed uniformly in the char particle and is not a shrinking particle reaction. Hence, the reaction with the char is somewhere in between.
3) The tar is represented by naphthalene as a model tar component. This assumption is justified by the comparison made in a previous work [9] between naphthalene and real tar conversion over char. It shows that naphthalene is a good representative for real tar.

Table 1 Reference values of the model parameters

Parameter	Symbol	Value
Pressure	P (Pa)	$1.01 \cdot 10^5$
Tar	$C_{10}H_8$	Naphthalene
Bulk temperature	T (°C)	850
Gas residence (space) time	τ (s)	0.3
Superficial gas velocity	u_o ($m \cdot s^{-1}$)	0.08
Char particle size	d_p (μm)	600
Standard gas composition (volume fraction)		
	CO	13 %
	CO_2	11 %
	H_2O	11 %
	H_2	6 %
	CH_4	4 %
	$C_{10}H_8$	0.2 %
	N_2	Balance

4) An overall first order kinetic rate of naphthalene conversion was assumed. This assumption is justified for two reasons. Firstly, the sensitivity of naphthalene conversion on the char for different gas compositions is low [9]. Secondly, the amount of naphthalene (< 0.5 vol. % of the producer gas) compared with the other gases is small. Therefore, in the kinetics of naphthalene conversion, the other gases were assumed to be in excess and lumped in the global pseudo-first order kinetics: $(\mathbf{k} = \boldsymbol{k'} \cdot \boldsymbol{C_{H_2O}^{a}} \cdot \boldsymbol{C_{CO_2}^{b}} \cdot \boldsymbol{C_{H_2}^{c}})$.

Naphthalene can be converted by parallel reactions with CO_2, steam and to a less extent with H_2. The extent of these reactions is not known. Therefore, for the mass balance of the different species, naphthalene was assumed to be converted because of its reaction with steam. This assumption is justified for the reasons given in the previous assumption.

Table 2 Modeled tar and char gasification reactions

Reaction	***Rate of reaction ($kJ \cdot mol^{-3} \cdot s^{-1}$)***	***Reference***
$C_{10}H_8 + 10H_2O \rightarrow 10CO + H_2$	$-r_1 = \eta \cdot 10^{-4} e^{-61{,}000/(RT)} C_n \cdot a$	[7]
$C + H_2O \rightarrow CO + H_2$	$-r_2 = \eta \cdot 2.62 \cdot 10^8 e^{(-\frac{237{,}000}{RT})} \cdot \left(C_{H_2O}\, RT\right)^{0.57} \cdot a \cdot \frac{w_c \rho_c}{M_c}$	[10, 11]
$C + CO_2 \rightarrow 2CO$	$-r_3 = \eta \cdot 3.1 \cdot 10^6 e^{(-\frac{215{,}000}{RT})} \cdot \left(C_{CO_2}\, RT\right)^{0.38} \cdot a \cdot \frac{w_c \rho_c}{M_c}$	[10, 11]
$C + 2H_2 \rightarrow CH_4$	$-r_4 = \eta \cdot 9.14 \cdot 10^{-10} e^{(-\frac{149{,}050}{RT})} \cdot \left(C_{H_2} RT\right) \cdot S_o \cdot a \cdot w_c \cdot \rho_c$	[12]
$H_2O + CO \rightleftharpoons H_2 + CO_2$	$r_5 = 2.78 \cdot 10^3 \cdot e^{(-\frac{12{,}5060}{RT})} C_{H_2O} C_{CO} - 1.05 \cdot 10^5 \cdot e^{(-\frac{-45{,}466}{RT})} C_{H_2} C_{CO_2}$	[13]

Kinetics

The main reactions and their kinetic rate equations that occur in the char particle are presented in Table 2. The considered kinetic constants are only those valid for producer gas in the temperature range of 700 to 900 °C at atmospheric pressure.

Steam reforming reaction of naphthalene is an endothermic and heterogamous reaction. The char catalyzes this reaction. The kinetics proposed in a previous work [7] was used here.

Steam and dry gasification reactions of char are endothermic and heterogeneous reactions. The gasification reactions of biomass char are not studied extensively in literature as coal char reactions. The nth order kinetics found by Barrio et al. [10, 11] were adopted here since they were estimated for biomass char (birch wood) in the temperature range 750 to 950 °C. The type of biomass and gasification temperature range is close to our experimental conditions. The overall rate of the steam gasification reaction is expressed in grams of carbon gasified per second per gram of carbon present. Similarly, the overall rate of the dry gasification reaction is expressed in grams of carbon gasified per second per original gram of carbon present. It was found that the rate of the char gasification is not constant but decreases with time [9] because of the changing surface area of the char particle with time. Thus, the effect of the relative internal surface area of the char particle is included in the model for the gasification reactions of the carbon. Modifications were made on the steam and dry gasification equations in order to be compatible with our findings and the model units. The units of the equations were converted to ($kmol \cdot m^{-3} \cdot s^{-1}$): multiplying by the concentration of the char (C_C). The concentration of the char is expressed in terms of the char particle density ($C_c = w_c \cdot \rho_c / M_C$). The gas component pressure was expressed in terms of concentration ($P_i = C_i \cdot R \cdot T$). The rate constant is multiplied by the relative surface area (a) which is function of carbon conversion (X_c).

The hydrogasification reaction is an exothermic reaction. The first order kinetic data obtained for coconut chars is used where the kinetic constant is based on the internal surface area available for the reaction [12]. Modifications were made on the hydrogasification equation to be compatible with our model. The rate equation was based on a unit volume of the particle by multiplying with ($S_o \cdot \rho_C \cdot w_b$) and the rate constant is multiplied by the relative surface area (a). The pressure of H_2 was based on concentration ($P_i = C_i \cdot R \cdot T$).

The water gas shift reaction is a homogeneous reaction and exothermic in the direction of the forward reaction. This reaction is kinetically limited as it is not catalyzed by the char. The forward and backward reactions are used in the model mass balance calculations [13].

Mass and Energy Balance

The mass and energy balance equations were solved simultaneously to obtain the temperature gradient inside the particle. In addition, the effect of mass transfer were calculated at different temperatures, particle sizes and times on stream.

The species concentration and temperature profiles inside the particle were estimated by solving mass and energy balance equations as given in Table 3.

Table 2 Summary of the equations used to model the tar and char conversion of a single char particle

Type of equation	Equation	Reference
Gas mass balance	$\frac{\partial C_i}{\partial t} + \frac{1}{r^2}\frac{\partial}{\partial r}\left[D_e r^2 \frac{\partial C_i}{\partial r}\right] = R_i$ $\sum_{i=1}^{n} x_i$	[15]
Solid (carbon) mass balance	$\frac{\partial \rho_c}{\partial t} = M_c R_c$	[15]
Energy balance	$\frac{\partial}{\partial t}\left[\rho_{p,t}(1-\varepsilon)C_{p,c}T\right] = \frac{1}{r^2}\frac{\partial}{\partial r}\left[\lambda_e r^2 \frac{\partial T}{\partial r}\right] - \sum -r_j\left(-\Delta H_j\right)$	[16]

Table 3 Initial and boundary conditions of mass and energy balance equations

Condition	Mass balance		Energy balance
	Gas phase	Solid phase	
$t = 0$	$C_i = 0$	$\rho_c = \rho_{c,o}$	$T = T_b$
$r = 0$	$\frac{\partial C_i}{\partial r} = 0$	-	$\frac{\partial T}{\partial r} = 0$
$r = R_o$	$D_{e,i}\frac{\partial C_i}{\partial r} = k_{g,i}\left(C_{i,b} - C_{i,s}\right)$	-	$\lambda_e \frac{\partial T}{\partial r} = h(T_s - T_b)$

The convection inside the particle is assumed to be negligible [14]. The gas mass balance equations represent mass balances for the gas components naphthalene, H_2O, H_2, CO, CO_2, and CH_4. Further, a total mass balance (including the mentioned components and N_2) can be formulated where the sum of the mole fractions of the gas components is equal to one. In addition, the carbon mass balance of the char particle is given. The carbon concentration in the particle was expressed in terms of carbon density. On the other hand, the temperature profile inside the char particle can be found from the energy balance equation.

The initial and boundary conditions applied to solve these equations are given in Table 4. The initial concentration of all the gaseous components inside the particle is zero. Further, the derivative of the concentration of the gas component in the center of the particle is zero because of the particle symmetry. The rate of diffusion at the surface of the particle is equal to the rate of mass diffusion in the gas film surrounding the particle. On the other hand, the initial temperature inside the particles was assumed similar to the bulk temperature in the reactor as, during the experiments, char particles were heated inside the reactor until they reach the desired temperature. At any time, the temperature gradient in the center of the particle is zero because of the particle symmetry. At the surface of the particle, the heat flux by conduction in the particle is equal to the heat flux by external convection.

Internal and External Diffusion

To estimate the effect of the pore diffusion resistance on the overall reaction rate, the effectiveness factor was used. It is defined as the ratio of the actual reaction rate including the pore diffusion resistance to the reaction rate excluding pore diffusion resistance [17]. On the other hand, the Biot number is used to estimate the effect of the external diffusion [15]. This number represents the ratio between the external and the internal diffusion. The correlations are given in Table 5.

Table 4 Internal and external mass transfer correlations

Property	Correlation	Reference
Effectiveness factor	$\eta = \frac{1}{\phi_T}\left(\frac{1}{tanh\,(3\phi_T)} - \frac{1}{3\phi_T}\right)$	[14]
Thiele Modulus	$\phi_T = L\sqrt{\frac{(n+1)}{2}\frac{kC_s^{n-1}}{D_e}}$	[17]
Sherwood number	$Sh_i = \frac{k_{g,i} \cdot d_p}{D_i}$ $Sh_i = \frac{\phi_s}{6(1-\varepsilon_b)\xi}\frac{u_o d_p}{D_i}$	[18]
Biot number	$Bi_m = L\frac{k_g}{D_e}$	[15]
Kinetics control	$\phi_T \ll 1$	[16]
Internal diffusion control	$\phi_T \gg 1 \quad and \quad Bi_m \gg \phi_T$	[16]
External diffusion control	$\phi_T \gg 1 \quad and \quad Bi_m \ll \phi_T$	[16]

Correlations and parameters estimation

The physical properties and the correlations used for estimating parameters used in the model are presented in Table 6.

Numerical Simulation

The model was solved numerically using the chemical engineering module within the software package Comsol [28]. The gas and solid mass balance equations and the energy balance equation were applied in the software as three application modes in one dimensional geometry. The convective and diffusion mode was used for solving the mass balance equations. On the other hand, the convective and conduction mode was used for solving the energy balance equation.

In each application mode, the dependent variables were set. These variables represent the local concentration of the gas species inside the particle. For the gas mass balances, the following dependent variables were set: H_2O, H_2, CO, CO_2, CH_4, and Naphthalene, respectively. Further, a total mass balance equation was used where the sum of the mole fractions of the gas components is equal to one.

For the char mass balance, the apparent density of the char particle was set as the dependent variable. For the energy balance, the particle temperature was set as the dependent variable. For the three balances (application modes), the independent variable was set as the particle radius. The three equations were solved together using a time-dependent solver. The applied time step was the default value of the solver. The geometry of the model is 1-D divided into smaller intervals (or mesh elements). The maximum element size was $1 \cdot 10^{-5}$.

The rate equations of the steam and dry gasification of the char contain powers. These powers were a source of instability because powers of negative numbers can occur when the concentration is close to zero. The problem was overcome by using absolute values for the concentrations raised to powers.

Table 5 Correlations and values used in the single char particle model

Property	**Correlation**	**References**
Effective diffusion coefficient	$D_e = \varepsilon^2 D$	[14, 19]
Particle voidage	$\varepsilon = 1 - \frac{\rho_p}{\rho_{p,t}}(1 - \varepsilon_o)$	[20]
	$\varepsilon_o = 0.75$	
Relative surface area of char particle	$a = \frac{S_p}{S_{p,o}}$	[21]
	$a = -4.361X_C^3 + 3.752X_C^2 - 0.391X_C + 1.0$	[9]
Carbon conversion	$X_C = 1 - \frac{m}{m_o}$	
Nusselt number	$Nu = \frac{hd_p}{\lambda_g}$	[18]
	$Nu = \frac{\phi_s}{6\xi(1 - \varepsilon_b)} \frac{u_o d_p \rho_g C_{p,g}}{\lambda_g}$	
Rates of chemical production/consumption of gas component	$R_i = \sum \alpha_{ij} r_j$	[15]
	$R_n = -r_n$	
	$R_{H_2O} = -r_2 - r_5 - 10r_1$	
	$R_{H_2} = r_2 - 2r_4 + r_5 + 14r_1$	
	$R_{CO} = r_2 + 2r_3 + r_5 + 10r_1$	
	$R_{CO_2} = -r_3 + r_5$	
	$R_{CH_4} = r_4$	
Rates of chemical consumption of solid carbon	$R_C = -r_2 - r_3 - r_4$	[15]
Heat of reaction (kJ.kmol^{-1})	$\Delta H_j = \sum_i \alpha_{ij} H_i$	[15]
	$\Delta H_1 = 14H_{H_2} + 10H_{C0} - 10H_{H_2O} - H_n$	
	$\Delta H_2 = H_{H_2} + H_{C0} - H_{H_2O} - H_C$	
	$\Delta H_3 = 2H_{C0} - H_{CO_2} - H_C$	
	$\Delta H_4 = H_{CH_4} - 2H_{H_2} - H_C$	
	$\Delta H_5 = H_{C0_2} + H_{H_2} - H_{H_2O} - H_{CO}$	
Molar enthalpy (kJ.kmol^{-1})	$H_i = H_{f,i} + M_i H_{s,i}$	[22]
	$H_{f,C}^{o} = H_{f,H_2=}^{o}0$	
	$H_{f,n_{(g)}}^{o} = 1.56 \cdot 10^5$	
	$H_{f,H_2O}^{o} = -2.41826 \cdot 10^5$	
	$H_{f,CO}^{o} = -1.10603 \cdot 10^5$	
	$H_{f,CO_2}^{o} = -3.93777 \cdot 10^5$	
	$H_{f,CH_4}^{o} = -7.48977 \cdot 10^4$	
Sensible heat (kJ.kg^{-1})	$H_{s,i} = \int_{T_o}^{T} C_{p,i} dT$	[15]

Table 6 (continued)

Property	**Correlation**	**References**
Heat capacity (kJ.kg^{-1}.K^{-1})	$C_{p,g}=\frac{\sum_i C_{p,i}\rho_i}{\rho_g}$ $C_{p,n}=-0.10+4.4\cdot10^{-3}T-1.57\cdot10^{-6}T^2$ $C_{p,H_2O}=1.67+6.4\cdot10^{-4}T$ $C_{p,H_2}=13.925+1.3\cdot10^{-3}T$ $C_{p,CO}=0.767+4\cdot10^{-4}T$ $C_{p,CO_2}=0.9784+2\cdot10^{-4}T$ $C_{p,CH}=1.508+2.9\cdot10^{-3}T$ $C_{p,N}=0.97+2\cdot10^{-4}T$ $C_{p,C}=0.42+2.09\cdot10^{-3}T+6.85\cdot10^{-7}T^2$	[23, 24]
Thermal conductivity (kJ.s^{-1}.m^{-1}.K^{-1})	$\lambda_e=(1-\varepsilon)^2\lambda_C+\varepsilon^2\lambda_g$ $\lambda_C=1.85\cdot10^{-3}$ $\lambda_g=\frac{1}{2}\left[\sum_i x_i\lambda_i+\left(\sum\frac{x_i}{\lambda_i}\right)^{-1}\right]$ $\lambda_n=2.15\cdot10^{-5}+1.039\cdot10^{-7}T$ $\lambda_{H_2O}=-2.694\cdot10^{-5}+1.253\cdot10^{-7}T$ $\lambda_{H_2}=-3.988\cdot10^{-5}+4.861\cdot10^{-7}T$ $\lambda_{CO}=1.234\cdot10^{-5}+5.055\cdot10^{-8}T$ $\lambda_{CO_2}=-5.031\cdot10^{-6}+7.617\cdot10^{-8}T$ $\lambda_{CH_4}=-5.77\cdot10^{-6}+1.50\cdot10^{-7}T$ $\lambda_{N_2}=1.00\cdot10^{-5}+5.562\cdot10^{-8}T$	[22, 25-27]

3. CONCLUSIONS

This model focused on naphthalene conversion over the active biomass char particle besides the simultaneous carbon conversion of the particle. The char particle was assumed a sphere surrounded by a gas film in a bulk gas composition of naphthalene, nitrogen, steam, hydrogen, carbon monoxide, carbon dioxide and methane. The model treated the heterogeneous gasification reaction kinetics, external and internal mass transfer and changing particle properties during gasification. The effect of kinetics and internal and external mass transfer on naphthalene and carbon conversion will be investigated. The investigated parameters were particle size, bulk gas temperature and time on stream. The gas and solid (carbon) mass balance and energy balance equations will be solved in the radial direction using a set of initial and boundary conditions. Later, the model results will be presented. Then, the model will be transformed to a fixed bed reactor model and verified through laboratory experiments.

NOMENCLATURE

a	Relative surface area of the char particle	m^2.m^{-2}
Bi_m	Biot number	—
C_i	Gas component concentration	kmol.m^{-3}
$C_{i,b}$	Bulk concentration of the gas component	kmol.m^{-3}
$C_{i,s}$	Surface concentration of the gas component	kmol.m^{-3}
C_n	Naphthalene concentration	kmol.m^{-3}
$C_{p,C}$	Specific heat capacity of char	kJ.kg^{-1}.K^{-1}

$C_{p,i}$	Specific heat capacity of gas component i	kJ.kg^{-1}.K^{-1}
$C_{p,g}$	Specific heat capacity of the gas	kJ.kg^{-1}.K^{-1}
C_s	Surface concentration of gas	kmol.m^{-3}
D_e	Effective diffusion coefficient	m^2.s^{-1}
D_i	Binary diffusion coefficient of the gas component i	m^2.s^{-1}
d_p	Particle diameter	m
h	Heat transfer coefficient in the gas film around the particle	kJ.m^{-2}.s^{-1}.K^{-1}
H_f	Molar heat of formation	kJ.kmol^{-1}
H_i	Molar enthalpy	kJ.kmol^{-1}
H_s	Specific sensible heat	kJ.kg^{-1}
k	Kinetic rate constant	s^{-1}.(kmol.m^{-3})$^{n-1}$
k'	Intrinsic rate coefficient on area basis	kg.m^{-2}.s^{-1}
k_{app}	Apparent kinetic rate constant of the naphthalene reforming reaction	s^{-1}
$k_{g,i}$	Mass transfer coefficient of the gas component i in the gas film	m.s^{-1}
L	Characteristic length; equivalent to (dp/6) for a sphere	m
M_i	Molecular weight of the ith gas component	kmol.kg^{-1}
M_C	Molecular weight of the carbon	kmol.kg^{-1}
N	Apparent reaction order with respect to the gas component	—
Nu	Nusselt number	—
P_i	Partial pressure of component i	Pa, atm
r	Radius coordinate	m
r	Intrinsic volumetric gasification rate	kmol.m^{-3}.s^{-1}
r_n	Rate of naphthalene conversion reaction	kmol.m^{-3}.s^{-1}
R	Gas constant	kJ.kmol^{-1}.K^{-1}
R_C	Sum of the rates of the chemical carbon conversion	kmol.m^{-3}.s^{-1}
R_i	Sum of the rates of the chemical production of gas component i	kmol.m^{-3}.s^{-1}
R_o	Outside particle diameter	—
S	Effective surface area of the char at a certain carbon conversion	m^2.kg^{-1}
S_o	Initial total surface area of the char	m^2.kg^{-1}
Sh	Sherwood number	—
t	Time on stream	s
T	Local particle temperature	K
T_b	Temperature of the bulk gas	K
T_c	Temperature of the center of the particle	K
T_s	Temperature of the surface of the particle	K
u_o	Superficial gas velocity	m.s^{-1}
w_C	Carbon content in the char particle (0.88)	—
x_i	Gas component mole fraction	—
X_C	Carbon conversion at a certain time on stream	—

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