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## Multi-fluid reactive modeling of fluidized bed pyrolysis process

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## HIGHLIGHTS

- Biomass pyrolysis in a bubbling fluidized bed reactor has been studied.
- Effect of operating temperature on product yield has been analyzed.
- Effect of gas velocity and particle size has also been examined.

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## ABSTRACT

A multiphase reactive model of biomass pyrolysis process has been implemented by integrating the reaction kinetics of the thermo-chemical decomposition of biomass with the hydrodynamics of the fluidized bed. The model was validated with the experimental data of biomass pyrolysis in the presence of a sand bed. The simulation results were examined to analyze the effect of reactor temperature, superficial gas velocity and biomass particle size on the bed hydrodynamics and product yields. It was found that at temperatures higher than 500 °C, there was a significant conversion of primary tar into NCG (non-condensable gases) due to thermal cracking inside the reactor. However, the increase in superficial gas velocity led to higher concentration of tar due to lower residence time for tar cracking reactions. Any increase in biomass particle size reduced the yield of volatile products due to decrease in the rate of heat transfer, which in turn increased the yield of biochar.

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## 1. Introduction

Pyrolysis is one of the significant processes for producing energy from biomass (Chakravarti et al., 2012), where a set of thermo-chemical decomposition processes are used to convert the organic materials in biomass into carbon-rich solid and volatile matters by heating in the absence of oxygen (Demirbas and Arin, 2002). The solid content of pyrolysis products is known as the biochar or char, and is generally high in carbon content. The volatile contents are partly condensed to give a liquid fraction called tar or bio-oil (high molecular weight compounds) along with a mixture of the non-condensable gases (H<sub>2</sub>, CO, CO<sub>2</sub>, and C1–C4 hydrocarbons) (Sharma et al., 2014). The formation of these products is from both primary decomposition of the solid biomass as well as secondary reactions of condensable volatile organic products into low-molecular weight gases, secondary tar and char,

while transporting through particle and reactor gas environment (Di Blasi, 2008).

Computational Fluid Dynamics (CFD) modeling is vastly used for studying the thermo-chemical reactions with heat and mass transfer in various reactor configurations. Different thermo-chemical processes such as combustion and gasification (Fletcher et al., 1998; Fletcher et al., 2000; Jin, 2013; Jones et al., 2000; Wu et al., 2013; Xue and Fox, 2014; Yan and Lai, 2006) have been simulated using CFD techniques. CFD models for studying biomass pyrolysis have also been developed. Papadikis et al. (2009) studied the pyrolysis of biomass inside a fluidized bed reactor with Euler-Euler-Lagrangian approach of CFD modeling. Oevermann et al. (2009) developed an Euler-Lagrangian model for wood gasification in a bubbling fluidized bed reactor. Gerber et al. (2010) proposed an Euler-Euler model for wood gasification in bubbling fluidized bed using char as bed material. An Euler-Euler model for fast pyrolysis of biomass in fluidized bed was proposed by Xue et al. (2011). The results for apparent biomass density, vertical velocity, average temperature and product yields were also compared for different 2-D grid sizes and between 2-D and 3-D domain along with experimental data (Xue et al., 2012).

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Bruchmüller et al. (2012) studied the thermo-chemical decomposition of biomass using Computational Fluid Dynamics– Discrete Element Model (CFD– DEM) approach. The effect of fluidization velocity, temperature and moisture content on product yield and composition was analyzed and compared with experimental results. However, most of these studies (Bruchmüller et al., 2012; Papadikis et al., 2009) were carried out for considerably less amount of time not sufficient for approaching the steady state inside the reactor. Furthermore, the lumped reaction model for biomass decomposition and tar cracking needs to be studied during biomass pyrolysis in the large scale reactors.

The objective of this study was to analyze both primary and secondary pyrolysis reactions in dense bed and in freeboard region in the bubbling fluidized bed reactor which allows better mixing of different phases during biomass degradation, and also favors the separation of biomass and biochar particles from other volatile products. In this study, a CFD model was developed for biomass pyrolysis for analyzing the effect of certain parameters such as reactor temperature and particle size on product yield in the fluidized bed conditions. The model did not consider the intra-particle heat and mass transfer effects for the particle range (< 1 mm) analyzed in this study. However, it considered the inter-particle heat transfer in the bed and in the freeboard region with particle degradation, tar cracking reactions and variation in thermo-physical properties with change in biomass composition. The mass transfer mechanisms inside the reactor were treated using kinetic reactions given for the process. Furthermore, the product distribution was analyzed quantitatively, as the products were lumped into gas, tar and biochar in the model.

## 2. Model description

The kinetic mechanism with particle scale degradation of biomass and the hydrodynamic model were utilized for developing a reactive multi-phase CFD model. The model also considered the bed hydrodynamics and mixing/segregation of the solid phases, as well as heat and mass transfer effects during biomass thermo-chemical decomposition in a fluidized bed reactor.

### 2.1. Kinetic model

The reaction mechanism proposed for the biomass pyrolysis is given in Fig. 1. During pyrolysis process in the bubbling fluidized bed, the gas mixture (*g*), bio- mixture (*s1*) and bed particles (*s2*) such as sand or a catalyst are present in the reactor. The gas mixture contains primary tar (*T1*), secondary tar (*T2*), non-condensable gases (*G*), water vapor (*W*) and inert gas (*I*) such as nitrogen. The bio-mixture contains biomass (*B*), biochar or char (*C*) and moisture (*M*) content. As per this lumped model, the biomass decomposes to give non-condensable gases, primary tar (bio-oil) and char (solid carbon product containing mineral matter). The primary tar (containing oxygenated compounds such as acetols) further reacts homogeneously in the gas phase as well as heterogeneously on the surface of catalytically active sites (such as on char surfaces) to convert into NCG and higher molecular weight secondary tar (such as Poly-aromatic Hydrocarbons). The secondary tar also re-polymerizes heterogeneously into char particles in the presence of thermal catalytic reactor environment. The bio-mixture is assumed to be filled with gas in the pores available inside the char particles. This leads to lower density of char particles produced after removal of volatiles from the biomass. The moisture converts into water vapor during drying of biomass particles in the reactor. This mechanism has been considered using a separate reaction scheme as given by Chan et al. (1985). The bed particles do not react, and hence, no kinetic scheme given in the

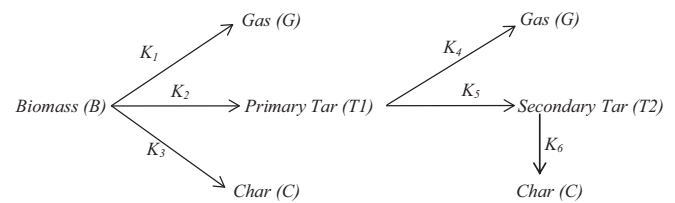


Fig. 1. Biomass thermo-chemical decomposition mechanism (Sharma et al., 2014).

Table 1  
Kinetic rate parameters.

Reaction constant, $K_i$	Pre-exponential factor, $A$ ( $s^{-1}$ )	Activation energy, $E$ (J/kmol)	References
$K_1$	$4.38 \times 10^9$	$1.527 \times 10^8$	Di Blasi and Branca (2001)
$K_2$	$1.08 \times 10^{10}$	$1.48 \times 10^8$	Di Blasi and Branca (2001)
$K_3$	$3.27 \times 10^6$	$1.117 \times 10^8$	Di Blasi and Branca (2001)
$K_{vap}$	$5.13 \times 10^{10}$	$8.8 \times 10^7$	Bryden and Hagge (2003)

model. All the reactions in the solid and gas phase are first order in nature with rate constants given using Arrhenius kinetic expressions.

The rate of generation or consumption for all the solid and gas phase species is given as:

$$\text{For Biomass, } S_{s1,B} = -(K_1 + K_2 + K_3) \alpha_{s1} \rho_{s1} X_{s1,B} \quad (1)$$

$$\text{For Char, } S_{s1,C} = K_3 \alpha_{s1} \rho_{s1} X_{s1,B} + K_6 \alpha_g \rho_g X_{g,T2} \quad (2)$$

$$\text{For Moisture, } S_{s1,M} = -K_{vap} \alpha_{s1} \rho_{s1} X_{s1,M} \quad (3)$$

$$\text{For Non-Condensable Gas, } S_{g,G} = K_1 \alpha_{s1} \rho_{s1} X_{s1,B} + K_4 \alpha_g \rho_g X_{g,T1} \quad (4)$$

$$\text{For Primary Tar, } S_{g,T1} = K_2 \alpha_{s1} \rho_{s1} X_{s1,B} - (K_4 + K_5) \alpha_g \rho_g X_{g,T1} \quad (5)$$

$$\text{For Secondary Tar, } S_{g,T2} = K_5 \alpha_g \rho_g X_{g,T1} - K_6 \alpha_g \rho_g X_{g,T2} \quad (6)$$

$$\text{For Water Vapor, } S_{g,W} = K_{vap} \alpha_{s1} \rho_{s1} X_{s1,M} \quad (7)$$

here  $K_1$ ,  $K_2$  and  $K_3$  are the kinetic constants given for the reactions of biomass pyrolysis in Table 1.  $K_{vap}$  is the kinetic constant for the conversion of moisture available inside the biomass. The heats of reactions used for the biomass pyrolysis reactions are given in Table 2.  $K_4$ ,  $K_5$  and  $K_6$  are the kinetic constants for the tar cracking reactions and their application would be discussed later in the results and discussion section. The heat of reaction for the tar cracking reactions is taken as  $-42000$  J/kg (Di Blasi, 1993).

### 2.2. Multi-phase reactor model

The Eulerian-Eulerian (EE) approach was applied for this multi-phase reactor model. The gas mixture phase treated as the primary continuous phase, whereas the solid phases considered as the secondary dispersed phases in the reactor. The continuity, momentum and energy equations were solved for each phase, and a single pressure was shared by all phases. In this model, the turbulence was not resolved both in the dense bed and in the freeboard region due to consideration of Reynolds number (Re) in the range of laminar flow (van Wachem et al., 2001). The model equations for both gas and solid phase hydrodynamics as well as for associated constitutive relations are described below, details of

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