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Modeling the impact of bubbling bed hydrodynamics on tar yield and its fluctuations during biomass fast pyrolysis ☆

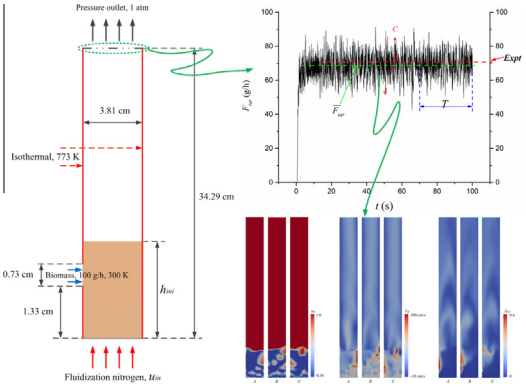
Qingang Xiong [a,*], Fei Xu [b], Emilio Ramirez [a], Sreekanth Pannala [c], C. Stuart Daw [a,*]

[a] Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA
[b] Department of Mechanical Engineering, Iowa State University, Ames, IA 50011, USA
[c] Saudi Arabia Basic Industries Corporation Americas, Sugar Land, TX 77478, USA

HIGHLIGHTS

- A bubbling bed for biomass fast pyrolysis was simulated using CFD.
- The effects of bubbling bed hydrodynamics on tar production were investigated.
- Effects of operating conditions on oscillation of tar yield were revealed.
- Slugging was found to be an important issue for oscillation of tar yield.

GRAPHICAL ABSTRACT



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ABSTRACT

The impact of bubbling bed hydrodynamics on temporal variations in the exit tar yield for biomass fast pyrolysis was investigated using computational simulations of an experimental laboratory-scale reactor. A multi-fluid computational fluid dynamics model was employed to simulate the differential conservation equations in the reactor, and this was combined with a multi-component, multi-step pyrolysis kinetics scheme for biomass to account for chemical reactions. The predicted mean tar yields at the reactor exit appear to match corresponding experimental observations. Parametric studies predicted that increasing the fluidization velocity should improve the mean tar yield but increase its temporal variations. Increases in the mean tar yield coincide with reducing the diameter of sand particles or increasing the initial sand bed height. However, trends in tar yield variability are more complex than the trends in mean yield. The standard deviation in tar yield reaches a maximum with changes in sand particle size. The standard deviation in tar yield increases with the increases in initial bed height in freely bubbling state, while reaches a maximum in slugging state.

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* Corresponding authors.
E-mail addresses: xiongq@ornl.gov (Q. Xiong), dawcs@ornl.gov (C.S. Daw).

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1. Introduction

Biomass pyrolysis for producing high energy–density transportable bio-oil has attracted increased considerable interest in recent years [1]. Under external heating in the absence of oxidizer agent, lignocellulosic biomass is rapidly decomposed into three primary products: condensable liquids (also called tar), non-condensable gases, and biochar [2]. The non-condensable gases and biochar can be used to make high-value coproducts or used as fuel for process heat, while the condensable liquids contain hydrocarbons that can be used to produce liquid fuels via catalytic upgrading [3].

Because the potential output of bio-oil is directly dependent on the initial pyrolysis tar yield and composition, there is considerable interest in experimental [4–8] and computational [9–22] investigations of how tar yield is affected by the biomass type and also by the employed conditions for pyrolysis. This interest should include both the mean tar yield as well as its variability, since large fluctuations in the condensable hydrocarbon stream used for catalytic upgrading would significantly complicate the process control. With this in mind, we focused our attention in this study on simulating how the operation of a bubbling bed pyrolysis reactor might affect both mean tar yield and its variability.

Many versions of biomass fast pyrolysis have been implemented in bubbling fluidized-bed reactors because of their excellent solid mixing and heat transfer [4]. Some general guidelines regarding the effects of operating conditions on overall tar yields have been proposed in the literature [5,6]. The effects of inlet gas velocity [16], sand particle size [23], biomass particle size [19], reactor temperature [24], reaction atmosphere [22,25], etc., on overall tar yields have been extensively investigated. However, it appears that the issue of temporal fluctuations in the exit gases from such reactors has rarely been addressed. Since the hydrodynamics and transport processes in bubbling bed reactors include inherent fluctuations associated with the multi-phase flow, one would expect this to create some degree of fluctuations in the flow and composition of the pyrolysis products leaving the reactor. This latter issue is of particular concern in this study because of its potential impact on processes involving direct catalytic upgrading of the chemically unstable products in raw bio-oil.

2. Modeling approach

In this study, the effects of different operating parameters on the fast pyrolysis products exiting the reactor were simulated using computational fluid dynamics (CFD) to account for the multi-phase hydrodynamics. Compared with time-consuming and costly experiments, multi-phase CFD is able to provide detailed information on the spatiotemporal variations in species concentrations, flows, and temperatures within the reactor with reduced effort and developing circle. This makes CFD a promising platform to account for physical relationships that are beyond the capability of direct experimental measurements [7–15]. To make sure that the simulated reactor conditions were relevant, we based the simulations in the present study on an experimental laboratory-scale bubbling fluidized-bed pyrolyzer located at Iowa State University [16]. Assumptions concerning inlet gas flow, biomass feed rate, initial bed height, and bed solids properties were based on realistic variations about the published nominal operating conditions for this reactor.

Following previous experience with similar reactor simulations, we adopted the multi-fluid modeling (MFM) approach for CFD [17–21,26,27] for this study. Compared with discrete particle models [28–30], MFM models treat all phases as continua through volume fraction averaged conservation equations, hence considerable computational effort is saved. The relevant conservation equations for MFM can be found in our previous papers [17,23], and thus they are not described here. The momentum and energy transfer coefficients between gas and solids were modeled by the Gidaspow drag model [31] and Gunn heat transfer model [32], respectively.

Table 1
Reaction rate constants for biomass fast pyrolysis reactions.

Components	Reaction	Y	A (s^{-1})	E (J/mol)
Cellulose	k_{1c}		2.8×10^{19}	2.424×10^5
	k_{2c}		3.28×10^{14}	1.965×10^5
	k_{3c}	0.35	1.3×10^{10}	1.505×10^5
Hemicellulose	k_{1h}		2.1×10^{16}	1.867×10^5
	k_{2h}		8.75×10^{15}	2.024×10^5
	k_{3h}	0.6	2.6×10^{11}	1.457×10^5
Lignin	k_{1l}		9.6×10^{8}	1.076×10^5
	k_{2l}		1.5×10^{9}	1.438×10^5
	k_{3l}	0.75	7.7×10^{6}	1.114×10^5
Tar	k_4		4.25×10^{6}	1.08×10^5

The multi-component, multi-step devolatilization scheme proposed by Miller and Bellan [33] was employed to model the biomass fast pyrolysis reactions, since it includes both the effects of the initial biomass composition and secondary tar cracking [34]. The Miller and Bellan devolatilization scheme includes the following major reactions:

$$\text{Activation}: \quad \text{Virgin biomass} \xrightarrow{k_1} \text{Active biomass} \tag{1}$$

$$\text{Primary}: \quad \text{Active biomass} \xrightarrow{k_2} \text{Tar} \tag{2a}$$

$$\text{Active biomass} \xrightarrow{k_3} Y\ \text{Biochar} + (1 - Y)\ \text{Gas} \tag{2b}$$

$$\text{Secondary}: \quad \text{Tar} \xrightarrow{k_4} \text{Gas} \tag{3}$$

Virgin biomass is assumed to consist of cellulose, hemicellulose, and lignin with prescribed mass fractions. In this reaction network, each reaction is assumed to be first-order irreversible, and detailed data on the reaction rate constants and heats are listed in Table 1.

The open-source code MFIX developed at National Energy Technology Laboratory was used to conduct the numerical study [35]. The stiff ordinary chemistry solver was launched each time step to integrate the reaction kinetics and a time-split scheme [36] was utilized to couple the multiphase flow solver with the chemical reaction solver.

3. Simulation details

The experimental laboratory-scale bubbling fluidized-bed reactor at Iowa State University [16] on which the present simulations were based is illustrated schematically in Fig. 1. This reactor has been used in many CFD studies [19,26,37] to provide high-fidelity comparisons with computational predictions. The inner reactor diameter and total height are 3.81 cm and 34.29 cm, respectively. The bulk of the reactor bed is composed of sand particles with density of 2.649 g/cm^3 and diameter (d_s) of 0.052 cm. These are typically added to the reactor vessel to an initial height (h_{ini}) of 5.5 cm and a void fraction of 0.41. Biomass particles are injected into the reactor 1.33 cm above the reactor bottom. The reactor exit is open to atmospheric pressure where products are collected for post-processing. The temperature of the side wall is maintained at 773 K. The assumed biomass feed in this study was red oak, with a mean diameter of 0.04 cm and a composition of 41% cellulose, 32% hemicellulose, and 27% lignin on mass basis [16].