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CFD-based Biomass Fast Pyrolysis Simulations in a Gas-Solid Vortex Reactor demonstrating Process Intensification

Shekhar R. Kulkarni[a], Arturo González Quiroga[a], Patrice Perreault[b], Hitesh Sewani[c], Geraldine J. Heynderickx[a], Kevin M. Van Geem[a]*, Guy B. Marin[a]

[a] Laboratory for Chemical Technology, Ghent University Technologiepark 914, 9052 Gent, Belgium
[b] Universidad Autónoma de Yucatán, Calle 60 491A, Centro, 97000 Mérida, Yuc., Mexico
[c] Indian Institute of Technology Roorkee, Uttarakhand-247667, India
Kevin.VanGeem@Ugent.be

Gas-Solid Vortex Reactors (GSVRs) are a new generation of rotating fluidized beds. The solids beds are closely packed. Gas-solid contact improves, resulting in better heat and mass transfer. It makes GSVRs appropriate for thermally intensive reactions like biomass fast pyrolysis. In this work, results of Computational Fluid Dynamics (CFD) simulations demonstrate various salient features of the GSVR. Biomass fast pyrolysis simulations predict high yields of liquid bio-oil when performed in a GSVR. Additionally, the GSVR is found to radially segregate biomass and char, resulting in a higher entrainment tendency of char particles. GSVR is thus found to be a viable alternative for biomass fast pyrolysis, resulting in a considerable process intensification.

1. Introduction

Although over the last century, the global energy sources are dwindling, the need for energy and fuels keeps growing and will have to be met with newer and more efficient sources like solar energy, geothermal energy, etc. High value chemicals (like furfurals, levoglucosan) and fuel grade liquid (commonly referred to as bio-oil) can be produced via fast pyrolysis of lignocellulosic biomass (Zhang, 2009). A successful conversion of biomass however, is controlled by a few important factors. An effective separation of generated char particles from the product vapors will reduce the secondary product degradation. A rapid removal and instantaneous condensation of the product vapors and an effective temperature control of the reaction will maximize the yields of valuable products (Bridgwater, 2012). The effectiveness of biomass fast pyrolysis as a dependable process to produce energy and fuel thus hinges on, among others, a suitable reactor configuration simultaneously satisfying the above criteria (Mettler et al., 2012).
In chemical engineering, fluidized bed reactor technology is commonly preferred for processes involving multiphase gas-solid flows. In these reactors, a solids bed is formed under the counteracting influence of a drag force exerted by the gas flow, and the gravitational force. Low pressure drops, moderate slip velocities along with simplicity in design and ease in operation are some of the reasons for these reactors being preferred in both chemical engineering research and industry. However, there are key shortcomings which make their application potentially restricted. Firstly, the fluidized beds are loosely packed as compared to the packed beds with solid volume fractions in the range of 0.1 – 0.2. Secondly, the gas flow rate is limited as the bed density decreases with increasing gas flow rates (Zhang, 2009). These limitations necessitate a need to consider a more efficient breed of multiphase fluidized bed reactors which could simultaneously satisfy the process requirements mentioned earlier, especially for biomass fast pyrolysis (Bridgwater, 2012).
An interesting alternative is offered by replacing the gravitational force by a centrifugal force, as realized in a rotating fluidized bed. In a GSVR, the rotating bed is realized by feeding gas at high velocity in almost azimuthal direction through small rectangular openings called slots. In the GSVR, momentum transfer makes the particles rotate azimuthally. A rotating solids bed, fluidized in the radial direction is formed in the centrifugal field. Replacing the gravitational force by the centrifugal force results into a few salient features.

The solids bed exhibits denser packing, the interphase slip velocities are higher and the gas residence time in the reactor is of the order of a few milliseconds (De Wilde, 2014). Moreover, unlike the gravitational fluidized beds, the centrifugal fluidized bed becomes more compact when the gas flow rate increases. The GSVR thus becomes a worthy candidate to study thermally intensive and potentially beneficial reactions like biomass fast pyrolysis. Owing to these properties, GSVR has been a topic of active research into mass transfer (Volchkov et al., 1993), heat transfer (Eliaers and De Wilde, 2013), etc.

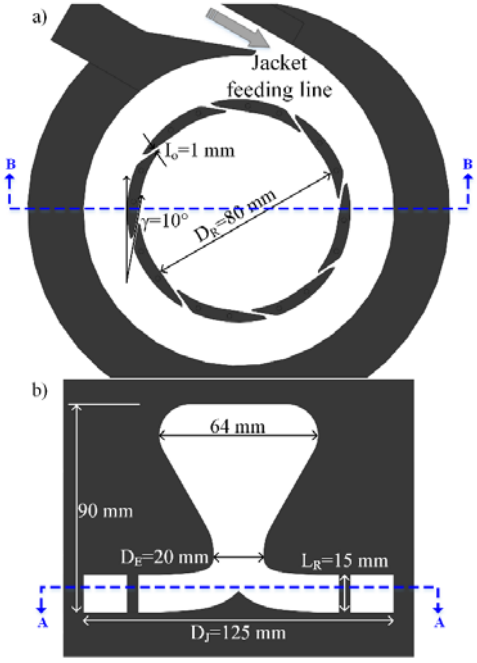


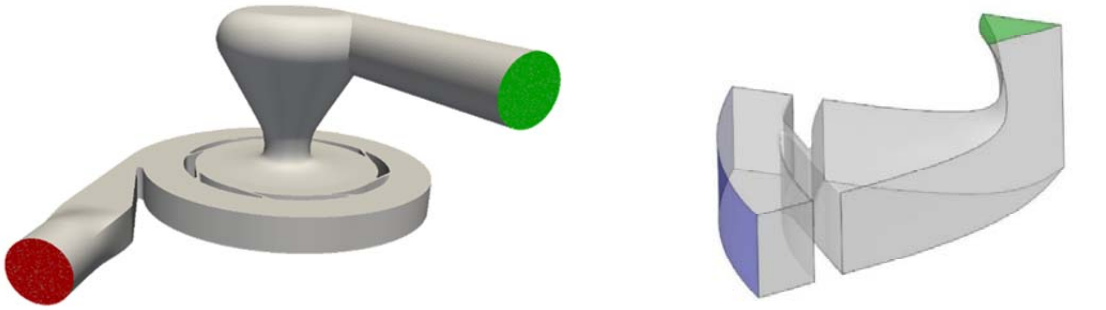
Figure 1 : (a) Top and (b) front views of the Gas-Solid Vortex Reactor highlighting key design features like bottom plate cone and exhaust design (taken from (Gonzalez-Quiroga et al., 2017)).

At the Laboratory for Chemical Technology (Ghent University), experimental and numerical vortex technology research has contributed to this upcoming research area in single phase and multiphase flows (Kovacevic et al., 2014), (Niyogi et al., 2017), (Friedle et al., 2017). The next stage in this research is the demonstration of a GSVR designed to handle heat sensitive reactions like biomass fast pyrolysis. In the current GSVR design, as shown in
Figure ***1*** the gas enters the jacket surrounding the GSVR, through a single inlet and is further distributed into the main reactor chamber via 8 rectangular, azimuthally inclined slots. These slots of 1 mm width each are positioned at an angle of 10° with the tangent to the reactor chamber. The chamber diameter and height are 80 mm and 15 mm respectively. The gas exits the reactor through a central exhaust of 20 mm diameter. The exhaust is uniquely shaped to retain the kinetic energy of the exiting gas and to minimize the pressure drop across the unit. In order to reduce the effect of backflow of gas in the central exhaust, the bottom plate of the reactor is conically shaped. There is no separate solids outlet. The solids are drained from the reactor through entrainment, whenever required. This design of the reactor was optimized through various single-phase CFD simulations (Gonzalez-Quiroga et al., 2017). In the present study, there is first an emphasis on selecting the appropriate simulation domain to calculate reactor hydrodynamics in the 3D GSVR geometry. Once this domain is identified, a lumped kinetic model for biomass fast pyrolysis is implemented. Calculated product yields are compared with the values obtained in a previous study by (Ashcraft et al., 2012). The latter performed simulations on a 2D reactor domain. The effects of upgrading 2D domain to 3D are identified.

2. Non-reactive Two-phase Flow Simulations

To perform non-reactive CFD simulations in the GSVR, two geometries are considered: the full geometry and a pie geometry. The full geometry mimics the actual size and shape of the reactor construction, along with the unique inlet and outlet sections. Due to the presence of the latter design features, there is no rotational periodicity in the geometry. Restricting the computational geometry to a smaller, periodic section requires that some liberties are taken. The inlet is assumed to be the entire circumferential wall and the outlet is reduced to

the bell only. These simplifications thus allow to perform a simulation on a sectional pie geometry consisting of 1 gas inlet slot (45°) and assuming rotational periodicity along the axial boundaries. If both the geometries predict similar hydrodynamics inside the GSVR, the pie geometry can be used for further research. Mesh independency studies are separately performed on both these geometries (not shown) resulting in meshes with 2.3 million cells and 0.25 million cells as shown in Figure 2. Non-reactive gas-solid flow simulations are performed in both geometries for an Air – Aluminium system. The simulation settings are given in Table 1.



(a) Full Geometry (~ 2.3 million cells) (b) Pie Geometry (~ 0.25 million cells)

Figure 2: *Computationally studied geometries of the GSVR: (a) full geometry including the gas inlet and outlet section. (b) visualized pie section of the geometry containing one slot and part of the gas inlet and outlet section.*

Table 1 : Operating conditions for the non-reactive GSVR simulations.

Air inlet temp (K)	289
Air inlet flow ($Nm^3\ hr^{-1}$)	55
Aluminium loading (kg)	0.01239
Aluminium density ($kg\ m^{-3}$)	2700
Aluminium d_p (mm)	0.5
Turbulence model	SST-kω

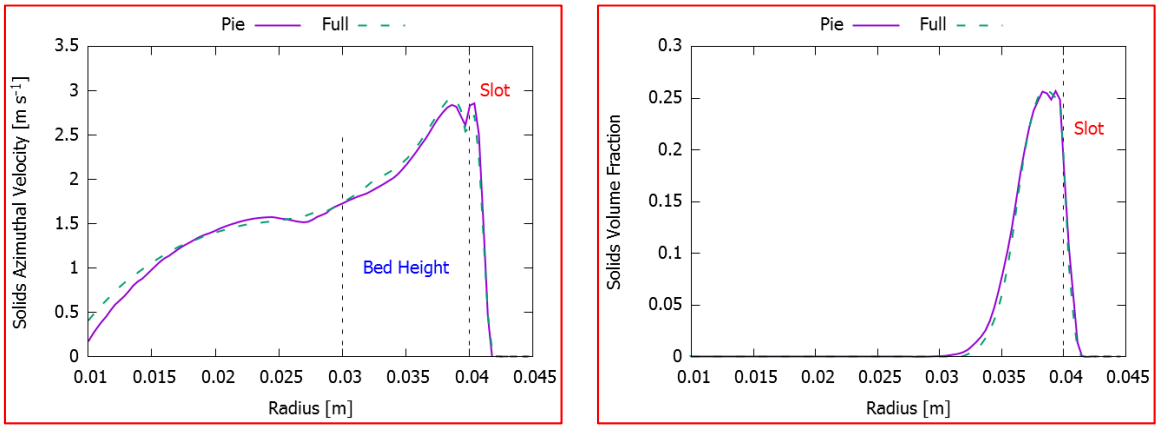


Figure 3 : Comparisons of azimuthal solids velocity profiles and solids volume fractions for full and pie geometries for the conditions described in Table 1. These profiles are displayed for an axial plane at 10 mm.

Eulerian – Eulerian CFD simulations are performed using ANSYS® Fluent® (v18). Solid feeding in both geometries is achieved by using a UDF (User Defined Function) in Fluent® where the solids are added as a mass source term in the flow equations within the GSVR at radius 0.038 – 0.039 m.
Comparison of key parameters in both these geometries is displayed in Figure 3. Both geometries predict a quite compact Aluminium bed, with a bed height of 10 mm. The bed rotates with azimuthal velocity of 1.5-3 m

s^{-1}. The simulation results for both geometries agree well. The pie geometry will be considered for further reactive simulations.

3. Fast Pyrolysis Simulations

Kinetic models describing biomass thermal degradation have been in active research for several decades. The complexity of and inherent variations in the biomass feedstock composition result in a variety of kinetic models available. Over the years, several researchers have proposed models ranging from lumped to detailed, that is with increasing complexity. Lumped models (Shafizadeh and Chin, 1977) consider reactions of biomass components into gaseous (permanent gases), vapor (typically bio-oil) and solid (char) products but give no detailed product distribution. Detailed models (Vinu and Broadbelt, 2012, Zhou et al., 2014) on the other hand provide a more complete and detailed product distribution by accounting for the thermal degradation of all biomass components individually.

Although the detailed models are attractive to understand the fast pyrolysis process, their application in a CFD framework is computationally intensive and not needed for a first study of the performance of new reactor technology like GSVR. Thus, for the present study, a lumped kinetic model is considered. The kinetic parameters used are based on the research of (Xue et al., 2011). This lumped model was used by (Ashcraft et al., 2012) for 2-D reactive simulations of a GSVR. The kinetic model and kinetic parameters are given in Table 2.

Table 2: Lumped model for biomass pyrolysis with kinetic parameters for the reactions considered.

Reaction	ΔH_{rxn} (kJ kg^{-1})	A_f (s^{-1})	E_A (kJ mol^{-1})
v. cellulose → a. cellulose	0	2.80×10^{19}	242.4
v. hemicellulose → a. hemicellulose	0	2.10×10^{16}	186.7
v. lignin → a. lignin	0	9.60×10^{8}	107.6
a. cellulose → bio-oil	255	3.28×10^{14}	196.5
a. hemicellulose → bio-oil	255	8.75×10^{15}	202.4
a. lignin → bio-oil	255	1.50×10^{9}	143.8
a. cellulose → 0.35 $char_{cellulose}$ + 2.6 Pgas	-20	1.30×10^{10}	150.5
a. hemicellulose → 0.6 $char_{hemicellulose}$ + 1.6 Pgas	-20	2.60×10^{11}	145.7
a. lignin → 0.75 $char_{lignin}$ + Pgas	-20	7.70×10^{6}	111.4
bio-oil → Pgas	-42	4.25×10^{6}	108.0

$(v.: virgin\ species\ ;\ a.: activated\ species\ ;\ Pgas: non\text{-}condensable\ pyrolysis\ gases)$

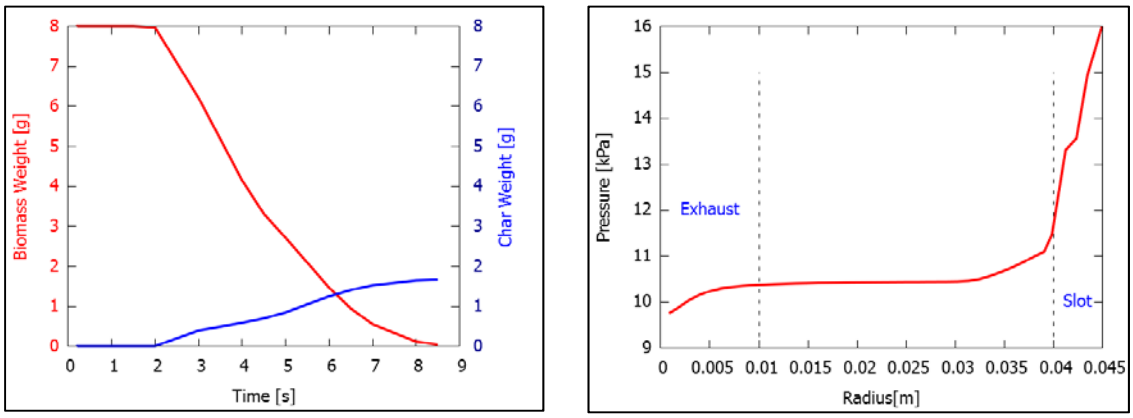
The simulations in the 3D pie-shaped geometry of the GSVR are performed stepwise. From 0-0.1 s flow time, gas only simulations are performed with hot nitrogen entering the reactor at 842 K. Cold biomass particles (298 K; d_b = 0.5 mm; ρ_b = 500 kg m^{-3}; biomass fed to the reactor = 8 g) are fed from 0.1 s onwards near the circumferential wall of the slot. The biomass reactions are enabled from 0.3 s onwards till the entire biomass is converted into products. The delay in the start of the reactions is instituted to facilitate the bed formation and stabilization. In the performed simulations, 3 phases are defined in the GSVR. Nitrogen and product gases contribute to the gas-phase; biomass particles, both virgin and activated, constitute the first solid phase and various char particles generated from biomass components constitute the second solid phase (particle properties: d_c = 0.2 mm; ρ_c = 450 kg m^{-3}). Considering two separate solid phases allows to individually control different phase properties, at the same time allowing to study their interactions. The effect of gravity is accounted for in the simulations. Various interphase interactions like drag and heat transfer are accounted for using the models proposed by (Gidaspow, 1994) and (Gunn and De Souza, 1974) respectively.

4. Results and Discussions

4.1 Product yields

The time-dependent change in biomass and char amounts in the GSVR, as shown in Figure 4, indicate 8 s are needed for a complete biomass conversion into products. The corresponding product yields are 19-21 wt % char, 65-69 wt % bio-oil and 10-12 wt % gases (yields are defined as wt. % of fed biomass being converted to the respective products). The bio-oil yields are high compared to the typically preferred reactor technologies like conical spouted bed (~ 60 wt %), gravitational fluidized bed (~ 50 wt %) or microwave pyrolysis (~ 40 wt

%) (Guedes et al., 2017). When the bio-oil yields obtained are compared to the results presented in the work of (Ashcraft et al., 2012) the char amounts are found to be somewhat higher in the current study. The presence of end wall effects in this 3-D CFD simulations, as compared to their absence in the 2-D CFD study by (Ashcraft et al., 2012) could be a possible reason for this higher char yield. 2-D simulations are typically performed on an axial plane of the reactor, without taking into account the end-wall and fluid/solid interactions. For a reactor such as GSVR, these interactions are important and hence difference in char yields could be explained between 3-D and 2-D simulations. Furthermore, biomass is seen to hardly react in the first 2 s, corresponding to the time required for the cold biomass particles to attain the reaction temperature in the hot Nitrogen gas.

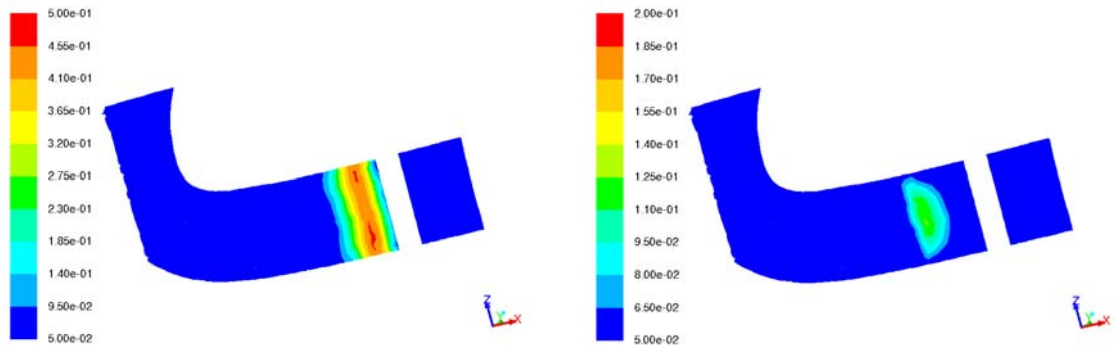


(a) (b)

Figure 4: (a) Time-dependent biomass and char weight in the reactor indicating biomass degradation and char formation, (b) Pressure profile in the reactor indicating a low pressure drop of 4-5 kPa across the slots and 1-2 kPa over the solids bed.

4.2 Radial product segregation

The radial position of a particle in the GSVR is determined by the balance of centrifugal and drag force. When moving towards the gas outlet, the gas velocity and subsequently the drag on the solid particles increases. The effect of this force balance is seen in action via the solids volume fraction fields in the GSVR as shown in Figure *5* . A biomass and a char bed are formed at different radial positions. Since their density ratio of 1.1 is close to unity, the segregation is primarily realized by the particle diameter ratio of 2.5. The char bed is occupying a position closer to the outlet, indicating its likeliness to be entrained. This will effectively reduce its contact time with the generated product vapors, reducing the catalytic activity of the char particles and restricting further degradation of primary products thereof.



(a) biomass (b) char

Figure 5: Instantaneous solids volume fraction fields in the GSVR indicating radially segregated biomass and char layers .

However, segregation is a transient phenomenon as the biomass bed shrinks in time due to reaction, while the char bed grows and closes in to the outer wall with time. Thus, to maintain segregation and yield the maximum

benefits from this process intensification in GSVR, it is required to continuously feed biomass to and remove char from the reactor.

5. Conclusions

3-D CFD simulations of biomass fast pyrolysis are performed in a pie geometry of a GSVR. A lumped reaction model for biomass thermal degradation is used to assess the performance of the GSVR. As compared to the reactors that are preferably used, the bio-oil yields in the GSVR are as high as 70 wt %, which indicate the superiority of the technology that could be used for harnessing maximum benefits from a potentially dependable energy and fuel source. Additionally, particle diameter based radial segregation can be achieved according to the simulations, making GSVR a potential candidate for (studying) biomass fast pyrolysis.

Acknowledgments

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References

Ashcraft, R. W., Heynderickx, G. J. & Marin, G. B. 2012. Modeling fast biomass pyrolysis in a gas–solid vortex reactor. *Chemical Engineering Journal,* 207-208**,** 195-208.

Bridgwater, A. V. 2012. Review of fast pyrolysis of biomass and product upgrading. *Biomass and Bioenergy,* 38**,** 68-94.

De Wilde, J. 2014. Gas–solid fluidized beds in vortex chambers. *Chemical Engineering and Processing: Process Intensification,* 85**,** 256-290.

Eliaers, P. & De Wilde, J. 2013. Drying of Biomass Particles: Experimental Study and Comparison of the Performance of a Conventional Fluidized Bed and a Rotating Fluidized Bed in a Static Geometry. *Drying Technology,* 31**,** 236-245.

Friedle, M., Niyogi, K., Torregrosa, M. M., Marin, G. B. & Heynderickx, G. J. 2017. A drag model for the gas-solid vortex unit. *Powder Technology,* 312**,** 210-221.

Gidaspow, D. 1994. *Multiphase flow and fluidization: continuum and kinetic theory descriptions*, Academic press.

Gonzalez-Quiroga, A., Reyniers, P. A., Kulkarni, S. R., Torregrosa, M. M., Perreault, P., Heynderickx, G. J., Van Geem, K. M. & Marin, G. B. 2017. Design and Cold Flow Testing of a Gas-Solid Vortex Reactor Demonstration Unit for Biomass Fast Pyrolysis. *Chemical Engineering Journal*.

Gunn, D. & De Souza, J. 1974. Heat transfer and axial dispersion in packed beds. *Chemical Engineering Science,* 29**,** 1363-1371.

Kovacevic, J. Z., Pantzali, M. N., Heynderickx, G. J. & Marin, G. B. 2014. Bed stability and maximum solids capacity in a Gas–Solid Vortex Reactor: Experimental study. *Chemical Engineering Science,* 106**,** 293-303.

Mettler, M. S., Vlachos, D. G. & Dauenhauer, P. J. 2012. Top ten fundamental challenges of biomass pyrolysis for biofuels. *Energy & Environmental Science,* 5**,** 7797-7809.

Niyogi, K., Torregrosa, M. M., Pantzali, M. N., Heynderickx, G. J. & Marin, G. B. 2017. Experimentally validated numerical study of gas-solid vortex unit hydrodynamics. *Powder Technology,* 305**,** 794-808.

Shafizadeh, F. & Chin, P. P. S. 1977. Thermal Deterioration of Wood. 43**,** 57-81.

Vinu, R. & Broadbelt, L. J. 2012. A mechanistic model of fast pyrolysis of glucose-based carbohydrates to predict bio-oil composition. *Energy & Environmental Science,* 5**,** 9808.

Volchkov, E. P., Terekhov, V. I., Kaidanik, A. N. & Yadykin, A. N. 1993. Aerodynamics and Heat and Mass Transfer of Fluidized Particle Beds in Vortex Chambers. *Heat Transfer Engineering,* 14**,** 36-47.

Xue, Q., Heindel, T. J. & Fox, R. O. 2011. A CFD model for biomass fast pyrolysis in fluidized-bed reactors. *Chemical Engineering Science,* 66**,** 2440-2452.

Zhang, W. 2009. A review of techniques for the process intensification of fluidized bed reactors. *Chinese Journal of Chemical Engineering,* 17**,** 688-702.

Zhou, X., Nolte, M. W., Mayes, H. B., Shanks, B. H. & Broadbelt, L. J. 2014. Experimental and Mechanistic Modeling of Fast Pyrolysis of Neat Glucose-Based Carbohydrates. 1. Experiments and Development of a Detailed Mechanistic Model. *Industrial & Engineering Chemistry Research***,** 140816080116006.