# **BIOMASS TORREFACTION IN A PULSED FLUIDIZED BED**

## REACTOR

by

Ruixu Wang

B.Sc., China University of Petroleum, Beijing, 2012

# A THESIS SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE OF

## DOCTOR OF PHILOSOPHY

in

### THE FACULTY OF GRADUATE AND POSTDOCTORAL STUDIES

(Chemical and Biological Engineering)

THE UNIVERSITY OF BRITISH COLUMBIA

(Vancouver)

October 2021

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The following individuals certify that they have read, and recommend to the Faculty of Graduate and Postdoctoral Studies for acceptance, the dissertation entitled:

Biomass torrefaction in a pulsed fluidized bed reactor

submitted by	Ruixu Wang	in partial fulfillment of the requirements for
the degree of	Doctor of Philosophy	
in	Chemical and Biological Enginee	ering

### **Examining Committee:**

Xiaotao Bi, Chemical and Biological Engineering, UBC

Supervisor

C. Jim, Lim, Chemical and Biological Engineering, UBC

Co-supervisor

Shahabaddine Sokhansanj, Chemical and Biological Engineering, UBC

Supervisory Committee Member

A. Paul Watkinson, Chemical and Biological Engineering, UBC

University Examiner

Edouard Asselin, Materials Engineering, UBC

University Examiner

### **Additional Supervisory Committee Members:**

Anthony Lau, Chemical and Biological Engineering, UBC

Supervisory Committee Member

Xuantian Li, Nanjing Tech University, China

Supervisory Committee Member

### Abstract

Torrefaction at 200–350 °C in an inert or oxygen-deficient environment is a promising pretreatment to improve the properties of biomass for energy utilization. Fluidized beds with enhanced heat and mass transfer could be a candidate for biomass thermal treatment. Gas pulsation has been proven effective in fluidizing biomass particles with unconventional natures that caused problems in fluidization. A novel pulsed fluidized bed (PFB) reactor for continuous biomass torrefaction was designed and commissioned in this work. The bed has a length-to-width ratio of 10:3 to limit the solids residence time distribution (RTD), leading to uniform solids products.

Biomass particles transport and backmixing were studied by solids RTD measurement. A 2D axial dispersion model with an exchange flow between the active and stagnant zones was proposed to quantify solids backmixing by fitting the model to RTD curves. Horizontal dispersion coefficient representing the backmixing degree were slightly higher in the deeper bed, increased with increasing particle velocity in the shallow bed, and increased greatly with increased gas velocity. Less backmixing was obtained when the bed was operated under a gas pulsation frequency close to the bed natural frequency. Horizontal solids dispersion coefficients obtained in horizontal PFB were significantly smaller than values calculated from literature. A correlation of horizontal dispersion coefficients for biomass particles in the horizontal PFB of shallow beds was established.

Continuous biomass torrefaction was successfully performed in the PFB unit with a feed rate of up to 2 kg/h. Maximum weight loss was identified at the frequency range of 2–4 Hz for feed rates of 1 kg/h and 1.5 kg/h. The temperature near the reactor exit exhibited the most significant influence on the biomass weight loss and the product properties, i.e., higher heating value (HHV),

contents of proximate analysis components, and elemental carbon content, whereas the feed rate had a slight effect. Torrefied biomass obtained in fluidizing gas with 3–6 vol.% oxygen concentrations showed similar HHV with that of non-oxidative torrefaction. Compared to other continuous torrefaction technologies, the PFB torrefier utilized small particles with low residence time.

## Lay Summary

Torrefaction is a thermal process that increases the energy density and shelf life of biomass. A novel reactor system was built to produce torrefied biomass continuously. By evaluating the performance, the reactor technology developed is proven to be efficient in biomass thermal treatment. It has the potential to be commercialized to promote utilization of local renewable biomass resources and contribute to mitigating greenhouse gas emissions via displacing fossil fuels.

### Preface

The work in this thesis, including literature review, reactor design and setup, experimental design, data analysis, and thesis writing were completed by Ruixu Wang under the supervision of Professor Xiaotao Bi, Professor C. Jim Lim, and Professor Shahabaddine Sokhansanj at the Department of Chemical and Biological Engineering, the University of British Columbia, Vancouver campus. Elemental analysis of raw and torrefied sawdust reported was conducted at the Department of Chemistry, the University of British Columbia or Econotech Services Ltd., Delta, British Columbia. Composition analysis of raw biomass material was conducted at State Key Laboratory of Biobased Material and Green Papermaking, Qilu University of Technology, China.

Part of Chapter 3 was submitted for publication. Wang, R., Wang, Z., Bi, X., Lim, C.J. and Sokhansanj, S., Residence Time Distribution and Solids Mixing of Sawdust in a Horizontal Pulsed Fluidized Bed. Powder Technology (submitted). I conducted all the experiments and analysis, and wrote the manuscript under the guidance of Dr. Bi, Dr. Lim, and Dr. Sokhansanj. Dr. Ziliang Wang assisted in experimental design, participated in the experiments, and reviewed the manuscript.

Part of Chapter 3 was published in conference proceedings. Wang, R., Wang, Z., Bi, X., Lim, C.J., Sokhansanj, S. and Kim, C.S., (2021) Measurement of Residence Time Distribution of Sawdust in a Horizontal Fluidized Bed with Gas Pulsation. Proceedings of 13<sup>th</sup> International Conference on Fluidized Bed Technology. I conducted all the experiments and analysis, and wrote the manuscript under the guidance of Dr. Bi, Dr. Lim, Dr. Sokhansanj, and Dr. Chang Soo Kim. Dr. Ziliang Wang assisted in experimental design, participated in the experiments, and reviewed the manuscript. Part of Chapter 4 has been presented at the following conferences:

Wang, R., Wang, Z., Bi, X., Lim, C.J., and Sokhansanj, S. (2018) Biomass Torrefaction in a Pulsed Fluidized Bed. AIChE Annual Meeting, Pittsburgh, US.

Wang, R., Wang, Z., Bi, X., Lim, C.J., and Sokhansanj, S. (2018) Performance of a Prototype Pulsed Fluidized Bed Reactor for Biomass Residues Torrefaction. BioProducts Institute Research Day, Vancouver, Canada.

Wang, R., Peng, J., Jia, D., Bi, X., Lim, C.J., Sokhansanj, S., and Kim, C.S. (2016). Development of a Biomass Torrefaction and Densification Process. Canada-Korea Conference on Science and Technology, Ottawa, Canada.

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# List of Symbols

<i>a</i> *	Spectrophotometer parameter
Α	Bed cross-section area, m <sup>2</sup>
<i>b</i> *	Spectrophotometer parameter
C <sub>b</sub>	Conductivity of the solution of the solids remained in the bed, $\mu$ S/cm
c <sub>i</sub>	Conductivity of the $i^{th}$ sample solution, $\mu$ S/cm
<i>C</i> <sub>1</sub>	Tracer concentration in the active zone
<i>C</i> <sub>2</sub>	Tracer concentration in the stagnant zone
C(t)	Tracer concentration function
$d_b$	Bubble diameter, m
$d_{b,in}$	Initial bubble size, m
$d_{b,m}$	Maximum bubble size, m
$d_p$	Particle diameter, m
$D_{x}$	Horizontal dispersion coefficient, m <sup>2</sup> /s
$D_y$	Vertical dispersion coefficient, m <sup>2</sup> /s
D <sub>sr</sub>	Particle diffusion coefficient, m <sup>2</sup> /s
E(t)	Residence time distribution function
$f_n$	Natural frequency, Hz
$f_p$	Gas pulsation frequency, Hz
F	Feed rate, kg/h
g	Gravitational acceleration, 9.81 m/s <sup>2</sup>
$H_w$	Weir height, m

$H_{bed}$	Bed height, m
$H_{mf}$	Bed height at minimum fluidization velocity, m
K	Apparent mass transfer coefficient between the active and stagnant zones, m/s
L	Bed length, m
$L^*$	Spectrophotometer parameter
$m_0$	Mass of the sample dissolved for conductivity measurement, g
$m_b$	Mass of the solids remained in the bed, g
$m_i$	Mass of the i <sup>th</sup> sample, g
<i>M<sub>carbon</sub></i>	Carbon molar mass, 12.0 g/mol
Ν	Lumped transfer number, 1/s
N <sub>total</sub>	Total sample number
$\mathbf{P}_1$	Absolute pressure before the buffer tank, kPa
P <sub>2</sub>	Absolute pressure in the freeboard, kPa
P <sub>3</sub>	Pressure drop across the bed and distributor, Pa
P4	Pressure drop of the filter, kPa
P <sub>5</sub>	Pressure drop of the combustor, kPa
Pe <sub>eff</sub>	Effective Peclet number
$Q_{CO_2}$	Carbon dioxide flowrate, SL/h
S	Transfer area per unit length between the active and stagnant zones, $m^2/m$
t	Time, s
$t_m$	Mean time, s
t <sub>peak</sub>	Peak time, s

$T_1$	Temperature of the preheated gas, °C
T <sub>2</sub>	Temperature of the windbox, °C
<b>T</b> <sub>3</sub>	Temperature of the dense bed (feed side), °C
T4	Temperature of the dense bed (middle), °C
T <sub>5</sub>	Temperature of the dense bed (discharge side), °C
T <sub>6</sub>	Temperature of the torrefaction gas, °C
T <sub>7</sub>	Temperature of the catalyst bed (bottom), °C
T <sub>8</sub>	Temperature of the catalyst bed (middle), °C
T9	Temperature of the catalyst bed (top), °C
T <sub>10</sub>	Temperature of the flue gas, °C
$T_{\infty}$	Temperature of the imaginary fluid, °C
T <sub>ave</sub>	Average temperature of the bed, °C
T <sub>bed</sub>	Bed temperature, °C
$u_H$	Maximum solids velocity, m/s
$u_x$	Solids velocity, m/s
$u_x(y)$	Solids velocity function
$\bar{u}_x$	Average solids velocity, m/s
$\bar{u}_{x,eff}$	Effective average solids velocity, m/s
U	Superficial gas velocity, m/s
U <sub>elu</sub>	Elutriation gas velocity, m/s
$U_{mf}$	Minimum fluidization velocity, m/s
$V_m$	Standard molar volume, 22.4 L/mol

<i>W<sub>hi</sub></i>	Initial raw biomass weight in the hopper, kg
<i>W</i> <sub>hr</sub>	Remaining raw biomass weight in the hopper, kg
w <sub>p</sub>	Torrefied biomass weight collected in the product receiver, kg
W <sub>r</sub>	Torrefied biomass weight remained in the reactor, kg
W <sub>s</sub>	Torrefied biomass weight collected from the sample port, kg
W	Bed width, m
$W_{bed}$	Bed weight, kg
$W_L$	Weight loss, %
$W_L^*$	Corrected weight loss, kg
x	Horizontal position, m
<i>X</i> 02	Oxygen concentration, vol.%

# Greed symbols

α	Fraction of the active volume
β	Ratio of boundary layer thickness to weir height
δ	Boundary layer thickness, m
$\delta_b$	Bed bubble fraction
∆a*	Change in spectrophotometer parameter <i>a</i> *
$\Delta b^*$	Change in spectrophotometer parameter $b^*$
$\Delta E$	Color value change
$\Delta L^*$	Change in spectrophotometer parameter $L^*$
ε	Bed voidage

$\mathcal{E}_{mf}$	Bed voidage at minimum fluidization velocity
$\mu_g$	Gas viscosity, Pa·s
$ ho_g$	Gas density, kg/m <sup>3</sup>
$ ho_p$	Particle density, kg/m <sup>3</sup>
τ	Mass turnover time, t

# List of Abbreviations

ADM	Axial dispersion model
ADMex	Axial dispersion model with exchange flow
BET	Brunauer-Emmett-Teller
CSTR	Continuous stirred-tank reactor
FFT	Fast Fourier transform
FIR	Finite impulse response
HHV	Higher heating value
HPFB	Horizontal pulsed fluidized bed
MB	Moving bed
PFB	Pulsed fluidized bed
PID	Proportional-Integral-Derivative
RD	Rotary drum
RTD	Residence time distribution
SB	Spouted bed
SC	Screw conveyor
SPF	Spruce-Pine-Fir
TGA	Thermogravimetric analysis

## Acknowledgements

I would like to thank my supervisors Dr. Xiaotao Bi, Dr. C. Jim Lim, and Dr. Shahabaddine Sokhansanj for continuous support and guidance throughout the years. I am most grateful to my primary supervisor, Dr. Bi, who has always been kind, resourceful, and knowledgeable. It would not have been possible to write this dissertation without his enormous help and patience in my research.

I would also like to thank the rest of my thesis committee members, Dr. Anthony Lau and Dr. Xuantian Li, for their valuable comments and suggestions.

I am grateful to the Natural Science and Engineering Research Council of Canada, Mitacs, and Korea Institute of Science and Technology for their financial support of this project.

I would like to acknowledge the support of the technical and administrative staff at the Department of Chemical and Biological Engineering. Thanks to Doug, Serge and Charles for consultations, reactor fabrication and unit construction. Thanks to Marlene, Kristi, Brittany, and Amber for the administrative support.

I am thankful to my colleagues and visiting scholars who inspired me and offered their help in the lab. I am also thankful to my friends who made my time at UBC memorable.

Last but not least, my deep gratitude is owed to my parents for giving me unequivocal support throughout my years of education and my life.

### **Chapter 1: Introduction**

Biomass is a sustainable resource, usually referring to lumber mill and harvest residues, agricultural residues, and municipal solid waste, which are increasingly utilized to produce clean fuel. By developing efficient biofuel technologies, the renewable carbon in biomass could be used to generate energy to displace fossil fuels, hence reduce CO<sub>2</sub> emission (Hall et al., 1991; Kopetz, 2013).

Canada has abundant biomass resources. Wood pellets industry, for example, densifies waste sawdust or other forest residues into pellets to reduce the storage and transportation cost associated with the low bulk density biomass. Canada had 48 pellet plants with nearly 5 million tonnes annual production capacity as of January 2021 (Canadian pellet plants, 2021). However, total biofuels and waste account for only less than 5% of the total primary energy supply in Canada in 2019, according to the International Energy Agency's database. Particularly, British Columbia has the largest managed forest resources in Canada, which have the potential to supply almost half the province's energy consumption (Roach & Berch, 2014). Historically, biomass has been predominantly used for heating. However, advanced conversions enable biomass to serve diversified end users. Synthetic fuel, biomethane as well as bio-power can be generated via gasification and pyrolysis; other thermochemical processes produce solid fuels that can be co-fired with coal for power generation or burned for residential heating; alcohols as transport fuel substitutes are obtained by hydrolysis and fermentation; other renewable liquid fuels also can be produced by biological or chemical processes. Biomass could, therefore, play a major role in phasing out fossil fuels.

However, the intrinsic properties of biomass such as high moisture content, inherent hydrophilicity, and low calorific value compared to fossil fuels have raised a myriad of grinding, transportation and storage problems. Torrefaction as a promising pretreatment technology is proposed to address the issues that hinder the utilization of biomass as a solid fuel or as a feed to advanced applications (Basu et al., 2013; Bergman et al., 2005; Bergman & Kiel, 2005). In addition, torrefied wood pellets made from forest residues in British Columbia are found to be able to reduce primary energy consumption and carbon footprint by about 30% compared to conventional pellets (Yun et al., 2020). Moreover, an efficient reactor needs to be carefully designed to accommodate the distinctive properties of biomass particles for any thermal conversion process.

#### **1.1 Biomass torrefaction**

### 1.1.1 Process description

Torrefaction is a mild pyrolysis process, aiming at upgrading biomass into a commodity solid fuel with favorable properties in view of logistics and end-use. Particularly in substituting coal in the existing facilities without major modification, a complete replacement can be achieved, instead of only 8–15% replacement by untreated chips and pellets (Kuzmina et al., 2016). In torrefaction process, biomass is heated to 200–350 °C in an inert or low-oxygen environment, with a residence time ranging from several minutes to hours, depending on the particle size and heating rate (Campbell & Evitts, 2018; van der Stelt et al., 2011).

When biomass is torrefied, its dry matter is lost mainly because of the dehydration, decarboxylation, and deacetylation of hemicellulose components of the lignocellulose structure in biomass (Burhenne et al., 2013; Deng et al., 2009; Yang et al., 2007). Torrefied forest and

agriculture residues become darker in appearance. Moisture content is reduced from over 10% to less than 5% and even lower than 2% under some severe torrefaction conditions. The atomic O/C and H/C are reduced as a result of the oxygen and hydrogen removal, whereas the relative content of elemental carbon rises. There is an increase in the higher heating value (HHV) of raw biomass generally from the range of 15–20 MJ/kg to approximately between 16 and 29 MJ/kg (Chen et al., 2021; Chen et al., 2015b). After torrefaction, hydrogen bonding will not form, because the hydroxyl groups will be partly destroyed through dehydration (Yan et al., 2009). Thus, torrefied biomass is hydrophobic. Torrefied lignocellulosic biomass is less susceptible to biological degradation, due to the reduction in moisture and nutritive source (Hakkou et al., 2006; Kymäläinen et al., 2014). The grindability of torrefied material is also improved significantly, as the cell walls are destroyed (Arias et al., 2008; Phanphanich & Mani, 2011; Saleh et al., 2013). When combined with densification treatment (Figure 1.1), a high bulk density is acquired which further improves material handling and high energy-intensity applications.



Figure 1.1 Torrefaction and densification process

Organic compounds, carbon monoxide, carbon dioxide, and moisture mainly compose the hot gas released from torrefaction, called torgas. Tumuluru et al. (2012) summed up the analytical techniques for characterizing volatiles and other biomass torrefaction and pyrolysis products. Lê Thành et al. (2015) identified nearly 80 condensable species during torrefaction of pine. The major condensable species identified are water, acetic acid, 2-propanone, 1-hydroxy-, methanol, formic acid, furfural, formaldehyde, whereas the major non-condensable species are carbon dioxide,

carbon monoxide, and methane (Anca-Couce et al., 2016; Arteaga-Pérez et al., 2015; Chen et al., 2015a; Nocquet et al., 2014; Prins et al., 2006a). Yun et al. (2021) calculated that the HHV of torgas ranges from 2.23 to 8.03 MJ/kg depending on the operating conditions. When a large amount of carrier gas is used, the heating value of unit volume gas from the torrefier is expected to be even lower.

The direct recycling of carrier gas is economical and energy-efficient since autothermal operation can be achieved at typical torrefaction conditions (Haseli, 2018; Yun et al., 2021). After the removal of organic volatiles by oxidation/combustion/incineration, oxygen and carbon dioxide are inevitably present in the flue gas. The enforcement of inert conditions while recycling the flue gas results in high capital cost and increases operational complexity (Svanberg et al., 2013). Therefore, torrefaction under the oxidative environment with the presence of oxygen and carbon dioxide has been studied (Brachi et al., 2019b; Chen et al., 2014a; Chen et al., 2016; Eseltine et al., 2013; Rousset et al., 2012; Thanapal et al., 2014; Tran et al., 2016; Uemura et al., 2015; Wang et al., 2013; Wang et al., 2018; Zhang et al., 2019). The presence of oxygen, particularly, leads to operating challenges while the presence of inert carbon dioxide often does not. Joshi et al. (2015) observed combustive thermal runaway during oxidative torrefaction in a packed bed when the oxygen concentration was higher than 5% at 270 °C and 1% at 290 °C, respectively. Wang et al. (2019) tested up to 6% oxygen concentration at 270 °C and 3% at 300 °C for torrefaction in a spouted bed and suggested the existence of an upper limit of oxygen concentration in the carrier gas. In general, it is feasible to use oxygen-laden gas for biomass torrefaction, as properties of torrefied biomass and pellets from oxidative torrefaction are not modified significantly. Higher weight loss is reported compared to the inert torrefaction, owing to the biomass oxidation.

#### 1.1.2 Weight loss as torrefaction severity indicator

Though torrefaction alters many properties of biomass, the weight loss has been chosen to be an indicator representing the severity of the reaction by many researchers (Almeida et al., 2010; Chen et al., 2014a; Chen et al., 2014b; Chen et al., 2015c; Chiou et al., 2016; Ho et al., 2018; Li et al., 2012; Peng et al., 2013; Rago et al., 2018; Sabil et al., 2013). Li et al. (2012) found that mass loss determined the energy yield and higher heating value in both thermogravimetric analysis (TGA) and fluidized bed tests. Grigiante and Antolini (2015) showed similar results of fibers distribution and equilibrium moisture content of torrefied biomass with the same weight loss. Energy yield, carbon content, proximate composition, and the higher heating value was shown to be linearly related to weight loss. Moreover, it was pointed out that regardless of the temperature and residence time, torrefied materials with similar weight loss displayed similar properties. Thus, weight loss can be used in torrefaction modeling to predict the quality of torrefied materials (Devos et al., 2020; Repellin et al., 2010).

#### **1.1.3** Continuous biomass torrefaction operation

In the early studies, biomass torrefaction was carried out mostly by TGA and well-controlled batch reactors, where effects of different combinations of temperature and residence time on torrefaction product for each biomass species were studied. However, as operations move to large scales with continuous feeding and discharging, the operation becomes complex with operating parameters depending on specific experimental setups. For example, the average temperature of various locations of the bed (Ohliger et al., 2013; Wang et al., 2017b), maximum temperature (Shang et al., 2014), exit temperature (Barbanera & Muguerza, 2020; Nhuchhen et al., 2016b; Strandberg et

al., 2015) and reactor wall temperature (Nachenius et al., 2015b) were employed as the torrefaction temperature in various studies. On the other hand, residence time was measured separately at room temperature using the same reactor (Nachenius et al., 2015a; Nhuchhen et al., 2016a) or calculated theoretically (Campbell et al., 2018). Even so, the parameters could not be isolated in some cases (Nachenius et al., 2015b) as it was known to be influenced by other operating conditions or reactor design parameters.

Existing reactor types have been explored for torrefaction. Continuously operated reactors developed for biomass torrefaction include rotary drums (Agar et al., 2015; Barbanera & Muguerza, 2020; Granados et al., 2017; Mei et al., 2015; Nhuchhen et al., 2016b; Strandberg et al., 2015), screw-type reactors (Chang et al., 2012; Keivani et al., 2018; Nachenius et al., 2015b; Ohliger et al., 2013; Poskart et al., 2018; Shang et al., 2014), moving bed reactors (Campbell et al., 2018; Nanou et al., 2016), spouted bed reactors (Wang et al., 2018; Wang et al., 2019; Wang et al., 2017b), and fluidized bed reactors with sand particles as the inert bed material (Fleig et al., 2021). However, no single reactor has been recognized as the most suitable when feedstock properties, product applications, and costs are taken into consideration (Chen et al., 2021).

#### **1.2** Application of fluidized beds

### **1.2.1** Fluidization of biomass particles

Fluidized bed reactor as a proven technology is a potential candidate for torrefaction because of its enhanced heat and mass transfer, excellent uniform heating and mixing. However, unconventional nature of biomass, such as high moisture content, wide particle size distribution, low density, and irregular shape is problematic and even causes defluidization. One common solution to facilitate fluidization of biomass particles is to use an inert material, most often sand (Brachi et al., 2019b; Fleig et al., 2021). However, segregation and attrition problems arise, which affect the reliability and efficiency of the fluidized bed process. Moreover, it increases ash content in biomass product, which causes corrosion, sintering, and slagging during its combustion application, leading to additional maintenance for the removal of deposits and even unscheduled shutdowns (Craig et al., 1995). Cui and Grace (2007) reviewed the research on the hydrodynamics and mixing of biomass particles in fluidized beds. It was pointed out that novel and effective configurations of reactors are needed to improve the mixing of biomass particles. Successful fluidization without inert bed materials can be obtained with the assistance of gas pulsation or special gas distributor design only with particles of a narrow particle size distribution.

### 1.2.2 Pulsed fluidized beds

Pulsed fluidization is used among various assisted fluidization techniques to improve the intrinsic performance of fluidized beds. In a pulsed fluidized bed, the gas flow rate is varied periodically with time in the form of regular or irregular patterns; e.g., rectangular wave, saw-tooth wave, or any other pattern (Akhavan et al., 2009). The pulsed fluidized bed has several advantages over the conventional fluidized bed: it reduces the formation of bubbles, slugs, and channels, decreases the minimum fluidization velocity and bed pressure drop, consumes less energy, and fluidizes cohesive particles, which are hard to fluidize in the conventional bed (Machado et al., 2014). Some studies have shown the improvement in the fluidization quality and the enhancement in gas particle contact with pulsating gas flow (Khanali et al., 2014; Nitz & Taranto, 2009; Prachayawarakorn et al., 2005; Reyes et al., 2008). Due to these merits, pulsed fluidized beds have been widely used in drying process (Bubnovich et al., 2008; de Souza et al., 2010; Reyes et al., 2012).

Specifically, biomass particles have been successfully fluidized with the assistance of gas pulsation generated by a solenoid valve in a batch fluidized bed (Jia et al., 2015). Gas-solid flow behaviours at different pulsation frequencies were observed in the transparent reactor. At pulsation frequency of 0.33 Hz, large voids formed at the bottom, quickly merged to slugs and reach the top of the bed. When the pulsation frequency increased to 1 Hz, gas passed the bed through various channels. At 3 Hz, voids turned into bubbles rising to the surface of the bed. As the pulsation frequency further increased to 6.67 Hz, bubble size reduced due to the less excessive gas available during each pulsation. Optimal drying occurred in the range of pulsation frequency between 0.75 Hz and 1.5 Hz, which is in the proximity of the natural frequency of the bed. As a batch reactor for biomass drying, the pulsed fluidized bed has been modified geometrically to improve mass and heat transfer (Jia et al., 2017a), with enhanced heat transfer supported by models (Jia et al., 2017b). However, a continuously operated reactor for biomass processing still needs to be examined.

### 1.2.3 Horizontal fluidized beds

Particles in deep fluidized beds are well-mixed with widely spread residence time distribution of particles during their movement in the continuous operation, resulting in the non-uniform properties of the particle products. To achieve uniform product quality, narrow residence time distribution or plug flow of particles in a continuous fluidized bed is desired (Babu & Setty, 2003). Different types of baffles can be added to divide the deep bed into several compartments, mimic CSTRs in series for particle flow. However, gas pulsation is more effective in shallow beds (Jia et al., 2016). Gas pulsation became weak and could not penetrate bed when the bed was deep. Alternately, horizontal fluidized beds, which features a long, narrow channel through which the fluidized solids flow horizontally, can be applied (Bachmann et al., 2017; Chen et al., 2018; Ramli

& Daud, 2007; Wan Daud, 2008). Bed lengths up to 20 m can be employed, with length-to-width ratios of 4:1 to 30:1 (Baker et al., 2006). Fluidized particles are well mixed vertically and move horizontally along the bed as a plug flow with limited backmixing.

Plug flow fluidized bed dryers have been modelled to predict the moisture content and temperature profiles of the particles. Ramli and Daud (2007) established a model, which was found to underestimate both the solids moisture content and temperature near the solids outlet. Khanali et al. (2013) presented a model of the plug-flow fluidized bed dryer under steady-state conditions and compared with experimental data. Both the simulated horizontal profiles of solid moisture content and temperature showed a sharp change at the dryer inlet as a result of solid back mixing.

By applying the pulsed fluidized bed with a horizontal configuration, a narrower biomass residence time distribution could be achieved for thermal conversion of biomass. As a result, reactor volume to treat the bulky biomass materials could be reduced, leading to reduction in capital cost at large scale.

#### **1.3** Solids transport and backmixing in horizontal fluidized beds

### **1.3.1** Residence time distribution

Solids transport and backmixing have been commonly studied by the measurement of solids residence time distribution (RTD). RTDs of biomass particles have been measured in different types of reactors, i.e., rotary drums (Bongo Njeng et al., 2015a; Colin et al., 2015; Renström, 2008), screw-type reactors (Chamberlin et al., 2018; Nachenius et al., 2015a; Sievers et al., 2016; Youssef et al., 2017), stirred bed reactors (Xi et al., 2015a), and horizontal fluidized bed reactors (Bachmann et al., 2016). Effects of design parameters, flowability of materials and operating

conditions on solids RTD profile and mean residence time have been investigated, which are essential for the design and optimization of the reactor. Experimental mean residence time for a pulse injection of tracer can be determined from RTD curves by Eqs. (1.1) and (1.2) (Fogler, 2017):

$$E(t) = \frac{C(t)}{\int_0^\infty C(t)dt}$$
(1.1)

$$t_m = \frac{\int_0^\infty tE(t)dt}{\int_0^\infty E(t)dt} = \int_0^\infty tE(t)dt$$
(1.2)

where E(t) is the residence-time distribution function, C(t) is the concentration of tracer at time t, and  $t_m$  (s) is mean residence time.

Solids holdup measurement is supplementary and often conducted along with solids RTD tests. In a horizontal stirred reactor, for example, Xi et al. (2015a) found different designs of mixing blades and flowability of materials considerably influenced the particle bed inclination angle and solids holdup. The solid holdup in a screw conveyor, which corresponds to the degree of filling, could be manipulated by adjusting the screw speed (Nachenius et al., 2015a; Waje et al., 2007). Bongo Njeng et al. (2015b) investigated the effects of internal lifter shape and exit dam height on mass holdup of sand and rice particles in a rotary kiln. On the other hand, in fluidized beds, differences in solids holdups with certain bed geometry mainly result from the various extent of bed expansion at different operating conditions (Bachmann et al., 2017; Geldart, 2004). Particularly in gas-solid crossflow configuration, a deeper bed with a higher holdup may impose more solids mixing by gas bubble movements through the bed. Increased lateral mixing and particle dispersion were reported with the increase in the bed height (Bachmann et al., 2017; Nilsson & Wimmerstedt, 1988; Sánchez-Prieto et al., 2017).

#### 1.3.2 Evaluation and modeling of solids mixing

Solids backmixing can be quantified through model fitting of RTD curves (Danckwerts, 1953). Tank-in-series model and axial dispersion model (ADM) have been widely applied for RTD curvefitting, accounting for the deviation of practical solids flow from the well-mixed and ideal plug flow, respectively. The ADM is written mathematically as

$$\frac{\partial C}{\partial t} = D_x \frac{\partial^2 C}{\partial x^2} - u_x \frac{\partial C}{\partial x}$$
(1.3)

where  $D_x$  (m<sup>2</sup>/s) denotes the particle dispersion coefficient and  $u_x$  (m/s) is the solids velocity. Admitting sometimes these two types of models are interchangeable (Levenspiel, 1979; Youssef et al., 2017), solids flow in horizontal shallow fluidized beds is better characterized by ADM with closed-closed boundary conditions (Bachmann et al., 2016; Kong et al., 2018; Nilsson & Wimmerstedt, 1988). Kong et al. (2018) conducted solids RTD tests of cristobalite powder in a horizontal shallow fluidized bed with and without baffles. ADM, tank-in-series model, and nonideal compartment model were used to fit the RTD data, and ADM appeared to give the best fit within tested range with Peclet number over 25.

Another merit of the ADM is that the dispersion term in Eq. (1.3) describes the deviation from the plug flow behaviour as backmixing and quantifies the mixing intensity. Several studies (Bachmann et al., 2016; Bongo Njeng et al., 2015a; Nachenius et al., 2015a; Nilsson & Wimmerstedt, 1988; Xi et al., 2015b) on solids RTDs in various reactors further established empirical correlations between dispersion coefficient and influencing factors, allowing reasonable estimations of solids backmixing behavior under chosen conditions for the specific case studied. Nilsson and
Wimmerstedt (1988) measured RTDs of sand and apatite in four sizes in a pilot-scale horizontal shallow fluidized bed. By fitting ADM to experimental RTD curves, values of horizontal dispersion coefficients were obtained, and a correlation was established as Eq. (1.4):

$$D_{x} = \frac{1.49 [0.01(H_{bed} - 0.05) + 0.00165 \rho_{g} (U - U_{mf})] u_{x}^{0.23}}{U_{mf}^{1/3}}$$
(1.4)

which takes the effects of bed height  $H_{bed}$  (m), gas density  $\rho_g$  (kg/m<sup>3</sup>), minimum fluidization velocity  $U_{mf}$  (m/s), gas velocity U (m/s) and solids flow rate into account. Bachmann et al. (2017); Bachmann et al. (2016) tested RTDs of gamma alumina particles with two sizes in lab-scale and pilot-scale horizontal fluidized beds with and without baffles. ADM was also used. Eq. (1.5) was deduced with the effect of bubble size being considered. In Eq. (1.5), L (m) and W (m) are the length and width of the bed.  $d_{b}$ ,  $d_{b,in}$ ,  $d_{b,m}$  (m) denote bubble size, initial bubble size at the distributor plate, maximum attainable bubble diameter, respectively. F (kg/s) is solids feed rate and  $U_{elu}$  (m/s) is the elutriation gas velocity.

$$D_{\chi} = u_{\chi}L \varepsilon \left(\frac{W}{L} \cdot \frac{H_{bed}}{L}\right)^{\varepsilon} \frac{d_b}{d_p} \left[\frac{A \rho_g (U - U_{mf})}{F}\right]^{1/2} \left(\frac{d_{b,in}}{d_{b,m}}\right)^{0.15} \left(\frac{U_{mf}}{U_{elu}}\right)^{0.75}$$
(1.5)

Considering the gas-solid cross flow, horizontal mixing in horizontal fluidized beds is analogous to the lateral mixing in the vertical bed, subjected to the impact of horizontal bulk solids flow. Horizontal mixing in vertical beds was studied by fitting the mixing curve of a batch tracer with a Fickian-type diffusion equation (Bellgardt & Werther, 1986; Liu & Chen, 2011; Niklasson et al., 2002; Pietsch et al., 2020; Sette et al., 2015). The horizontal dispersion coefficient reported in the literature scattered from  $1 \times 10^{-4}$  to  $0.1 \text{ m}^2/\text{s}$  (Hua et al., 2019). The complex horizontal mixing

phenomenon in fluidized beds is often attributed to the gas bubble motion. Bubbles moving upward and laterally through the bed, erupting at the surface of the bed, as well as gross solids movement and circulation in the bed, give rise to the horizontal mixing (Shen & Zhang, 1998; Shen et al., 1995; Shi & Fan, 1984).

The pulsed fluidized bed is proven to be effective in biomass drying, but it has not been demonstrated and validated for biomass torrefaction. Despite the extensive applications of horizontal fluidized beds, gas pulsation has never been incorporated to assist fluidization of irregularly shaped particles, e.g., biomass particles. Control of the bed temperature and solids residence time in the horizontal pulsed fluidized bed is also not clear due to the lack of understanding in solids transport and backmixing. Effects of gas pulsation frequency, temperature, and feed rate on biomass torrefaction performance in the unique reactor are unknown.

## 1.4 Research objectives

The objective of this research is to develop and evaluate a continuously operated horizontal pulsed fluidized bed torrefaction reactor system, for efficient production of high-quality torrefied material from local biomass resource. Specific research tasks are,

- Design a pre-pilot horizontal pulsed fluidized bed reactor for biomass torrefaction that can fluidize biomass particles with a wide size distribution, without inert bed materials.
- Construct and commission a continuously operated biomass torrefaction system, including preheaters, gas pulsation generation devices, a horizontal pulsed fluidized bed torrefier, a catalytic combustor to eliminate volatiles in torrefaction flue gas, cooling system, and control and data acquisition devices.

- Measure the solids residence time distributions at various operating conditions at room temperature to characterize solids mixing in horizontal pulsed fluidized bed.
- Develop a model to quantify the horizontal solids mixing and evaluate the effects of gas pulsation frequency, feed rate, and superficial gas velocity on solids mixing.
- Investigate the effects of gas pulsation frequency, temperature, and feed rate on biomass torrefaction performance of the pulsed fluidized bed system, so as to identify the most effective pulsation frequency and other operating conditions.
- Operate the system with fluidizing gas of different controlled oxygen concentrations, compare the torrefied product properties to assess the feasibility of using oxygen-lean flue gas as fluidizing gas for biomass torrefaction in the pulsed fluidized torrefier.

#### 1.5 Thesis layout

The thesis consists of five chapters. Chapter 1 reviews the biomass torrefaction process, explores the possibility of applying fluidized bed for biomass thermal conversion, identifies the knowledge gap, and outlines the objective and specific tasks of this work. Chapter 2 describes the experimental setup, material properties, experimental procedures, and characterization methods. Chapter 3 presents the experimental results of solids residence time distributions and a dispersion model to quantify solids backmixing in the horizontal pulsed fluidized bed. Minimum fluidization velocity and pressure fluctuations are also reported in this Chapter. Chapter 4 evaluates the performance of the pulsed fluidized bed torrefier, with the effect of key operating parameters such as pulsation frequency, temperature and biomass feed rate on product quality investigated. Finally, Chapter 5 concludes the key findings from this work and suggests future studies.

# **Chapter 2: Experimental Methods**

To achieve the research goal proposed in the previous chapter, experimental and modelling works were carried out. A prototype horizontal fluidized bed reactor for biomass torrefaction was fabricated by the department workshop. The fluidized bed torrefaction unit with continuous solid feeding and discharging was commissioned at the Clean Energy Research Center at the University of British Columbia. To better understand the product distribution, solid flow and mixing were characterized by solid residence time distribution measurement and modelling, with the effects of weir height, pulsation frequency, feed rate, and gas velocity investigated. Reactor performance was evaluated by a parametric study of sawdust torrefaction, with effects of bed temperature, feed rate, pulsation frequency, and oxygen content in the carrier gas investigated. Torrefaction products were characterized by proximate analysis, ultimate analysis, and high heating value measurement. Horizontal pulsed fluidized bed reactor

A fluidized bed reactor with a narrow rectangular cross-section was fabricated in carbon steel for biomass torrefaction. Dimensions of the reactor are illustrated in Figure 2.1. The reactor height was 521 mm, whereas the bed weir height was 63.5 mm for torrefaction tests. The cross-section was 254 mm×76.2 mm. The high length to width ratio was selected to create a horizontal solid flow close to a plug flow. Biomass particles entered from the feed port and exited through the overflow weir at the opposite end. Particles then dropped through a 362 mm long discharge leg into a product receiver. Measurement and observation ports were reserved on the top plate.







Figure 2. 1 Dimensions of the horizontal pulsed fluidized bed reactor. (a) 3D view and features of the reactor; (b) Top view, section view, and front view of the reactor.

Windbox was packed with 9.5 mm ceramic spheres to aid the distribution of fluidizing gas. Meanwhile, reduced gas volume in the windbox had a low damping effect on gas pulsation. A perforated plate with 4.8 mm diameter orifices and 26% opening ratio was fabricated and installed between the bed and windbox as the gas distributor. A fine mesh was pressed by a rectangular ring to the distributor to prevent small particles from falling into the windbox. The unorthodox high opening ratio, thus a low-pressure drop, was chosen again to minimize the damping of gas pulsation. Since the gas was introduced to the windbox through a single port located in the middle of the windbox bottom plate, the orifices were not arranged uniformly on the distributor plate to achieve a relatively even gas flow across the entire bed cross-section. Local opening ratios of different distributor sections were determined by preliminary simulation (Appendix A1), considering of the effects of the distributor and the windbox. Arrangement of the triangle-patterned orifices is shown in Figure 2.2, with the lowest opening ratio of 4% in the central region and up to 35% opening ratio at two ends of the bed.



Figure 2. 2 Orifice arrangement on the distributor.

#### 2.1 Fluidized bed torrefaction unit

The fluidized bed torrefaction unit with the horizontal pulsed fluidized bed reactor is depicted in Figure 2.3. Liquid nitrogen at 100 PSI was used to provide the main carrier gas. Building compressed air after being regulated was mixed with nitrogen gas to provide the oxygen content. Rotameters (FL4513-V for N2 and FL4511-V for air, Omega Engineering, USA) with needle valves were installed in both the nitrogen and air gas lines to monitor and control the flow rates. A 75.7 L (20 Gallon) buffer tank (ASME-Code, max. pressure: 200 PSI) was placed between the rotameters and the solenoid valve to eliminate the oscillation at the rotameters and sustain the gas supply for pulsation. The gas was then heated by a pair of electric semi-cylinder heaters (Watlow VS105A18S, Thermal Devices, USA). The heated gas line was wrapped with heavy-duty insulated heating tapes (STH052-100, Omega Engineering, USA). A solenoid valve (DN10, normally closed, model number: AXDC-510110M-H, Xi'An Aoxin Automation Instrument, China) that could stand a temperature as high as 350 °C was installed to generate gas pulsations. The distance between the solenoid valve and windbox was kept at a minimum to preserve the intensity of pulsating gas.





(b)

Figure 2. 3 (a) Schematic of the pulsed fluidized bed torrefaction unit; (b) Photo of the pulsed fluidized bed torrefaction unit.

Biomass particles were fed by a screw feeder (WLS-14\*400 mm, Zhenjiang Jincan Machinery, China), with a linear correlation of feed rate and motor frequency calibrated at room temperature. A small amount nitrogen gas stream was introduced to the feeder hopper to maintain slightly positive pressure inside, and along with a water-cooling jacket to prevent heat in the reactor from flowing back to the feeding screw and the hopper. Biomass particles moved horizontally, suspended with the fluidizing gas, and reacted. Solid products overflowed from the bed and collected to the product receiver, a 37.9 L (10 Gallon) steel drum. A solid product sampling port was available on the discharging pipe. Fine particles entrained were retained by a pleated sintered metal filter (customized, Anping KUNYA Wire Mesh Products, China) installed at the gas exit inside of the reactor, which was purged periodically by nitrogen gas with a back pressure of 50 PSI. The purging gas line was inserted into the filter and equipped with a solenoid valve (3/8 in, ASCO 8210G073, Pacific Controls, Canada). In order to further heat up the bed and compensate for the heat loss of the reactor, two pairs of flat electric heaters (Watlow VF514A18S for the bed; Watlow VF704A06S for the windbox, Thermal Devices, USA) were placed on the front and back sides of the bed and windbox. All heated parts were wrapped with one-inch-thick ceramic fibre insulation layer to reduce the heat loss.

Torrefaction gas entered a tubular reactor, located immediately after the gas exit of the torrefier, with 580 g palladium on alumina catalyst (Strem Chemicals, USA) to aid combustion of volatile organic compounds released from biomass torrefaction. Catalyst properties are listed in Table 2.1. Bulk density was determined by a 100 ml glass gradual cylinder. BET surface area was measured by an ASAP 2020 analyzer (Micromeritics, USA). The reactor is 102 mm (4 in) in diameter and 508 mm (20 in) high, with meshes at each end to prevent packing materials from falling to the pipe

or being carried out by the gas. The lower 127 mm (5 in) of the reactor was packed with 9.5 mm (3/8 in) ceramic balls, and the rest was filled with catalyst particles dispersed in 6×6 mm ceramic Raschig rings. Auxiliary air was injected at 100 mm above the reactor bottom to provide oxygen for the combustion. The catalytic combustor was also heated by a pair of electric heaters (Watlow VS105A12S, Thermal Devices, USA) at the start-up stage of each run to ignite the gas. The water-cooling system then cooled flue gas with a condenser to trap water and tars that had not been destroyed before ventilation.

Table 2. 1 Properties of catalyst in the combustor.

Catalyst	Looding	Shana	Bulk density, BET su	
Catalyst	Loading	Snape	kg/m <sup>3</sup>	area, m²/g
Dalladium on alumina	0 5 wt %	3.175 mm	085	101.9
i anadium on aiumma	0.5 wt.70	(1/8 in) pellet	985 1/8 in) pellet	

## 2.2 Instrumentation and data acquisition

To monitor and record the operation of the pulsed fluidized bed torrefaction unit, thermocouples and pressure sensors were installed at various locations on the unit, and data were collected through the data acquisition system by a computer. In the meantime, the actuation of solenoid valves for fluidizing gas pulsation and filter purging gas was controlled by the computer as well. Each heating element was separately controlled. Gas sampling and analysis instruments were attached to the system to analyze the gas composition online.

# 2.2.1 Temperature measurement

The temperatures of preheated gas, windbox, biomass bed, catalyst bed, and flue gas were measured by K-type thermocouples, as labelled in Table 2.2.  $T_1-T_6$  were measured by single point thermocouples, whereas  $T_8-T_9$  were measured by a profile thermocouple with the three measurement points located 102 mm (4 in), 203 mm (8 in), and 305 mm (12 in) from the top of the combustor, respectively. The thermocouples were 3.175 mm (1/8 in) in diameter so as to maintain the structural strength with an acceptable response time. Thermocouple specifications are summarized in Table 2.3. A PCI-DAS08 analog input board (Measurement Computing, USA) read and converted the voltage signals from the thermocouples to digital forms through an analog multiplexer board (CIO-EXP16, Measurement Computing, USA), which amplified the original voltage signal one hundred times. Temperatures were displayed in real-time by a LabVIEW program (Appendix A2).

Label	Temperature	Label	Temperature
T <sub>1</sub>	Preheated gas	T <sub>6</sub>	Torrefaction gas
$T_2$	Windbox	$T_7$	Bottom region of the catalyst bed
$T_3$	Dense bed, ~13 mm from the solid inlet	$T_8$	In the middle of the catalyst bed
$T_4$	Dense bed, in the middle	<b>T</b> 9	Top region of the catalyst bed
<b>T</b> <sub>5</sub>	Dense bed, ~13 mm from the weir	$T_{10}$	Flue gas

Table 2. 2 Temperature measurements in the pulsed fluidized bed torrefaction unit

Sensor	Range	Error	Response time	Sampling rate	
K type	-200 – 1250 °C	Greater of 2.2 °C	< 0.75 s in water	1 Hz	
thermocouple	-200 1250 C	or 0.75%	< 0.75 5 III water	1 112	

# 2.2.2 Pressure measurement and data processing

Pressure transducers were installed at various locations to measure either absolute gas pressure or pressure drop across a segment of the unit. Model numbers (Omega Engineering, USA) for each measurement are listed in Table 2.4. All transducers were calibrated with a U-tube monometer prior to installation. For the transducers of  $P_2$ – $P_4$ , a fine mesh was inserted in the connecting tube to prevent particles from entering the sensor. Pressure data were logged by NI USB-6008 DAQ device (National Instrument, USA), which was connected to a computer. Unlike thermocouples, pressure transducers have a fast response time of 1 ms without compromising the robustness. A much higher sampling rate of 200 Hz was used to capture the dynamic pressure fluctuations in the fluidized bed, which also enabled early detection of an instantaneous fluctuation in gas and solid flow. Besides the pressure transducers, pressure gauges were also installed after the rotameters for flowrate calibration; on the torrefier to aid the pressure monitoring, in case of any transducer failure; on the screw feeder hopper and the product receiver to be checked regularly during the test to ensure proper solid flow. In addition, pressure relief valves were installed on the buffer tank, the hopper, and the reactor with settings of 50 PSI, 10 PSI, and 10 PSI, respectively.

Label	Measurement	Range	Model number
$\mathbf{P}_1$	Absolute pressure before the buffer tank	0–30 PSI	P142-030A5V
$P_2$	Absolute pressure in the freeboard	±2.5 PSI	P143-2.5BD5V
<b>P</b> <sub>3</sub>	Pressure drop between the windbox and free board	±0.36PSI	PX164-010BD5V
<b>P</b> <sub>4</sub>	Pressure drop of the filter	±1PSI	PX143-01BD5V
<b>P</b> <sub>5</sub>	Pressure drop of the combustor	±1PSI	PX143-01BD5V

Table 2. 4 Pressure measurements in the pulsed fluidized bed torrefaction unit

Finally, a LabVIEW program (Appendix A2), which included the pressure transducer calibration function and flowrate calibration function according to the manufacturer with an input of pressure reading from the gauges, was written to calculate superficial gas velocity under certain pressure and temperature, and display pressure readings in real-time.

Due to the difficulty of setting up pressure measurement just above the distributor in the continuously operated bed without disturbing the solid flow, bed pressure drop, which is of great interest in fluidized beds, was not directly measured. Instead, the bed pressure drop was calculated as the difference of measured pressure drop across the distributor and the bed (P<sub>3</sub>) and the pressure drop of the empty bed at the same gas velocity. The original P<sub>3</sub> data contained the noise from the freeboard and product container, as shown in Figure 2.4. A Fast Fourier transform (FFT) of the original signal was performed, and results showed components at over 20 Hz with an amplitude comparable to the dominant pulsation frequency. A low-pass FIR filter with Hanning window function in the DIAdem software (National Instruments, USA) was thus applied to filter the signals with a frequency higher than 15 Hz. After the adjustment, the pressure fluctuation pattern of the batch fluidized bed was very similar to the result reported by Jia et al. (2016).



Figure 2. 4 Eliminating noises from the measured pressure signal.

# 2.2.3 Solenoid valve

Two different LabVIEW programs were created to control the main gas pulsation valve and the filter purging gas valve, respectively. Control of solenoid valve actuation is illustrated in Figure 2.5. High and low state digital outputs corresponded to the solenoid valve open and close states were sent to an electrical relay through NI-6051DAQ device (National Instrument, USA).

The open and close time for each pulse was set to be the same for the gas pulsation valve at any pulsation frequency. As to the filter purging gas valve, it opened every 30 s for 0.2 s.



Figure 2. 5 Solenoid valve control diagram.

#### 2.2.4 Heater

As shown in Figure 2.3 and stated in Section 2.2, in order to heat the unit and make up for the heat loss, ceramic fiber heaters were placed on the preheater, windbox, fluidized bed and combustor, and heating tapes were employed for gas lines. Ceramic fiber heaters were controlled by temperature controllers (1/16 DIN controllers: 2×CN77553, Omega Engineering, Canada, 2×CAL #9300, Cole-Parmer, Canada) integrated into a control box as shown in Figure 2.3. Each heating tape was controlled by a portable controller (LOVE CONTROLS SERIES 2500, 2×LOVE CONTROLS SERIES 4B, DWYER Instruments, USA), as shown in Figure 2.6. The surface temperature of each heated fragment was set up as the input. PID parameters for each controller were set by running autotune function at a typical experimental operating condition.



Figure 2. 6 Portable controllers for heating tapes.

## 2.2.5 Gas sampling and analysis

Although gas is not the primary product of this study, analysis of permanent gas contents can provide information on the combustor's performance. The gas sampling and analysis system attached to the unit is illustrated in Figure 2.7. An empty 500 ml Erlenmeyer flask was used as a safety buffer between the cold-water trap in a 1000 ml Erlenmeyer flask and the gas inlet. After being washed and dried, torrefaction gas or combustion flue gas, only one gas at a time, was conditioned by a HORIBA sampling unit and fed to a flue gas analyzer (PG-250, HORIBA, Japan) at a rate of 0.25 L/min. Sufficient time was given for each switch between input gases to purge the buffer flask. In order to confirm the oxygen concentration controlled by adjusting the nitrogen to air flowrate ratio in the fluidizing gas, the windbox was connected directly to the gas analyzer with a fast response.



Figure 2. 7 Schematic of gas sampling and analysis system.

#### 2.3 Materials

Two locally sourced biomass species were used in this work: spruce-pine-fir (SPF) sawdust obtained directly from the Tolko's sawmill at Merritt, BC and hemlock sawdust provided by Teal-Jones Group, Surrey, BC.

## 2.3.1 Preparation and characterization

Received samples were first ground by a hammer mill (10HMBL, Glen mills, Clifton, NJ, USA) with a 3.18 mm (1/8 in) screen, then dried in an oven (LHT6, Carbolite Gero, Sheffield, UK) at 105 °C for 24 h and stored in a zip bag to keep the moisture out. Treated samples are shown in Figure 2.8.



Figure 2. 8 Ground and dried (a) SPF sawdust and (b) hemlock sawdust.

Properties of raw materials are listed in Table 2.5. Bulk density was measured by a 500 ml graduated glass cylinder (ASTM D7481-09). Particle size distribution was determined by sieving, and the averaged result of three parallel tests for each material is shown in Figure 2.9. It is worth noting that the ground particles are elongated disc-shaped, as shown in Figure 2.8. The particle size determined by sieving is close to the particle width (Jafari Naimi, 2016). Sauter mean diameter was calculated by Eq. (2.1),

$$d_p = \sum_{i=1}^{n} x_i d_{pi}^3 / \sum_{i=1}^{n} x_i d_{pi}^2$$
(2.1)

where  $x_i$  is the mass fraction of particles with a nominal diameter  $d_{pi}$ . Hemicellulose, cellulose, lignin, and extractives in the raw samples were determined followed TAPPI methods at State Key Laboratory of Biobased Material and Green Papermaking at Qilu University of Technology, China. Higher heating value (HHV), proximate analysis, composition analysis, and ultimate analysis are described in Section 2.5.2.

	SPF	Hemlock
Bulk density, kg/m3	205	187
Sauter mean diameter, mm	0.59	0.70
HHV, MJ/kg	19.60	19.62
Composition analysis <sup>d</sup>		
Hemicellulose, wt.%	26.3	32.3
Cellulose, wt.%	45.2	38.9
Lignin, wt.%	24.9	27.8
Extractives, wt.%	3.2	0.7
Proximate analysis <sup>d</sup>		
Volatile matter, wt.%	82.6	83.4
Fixed carbon*, wt.%	16.8	16.3
Ash, wt.%	0.40	0.30
Ultimate analysis <sup>d</sup>		
C, wt.%	49.8	50.2
H, wt.%	6.3	6.5
O*, wt.%	43.9	43.3
N, wt.%	< 0.1	< 0.1

Table 2.5 Properties of raw materials.

<sup>d</sup> Dry basis \* Value was determined by difference.



Figure 2. 9 Particle size distribution of tested materials.

## 2.3.2 Apparent minimum fluidization velocity

Minimum fluidization velocity is an essential parameter for fluidization and crucial for bed behaviour analysis. Meanwhile, the measured pressure drop versus gas velocity curves can be used to diagnose bubbling fluidized bed conditions (Yang et al., 2003). A decrease in minimum fluidization velocity was found in pulsed fluidized beds, compared to that in steady gas flow beds (Ireland et al., 2016). However, severe channelling was observed when fluidization velocity was only measured in this study with steady gas flow. Therefore, minimum fluidization velocity was only measured with gas pulsation using mean pressure drop and gas velocity, because both pressure drop and gas velocity oscillate. Mean pressure drop was used in most of the studies (Al-Ghurabi et al., 2020; Khosravi Bizhaem & Basirat Tabrizi, 2013; Reyes et al., 2007), while peak pressure (Jia, 2017), mean pressure of pulsation on-period (Ali & Asif, 2012), and calculated  $U_{mf}$  (Liu et al., 2017) were also used in previous studies. In this work, time-averaged pressure drop was plotted 33

against the time-averaged superficial gas velocity calculated by dividing the measured flow rate by the bed cross-section.

Hemlock was tested under both batch and continuous modes. For the batch test, pressure drop across the bed was directly measured by a differential pressure transducer (PX164-010BD5V, Omega Engineering, USA). Initial biomass load of 200 g and 400 g and pulsation frequency of 1, 2, 4 Hz were used. For biomass particles with a wide size distribution as shown in Section 2.4.1, the pressure drop gradually changes from the fluidization condition to the fix bed condition as the gas velocity decreases. The minimum fluidization velocity could be defined as the intersection of the linear extrapolation of the two sections at low and high gas velocities (Chiba et al., 1979). In the continuous mode, where sawdust was fed from one side and discharged at the opposite side continuously, the minimum fluidization velocity was identified at peak pressure drop. Both fluidization (velocity ascending) and defluidization (velocity descending) procedures were performed for each case in both modes. After adjusting the gas flowrate, sufficient time, at least 5 min, was given for the bed to stabilize, and 2 min data were recorded subsequently for the averaged values to be calculated. Results from the two modes were consistent, with a relative error of 2.3%. Since the continuous mode was close to the torrefaction test conditions, results from the continuous mode were adopted, which was 0.085 m/s for hemlock and 0.094 m/s for SPF. Detailed experimental results and discussion can be found in Chapter 3, Section 3.1.1.

## 2.4 Torrefaction test

To investigate the effects of operating conditions on reactor performance, torrefaction of SPF sawdust was conducted under various biomass feed rates, reactor temperatures, and gas pulsation frequencies, whereas hemlock was used to study the effect of oxygen content.

## 2.4.1 Procedure

Prior to each run, ~6 kg sawdust was loaded in the hopper, noted as  $w_{hi}$ , after which the hopper purging nitrogen was kept at 4 SCFM and water was kept running through the screw cooling jacket all the time until the run was finished, and the unit was cooled. The unit without particles was preheated by pulsating air for 2–2.5 hours. The empty bed was preheated to a temperature 15– 25 °C higher than the target bed temperature. Once the temperatures stabilized at the set points, carrier gas was switched to nitrogen and purged for 10 min to replace the air in the system. Auxiliary air to the combustor was introduced, and filter back purging gas was turned on before the feeding of particles.

Biomass was fed at time zero with a pre-set feed rate. Each run lasted for 150 min, which was limited by the hopper capacity. If the bed temperature  $T_{bed}$ , defined as the average temperature of T<sub>4</sub> and T<sub>5</sub> located in the middle of the bed and the end of the bed, did not reach the target range, temperature settings of the preheater and reactor heaters could be adjusted. Extra time was given for the bed to stabilize after heater adjustments. The gas flowrate was adjusted correspondingly to the bed temperature to keep the average superficial gas velocity constant. A total of 15 solid product samples of around 2 g each time were drawn through the solid sampling port at 5 min intervals after 15 min of feeding and 10–20 min intervals as the experiment progressed. Besides

the torrefaction bed temperatures, the fixed bed combustor temperatures were also closely monitored to prevent the catalyst bed from severe sintering. Once the combustor temperatures exceed 600 °C (Argyle & Bartholomew, 2015), air flow was reduced to limit the combustion.

In the oxidative torrefaction test, air was mixed in the nitrogen gas to make up for the target oxygen content. The volume ratio of air to nitrogen was varied from 16% to 40%, corresponding to the oxygen concentration of 3–6 vol.% in the fluidizing gas. The oxygen content of the mixed gas at the windbox was measured through a gas sampling system (details are provided in Section 2.3.5). In order to avoid the heat runaway in the torrefier, heaters on the fluidized bed were turned off when the reaction began. Gas sampling, where O<sub>2</sub>, CO<sub>2</sub>, and CO contents were determined, started when the process stabilized, typically after 30 min.

At the end of the run, heaters were turned off, and gas and solid feed stopped. The solid product in the product receiver was weighed as  $w_p$ , most of which was stored in heavy-duty air-tight plastic bags, with a small amount sealed in a jar for future characterization. Nitrogen gas was reduced and directed to the steady bypass line to cool the reactor, while the combustor was kept at an elevated temperature to burn off volatiles from the residues. Nitrogen gas was cut off when the bed temperature became lower than 200 °C. After the reactor was cooled to room temperature overnight, the remaining raw sawdust in the hopper was taken out and weighed as  $w_{hr}$ . The actual feed rate was determined by the amount fed divided by the duration of feeding. Particles in the reactor were also taken out and weighed as  $w_r$ . Therefore, the weight loss of each run was defined as,

$$W_L = (1 - \frac{w_p + w_r + w_s}{w_{hi} - w_{hr}}) \times 100\%$$
(2.2)

where  $w_s$  is the weight of all solid samples taken during the run.

All parameters studied in the torrefaction test are summarized in Table 2.6. Preliminary tests showed that severe channelling and bridging occurred when the bed was operated with steady gas flow. Thus, all tests were conducted with pulsating gas flow. Gas pulsation frequency  $f_P$  of 1–5 Hz was chosen to be around the natural oscillation frequency of 2.0 Hz. The natural frequency  $f_n$  (Hz) was calculated using Eq. (2.3) given by Hao and Bi (2005),

$$f_n = \frac{1}{2\pi} \sqrt{\frac{g}{H_{mf}}}$$
(2.3)

treating fluidized bed system as a second-order vibration system. Bed height at minimum fluidization velocity  $H_{mf}$  (m) was approximated as the weir height. Bed temperature  $T_{bed}$  was defined as the average temperature of T<sub>4</sub> and T<sub>5</sub>, located in the middle of the bed and the end of the bed, and used for the superficial gas velocity calculation. Feed rate *F* was set to be 1, 1.5, and 2 kg/h. The superficial gas velocity *U* (m/s) used in all experiments was kept over two times the measured minimum fluidization velocity at room temperature. According to Eq. (2.4) (Kunii et al., 1991),

$$U_{mf} = \frac{d_p^{\ 2}(\rho_p - \rho_g)g}{1650\mu_g} \tag{2.4}$$

for small particles, as temperature increases, gas density  $\rho_g$  (kg/m<sup>3</sup>) decreases, gas viscosity  $\mu_g$  (Pa·s) increases, and the difference between particle density and gas density does not change considerably. Hence, the minimum fluidization velocity is expected to be reduced. The gas velocity was selected to be high enough to ensure a good fluidization quality without severe channelling and partial defluidization. The operating range of oxygen content *Xoz* was also reasonably selected to represent the typical flue gas, which has an oxygen content of 2–5 vol.%, depending on the fuel used (Song et al., 2004). Certain experiments were repeated twice to check the reproducibility.

Parameter	Material	Operating conditions
Pulsation frequency <i>f</i> <sub>p</sub>	SPF	$f_P=1, 2, 3, 4, 5$ Hz; $T_{bed}=270$ °C; $F=1, 1.5$ kg/h; $U=0.2$
		m/s, $X_{02} = 0$ vol.%
Bed temperature <i>T<sub>bed</sub></i>	SPF	$T_{bed}$ =240, 270, 300, 330 °C; $F$ =1, 1.5, 2 kg/h; $f_P$ =2 Hz;
		$U = 0.2 \text{ m/s}, X_{02} = 0 \text{ vol.\%}$
	Hemlock	$T_{bed}$ =240, 270, 300 °C; $F$ =1.5 kg/h; $f_P$ =2 Hz;
		$U = 0.2 \text{ m/s}; X_{02} = 0 \%$
Feed rate F	SPF	$F = 1, 1.5, 2 \text{ kg/h}; T_{bed} = 240, 270, 300, 330 ^{\circ}\text{C};$
		$f_P = 2$ Hz; $U = 0.2$ m/s, $X_{O2} = 0$ vol.%
Oxygen content X <sub>02</sub>	Hemlock	$F=1.5$ kg/h; $f_P=2$ Hz; $U=0.2$ m/s, $X_{02}=0, 3, 6$ vol.%

Table 2. 6 Summary of experimental operating conditions for the torrefaction test

## 2.4.2 Solid product characterization

Solid product, the primary product of torrefaction, was characterized by proximate analysis, element analysis, and HHV, to evaluate the performance of the reactor. The volatile matter of the biomass samples was measured either by a SHIMADZU TA-60WS TGA or a TGA 550 (TA Instruments, USA), with a heating program of heating the sample to 110 °C at a rate of 10 °C/min, holding for 10 min to remove the moisture, heating to 800 °C at 40 °C/min, and holding for 30 min. The total weight loss of the dried sample represented the volatile matter content. Ash content was determined in a muffle furnace, where the sample was heated up to 575 °C with a heating program following NREL/TP-510-42622 method and kept there for 3 h. Fixed carbon content was subsequently determined by difference. Ultimate analysis, which reports elemental weight percentages of carbon, hydrogen, nitrogen, and oxygen (by difference) on dry basis, was measured by a Carlo Erba EA 1108 elemental analyzer in the Department of Chemistry at UBC or carried out at Econotech Services, Delta, BC. The elemental analysis is based on the complete and instantaneous oxidation of the sample. The HHV of the samples was determined by a Parr 6100 bomb calorimeter, with dried pellets made of raw and torrefied biomass particles. The color of the solid samples collected during the experiment was quantified by a KONICA MINOLTA CM-5 spectrophotometer using three color parameters (L\*, a\* and b\*). The color value change for each torrefied sample compared to the raw sawdust was calculated by Eq. (2.5),

$$\Delta E = \left[ (\Delta L *)^2 + (\Delta a *)^2 + (\Delta b *)^2 \right]^{\frac{1}{2}}$$
(2.5)

where  $\Delta E$ ,  $\Delta L^*$ ,  $\Delta a^*$  and  $\Delta b^*$  represent overall color value change and color value change of three parameters (L\*, a\*, and b\*).

Each characterization experiment was conducted at least twice with the averaged value reported. Due to the long-time span of all the pre-pilot torrefaction tests, two different instruments were used for the measurement of some properties. To ensure consistency, both instruments were calibrated and cross-checked.

### 2.5 Solid residence time distribution measurement

A simple method to measure the solid residence time distribution (RTD) under room temperature, tailored for hemlock sawdust particles in the pulsed fluidized bed reactor, was developed. A solid tracer was prepared from the raw material, and tracer concentration at the outlet was measured offline. Effects of weir height, pulsation frequency, feed rate, and gas velocity on RTD curves and solid mixing were investigated.

## 2.5.1 Tracer preparation and detection

Tracer was prepared by impregnating dried sawdust with solutions containing 30 g sodium chloride per 100 mL deionized water and then dried at 105 °C for 24 h again. Preliminary tests showed impregnated sawdust generally gained around 18% of the original weight with a similar weight ratio of large, medium, and small particles (Appendix B). Therefore, the physical property of the dedicated tracer was close to that of the raw material.

Each collected sample was well mixed, 10 g of which was taken and soaked in 100 ml deionized water to dissolve salt carried by the tracer. Sawdust particles were filtrated out to eliminate the

noise in the subsequent conductivity measurement. The conductivity of the same amount of dried raw sawdust solution without tracer was measured by a conductivity transmitter (CDTX90 with CDCE-90-01 sensor, Omega Engineering, USA) as the baseline. Collected sample solutions were corrected by subtracting the baseline. Despite that the conductivity of NaCl aqueous solutions does not have a linear relationship with the mass concentration (Rumble & ChemnetBase, 2018), the relationship between tracer mass percentage and solution conductivity was assumed to be linear over the range of this work, according to the preliminary test results shown in Appendix B. Thus, the measured solution conductivity could represent the tracer concentration in each sample.

### 2.5.2 Experimental setup

The modified PFB setup for solid RTD tests is depicted in Figure 2.10. This setup was also used for hydrodynamic tests. Only compressed air was used as the fluidizing gas. Sawdust was fed and discharged in the same manner described in Section 2.2. Instead of being collected by the product receiver, sawdust at the outlet fell into sample collection containers. The initial bed weir was 63.5 mm and was later adjusted to 127 mm, by inserting a plate on top of the original weir. The combustor was not connected to the reactor. Fine particles entrained were captured by a filter bag installed at the gas exit of the reactor. A camera and a light bulb were placed on top and inside the reactor, through the observation ports, to monitor and record the bed behavior to ensure proper tracer injection.



Figure 2. 10 Schematic of the PFB unit for solid RTD tests.

# 2.5.3 Procedure

Once both gas and solid flow reached steady state under testing conditions, for lower weir height, 20 g of tracer was poured onto the top of the feeding end of the bed by flipping the tracer box, whereas 30 g of tracer was used for weir height of 127 mm. Both amounts of tracer, which were determined by preliminary tests to ensure a good concentration resolution in following conductivity analysis, were less than 10% of the solids holdup of each case. By checking on the videos recorded during the experiments, the time of injection for lower weir height was generally less than 1 s and less than 1.5 s for the higher weir height and was short enough to consider the injection to be a pulse input, comparing with the theoretical mean residence time of 5–12 min. Discharged samples were collected starting at the exact moment of injection and continuing at a regular time interval. Detailed sampling schemes for different weir heights  $H_w$  and feed rates F and total sample number per run  $N_{total}$  are summarized in Table 2.7. Samples were taken normally every 30 s during the first half of each run, where concentration changed quickly. For the latter

half of the run, the sampling rate slowed down to lessen the workload. At a feed rate of 1 kg/h, a longer sample interval of 40 s at the quick sampling period was selected to ensure enough sawdust was collected for further analysis. At the lowest feed rate for each weir height, the measurement was prolonged to 40 min to fully capture the peak shape.

At the end of each RTD experiment, particle feed and gas were turned off at the same time. Solids remained in the reactor were taken out and weighed to determine the bed inventory, tapped bulk density of which was also determined. The amount of remaining tracer in the bed was measured in the same manner described in Section 2.6.1.

H <sub>w</sub> ,	F,	N	Qu	lick sampling	g Slow sampling			
mm	kg/h	<sup>1</sup> \total	Duration	Interval, s	Number	Duration	Interval, s	Number
63.5	1	49	1–18 min	40	27	19–40 min	60	22
63.5	1.5, 2	40	1–13 min	30	26	14–27 min	60	14
127	2	53	1–13 min	30	26	14–40 min	60	27
127	3, 4	40	1–13 min	30	26	14–27 min	60	14

Table 2. 7 Solid RTD sampling schemes.

Investigated operating conditions are summarized in Table 2.8. Gas pulsation frequency of 1–4 Hz was chosen to be around the natural oscillation frequency of 2.0 Hz for low weir height cases and 1.4 Hz for high weir height ones calculated by Eq. (2.3). The lower weir height was the same as the one used in the torrefaction test in Section 2.5. To study the effect of the bed height, a weir height twice as high as the previous one was used. As the weir height doubled, the feed rate was also increased to match the horizontal solid flow velocity. Two to three experiments were conducted to ensure reproducibility.

	Parameter	Operating conditions
Weir height	Pulsation frequency $f_P$	$f_P = 1, 2, 3, 4$ Hz; $F = 1.5$ kg/h; $U = 0.17$ m/s
H=63.5  mm	Feed rate, F	$F$ = 1.0, 1.5, 2.0 kg/h; $f_P$ = 1, 2, 4 Hz; $U$ = 0.17 m/s
	Gas velocity, U	$U = 0.14, 0.17, 0.20, 0.24 \text{ m/s}; f_P = 2 \text{ Hz}; F = 1.5 \text{ kg/h}$
Weir height	Pulsation frequency $f_P$	$f_P = 1, 2, 3, 4$ Hz; $F = 3.0$ kg/h; $U = 0.17$ m/s
<i>H</i> =127 mm	Feed rate, F	$F = 2.0, 3.0, 4.0 \text{ kg/h}; f_P = 1, 2 \text{ Hz}; U = 0.17 \text{ m/s}$
	Gas velocity, U	$U = 0.17, 0.20, 0.24 \text{ m/s}; f_P = 2 \text{ Hz}; F = 3.0 \text{ kg/h}$

Table 2. 8 Summary of experimental operating conditions for the solid residence time distribution test

# Chapter 3: Hydrodynamics and Solids Backmixing in a Horizontal Pulsed Fluidized Bed

# 3.1 Hydrodynamics of a horizontal pulsed fluidized bed

Prior to torrefaction experiments, simple tests on the hydrodynamics of the continuously operated pulsed fluidized bed were performed to characterize the gas and solid flow. Apparent minimum fluidization velocity was measured, and pressure fluctuation patterns under gas pulsation were inspected and compared to the previous study on a batch-operated pulsed fluidized bed.

# 3.1.1 Apparent minimum fluidization velocity

Measurement of apparent minimum fluidization velocity of sawdust is illustrated in Figure 3.1. The minimum fluidization velocities were identified at the intersection of the linear extrapolation of the two pressure drop sections at low and high gas velocities. Pressure drop sections at high gas velocities for all cases had smaller but never zero slopes, which could be caused by the imperfect distributor design and dead zone on the grid (Yang et al., 2003). Data points where the pressure drop steadily increased as velocity increased were used for linear extrapolation.








Figure 3. 1 Determination of apparent minimum gas velocity with different initial sawdust loads and pulsation frequencies. (a. 200 g, 1 Hz; b. 400 g, 1 Hz; c. 200 g, 2 Hz; d. 400 g, 2 Hz; e. 200 g, 4 Hz; f. 400 g,

For cases operated under 4 Hz, the pressure drop increased faster as the gas velocity increased at the high velocity region, indicating a larger dead volume at 4 Hz than those at 1 Hz and 2 Hz. The highest pressure drops were much lower than the total pressure drop (see Table 3.1), which is expected when all bed particles are fully suspended, because the pressure drop was a time-averaged value, and the dynamic pressure drop could be as low as zero during the gas 'off' period. For 200 g bed, the difference was 41% at 1 Hz and 2 Hz and decreased to 21% at 4 Hz; for 400g bed, 47% at 1 Hz, 31% at 2 Hz, and 20% at 4 Hz. Similar bed pressure increase with an increased gas pulsation frequency was summarized by Ireland et al. (2016), which could be explained by the shorter and eventually disappeared 'off' period with increasing pulsation frequency.

 Table 3. 1 Apparent minimum fluidization velocities with different initial sawdust loads and pulsation frequencies.

		Minimum fluidization velocity, m/s			
Bed weight, g	Total pressure, Pa	1 Hz	2 Hz	4 Hz	Average
200	101.4	0.0787	0.0835	0.0928	0.0850
400	202.7	0.0751	0.0709	0.1005	0.0821

Results are summarized in Table 3.1, with the average of the measured velocities from increasing and decreasing gas velocity in each case. The apparent minimum fluidization velocities measured at 4 Hz appeared to be higher than those at 1 Hz and 2 Hz for both loads. However, it should be noted that the extrapolation method used was somewhat arbitrary; the gas distributor was not perfect; the possible presence of channelings and dead volume brought uncertainties in the results. It was also argued that the pulsation frequencies applied (under 5 Hz) did not significantly influence the minimum fluidization velocity (Ali et al., 2016; Ali & Asif, 2012). Therefore, the average value of velocities tested over three frequencies for each sawdust load was calculated: 0.0850 m/s for 200 g bed and 0.0821 m/s for 400 g bed.

Minimum fluidization velocities of hemlock and SPF sawdust were also measured in continuous mode, as shown in Figure 3.2. Since no clear influence of pulsation frequency was found in the previous batch test, only 2 Hz was applied in this measurement. Higher peak pressure drop was seen in the gas velocity descending process, opposite to what has been observed in batch tests. Based on the recorded video, it was seen that when the gas velocity decreased to low velocities, the bed was fully or partially defluidized, and particles started to pile up on the feeding side and gradually spread to the entire bed area, corresponding to the faster pressure drop increase as the gas velocity decreased from 0.15 m/s to 0.1m/s. On the contrary, when the gas velocity increased, the bed height appeared to level itself and became evener. This process was somewhat random with continuous particle feed, resulting in more fluctuations at low velocities in the gas velocity ascending process. At high gas velocities, pressure drops in both gas velocity ascending and descending processes converged and steadily decreased as the gas velocity increased due to the steady reduction in the bed solids holdup. Average of the velocities corresponding to the highest pressure drops measured by increasing and decreasing gas velocity was adopted as the minimum fluidization velocity, which was determined as 0.085 m/s for hemlock and 0.094 m/s for SPF. The minimum fluidization velocity measured for hemlock in the continuous mode is very close to that measured in the batch mode.



Figure 3. 2 Measurement of minimum fluidization velocities in continuous mode for (a) hemlock particles and (b) SPF sawdust.

### 3.1.2 Pressure fluctuations

The gas pulsating behaviour is featured by 'flow-on' and 'flow-off' periods in each cycle, which creates different bed pressure fluctuation patterns. Typical sawdust bed pressure fluctuations as a function of time in batch mode and in the continuous mode are displayed in Figure 3.3 and Figure 3.4. In all tests, gas 'on' and gas 'off' periods were set to be equal.

In Figure 3.3, during the 'on' period, the pressure drop cycle started with a sharp peak caused by the highest initial gas velocity, followed by a section of rather stable pressures, with some not so distinctive damped oscillations and small fluctuations due to the bubble eruption. During the off period, a pressure recovery peak caused by the compression of the falling particles is clear for pulsation frequency of 1 Hz and 2 Hz. The pressure was then relaxed gradually. An evident flat section can be seen in the 1 Hz case. As the gas pulsation increased to 2 Hz, the pressure declined gradually after the recovery peak to the end of the 'off' period. When the pulsation frequency increased to 4 Hz, the 'off' period could hardly be identified, and the pressure drop was never lower than zero, which means the bed did not have time to settle on the distributor before it was lifted by the gas in the next cycle. The pressure drop pattern in the batch mode is similar to that reported by Jia et al. (2015), but with a more distinctive dominant peak due to the larger buffer tank for pulsating gas supply.



Figure 3. 3 Pressure fluctuations in batch mode at various pulsation frequencies.



Figure 3. 4 Pressure fluctuations in continuous mode at various pulsation frequencies.

The pressure drop profile in the pulsed fluidized bed with continuous feeding and discharging was somewhat different from the batch test and exhibited a pattern more like a square waveform. The initial peak was not as distinctive, and the pressure declined right after the gas was turned off. Instead of a pressure recovery at the beginning of the 'off' period, the ''on'' period was prolonged. The different pressure drop patterns in the continuous mode were caused by the particle overflow from the weir.

# 3.2 Solids residence time distributions in a horizontal pulsed fluidized bed

### **3.2.1** Operating conditions

Experimental operating conditions for solids RTD tests in the horizontal pulsed fluidized bed (HPFB) are listed in Table 3.2. All experiments were carried out under room temperature with air as the fluidizing gas. The letter h or H in case names means bed height of 63.5 mm or 127 mm, respectively, followed by the nominal value of gas pulsation frequency  $f_p$ , average superficial gas velocity U and solids feed rate F. The actual solids feed rate was calculated by the fed amount divided by the feeding duration. For each condition, two to three experiments were conducted to check the reproducibility except for case #H-4-0.17-3.0, where substantial measurement error in solids holdup was observed and a highly scattered RTD curve was obtained. Errors in the actual feed rate and the subsequent results were the mean absolute deviation, if not otherwise specified.

#	H <sub>w</sub> ,	f <sub>p</sub> ,	∐ m/s	F, kg/h	
π	mm	Hz	0, 11/3		
Δ	-	-	$\pm 0.003$	-	
h-1-0.17-1.0	63.5	1	0.172	$1.05 \pm 0.03$	
h-1-0.17-1.5	63.5	1	0.169	$1.49{\pm}0.04$	
h-1-0.17-2.0	63.5	1	0.170	$2.02 \pm 0.00$	
h-2-0.17-1.0	63.5	2	0.170	$1.03 \pm 0.01$	
h-2-0.17-1.5	63.5	2	0.170	$1.47{\pm}0.02$	
h-2-0.17-2.0	63.5	2	0.172	$2.01 \pm 0.01$	
h-2-0.14-1.5	63.5	2	0.140	$1.51 \pm 0.02$	
h-2-0.20-1.5	63.5	2	0.202	$1.50{\pm}0.01$	
h-2-0.24-1.5	63.5	2	0.240	$1.45 \pm 0.00$	
h-3-0.17-1.5	63.5	3	0.170	$1.46 \pm 0.04$	
h-4-0.17-1.0	63.5	4	0.170	$1.03 \pm 0.03$	
h-4-0.17-1.5	63.5	4	0.169	$1.44{\pm}0.01$	
h-4-0.17-2.0	63.5	4	0.172	2.03±0.01	
H-1-0.17-2.0	127	1	0.170	2.01±0.05	
H-1-0.17-3.0	127	1	0.170	$3.04{\pm}0.05$	
H-1-0.17-4.0	127	1	0.170	$4.07 \pm 0.01$	
H-2-0.17-2.0	127	2	0.171	$2.00 \pm 0.02$	
H-2-0.17-3.0	127	2	0.170	$3.01{\pm}0.03$	
H-2-0.17-4.0	127	2	0.170	$4.04{\pm}0.09$	
H-2-0.20-3.0	127	2	0.201	$3.06 \pm 0.03$	
H-2-0.24-3.0	127	2	0.240	3.09±0.01	
Н-3-0.17-3.0	127	3	0.171	$3.01 \pm 0.04$	
H-4-0.17-3.0	127	4	0.171	3.02	

Table 3. 2 Experimental conditions of solids RTD tests.

## 3.2.2 Solids holdup and mass turnover time

For a certain weir height, assuming all sizes of the particles exiting the bed at the same velocity, larger solids holdup means more particles are in the bed, increasing the degree of mixing. Bed mass at the end of the solids RTD test is reported in Figure 3.5 as solids holdup. The high weir height was twice as high as the low one, and tested feed rates in the high weir bed were also doubled to maintain the same apparent solids resident time. However, the solids holdups in Figure 3.5(b) were less than double the corresponding ones in Figure 3.5(a). This may indicate that the bed expanded over the weir height and the shallow bed expanded more than the deeper bed did. For both weir heights, as the pulsation frequency increased from 1 Hz to 4 Hz, in spite of that the time-averaged gas velocity was kept constant, the instantaneous gas velocity decreased, leading to less bed expansion. With a fixed weir height, the bed mass holdup increased as bed expansion decreased. A similar effect of pulsation frequency on bed expansion was described in Jia et al. (2015). Such impact of gas velocity was also seen in Figure 3.5(c). As the average gas velocity increased from 0.17 m/s to 0.24 m/s, solids hold up decreased by 19.9% and 21.3% for low and high weirs, respectively. The only exception was the 4 Hz case with high weir when the particles were observed to stack up at the exit periodically, bringing in substantial measurement error. Solids holdup remained about the same for shallow bed cases when the solids feed rate increased from 1 kg/h to 1.5 kg/h. As the feed rate increased to 2 kg/h, an increase of 4.6-8.9% was observed. In the deep bed, solids holdup increased accordingly, as the feed rate increases from 2 kg/h to 4 kg/h. Nevertheless, errors were more noticeable in the latter case.



Figure 3. 5 Solids holdups at different operating conditions (a)  $H_w = 63.5 \text{ mm}$ , U = 0.17 m/s, (b)  $H_w = 127 \text{ mm}$ , U = 0.17 m/s, (c)  $f_p = 2 \text{ Hz}$ .



Figure 3. 6 Mass turnover time at different operating conditions (a)  $H_w = 63.5 \text{ mm}$ , U = 0.17 m/s, (b)  $H_w = 127 \text{ mm}$ , U = 0.17 m/s, (c)  $f_p = 2 \text{ Hz}$ .

The mass turnover time defined by Eq. (3.1)

$$\tau = \frac{Bed \ mass}{Mass \ feed \ rate} \tag{3.1}$$

under various operating conditions is shown in Figure 3.6. The effect of pulsation frequency and gas velocity on mass turnover time followed the same trends as on solids holdup at a constant feed rate. As the solids feed rate increased, the solids holdup increased less than 10% for the low weir cases and 3.8–13.5% for the high weir cases, at a constant pulsation frequency. On the contrary, the increase in the solids feed rate affected the mass turnover time more significantly. It is noted that the bed weight was 43% higher at 4 Hz and 2 kg/h than the one at 1 Hz and 1.5 kg/h, so the mass turnover time was higher as well, even at a higher feed rate in Figure 3.6(a). Thus, combinations of pulsation frequency and feed rate need to be carefully selected to meet the targeted mean particle residence time.

## 3.2.3 Effects of operating conditions on RTD curves

Normalized concentration of tracer in each sample was calculated by Eq. (3.2),

$$C = \frac{c_i m_0}{\sum c_i m_i + c_b m_b} \tag{3.2}$$

where  $c_i$  (µS/cm) is the conductivity of i<sup>th</sup> sample solution,  $m_0$  (g) is the sample mass dissolved,  $m_i$  is the mass of the i<sup>th</sup> sample, and  $c_b$  and  $m_b$  are conductivity and mass of the solids remained in the bed at the end of each run. RTD curves were obtained by plotting the normalized concentration of each sample against the midpoint of the sampling period. Experiments were repeated for each case to check the reproducibility of RTD measurement. RTD curves of repeated experiments of case h-1-0.17-1.0 and H-2.0-0.17-3.0 are compared in Figure 3.7. The curves of the same case are very close to each other. In general, curves of cases with the high weir differ from each other more than the ones with the low weir, which contributes to the larger errors seen in the fitted results. Although the shape of the RTD curves varies for each case, the peak time of RTD curves is greatly consistent, which is a critical parameter used in the subsequent modelling.

Figures 3.8–3.10 show the effects of operating conditions on RTD curves in two difference weir heights. One curve was chosen to represent each combination of operating conditions. Long tails were spotted in all cases, possibly due to the presence of a stagnant zone, which was also observed to be 5–7% in other fluidized beds (Hua et al., 2019). Jia et al. (2017a) used a tapered bottom to enhance the heat and mass transfer performance, also confirmed the existence of the dead zone, which cannot be neglected, in a rectangular bed. The tailing phenomenon could also be a result of a partially well-mixed bed (Geng et al., 2017; Hofer et al., 2019; Hua et al., 2019; Pietsch et al., 2020). Most of the RTD curves exhibited a single distinct peak. Only for pulsation frequency of 3 Hz and 4 Hz, shown in Figure 3.8, and gas velocity as low as 0.14 m/s in Figure 3.9, the curves had multiple peaks or substantial fluctuations over the measured period. Moreover, the peak shifted, responding to the changes of operating conditions tested. Both peak shift and shape of the curve reflected the spread of the tracer, and ultimately the solids mixing.



Figure 3. 7 Repeated measurements of case (a) h-1-0.17-1.0, (b) H-2.0-0.17-3.0.



Figure 3. 8 Effect of pulsation frequency on RTD curves with a weir height of (a) 63.5 mm and (b) 127 mm.



Figure 3. 9 Effect of gas velocity on RTD curves with a weir height of (a) 63.5 mm and (b) 127 mm.



Figure 3. 10 Effect of solids feed rate on RTD curves with a weir height of (a) 63.5 mm and (b) 127 mm.

Figure 3.8 shows RTD curves at different pulsation frequencies with constant average gas velocity and feed rate. In Figure 3.8(a), the peak slightly shifted to the right with the increase of pulsation frequency, caused by the lower solid hold-up at a higher pulsation frequency and possibly the wider spread of tracer. In Figure 3.8(b), the peak of the 2 Hz curve shifted to the left and is slightly broader than the one of the 1 Hz curve. Considering the average solids velocity, defined as the quotient of bed length and mass turnover time, was lower at 2 Hz than that at 1 Hz, changes in tracer spreading caused by mixing were more significant than that caused by solids flow.

Figure 3.9 shows the effect of gas velocity on RTD curves. Curves with the same weir height had similar shapes, except for the case of a gas velocity of 0.14 m/s, which resulted in scattering of RTD signals. A more fluctuated signal at high weir height could also be expected and hence was avoided. The peak appeared earlier at higher gas velocity, because of higher bed expansion and thus lower solids hold-up. Peaks appeared earlier in small increments as the gas velocity increased. Solids velocity increase due to the decreased solids holdup and enhanced particle mixing induced by increased gas velocity could both contribute to the peak shifting.

As the feed rate increased, curves in Figure 3.10 became clearly narrower due to the increased solids velocity, and the peak moved slightly to the left. Effects of gas velocity and feed rate seemed to be similar in both cases with different weir heights.

#### 3.2.4 Mean residence time

Experimental mean residence time for each run was determined from RTD curves by Eqs. (3.3) and (3.4) (Fogler, 2017) for a pulse injection of tracer:

$$E(t) = \frac{C(t)}{\int_0^\infty C(t)dt}$$
(3.3)

$$t_m = \frac{\int_0^\infty tE(t)dt}{\int_0^\infty E(t)dt} = \int_0^\infty tE(t)dt$$
(3.4)

where E(t) is the residence-time distribution function,  $t_m$  (s) is mean residence time, and C(t) is the concentration of tracer at time t. Note that because of the long tailing of the RTD curves shown in previous section,  $t_m$  was affected by the cut-off time of each run.

Figure 3.11 compares the mean residence time tm and peak time of each RTD curve,  $t_{peak}$ , to the mass turnover time,  $\tau$ . Mean residence time in the low weir height bed appeared to be smaller than  $\tau$  with differences of 5.1–25.8%, which could be caused by the existence of internal circulation or less active zones, while  $t_m$  in the high weir bed fluctuated along the diagonal line. Mass turnover time is determined based on the mass balance of the process. Mean residence times larger than the mass turnover times are supposed to be due to the error in holdup or RTD measurements (Fogler, 2017). Another possible explanation in this study was that mass turnover time calculation was based on the overall bed mass without consideration of the segregation between coarse and fine particles. Segregation has been commonly observed in gas pulsating fluidized beds using particles with a wide size distribution and pulsed fluidized beds could even be utilized to perform particle

separation (Dong et al., 2016; Saidi et al., 2014). In this work, coarser particles were observed to accumulate in the bed, and bed bulk density was increased by 22.3–35.6%, supporting the segregation speculation. In addition, the stagnant zone might be relatively smaller when compared to the larger total bed volume in the deeper bed, which led to a less reduction in measured  $t_m$ . Peak time is closely linked to the solids flow condition and is highly sensitive to the variation in operating conditions, as discussed in Section 3.2.3. The peak time of all cases was much smaller than  $\tau$ , indicating a stagnant volume or the short circuit of fast-moving tracer particles at the bed surface, considering that the bed was subjected to a velocity profile.



Figure 3. 11 Comparison of mean residence time/peak time with mass turnover time.

# 3.3 Modelling of residence time distribution curves

As discussed in the previous sections that solids transport and backmixing cannot be directly inferred from the RTD curves, a modified axial dispersion model was developed. Dispersion parameters were obtained by fitting the model to the RTD curves.

## 3.3.1 Model description

The obtained RTD curves were modelled by a 2D axial dispersion model with an exchange flow (ADMex) between the active zone and stagnant zone, according to Eq. (3.5). Schematic of the model is depicted in Figure 3.12.

$$\begin{cases} \frac{\partial C_1}{\partial t} = D_x \frac{\partial^2 C_1}{\partial x^2} + D_y \frac{\partial^2 C_1}{\partial y^2} - u_x(y) \frac{\partial C_1}{\partial x} + N \frac{(C_2 - C_1)}{\alpha} \\ \frac{\partial C_2}{\partial t} = \frac{N(C_1 - C_2)}{1 - \alpha} \end{cases}$$
(3.5)



Figure 3. 12 Schematic of the ADMex model

The active zone volume fraction is  $\alpha$ , where the solids bulk flow is along the horizontal direction with a vertical velocity profile. The tracer concentrations in active zone  $C_I$  (dimensionless) and stagnant zone  $C_2$  (dimensionless) are functions of time and position in the bed. Mass transfer rate between active and stagnant zones is  $KS\Delta C/A$ , where K (m/s) is the apparent mass transfer coefficient;  $S(m^2/m)$  is the transfer area per unit length between active and stagnant zones; and A(m<sup>2</sup>) is the cross-section area of the bed. Transfer number N = KS/A is defined for simplicity. Despite the solids exchange between two zones, solids in the stagnant zone are assumed to circulate inside the stagnant volume and do not exit the system. Tracer is carried over by the bulk flow and spreads in both horizontal and vertical directions with dispersion coefficients of  $D_x$  and  $D_y$  (m<sup>2</sup>/s). Dispersion coefficients are assumed to be constant throughout the bed. Thus, tracer concentration detected at the exit of the system corresponds to average  $C_I$  at the end of the active zone. Dispersion coefficients and transfer number N are obtained by fitting the ADMex model established above to tracer data of each trial.

## 3.3.2 Parameter specifications

Average solids velocity is defined as Eq. (3.6),

$$\bar{u}_x = \frac{L}{\tau} \tag{3.6}$$

where  $\tau$  (s) is the mass turnover time. Due to the existence of the stagnant volume, the effective average solids velocity is expressed as Eq. (3.7),

$$\bar{u}_{x,eff} = \frac{L}{\alpha\tau} \tag{3.7}$$

where L (m) is the bed length. Velocity profile in Poiseuille flow boundary layer is adopted as vertical solids velocity profile to solve Eq. (3.5) (Spurk & Aksel, 2019). Velocity profile expressions are arranged as a function of the effective average solids velocity and vertical position in the bed, which vary as the thickness of the hypothetical boundary layer  $\delta$  changes. When the boundary layer is thicker than the bed height, as given by  $\beta = \delta/H_w \ge 1$ , horizontal velocity is expressed as Eqs. (3.8) and (3.9)

$$u_x(y) = \frac{6\delta y - 3y^2}{3\delta H_w - {H_w}^2} \bar{u}_{x,eff}$$
(3.8)

When  $\beta < 1$ ,

$$\begin{cases} u_{H} = \frac{\overline{u}_{x,eff}}{\frac{2}{3}\beta + (1 - \beta)} \\ u_{x}(y) = \begin{cases} u_{H} & (y > \beta H) \\ \left\{\frac{y}{\beta H_{w}} - \frac{1}{2}\left(\frac{y}{\beta H_{w}}\right)^{2}\right\} u_{H} & (y \le \beta H) \end{cases}$$
(3.9)

where  $u_H$  is the maximum velocity.

Instantaneous tracer injection rate is approximated by a Gaussian function with the total amount of unity at the point (x=0, y= $H_w$ ). Injection time is assumed to be 1 s for cases with a weir height of 63.5 mm and 1.5 s for 127 mm. The initial condition, zero flux boundaries, and closed-closed boundary conditions are applied as follows:

$$\begin{cases} C_1(0, x, y) = 0\\ D_y \frac{\partial C_1(t, x, 0)}{\partial y} = D_y \frac{\partial C_1(t, x, H)}{\partial y} = 0\\ D_x \frac{\partial C_1(t, 0^-, y)}{\partial x} = 0\\ u_x(y)C_1(t, 0^+, y) = D_x \frac{\partial C_1(t, 0^+, y)}{\partial x}\\ D_x \frac{\partial C_1(t, 1, y)}{\partial x} = 0 \end{cases}$$
(3.10)

Active zone fraction  $\alpha$  and boundary thickness  $\delta$  cannot be directly measured in the experimental setup. Before finding the dispersion coefficients, an experimental curve was fitted with the model by sweeping  $\alpha$  and  $\beta$ . The two parameters were selected while the best fit was achieved. The tested range of active zone fraction was around the tracer recovery fraction measured in each experiment with an interval of 0.05.  $\beta$  value ranged from 0.6–3, as values larger than 3 did not significantly impact on the fitted curves. Once the dispersion coefficients and transfer number were fitted,  $\alpha$  value was verified again with the updated parameters.

## 3.3.3 Model validation

#### 3.3.3.1 Goodness-of-fit

Dispersion coefficients and transfer number *N* were determined by minimizing the least square objective value. The overall agreement for RTD curves obtained with gas pulsations of 1 Hz and 2 Hz and gas velocity  $\ge 0.17$  m/s is satisfactory. The model fitted excellently to 66.7% of individual RTD curves, with a mean squared error less than 5×10<sup>-6</sup>. Figure 3.13 exemplifies the goodness-of-fit of the proposed ADMex model. The mean squared errors were  $1.42 \times 10^{-6}$  and  $7.73 \times 10^{-7}$  for presented case h-2-0.17-1.5 and case H-1-0.17-3.0, respectively. However, the value could not

give the information of the discrepancy location on the curve. As described in the following section, the peak of the curve was primarily fitted. It is noteworthy that, while the overall fitness value is not ideal for some cases, the fitted curve successively captured the peak time and peak shape of the RTD curves.

#### **3.3.3.2** Sensitivity analysis of dispersion coefficients

 $D_x$ , ×10<sup>-4</sup> m<sup>2</sup>/s

Case h-1-17-1.5 is used as an example in this section to illustrate the relationship of dispersion coefficients and the simulated curve by the ADMex model. Figure 3.14(a) compares the simulated curves with different horizontal dispersion coefficients of the same order magnitude. Peaks shifted left evidently as horizontal dispersion coefficients increased. In contrast, values of vertical dispersion coefficients used in the fitting were varied by orders of magnitude. Only when  $D_y$  was in the same order of magnitude as  $D_x$ , the shift in curve peaks could be seen. As  $D_y$  was larger than  $D_x$  by at least one order of magnitude, the curve change was negligible. This suggests that the ADMex model is sensitive to the change in  $D_x$  but not sensitive to the change in  $D_y$ .

The impact of  $D_y$  in fitting  $D_x$  by the ADMex model was further examined.  $D_x$  was solved with different  $D_y$  values, shown in Table 3.3. Once  $D_y$  became at least one order of magnitude larger than  $D_x$ , the change in  $D_x$  was generally less than 5%. It can be concluded that  $D_y$  should be always at least one order of magnitude larger than  $D_x$ , and the ADMex model cannot be used to accurately evaluate  $D_y$ .

 $\frac{D_{y}, \times 10^{-4} \text{ m}^2/\text{s}}{0.1 \text{ 1}} 10 100$ 

0.422

Table 3. 3  $D_x$  values solved with different Dy using the case h-1-17-1.5.

0.324

0.322

0.322



Figure 3. 13 Comparison of experimental data and model fitted curve for case (a) h-2-0.17-1.5, (b) H-1-0.17-3.0.



Figure 3. 14 Comparison of simulated curves with different (a) horizontal dispersion coefficients and (b) vertical dispersion coefficients.

## 3.4 Effects of operating conditions on dispersion coefficient

Since the vertical dispersion coefficient is at least one order of magnitude larger than in the HPFB and could not be accurately fitted by the ADMex model, only horizontal dispersion coefficient results are discussed in this section.

The horizontal dispersion coefficient is plotted against average horizontal solids velocity in Figure 3.15. The average solids velocity was defined as the quotient of bed length and mass turnover time, which is determined by the feed rate and solids holdup discussed in Section 3.2.2. Effects of pulsation frequency and solids velocity on the horizontal dispersion coefficient are shown in Figure 3.15(a) for the shallow bed and in Figure 3.15(b) for the deep bed. Overall, dispersion coefficients for the deep bed were slightly higher than those of the shallow bed. Nevertheless, they were all of the same order of magnitude for pulsation frequencies of 1 Hz, 2 Hz and 4 Hz and gas velocity of 0.17 m/s.

For a constant pulsation frequency in Figure 3.15(a), the horizontal dispersion coefficient generally increased as solids velocity increased. Nevertheless, curves at different pulsation frequencies overlapped with each other as solids velocity had different degrees of impacts on  $D_x$ . At a pulsation frequency of 1 Hz, the gas pulsation was strong enough to facilitate the bed mixing at a relatively low solids velocity. In addition, solid velocity did not have substantial impact in the tested range. When the HPFB was operated at 4 Hz, less gas was introduced during each pulsation cycle, and smaller bubbles were formed compared to the ones generated at lower frequencies, leading to a less agitated bed and substantially reduced intensity of gas pulsation. Consequently, the bed was not as active as it was at lower pulsation frequencies, and the horizontal mixing was enhanced

largely by increasing average solids velocity. It is noteworthy that the effect of average solids velocity on  $D_x$  at pulsation frequency of 2 Hz changed over the tested range of particle velocity.  $D_x$  first increased as slowly as it did at 1 Hz and then increased rapidly when the average solids velocity increased over  $4.79 \times 10^{-4}$  m/s. The sudden increase in Dx might be partially attributed to the noticeable increase in the holdup discussed in Section 3.2.2.

Unsurprisingly,  $D_x$  in the deep bed was observed to follow a different trend from the shallow bed, as shown in Figure 3.15(b), and more uncertainties emerged when the weir height increased. Bubbles are generated at the bottom of the bed, and as the bed height increases, their motion and subsequent interaction while passing through the bed become poorly predictable (Francia et al., 2021). It is seen that the more intensive gas pulsation at 1 Hz led to lower horizontal dispersion coefficients, probably because that the bed was operated at a frequency closer to the natural frequency of the bed. Jia et al. (2015) found that the dominant frequency of pressure fluctuations in a pulsed fluidized bed was the same as the gas pulsation frequency. When the inherent pressure wave frequency coincided with the gas pulsation frequency, the effect of gas pulsation was at its peak due to the resonance effect (Jia et al., 2016). Bubbles were generated more orderly, reducing the chance of mixing. A rapid increase in  $D_x$  was seen again as solids velocity increased to 7.06×10<sup>-</sup> <sup>4</sup> m/s.

The horizontal dispersion coefficient was highly dependent on gas velocity, as shown in Figure 3.15(c) where the pulsation frequency was kept at 2 Hz to ensure sufficient bed mobility. As gas velocity increased from 0.17 m/s to 0.24 m/s,  $D_x$  demonstrated an over threefold increase in both the shallow and the deep beds. It was expected that the available excessive gas to form bubbles greatly enhanced horizontal particle movement and thus mixing.



Figure 3. 15 Horizontal dispersion coefficients at different operating conditions (a)  $H_w = 63.5$  mm, U = 0.17 m/s, (b)  $H_w = 127$  mm, U = 0.17 m/s, (c)  $f_p = 2$  Hz (square:  $H_w = 63.5$  mm, F = 1.5 kg/h, circle:  $H_w = 127$  m, F = 3.0 kg/h).

#### 3.5 Discussion on horizontal dispersion coefficient

Solids mixing in HPFB in terms of the horizontal dispersion coefficient can be compared with other fluidized beds. Based on their bubbling bed model, Kunii and Levenspiel (1969) assumed that solids drawn into the bubble wake are completely mixed and derived a theoretical equation Eq. (3.11)

$$D_{sr} = \frac{3}{16} \left( \frac{\delta_b}{1 - \delta_b} \right) \frac{U_{mf} d_b}{\varepsilon_{mf}}$$
(3.11)

for solids lateral dispersion coefficient, where  $\delta_b$  is the bubble fraction of the bed.

Shi and Fan (1985) further assumed that solids were pushed by successive rising bubbles. By performing a dimensionless analysis incorporating the bulk solids flow effect on lateral mixing in shallow fluidized bed, they obtained Eq. (3.12)

$$D_x = \frac{u_x H_{bed}}{0.730} \left(\frac{U - U_{mf}}{u_x}\right)^{0.290} \left(\frac{\sqrt{gH_{bed}}}{U - U_{mf}}\right)^{0.0725}$$
(3.12)

for the horizontal dispersion coefficient.

Horizontal dispersion coefficients in HPBF obtained in this work were compared with dispersion coefficients calculated by literature correlations in Table 3.4. Eq. (1.4) by Nilsson and Wimmerstedt (1988) and Eq. (1.5) by Bachmann et al. (2016) are correlations established in other horizontal fluidized beds with continuous solids flow. Eq. (3.12) of Shi and Fan (1985) discussed above also took solids flow into account. The values calculated from Eq. (1.4), Eq. (1.5) and Eq.

(3.12) are generally 5–20 times of the data in this work. On the other hand, dispersion coefficients calculated by Eq. (3.11) of Kunii and Levenspiel (1969) are around the same range of the value measured in HPFB from this study, although the Kunii and Levenspiel correlation was often found to underestimate the lateral mixing by 5-10 times (Shi & Fan, 1985) because it overlooked the bubble lateral movements, bubble bursting on the surface and solid circulation inside the bed. Dsr values do not vary with various feed rate, which are around  $0.425 \times 10^{-4}$  m<sup>2</sup>/s in shallow bed and  $0.555 \times 10^{-4}$  m<sup>2</sup>/s in the deep bed, due to the Kunii and Levenspiel correlation does not consider the solids flow effect. There are several possible underlying reasons on why  $D_{sr}$  is close to our data in HPFB, instead of just a simple coincidence. It has been proven that gas pulsation transforms the deterministic chaotic bubble behavior into ordered patterns (Coppens & van Ommen, 2003; Francia et al., 2021; Wang & Rhodes, 2005). Bubbles rise in rows resulting in a reduction of bubble interaction and coalescence. Besides, the bed remained shallow so that the gas pulsation could be strong enough to assist fluidization, and bubble lateral movements were reduced as it quickly reached the surface. In the meantime, the bubble growth was restricted, and the solids carried by bubbles that burst on the surface of the bed were consequently limited and might be negligible. Furthermore, the average solids velocity might be too small to affect the dispersion coefficient substantially. Note that the average solids velocity in this study was  $2.53 \times 10^{-4} - 7.06 \times 10^{-4}$  m/s. lower than 7.2×10<sup>-4</sup>–6.1×10<sup>-3</sup> m/s in Nilsson and Wimmerstedt (1988) and 8.30×10<sup>-4</sup>–3.37×10<sup>-3</sup> m/s in Bachmann et al. (2016). It can thus be argued that the horizontal dispersion coefficients obtained in this work are close to the particle diffusion coefficient postulated by Kunii and Levenspiel. Nevertheless, as the gas velocity increased (for cases of h-2-0.20-1.5, h-2-0.24-1.5, H-2-0.20-3.0, H-2-0.24-3.0), the horizontal solids mixing started to increase significantly, resulting from increased gross solids circulation. Since the Kunii and Levenspiel correlation did not account

for gross solids circulation, it tends to underestimate the dispersion coefficient in HPFB at high gas velocities.

$D_x$ or $D_{sr}$ , $ imes 10^{-4}$	UDED	Nilsson and	Bachmann	Shi and	Kunii and
<b>m</b> <sup>2</sup> / <b>s</b>	III FD	Wimmerstedt	et. al	Fan	Levenspiel
h-1-0.17-1.0	0.255	1.64	1.47	1.66	0.436
h-1-0.17-1.5	0.289	1.76	2.92	2.15	0.422
h-1-0.17-2.0	0.360	1.87	2.80	2.59	0.425
h-2-0.17-1.0	0.209	1.59	2.66	1.55	0.428
h-2-0.17-1.5	0.236	1.72	2.64	1.98	0.428
h-2-0.17-2.0	0.460	1.83	2.47	2.34	0.436
h-2-0.14-1.5	0.152	1.33	1.55	1.61	0.265
h-2-0.20-1.5	0.457	2.11	3.65	2.22	0.601
h-2-0.24-1.5	1.05	2.62	5.67	2.61	0.801
h-3-0.17-1.5	0.284	1.65	2.27	1.78	0.425
h-4-0.17-1.0	0.116	1.53	2.24	1.37	0.428
h-4-0.17-1.5	0.186	1.64	2.22	1.73	0.422
h-4-0.17-2.0	0.300	1.77	2.10	2.10	0.439
H-1-0.17-2.0	0.453	5.21	4.70	3.74	0.555
H-1-0.17-3.0	0.328	5.68	4.51	5.72	0.551
H-1-0.17-4.0	0.582	5.97	4.20	6.36	0.551
H-2-0.17-2.0	0.553	5.19	4.65	3.22	0.558
H-2-0.17-3.0	0.468	5.53	4.05	5.11	0.552
H-2-0.17-4.0	0.556	5.87	3.92	6.07	0.555
H-2-0.20-3.0	0.928	6.14	6.33	5.21	0.776
H-2-0.24-3.0	1.95	6.75	9.09	6.23	1.06
H-3-0.17-3.0	0.345	5.33	3.45	4.19	0.558
H-4-0.17-3.0	0.133	5.39	3.62	4.37	0.562

Table 3. 4 Horizontal dispersion coefficients in this study and calculated by literature correlations.

#### 3.6 Correlation of horizontal dispersion coefficient

To integrate the effect of horizontal solids flow and gas velocity, dimensionless analysis of the relationship between horizontal coefficient and influencing variables was performed. In addition to the particle diffusion coefficient  $D_{sr}$  estimated by Kunii and Levenspiel (1969), solids velocity  $u_x$ , weir height H, pulsation frequency  $f_p$  and gas velocity U, are all included. A correlation in the shallow HPFB is derived and shown in Eq. (3.13):

$$\frac{D_x}{D_{sr}} = 1.006 \left(\frac{u_x H_w}{D_{sr}}\right)^{1.064} \left(\frac{f_p - f_n}{f_n}\right)^{-0.038} \left(\frac{U - U_{mf}}{U_{mf}}\right)^{1.644}$$
(3.13)

where  $f_n$  is the natural frequency of the bed and  $U_{mf}$  is the minimum fluidization velocity. Within the tested range, the exponent of the first term on the right side is close to 1, which corresponds to the modest impact of the solid flow velocity; the exponent of the second term is negative and close to 0, which implies that operating close to natural frequency may slightly intensify the mixing; lastly, the exponent of the third term corrects the dispersion coefficients at high gas velocities.

A comparison of dispersion coefficients calculated by Eq. (3.13) with the observed data in Figure 3.16 shows that they agree satisfactorily. Data points are located on both sides along the diagonal line, and the mean error is 14%. Thus, the correlation can reasonably predict the horizontal dispersion coefficient. It is worth mentioning that the correlation is based on experimental data over a rather limited tested range in the specific reactor. Validity needs to be tested following the same method before extrapolation and generalization.



Figure 3. 16 Comparison between calculated dispersion coefficients by Eq. (3.13) and the observed data.

#### 3.7 Other parameters

Besides the horizontal and vertical dispersion coefficients discussed in previous sections, other parameters obtained from the model or calculated accordingly are discussed in this section.

As seen in the RTD curves and described by the ADMex model, the stagnant part of the bed is evident, the amount of which was estimated by the model. The average active volume fraction  $\alpha$ used in each case is listed in Table 3.5. The average active volume fraction of all the cases with the low weir is 0.663, larger than the value 0.756 for high weir cases, which confirms the speculation in Section 3.2.4 that the stagnant zone is smaller for the deep bed. Cases h-2-0.14-1.5,
h-4-0.17-1.0, h-4-0.17-1.5, H-3-0.17-3.0, contain a particularly small active volume, which may explain the substantial fluctuations observed in the corresponding RTD curves in Section 3.2.3.

Transfer number N, which connects the active volume and stagnant volume of the bed, was also fitted by the model and is listed in Table 3.5. As defined in Section 3.3.1, the transfer number is proportional to the mass transfer coefficients between the active and the stagnant volumes. For cases with small active volume fractions, the transfer numbers are generally large to make up for the loss of tracer trapped in the stagnant volume. The transfer number is of the same order of magnitude with the solids flow velocity, and in some cases, even larger, which might indicate a more vigorous mass transfer between two zones than that by the bulk flow.

Finally, with the fitted horizontal dispersion coefficients, effective Peclet numbers of the reactor are calculated by Eq. (3.14):

$$Pe_{eff} = \frac{\bar{u}_{eff}L}{D_x} = \frac{L^2/\tau_{eff}}{D_x} = \frac{L^2/(\alpha\tau)}{D_x}$$
(3.14)

at various operating conditions and listed in Table 3.5. The Peclet numbers are not as high as those reported by Kong et al. (2018), who studied a reactor with a length to width ratio 8 times than that in this work, but are comparable with the values in Bachmann et al. (2016). Error in the effective Peclet number, shown in the last column of Table 3.5, is less than 15% for most of the cases.

#	α	<i>N,</i> ×10 <sup>-4</sup> 1/s	$\overline{u}_{eff}$ , ×10 <sup>-4</sup> m/s	Pe <sub>eff</sub>	∆ Pe <sub>eff</sub>
h-1-0.17-1.0	0.650	2.80	5.07	5.04	0.05
h-1-0.17-1.5	0.650	6.74	7.39	6.51	0.52
h-1-0.17-2.0	0.650	9.15	9.96	6.74	0.20
h-2-0.17-1.0	0.650	3.64	4.61	5.70	0.76
h-2-0.17-1.5	0.650	8.77	6.54	7.15	0.91
h-2-0.17-2.0	0.700	3.53	7.69	4.28	0.66
h-2-0.14-1.5	0.425	22.8	8.56	14.27	0.18
h-2-0.20-1.5	0.675	6.04	6.73	3.74	0.22
h-2-0.24-1.5	0.675	8.01	7.73	1.88	0.08
h-3-0.17-1.5	0.600	6.91	6.07	5.46	0.49
h-4-0.17-1.0	0.525	7.99	4.83	11.75	3.87
h-4-0.17-1.5	0.475	16.7	7.44	10.14	0.10
h-4-0.17-2.0	0.650	11.4	9.52	9.28	2.74
H-1-0.17-2.0	0.750	10.0	5.17	2.92	0.31
Н-1-0.17-3.0	0.725	20.7	7.96	6.12	0.47
H-1-0.17-4.0	0.775	26.6	9.11	4.12	0.70
H-2-0.17-2.0	0.750	16.3	5.08	2.34	0.17
H-2-0.17-3.0	0.700	16.2	7.21	3.96	0.55
H-2-0.17-4.0	0.750	22.2	8.67	3.99	0.36
Н-2-0.20-3.0	0.800	21.6	7.51	2.07	0.18
Н-2-0.24-3.0	0.800	5.07	8.48	1.28	0.53
Н-3-0.17-3.0	0.575	13.4	7.41	5.49	0.59
H-4-0.17-3.0	0.600	23.5	7.42	14.18	-

Table 3. 5 Active volume fraction, transfer number and effective Peclet number for all cases.

## 3.8 Conclusion

Residence time distributions of hemlock particles at different operating conditions were measured in a horizontal pulsed fluidized bed designed for biomass torrefaction. Solids holdups were measured for the determination of mass turnover time and solids flow velocity. RTD curves, along with the comparison of the peak time and mass turnover time, indicated the existence of a stagnant zone in the HPFB with a rectangular cross-section. A 2D axial dispersion model with an exchange flow between the active zone and stagnant zone was proposed for the HPFB and applied to extract solids mixing coefficients from the experimental data. Proximate instead of measured velocity profiles were adopted. The ADMex model was able to characterize the peak time and peak shape of the RTD curves. In addition, the model is found to be sensitive to the change in horizontal dispersion coefficient but not sensitive to the change in vertical dispersion coefficient. Horizontal dispersion coefficients were slightly higher in the deeper bed. In the shallow bed, when gas pulsation frequency was kept constant, horizontal dispersion coefficients increased with increasing solids velocity induced mainly by increased solids feed rate. Similar trend was not observed in the deeper bed. Lowered backmixing intensity was observed when the bed was operated under a gas pulsation frequency close to the bed natural frequency. The solids backmixing intensity was also found to be more dependent on gas velocity. Horizontal solids dispersion coefficients obtained in HPFB were significantly smaller than values calculated by literature correlations for horizontal fluidized beds. Gas pulsation might play an essential role in reducing the mixing by transforming the chaotic bubble behavior into regular patterns. A correlation of horizontal dispersion coefficients in HPBF of shallow beds was established with a fairly good agreement with the data, which can be used in modeling the HPFB reactor for biomass torrefaction.

This Chapter provides information on the solids mean residence times at different operating conditions, which directly impact the torrefaction performance, and gives insights on the solids flow and quantifies the solids backmixing. The model describing the solids flow can be applied in

a reactor model. By incorporating torrefaction kinetics, heat transfer, and particle dispersion, bed temperature profile and product weight loss at different operating conditions can be predicted. Particularly, the effect of horizontal dispersion coefficient, which closely links to the torrefaction performance yet cannot be a separate variable in experiments, can be evaluated.

# **Chapter 4: Performance of a Pulsed Fluidized Bed Torrefier**

This Chapter documents the performance of the pulsed fluidized bed for sawdust torrefaction with the investigated parameters of gas pulsation frequency, bed temperature, sawdust feed rate and oxygen content in the carrying gas. For each run, the reactor was operated continuously for 2.5 hours with a feed capacity of up to 2 kg/h. Torrefied product was characterized by its higher heating value (HHV), proximate analysis, and ultimate analysis, for its application as a solid fuel. Relationships between the weight loss of the process and the product properties were also explored. The performance metrics include weight loss, energy yield, and enhancement factor. Continuous densification of the torrefied biomass produced was also tested in Appendix D.

## 4.1 Operating conditions

As discussed in Chapter 1, temperature and residence time were the mostly manipulated parameters to produce torrefied materials with different properties. In this work, sawdust particles were heated up by the hot gas and the reactor walls as they traveled from the feed port to the overflow weir. Temperatures at the feeding side (T<sub>3</sub>), in the middle (T<sub>4</sub>), and at the discharging side (T<sub>5</sub>) of the sawdust bed were measured. A temperature difference as large as 200 °C was seen between the two ends of the bed. Initially, the bed temperature was defined as the arithmetic mean of T<sub>4</sub> and T<sub>5</sub>, similar to that defined by Ohliger et al. (2013), where the temperature at the solids inlet was not included in the calculation of mean temperature. The arithmetic mean temperature of all temperatures measured in the bed was also selected as the torrefaction temperature by Strandberg et al. (2015) and Wang et al. (2017b), where the maximum temperature gradient scale was ~40 °C, much lower than that in this work. Moreover, the middle section temperature T<sub>4</sub> was

observed to fluctuate significantly with the gas pulsation frequency and generally lower than expected. Therefore, by imagining the bed being heated up by a fluid with a temperature slightly higher than T<sub>5</sub>, an average bed temperature is calculated by Eq. (4.1),

$$T_{ave} = T_{\infty} - \frac{T_5 - T_3}{ln(T_{\infty} - T_3) - ln(T_{\infty} - T_5)}$$
(4.1)

using T<sub>3</sub> and T<sub>5</sub>, where T<sub>5</sub> is assumed to approach  $T_{\infty}$ , which is 1 °C higher than T<sub>5</sub>. The value of  $T_{\infty}$  does not significantly influence the average temperature if it is defined as less than 3 °C higher than T<sub>5</sub>. Feed rate (*F*, kg/h) instead of residence time was varied for each run by adjusting the rotating speed of the feeder screw. The average feed rate over the entire run was recorded as the actual feed rate. Residence time distribution at various operating conditions was measured and reported in Chapter 3.

Besides the temperature and residence time, the impact of pulsation frequency ( $f_p$ , Hz) was investigated, as gas pulsation is the unique feature of the studied reactor. Operating pulsed fluidized beds around the natural pulsation frequency of the bed can enhance mass and heat transfer (Jia et al., 2017b; Jia et al., 2015), consequently impacting the torrefaction performance.

Time-averaged gas velocity (U, m/s) at the bed temperature was kept constant and high enough to avoid severe channeling and partial defluidization. Higher gas velocity was not employed to save nitrogen gas consumption which is translated to lower operating costs in large-scale operations.

The four parameters mentioned above, in fact, interact with each other and could not be completely isolated. Pulsation frequency influences both the bed temperature and residence time. As the

pulsation frequency increases, instantaneous gas velocity decreases, resulting in larger solids holdup, hence lower bed temperature with same heating settings and longer mean residence time. The gas flow rate was adjusted according to the bed temperature to keep the superficial gas velocity constant. Since the preheated gas is one of the heat sources, changes in gas flow rate, in turn, influence the final bed temperature profile. In summary, each set of experiments is not a strictly single variable investigation.

In addition, to test the feasibility of recycling the flue gas as the fluidizing gas, the effect of oxygen concentration in the fluidizing gas is investigated as well.

Operating conditions for all investigations are listed in Table 4.1. Cases P1-F1.0 to P5-F1.5 were performed with sawdust to investigate the effect of pulsation frequency. The first number in the case name is the pulsation frequency tested ranging from 1 Hz to 5 Hz, and the second number is the nominal feed rate, 1.0 or 1.5 kg/h. To incorporate the impact of pulsation frequency on heat transfer in the reactor, all heater settings, and flow rate of the gas entering the bed were the same for each feed rate. Fluctuations in feed rate were less than 5%. Case P2-F1.0, P3-F1.0, P4-F1.0, and P2-F1.5 were repeated. Errors calculated from two parallel runs in feed rate under actual operating conditions and in subsequent product characterizations showed fairly good reproducibility of the experiments. Thus, each of the rest experiments is presented separately.

Case name	$f_p$ , Hz	F, kg/h	Tave, °C	X02, %	<i>U</i> , m/s
P1-F1.0	1	1.06	283	0	0.21
P2-F1.0*	2	$1.02 \pm 0.01$	$280\pm2$	0	0.20
P3-F1.0*	3	$1.04 \pm 0.00$	276±1	0	0.21
P4-F1.0*	4	$1.02 \pm 0.01$	275±1	0	0.20
P5-F1.0	5	0.96	281	0	0.21
P1-F1.5	1	1.48	286	0	0.21
P2-F1.5*	2	$1.44 \pm 0.03$	286±1	0	0.21
P3-F1.5	3	1.49	278	0	0.21
P4-F1.5	4	1.41	282	0	0.21
P5-F1.5	5	1.53	267	0	0.21
SPF-FT1	2	1.01	256	0	0.22
SPF-FT2 (P2-F1.0)	2	1.01	281	0	0.20
SPF-FT3 (P2-F1.0)	2	1.02	278	0	0.20
SPF-FT4	2	0.98	307	0	0.20
SPF-FT5	2	1.01	331	0	0.20
SPF-FT6	2	1.50	254	0	0.21
SPF-FT7 (P2-F1.5)	2	1.41	285	0	0.22
SPF-FT8 (P2-F1.5)	2	1.47	287	0	0.21
SPF-FT9	2	1.47	320	0	0.22
SPF-FT10	2	1.47	321	0	0.21
SPF-FT11	2	1.49	348	0	0.21
SPF-FT12	2	1.89	292	0	0.21
SPF-FT13	2	2.04	283	0	0.21
SPF-FT14	2	2.05	292	0	0.21
SPF-FT15	2	2.11	317	0	0.21
SPF-FT16	2	1.53	296	0	0.21
SPF-FT17	2	1.71	292	0	0.20

Table 4. 1 Actual operating conditions of all experiments.

Case name	f <sub>p</sub> , Hz	F, kg/h	Tave, °C	X02, %	U, m/s	
HEM-FT1	2	1.49	238	0	0.19	
HEM-FT2	2	1.34	278	0	0.20	
HEM-FT3	2	1.40	276	0	0.19	
HEM-FT4	2	1.47	279	0	0.20	
HEM-FT5	2	1.36	306	0	0.22	
HEM-O1	2	1.47	233	3.10	0.19	
HEM-O2	2	1.47	248	3.19	0.18	
HEM-O3	2	1.35	263	3.05	0.21	
HEM-O4	2	1.49	263	3.09	0.20	
HEM-O5	2	1.50	283	3.17	0.21	
HEM-O6	2	1.45	236	6.09	0.19	
HEM-O7	2	1.44	242	6.15	0.20	

Table 4. 1 Actual operating conditions of all experiments (continued).

Cases SPF-FT1 to SPF-FT17 were performed with SPF sawdust to investigate the effects of feed rate and temperature. Pulsation frequency was kept at 2 Hz, which is close to the natural frequency of the bed. Gas velocity was ~0.20 m/s with a narrow variation of 10 %. Cases SPF-FT1 to SPF-FT5 were tested with a feed rate of ~1.0 kg/h and temperature ranged from 250 °C to 330 °C; cases SPF-FT6 to SPF-FT11 with a feed rate of ~1.5 kg/h and temperature ranged from 250 °C to 350 °C; cases SPF-FT12 to SPF-FT15 with a feed rate of ~2.0 kg/h and relatively narrow temperature window from 280 °C to 320 °C. SPF-FT16 and SPF-FT17 were two special cases to access the impact of preheated gas temperature and fine particles, respectively.

Cases HEM-FT1 to HEM-FT5 were performed with hemlock sawdust as the reference cases. Cases HEM-O1 to HEM-O7 were performed with hemlock sawdust to investigate torrefaction in an oxygen-lean gas environment. By varying operating conditions, responses in torrefied product properties, such HHV, volatile and fixed carbon contents by proximate analysis, and elemental contents, were expected. Torrefied products were not extensively characterized as the torrefaction process itself and its use and performance are out of the scope of this work.

### 4.2 Temporal temperature and pressure profiles, and solid product evolution

Temporal sawdust bed temperature profiles for case SPF-FT2 (P2-F1.0) are shown in Figure 4.1 as an example.  $T_5$  at the discharge side was the highest, followed by  $T_4$  in the middle of the bed and  $T_3$  at the feed side. The temperature difference between  $T_3$  and  $T_5$  in this specific case was 116 °C. This value mostly was ~120–150 °C, with the lowest of 81 °C at the lowest feed rate and bed temperature (case SPF-FT1), and the highest of 216 °C when the feed rate and the bed temperature were close to the highest (case SPF-FT14). In general, the temperature difference increased as the feed rate increased and the bed temperature increased, which is associated with the increased heating demand of biomass traveling from left to the right. However, compared to case SPF-FT15, case SPF-FT14 was operated with a slightly lower feed rate and ~10 °C lower bed temperature, but with a much lower set point of the bed electric wall heater and similar gas preheated temperature ( $T_1$ ) resulted in a higher temperature difference. Therefore, it is suspected that different contributions from two heating methods could affect the spatial temperature gradient in the bed, i.e., a higher preheated gas temperature and a lower bed heater setting point led to a smaller temperature difference, associated with the heating of biomass when they travel from side to side.



Figure 4. 1 Typical bed temperature profiles over time.

Temperatures dropped and fluctuated more immediately after the SPF sawdust particle at room temperature was fed to the reactor at time 0. With minimal adjustments at the beginning of this run, after  $\sim$ 30 min all temperatures leveled off as torrefaction progressed. The last 30 min of the stable operation of each run was averaged over time, and subsequently used to calculate the average bed temperature listed in Table 4.1. T<sub>3</sub> and T<sub>4</sub> fluctuated around the time-averaged mean temperature with a margin of 6 °C, while T<sub>5</sub> with a narrower fluctuation of 2 °C. This is true for all cases, and the fluctuations of T<sub>3</sub> and T<sub>4</sub> varied depending on the pulsation frequency (details in Section 4.3) and the feeding conditions.

Temporal combustor bed temperature profiles for case HEM-FT1 are shown in Figure 4.2.  $T_7$ ,  $T_8$ , and  $T_9$  located at the lower, middle, and top regions of the catalyst bed. All temperatures fluctuated more after the sawdust entered the torrefier.  $T_7$  started to rise first several minutes after torrefaction took place, followed by the rise of  $T_8$  and  $T_9$ . Combustor heaters were turned off as the torrefaction

started. Thus, the temperature rise was solely due to the heat released by the togas combustion. The increase in combustor temperatures gradually slowed down as the torrefaction progressed, yet generally kept increasing towards the end of each run. For cases with high volatile organic compounds released, the temperatures went up to as high as 700 °C, even with the catalyst dispersed in inert packing materials. Air to the combustor was cut down in those cases to maintain the catalyst bed temperatures at constant to protect the noble metal catalyst.



Figure 4. 2 Typical combustor temperature profiles over time.



Figure 4. 3 Typical profiles of (a) pressures drop of bed and distributor, (b) reactor pressure, (c) filter pressure drop and (d) combustor pressure drop over 30s.

30 s pressure profiles for case HEM-FT3 are shown in Figure 4.3. Pressure drop across the bed and distributor, pressure in the reactor, pressure drops of the filter and combustor primarily fluctuated because of the gas pulsation. A sudden decrease in the filter pressure drop, or increase in the other three measurements, caused by the filter purging gas every 30 s, appeared at around the same time, after which the pressure returned to the regular fluctuation pattern.

Moving average over 1 min of the pressure data was used to smooth the curve so that the trends during the run could be identified easily, as shown in Figure 4.4. Pressure drop across the bed and distributor stabilized after ~30 min, similar to the temperature profiles in previous discussions. The reactor pressure increased faster at the first 30–40 min and slowed down afterward. It is noteworthy that the filter pressure drop kept increasing at an almost constant pace over the entire run due to the steady cake building up on the filter, which indicated that fine particles were entrained and captured by the filter. The combustor pressure drop also followed the same trend as the reactor pressure. However, the slow increase of pressure could result from the increasing combustion temperature or the tar fouling on the exhaust pipeline.

With the observations on the temperature and pressure profiles over each run, it can be concluded that torrefaction in the pulsed fluidized bed generally reached a relatively steady state after 30 min for each run. Therefore, the time-averaged temperature at each location was calculated using the data after 30 minutes, and mostly the data over the last 30 min of steady operation. Torrefied product from the last stage was collected and analyzed subsequently.



Figure 4. 4 Typical profiles of moving averaged (a) pressures drop of bed and distributor, (b) reactor pressure, (c) filter pressure drop and (d) combustor pressure drop over time.

Solid product samples were drawn from the discharging pipe, and the color change of each sample compared to the raw sawdust was subsequently measured. Color measurement as a non-destructive method was used to quickly characterize torrefied products based on the established relationship between the color value change ( $\Delta E$ ) and other properties of torrefied products (Huang et al., 2020). Color value change of solid samples of three non-interrupted cases: SPF-FT8 (P2-F1.5) with low torrefaction severity; SPF-FT10 with moderate torrefaction severity; and SPF-FT11 with high torrefaction severity are plotted in Figure 4.5. From 15 min, when the first sample was taken, no

substantial variations were displayed in the color value change to the end of the run, demonstrating the steady production of torrefied materials with constant properties.



Figure 4. 5 Color value change over time for cases SPF-FT8 (P2-F1.5), SPF-FT10 and SPF-FT11.

## 4.3 Effect of pulsation frequency

As mentioned in previous sections, the bed temperature varied differently at different pulsation frequencies. T<sub>3</sub>, T<sub>4</sub>, T<sub>5</sub> for cases P1-F1.0 to P5-F1.5 are plotted in Figure 4.6 to illustrate the variations. Error bars are the standard deviation of the temperature data over the last 30 min. It can be seen that T<sub>3</sub> and T<sub>4</sub> in all cases had more significant fluctuations than T<sub>5</sub>, particularly at high pulsation frequencies where gas pulsation was not intense enough to keep the mobility of particles in the bed. T<sub>5</sub> was rather constant across all cases at a relatively constant feed rate, whereas T<sub>3</sub> and

 $T_4$  decreased with the pulsation frequency.  $T_4$  and  $T_5$  overlapped at the cases of P5-F1.0 and P3-F1.5, and even flipped at pulsation frequency of 4 Hz and 5 Hz with a feed rate of 1.5 kg/h. This is probably due to the uneven hole arrangement of the distributor, leading to less hot gas coming through the middle section of the bed.



Figure 4. 6 Bed temperatures for cases (a) P1-F1.0 to P5-F1.0 and (b) P1-F1.5 to P5-F1.2.



Figure 4. 7 Effect of pulsation frequency on HHV of torrefied sawdust for cases (a) P1-F1.0 to P5-F1.0 and (b) P1-F1.5 to P5-F1.2.

Weight loss and HHV of the torrefied sawdust for cases P1-F1.0 to P5-F1.5 are shown in Figure 4.7 and Figure 4.8, respectively. Overall, pulsation frequency did not significantly impact weight loss and HHV of the product over the narrow test range. Weight loss was ~27% for cases with a feed rate of ~1.0 kg/h and ~28% for cases with a feed rate of ~1.5 kg/h. Considering the small errors presented, maximum weight loss was seen in the frequency range of 2–4 Hz. A slight decrease, by up to 8% away from the average value at 1 Hz and 5 Hz could be noted, despite the slightly higher T<sub>5</sub> at 1 Hz, as shown in Figure 4.6. The constant trend suggests the optimum performance can be expected within the range of 2–4 Hz, which is close to the bed natural frequency of ~2 Hz. When

the gas pulsation frequency matches the bed's natural frequency, dissipation caused by adding up two types of vibration, i.e., gas flow motion and the second-order vibration of the bed, is minimized and the gas pulsation energy input is intensified. HHV of the SPF sawdust increased approximately the same amount regardless of the pulsation frequency. The HHV ratio of torrefied sawdust over raw sawdust in all ten cases was  $\sim$ 1.05, which indicates a mild torrefaction process where changes in the product properties may not be appreciable (Keivani et al., 2018; Kung et al., 2019; McNamee et al., 2016).



Figure 4. 8 Effect of pulsation frequency on weight loss for cases (a) P1-F1.0 to P5-F1.0 and (b) P1-F1.5 to P5-F1.2.

In Figure 4.9, as expected, volatile matter in the torrefied SPF sawdust is lower, and fixed carbon and ash content are higher. Again, no significant variations in proximate analysis at different pulsation frequencies. Similar results are also seen in the ultimate analysis, hence they are not charted in this section.



Figure 4. 9 Effect of pulsation frequency on volatile matter, fixed carbon, and ash content for cases (a) P1-F1.0 to P5-F1.0 and (b) P1-F1.5 to P5-F1.2.

## 4.4 Effects of temperature and feed rate

Based on the previous observations, pulsation frequency was fixed at 2 Hz for the rest of the experiments. It is found that changes in torrefied product properties were more responsive to the highest bed temperature at the discharge side  $T_5$ . Therefore, contour graphs of weight loss and product properties of cases SPF-FT1 to SPF-FT16 with respect to  $T_5$  and feed rate are presented.

Figure 4.10 shows the effects of temperature and feed rate on the total weight loss of the torrefaction process. Each black point refers to one experiment. Weight loss of all tested cases in this section ranged from 16% to 64%. Four presumably extreme cases with weight losses over 45%, which might not be practical, were conducted to provide a comprehensive insight into the reactor performance. In addition, weight loss in fluidized beds might include the loss due to the entrainment of fine particles (Li et al., 2012), which also probably occurred in this study, suggested by the constant increasing filter pressure drop. Thus, the actual weight loss due to torrefaction could be less than the values reported here. As the temperature increased, weight loss increased dramatically, whereas weight loss increased slightly as the feed rate decreased. The strong dependency on temperature has been reported in the literature (Campbell et al., 2018; Granados et al., 2017; Kung et al., 2019; Mei et al., 2015; Ohliger et al., 2013; Strandberg et al., 2015; Wang et al., 2019). With the fixed bed volume and constant gas velocity, solids residence time decreased when feed rate increased, leading to lower weight losses. Responses to the change of feed rate were small, yet the trend was consistent throughout. Additionally, the residence time variation might be within a small range of less than 15 min, according to the mass turnover time calculated under similar operating conditions in the previous Chapter, and hence had less impact on the process.



Figure 4. 10 Effects of temperature and feed rate on weight loss.

HHV of torrefied sawdust shown in Figure 4.11 displays the same trend as that of the weight loss. A drastic increase of 19% to 24.96 MJ/kg was observed as  $T_5$  increased from 320 °C to 370 °C.



Figure 4. 11 Effects of temperature and feed rate on HHV of the torrefied sawdust.

Figure 4.12 shows the volatile matter and fixed carbon weight fractions of torrefied sawdust on dry basis. As torrefaction severity increased, volatile matter content decreased, and fixed carbon content increased. This trend agrees with the general understanding that torrefaction removes the volatile components in the biomass and generate more thermally stable compounds that fall into the fixed carbon classification (Nachenius et al., 2015b; Strandberg et al., 2015). Ash content and elemental compositions are summarized in Table 4.3. As the torrefaction severity increased, ash content increased as it was retained in the solid product. The ash content of cases SPF-FT1 and SPF-FT6 was slightly lower than the ash content in the raw SPF sawdust, which could also be evidence for the fine particle entrainment, as the SPF particles that are less than 250 µm had an ash content of 2.70%. Loss in the fine particles resulted in decreased ash content in the product.



Figure 4. 12 Effects of temperature and feed rate on volatile matter and fixed carbon weight fraction of torrefied biomass.

Carbon content generally increased, and hydrogen and oxygen contents decreased after torrefaction due to decarboxylation and dehydration of hemicellulose and cellulose (Wang et al., 2017a; Wang et al., 2016). However, the relationships of elemental contents with the torrefaction severity were not as clear as other product properties. Case SPF-FT5 had the highest carbon content with a weight loss lower than that of case SPF-FT11. This might be due to the fairly large errors in the carbon content measurement. Results of elemental C, H, and O contents are also usually presented by the Van Krevelen diagram, which is given in Section 4.5.

Case name	F,	T5,	Weight	Ash content,	C, wt. %	H, wt. %	0, wt. %
	kg/h	°C	loss, %	wt. %			
Raw SPF	-	-	-	0.64±0.03	49.83±1.12	6.31±0.14	43.24±1.26
SPF-FT1	1.01	273	17.4	$0.62 \pm 0.00$	51.85±0.21	5.84±0.16	41.69±0.38
SPF-FT2 (P2-F1.0)	1.01	304	29.1	$0.76 \pm 0.06$	53.28±0.70	5.77±0.04	$40.20 \pm 0.65$
SPF-FT3 (P2-F1.0)	1.02	304	27.6	$0.71 {\pm} 0.05$	52.98±1.14	$5.80 \pm 0.04$	40.52±1.10
SPF-FT4	0.98	331	43.3	$0.74 \pm 0.02$	55.48±0.62	$5.74 \pm 0.04$	38.05±0.57
SPF-FT5	1.01	354	57.2	$0.96 \pm 0.07$	61.32±0.02	5.17±0.03	32.55±0.05
SPF-FT6	1.50	276	16.1	$0.62 \pm 0.05$	51.60±0.31	5.88±0.01	41.90±0.32
SPF-FT7 (P2-F1.5)	1.41	312	29.6	$0.82{\pm}0.01$	52.96±0.48	$5.78 \pm 0.04$	40.45±0.43
SPF-FT8 (P2-F1.5)	1.47	311	28.1	$0.81{\pm}0.05$	53.76±0.63	$5.50 \pm 0.01$	39.93±0.62
SPF-FT9	1.47	345	48.5	1.14±0.06	57.09±1.71	5.41±0.14	36.36±1.57
SPF-FT10	1.48	348	49.6	$1.08 \pm 0.14$	58.70±0.27	5.37±0.02	34.86±0.26
SPF-FT11	1.49	371	64.0	$1.34 \pm 0.08$	58.01±1.95	5.46±0.17	35.40±0.33
SPF-FT12	1.89	325	32.2	$0.72 \pm 0.01$	52.78±0.01	$5.87 \pm 0.00$	40.63±0.01
SPF-FT13	2.04	318	30.8	$1.00{\pm}0.07$	52.08±0.28	5.81±0.02	41.11±0.26
SPF-FT14	2.05	331	35.2	$0.73 \pm 0.02$	53.60±0.68	$5.80 \pm 0.05$	39.87±0.62
SPF-FT15	2.11	351	45.9	1.28±0.11	57.11±0.65	5.46±0.09	36.16±0.56
SPF-FT16	1.53	335	40.5	0.73±0.01	54.56±0.19	5.80±0.11	38.90±0.30

Table 4. 2 Ash content and elemental C, H and O contents in torrefied sawdust.

### 4.5 Weight loss and product properties

Previous research has proven that product properties are closely linked to weight loss (discussed in Chapter 1), based on which several torrefaction indices were developed (Campbell et al., 2019). From the analysis of the effect of operating conditions on torrefied product, a strong correlation between weight loss and product properties is also seen in this work. To confirm and correct the weight loss caused by the fine particle entrainment, case SPF-FT17 was conducted. Fine particles less than 250  $\mu$ m were sieved out prior to being loaded into the feeder hopper. Weight loss of case SPF-FT17 was found to be 33.4%, lower than the value obtained from the contour map in Figure 4.10 (which was established from normal tests without fine particles removed from the raw biomass) by 7.6%, which was very close to the mass fraction of particles smaller than 250  $\mu$ m in the raw SPF sawdust, 7.89%. Therefore, the measured weight loss was corrected by subtracting 7.89%, to give a corrected weight loss, *WL*\*.

A linear relationship between weight loss and product properties was first explored, as it is simple and has been used in many studies (Almeida et al., 2010; Chen et al., 2015c; Grigiante & Antolini, 2015; Rago et al., 2018; Sabil et al., 2013). The weight loss in those studies was generally lower than 40%, which was the range expected from typical torrefaction pretreatment, whereas in this work, the corrected weight losses in two cases were over 45%. Linear regression of HHV, content of proximate components, and carbon content on corrected weight loss was performed with the data of cases except cases SPF-FT5 and SPF-FT11, shown in Figure 4.13–4.15. Case SPF-FT17 plotted against the actual weight loss is highlighted in red in all graphs and appears to be in agreement with other cases, validating the weight loss correction method. R<sup>2</sup> values are larger than 0.91 for HHV, volatile matter, and fixed carbon content, demonstrating strong linear 109 relationships, and 0.81 for carbon content, which has a relatively larger error in the measurement. In Figure 4.13 and Figure 4.14, the two outlining data points that are not included in the regression clearly deviate from the linear relationship. Additionally, the raw sawdust properties also deviate from the regression lines more than other data do.



Figure 4. 13 Correlation between weight loss and HHV for SPF with corrected weigh loss less than 50%.



(a)



Figure 4. 14 Correlation between weight loss and (a) volatile matter and (b)fixed carbon content for SPF cases with corrected weigh loss less than 50%.



Figure 4. 15 Correlation between weight loss and carbon content for SPF cases with corrected weigh loss less than 50%.

To improve the correlation while including the data for raw biomass and the results with high weight loss, the ratio of raw biomass weight over torrefied biomass weight, i.e.,  $\frac{1}{1-W_L^*}$  was used to correlate with torrefied product properties. Note that this weight loss ratio was also used by Nachenius et al. (2015b). Improved linear relationships are found between HHV, proximate component contents, and carbon content for all data with R<sup>2</sup> of 0.9670, 0.9643, 0.9611, and 0.8052, respectively, as shown in Figure 4.16–4.18. The only correlation that is not improved is for the carbon content. However, Figure 4.18 shows that the point (2.28, 58.01) appears to be an outliner with large error bars. Figure 4.19 re-plots the carbon content data without the outliner, and the R<sup>2</sup> value is improved to 0.9083.



Figure 4. 16 Correlation between the ratio of raw and torrefied sawdust weight and HHV for SPF cases.





Figure 4. 17 Correlation between the ratio of raw and torrefied sawdust weight and (a) volatile matter and (b) fixed carbon content for SPF cases.



Figure 4. 18 Correlation between the ratio of raw and torrefied sawdust weight and carbon content for SPF

cases.



Figure 4. 19 Correlation between the ratio of raw and torrefied sawdust weight and carbon content for SPF cases without an outliner.

Elemental analysis results of all SPF cases are also presented in the van Krevelen diagram (Figure 4.20), where the atomic hydrogen to carbon ratio is plotted against the atomic oxygen to carbon ratio of each case. In Figure 4.20, the red square refers to the raw SPF sawdust, while each circle represents the torrefied sawdust of one experiment, with color mapping with respect to the weight loss. As the weight loss increases, the atomic H/C and O/C ratios decreases, and the value in the van Krevelen diagram moves towards the bottom-left quadrant. A strong linear relationship is observed between the atomic H/C and O/C ratios. The trend is consistent with the results given in the literature (Keivani et al., 2018; Martín-Lara et al., 2017; Nachenius et al., 2015b; Peng et al., 2013; Zhang et al., 2018).



Figure 4. 20 The van Krevelen diagram for raw (red square) and torrefied sawdust (color circles) (colormap of weight loss, i.e., the darker the color the higher the weight loss is).

## 4.6 Oxidative torrefaction

#### 4.6.1 Solid product characterization

It is found that torrefaction in the oxidative environment results in higher weight loss, compared to that in the non-oxidative environment, due to the oxidative reactions the material underwent (Brachi et al., 2019b; Chen et al., 2013; Wang et al., 2013; Wang et al., 2018; Zhang et al., 2019). However, limited tests under an oxygen-laden environment were conducted in this work due to the difficulties in controlling the temperature and feed rate (further discussed in Appendix C). Thus, weight loss cannot be directly compared between cases. Nevertheless, since it is confirmed in the previous section that the weight loss and product properties are strongly correlated, the properties 116

of oxidative products are compared with those of non-oxidative products as a function of the weight ratio.

Figure 4.21 shows that the HHVs of both oxidative and non-oxidative products are in line with each other, which means the oxidative environment did not significantly affect product HHV over the tested range, where the highest weight loss was 20% with ~3% oxygen concentration. This result is not consistent with the constant lower HHV of torrefied material in oxidative environments reported by Brachi et al. (2019b), and a slight decrease in HHV at a temperature of 240 °C and 270 °C reported by Wang et al. (2018). Zhang et al. (2019) pointed out that oxidative torrefaction may have different mechanisms under different temperatures and oxygen contents. With a higher weight loss at a higher temperature and oxygen concentration, the HHV may deviate from non-oxidative products. Figure 4.22 shows that the oxidative torrefaction product has a slightly lower volatile matter and a slightly higher fixed carbon content, which was also found in the studies mentioned above. Note that the low content of volatile matter, even with the same HHV value, suggests a low reactivity (Brachi et al., 2019a).



Figure 4. 21 HHV of non-oxidative and oxidative torrefaction products as a function of the ratio of raw and torrefied sawdust weight.





Figure 4. 22 (a) Volatile matter and (b) fixed carbon of non-oxidative and oxidative torrefaction products as a function of the ratio of raw and torrefied sawdust weight.

### 4.6.2 Carbon balance

Permanent gas contents in the combustor flue gas were measured for five cases with a moderate feed rate of 1.35-1.47 kg/h and weight loss of 10-16%, hence with a manageable amount of torgas generated. CO content was negligible in all cases, indicating the complete combustion of the volatile organic compounds. Therefore, it could be considered that the carbon loss from the solid phase was completely converted to the final product, CO<sub>2</sub>, in the flue gas. A simple carbon balance can be carried out for each run to identify the carbon and mass distribution of the process. Carbon yield in the solid phase is calculated by Eq. (4.2),
$$Carbon \ yield_{s} = \frac{Carbon \ content_{torrefied} \times F \times (1 - W_{L})}{Carbon \ content_{raw} \times F} \times 100\%$$
(4.2)

where carbon content in the raw and torrefied sawdust was determined by ultimate analysis; F (kg/h) denotes the actual sawdust feed rate;  $W_L$  (%) is the measured weight loss of the process. Carbon yield in the gas phase is calculated as Eq. (4.3),

$$Carbon \ yield_{fg} = \frac{M_{carbon}(Q_{CO_2}/V_m)}{Carbon \ content_{raw} \times F} \times 100\%$$
(4.3)

where the carbon molar mass  $M_{carbon}$  equals 12.0 g/mol; carbon dioxide flowrate  $Q_{CO_2}$  (SL/h) is calculated by the measured CO<sub>2</sub> concentration multiplied by total gas flow rate; the standard molar volume  $V_m$  equals 22.4 L/mol. The total gas flow rate is assumed to be constant for the flue gas, as the volume of vapors released in the reaction is less than 2 vol.% of the total flue gas flow according to preliminary calculations based on the data reported by (Prins et al., 2006b).

Results are summarized in Table 4.3. Total carbon yields in solid and flue gas are generally higher than 95%. The discrepancy could stem from the fine particles captured on the filter and errors in the carbon content measurement in the solid product, which is highly scattering.

Case number	Carbon yields, %	Carbon yield <sub>fg</sub> , %	Total carbon yield, %
HEM-O1	91.9±0.7	3.4±0.2	95.3±0.7
HEM-O3	92.2±0.8	5.3±0.2	97.5±0.9
HEM-O4	89.9±0.6	$7.4{\pm}0.6$	97.3±0.8
HEM-O6	92.9±1.2	4.8±0.6	97.8±1.4
HEM-07	92.0±0.6	3.5±0.1	95.5±0.6

Table 4. 3 Carbon balance for cases HEM-O1, HEM-O3, HEM-O4, HEM-O6, and HEM-O7.

## 4.7 Comparison with other continuously operated torrefaction processes

In order to compare torrefaction performance across different reactor types, evaluation metrics in this work consists of weight loss, energy yield, and enhancement factor. The metrics are chosen because they are easy to be evaluated regardless of the operating conditions that may be defined variously among different reactors. The weight loss ( $W_L$ ) over the entire run was directly measured after each torrefaction experiment, as described in Chapter 2. Energy yield calculates the energy retained in the torrefied product, expressed as Eq. (4.4),

$$Energy \ yield = \frac{HHV_{torrefied} \times (1 - W_L)}{HHV_{raw}} \times 100\%$$
(4.4)

where  $HHV_{torrefied}$  (MJ/kg) is the HHV of the torrefied product. Enhancement factor defined by (Zhang et al., 2019) as the ratio of the HHV of the torrefied material and that of the dried raw materials as Eq. (4.5)

$$Enhancement \ factor = \frac{HHV_{torrfied}}{HHV_{raw}} \tag{4.5}$$

which was also used by (Kung et al., 2020) as a torrefaction severity index.

Original weight loss and corrected weight loss with corresponding energy yield results for nonoxidative torrefaction cases are given in Table 4.4. Energy yield based on corrected weight loss generally increased by ~8% for SPF and ~3% for hemlock because the mass fraction of hemlock less than 250  $\mu$ m is 3.8%, compared to 7.89% of SPF. The entrainment can be avoided by removing the fines. Thus, the corrected weight loss and energy yield are compared with other reactors later.

As shown in Figure 4.23, the energy yield decreases with increasing enhancement factor almost linearly. To increase the energy density by 15% (enhancement factor = 1.15), about 20% energy content in the feedstock will be lost, although this amount of energy may be used for drying and torrefaction. From design and optimization points of view, a suitable enhancement factor should be selected based on the requirement of energy for autothermal operation, and desired quality of pellets (e.g., energy density, hardness, water resistivity) in order to maximize the energy recovery of torrefied product.

Case name	WL, %	Energy yield,	WL*, %	Energy	Enhancement
		%		yield*, %	factor
SPF-FT1	17.4	84.1	9.5	92.1	1.02
SPF-FT2 (P2-F1.0)	29.1	74.8	21.2	83.1	1.05
SPF-FT3 (P2-F1.0)	27.6	76.1	19.7	84.4	1.05
SPF-FT4	43.3	63.2	35.4	71.9	1.11
SPF-FT5	57.2	50.4	49.3	59.7	1.18
SPF-FT6	16.1	84.3	8.2	92.3	1.01
SPF-FT7 (P2-F1.5)	29.6	73.8	21.7	82.1	1.05
SPF-FT8 (P2-F1.5)	28.1	75.4	20.2	83.6	1.05
SPF-FT9	48.5	59.0	40.6	68.1	1.15
SPF-FT10	49.6	57.7	41.7	66.7	1.14
SPF-FT11	64.0	45.8	56.2	55.8	1.27
SPF-FT12	32.2	72.6	24.3	81.0	1.07
SPF-FT13	30.8	73.4	23.0	81.8	1.06
SPF-FT14	35.2	71.1	27.3	79.8	1.10
SPF-FT15	45.9	62.3	38.0	71.3	1.15
SPF-FT16	40.5	66.0	32.6	74.7	1.11
SPF-FT17	33.4	73.6	33.4	73.6	1.10
HEM-FT1	16.2	87.7	12.4	90.7	1.04
HEM-FT2	21.9	82.5	18.1	85.6	1.05
HEM-FT3	20.2	83.6	16.4	86.7	1.04
HEM-FT4	19.8	83.5	16.0	86.5	1.03
HEM-FT5	33.2	74.4	29.4	77.9	1.10

Table 4.4 Weight loss, energy yield and enhancement factor of all non-oxidative torrefaction cases.

\* After correction for fines loss.



Figure 4. 23 Energy yield as a function of the enhancement factor.

One representative of each type of continuous torrefaction reactor in the literature was selected, namely, the rotary drum (RD) torrefaction reactor designed and constructed in collaboration among Umeå University, Swedish Agricultural University and BioEndev AB (Strandberg et al., 2015); the screw conveyor (SC) reactor investigated by Nachenius et al. (2015b); the moving bed (MB) reactor at ECN's research facilities (Nanou et al., 2016); the spouted bed (SB) reactor at Fluidization Research Center at UBC (Wang et al., 2019), to compare with the pulsed fluidized bed (PFB) torrefaction.

The MB reactor developed by ENC has the largest scale with a design capacity of 50 kg/ h, followed by the RD with a design capacity of 20 kg/h. SC, SB and PFB have a similar scale of around 2 kg/h. A similar kind of softwood torrefaction is chosen to enable comparison. However, the biomass sample size varies with the type of reactors. SB and PFB utilized smaller particles for successful spouting or fluidization, whereas MB and RD torrefied much bigger wood pieces.

Torrefaction temperatures were measured differently for different reactors, and short residence times were used in SC and PFB as a result of higher temperature applied and utilization of smaller particles. In the SB, products were collected from the cyclone with a shorter residence time and from the reactor with a longer residence time. Most of the experiments resulted in the weight loss range of  $\sim 5-40\%$ , with several extreme cases with weight loss around or over 50% in RD, SC, and PFB. Note that the weight loss reported in RD was calculated based on the constant ash content. All studies achieved ~60–99% energy yields, with over 98% yields in RD, SC, and SB. As to the enhancement factor, SB has slightly higher values than other cases, especially for products collected in the reactor with residence time longer than other studies. Overall, the PFB is proven to perform sawdust torrefaction with a comparable torrefied product as other explored reactor technology. Operating complexity, scale-up ability, energy consumption, and other aspects are not discussed here. In particular, the performance of PFB is very close to the SC in the way of utilizing small particles with low residence time. Nevertheless, PBF delivered the extremely high weight loss cases in an even shorter time than the SC did. Moreover, the SC employed a stack of two over 1.5 m long screws due to the limited degree of filling, whereas the PFB reactor was 0.375 m in length. Therefore, the PFB reactor would have an edge when performing severe reactions such as pyrolysis and at a larger scale.

Reactor	Rotary drum Screw conveyor		Moving bed	Spouted bed	Pulsed fluidized bed
Scale	3.8–12 kg/h, ~2–6 h	0.79–2.48 kg/h, at least 1 h	33.3 kg/ h torrefied product, 36.9 h	0.6–1.4 kg/h, 50 min or 120 min	1–2 kg/h, 2.5 h
Material	Spruce, $8 \times 8 \times 4.5$ – $25 \times 25 \times 4.5 \text{ mm}$	Pine, 100 % <6mm, 66 % >2 mm	Spruce, $2 \times 2 \times 2-40$ $\times 40 \times 15$ mm	SPF, 0.5–1.0 mm with a Sauter mean of 0.86 mm	SPF, < 2.8 mm with a Sauter mean of 0.59 mm
Temperature, °C	260–310 (biomass surface)	275–375 (wall)	260	244–304	273–371 (bed exit)
Residence time, min	8–25	5.75–16.5	32	Not specified	~6–13
Weight loss, %	3, 11, 20, 23, 54 <sup>daf</sup> (calculated)	~5–35 with an extreme case of 55	21	9.3–38.2	8.2–41.7, 49.3, 56.2 (corrected)
Energy yield, %	99, 94, 88, 84, 62	$\sim$ 75–98 with an extreme case of $\sim$ 60	86	73.3–99.6	66.7–92.3, 55.8, 59.7 (corrected)
Enhancement factor	1.01, 1.05, 1.09, 1.1 and 1.36	$\sim$ 1.03–1.13 with an extreme case of $\sim$ 1.33	1.08	1.07–1.13 in the cyclone, 1.11–1.42 in the reactor	1.01–1.15, 1.18, 1.27
Source	Strandberg et al. (2015)	Nachenius et al. (2015b)	Nanou et al. (2016)	Wang et al. (2019)	This work

Table 4. 5 Compariso	n of continuous	torrefaction i	performance of	different type	s of reactors in	the literature.
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daf Dry ash-free basis

# **Chapter 5: Conclusions and Future Work**

#### 5.1 Conclusions

This work demonstrates the potential application of a pulsed fluidized bed (PFB) for biomass thermal conversion. Narrow residence time distribution and limited solid backmixing were achieved in the horizontal configuration, leading to uniform quality of torrefied product. Solids transport and mixing were characterized by a modified axial dispersion model. By fitting the model to the solids residence time distribution curves measured in the PFB with continuous feeding and discharging biomass particles under room temperature, effects of weir height, gas pulsation frequency, feed rate, and gas velocity on solids mixing were investigated systematically. Continuous biomass torrefaction was successfully performed in a PFB unit to prove the concept with parameters such as gas pulsation frequency, temperature, feed rate and oxygen content in the carrying gas investigated.

Key findings of this work are listed below.

- Residence time distribution (RTD) curves of hemlock particles measured at different operating conditions, along with the peak time and mass turnover time, indicated the existence of a stagnant zone in the horizontal PFB of a rectangular cross-section.
- A 2D axial dispersion model with an exchange flow between the active zone and stagnant zone was proposed for the horizontal PFB and was applied to extract solids dispersion coefficients from the experimental data.

- Horizontal dispersion coefficients were slightly higher in the deeper bed. In the shallow bed, the coefficient increased with increasing particle velocity induced mainly by increasing solids feed rate. Lower degree of backmixing was observed when the bed was operated under a gas pulsation frequency close to the bed natural frequency. The horizontal solids mixing intensity increased greatly with increased gas velocity.
- Horizontal solids dispersion coefficients obtained in horizontal PFB were significantly smaller than values calculated from literature correlations for horizontal fluidized beds. A correlation of horizontal dispersion coefficients for biomass particles in the horizontal PFB of shallow beds was established with a fairly good agreement with the data.
- The horizontal PFB reactor proves to be a promising technology for biomass torrefaction, as the prototype unit successfully produced over 100 kg torrefied biomass materials with greater higher heating value (HHV), less volatile content, greater fixed carbon content, and lower hydrogen and oxygen contents.
- Temperature difference as much as 200 °C was observed from the solids feed side to the discharge side of the bed. Based on the temporal pressure and temperature profiles of the bed, steady state operation of the reactor was achieved after 30 min of uninterrupted operation.
- Greater temperature fluctuations were associated with pulsation frequency of 4 Hz and 5 Hz, where the intensity of the gas pulsation was weaker than that of the lower pulsation frequencies tested in this work. Maximum weight loss was identified at the frequency range of 2–4 Hz for feed rate of 1 kg/h and 1.5 kg/h. However, pulsation frequency did not influence significantly on the HHV of the product over the tested range.

- The temperature near the reactor exit exhibited the most significant influence on the biomass weight loss and on the product properties. Dependency of the properties on feed rate was weak, as the mean residence time varied in a narrow range under the tested operating conditions.
- For torrefaction of SPF using inert fluidizing gas, strong linear relationships were found between the product properties, i.e., HHV, proximate component contents, and carbon content and the ratio of raw biomass weight over torrefied biomass weight.
- Torrefaction of hemlock sawdust in fluidizing gas with 3–6 vol.% oxygen concentrations
  resulted in torrefied materials of similar HHV and less volatile matter content at the same
  biomass weight loss.
- Compared to other continuous torrefaction technologies, the PFB torrefier utilized small particles with short residence time and achieved similar enhancement factor.

### 5.2 Future work

- Although most of the experiments were successfully conducted, the PFB torrefaction unit was under continuous improvement and troubleshooting as more experiences in operating the system were gained. Problems encountered during the experiments are described in Appendix C, provided with possible solutions or recommendations.
- It is suggested in Chapter 3 that the bed contains a considerate amount of stagnant volume.
   To verify the postulation, more thermocouples could be carefully installed at the bed corners, where the stagnant volume would mostly form, without contacting the reactor wall.
   A higher temperature than that of the active zone could indicate the existence of the stagnant zone.

- In this work, a catalytic combustor installed at the exit of the PFB torrefier eliminated the volatiles with low heating values, which is an integral part of the unit to realize the recycling of heat to directly heat the reactor bed. To efficiently utilize the heat released from the torgas combustion, the performance of the combustor needs to be studied. The global kinetics of the catalytic combustion would then guide the design and operation of the torgas combustion unit. Furthermore, the use of oxygen provided in the carrier gas in the oxidative torrefaction for complete combustion of the volatiles in the catalytic combustor should be investigated to further simplify the operation.
- In scaling up the current reactor design, the reactor volume needs to be increased. However, the bed height should be maintained low to retain the effectiveness of gas pulsation. Therefore, the bed width and length should be increased to scale up the operation. To limit the solids mixing, a large length to width ratio should be kept. As the bed becomes wider, introducing biomass to the bed needs to be carefully designed to distribute the materials evenly. For long reactor, its footprint may be restricted. Alternatively, long channels can be curved or folded into different shapes with solids mixing reduced by the introduction of vertical baffles. Those possible designs should be explored in reactor scale-up.
- Besides the reactor scale-up, multiple aspects of the current PFB unit could be modified to improve the performance. Better gas distribution could be achieved with multicompartment design of the windbox, leading to a better control of bed temperature. A tapered bed bottom could be added to reduce the dead zone at the corners. To reduce the fine loss, an expanded free board could be applied. Gas pulsation was generated by a solenoid valve installed close to the gas inlet of the windbox in this work. Because the solenoid valve could only stand 300 °C, the bed had to be heated up further by the heaters

on the windbox and reactor. If other pulsation mechanism could be adopted, such as piston in each windbox compartment, gas with higher temperature could be fed into the windbox and no additional heating would be required. Biomass feed rate in this work was limited by the size of the hopper and the heating capacity. A secondary hopper could be installed on top of the existing hopper, with airlock device in between hoppers, so that the top hopper could be separately loaded and provide supply to the main hopper. If the flue gas could be recycled and heat could be integrated, the heating would not be an issue. Biomass particles were dried prior to the tests in this study. However, when designing a biomass thermal conversion process, the length of the PFB could be increased to allow for the drying and preheating of the particles in the same reactor.

• A steady state reactor model with horizontal solids dispersion should be developed to predict reactor performance and guide reactor design. The model needs to be validated by comparing with the experimental results, with properly selected model parameters under gas pulsation, before it is used to predict the reactor performance in many scenarios not tested in the current experiment. Those suggested reactor modifications, such as taper shaped bed to eliminate the dead zone, heating by carrier gas, could be simulated by the model to confirm the improvement. Operating with other feed stocks could be guided by the anticipated performance of the model with kinetic data and properties of other biomass materials applied. Extended operating window of the PFB could also be explored with the model.

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# Appendices

### Appendix A Additional system design

#### A.1 Gas distribution

Gas distribution for the current distributor design is estimated by a simplified COMSOL simulation using the Laminar Flow module. The windbox was modeled as Fluid and Matrix Domain with the estimated porosity of 0.375 and calculated permeability of  $2.499 \times 10^{-8}$  m<sup>2</sup>. The bed is simplified to a 2D geometry with height and length, and divided into 20 parts along the bed length. The distributor opening ratio of each part is calculated separately. The opening area modeled as Fluid Domain, was simplified as a rectangular slot with the same width as the bed and same area as the actual opening area. Gas flowrate distribution along the bed length of an empty bed with a superficial gas velocity of 0.2 m/s at 25 °C and 300 °C are shown in Figure A.1. Relatively even distribution was achieved with a lower gas flow rate near the center of the bed.



Figure A. 1 Gas distribution along the length of the bed at 25 °C (U =0.25m/s).



Figure A. 2 Gas distribution along the length of the bed at 300 °C (U =0.25m/s).

#### A.2 Instrumentation and data acquisition

This work includes reactor design and installation of reactor, tubing, instrumentation, and data acquisition system. The reactor drawings are shown in Figure 2.1 and Figure 2.2. The schematic and unit pictures are shown in Figure 2.3, where the process is simplified to show the key segments. A process and instrumentation diagram with all the details and labeling for each part is shown in Figure A.3. In addition, several LabVIEW programs were written to monitor and record the process. Figure A.4 shows the front panels with real-time display and inputs for rotameter and pressure readings. Figure A.5 and Figure A.6 show the block diagrams of the programs for temperature and pressure signals, respectively. Figure A.7 shows the block diagrams for solenoid valve control.



Figure A. 3 Process and instrumentation diagram of pulsed fluidized bed torrefaction unit.



Figure A. 4 Front panels of LabVIEW programs for this project



Figure A. 5 Block diagram of the temperature program



Figure A. 6 Block diagram of the pressure and flowrate program



(a)



Figure A. 7 Block diagram of the programs for (a) gas pulsation solenoid valve control (b) filter purging gas solenoid valve control

#### Appendix B Tracer particles for solids residence time distribution tests

In order to carry out solids residence time distributions tests, a sawdust tracer was prepared by impregnating NaCl on dry sawdust, as described in Section 2.6.1. Dried sawdust after soaking in high concentration NaCl solutions is shown in Figure B.1. No NaCl crystallization on the surface was observed after optimizing the drying process, and the prepared tracer was similar in appearance to the raw sawdust. The sawdust particles were divided into coarse, medium, and fine fractions, using two sieves with the opening of 1.18 mm and 0.085 mm, respectively, before and after the impregnation process (Table B.1). The ratio of the three fractions appeared to be similar, regardless of the impregnation process, indicating uniform weight gain for different size fractions.



Figure B. 1 Dried sawdust tracer.

Table B. 1 Weight	of coarse,	medium and	l small particles,	before and a	after impregnation.
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Condition	#	Coarse, g	Medium, g	Fine, g	Total weight, g	Ratio
Before	1	2.922	2.754	4.526	10.202	1:0.941:1.549
impregnation	2	2.952	2.962	4.224	10.138	11.003:1.431
Tracer	1	3.469	3.291	5.373	12.133	1:0.949:1.548
114001	2	3.477	3.451	4.963	11.891	1:0.993:1.427

Total 10 g of raw sawdust with different amounts of tracer samples were dissolved in 100 ml deionized water to mimic the residence time distribution measurement. The conductivity of the

solutions was subsequently measured and plotted against the tracer amount in the sample to construct a calibration curve, as shown in Figure B.2. It can be seen that the conductivity is linearly correlated with the tracer amount in the sample, with an R<sup>2</sup> of 0.9987. Moreover, the y axis interception is 177.6  $\mu$ S/cm, which is close to the conductivity measured with a pure raw sample of 158.3  $\mu$ S/cm. Therefore, the conductivity of the sample solution is corrected by subtracting the base conductivity of raw sawdust. The corrected conductivity is proportional to the weight of tracer and hence can be used to represent the tracer concentration in the collected sample.



#### Appendix C Operating issues and troubleshooting

#### C.1 Abnormal temperature rises during preheating using air

While the reactor was preheated with air, an abnormal temperature rise would occur at the gas exit of the pulsed fluidized bed or in the bed occasionally. As the gas exit temperature slowly increased and reached ~150 °C, a more rapid increase appeared, and the temperature usually plateaued before 300 °C. This event could be caused by combustion of the fine particles stuck on the filter or inbetween the filter and the reactor wall, as the pyrolysis biochar from softwood generated under 600 °C is prone to self-heating, because the fine biochar had a minimum ignition temperature lower than 200 °C as measured in the basket experiments (Restuccia et al., 2019). As the material for combustion was limited, the combustion temperature run-away at the gas exit was not a severe issue if the filter and gas exit area were fully cleaned after each run. On the contrary, a drastic bed temperature increased to over 600 °C happened sometimes when the bed temperature exceeded 200 °C caused by the combustion of particles, which could lead to temperature run-away. Particularly, if the combustion run-away took place closer to the feed, there is a chance to have the sawdust in the feeder ignited. Particles could be left on the distributor if the bed cleaning was not thorough, fall from the feeding port, or be blown into the reactor if the hopper purging gas was too strong. Thus, loose particles on the feed port were removed before each run, and nitrogen purging gas to the hopper was kept low and on at all times. In the case that the temperature was out of control, carried gas was switched to nitrogen early to finish the preheating.

## C.2 Feeding disturbances

It is not uncommon to experience difficulties in feeding biomass particles, as they are bulky, irregular in shape and size, and have poor flowability. As the time-averaged feed rate was calculated after each run, minor disturbances during the run could not be detected. However, the time-averaged feed rate of hemlock sawdust under elevated temperatures was not consistent with a constant screw speed setting, accompanied by noises from the increased friction. The disturbance could also cause bed temperature fluctuations during the run, as shown in Figure C.1. T<sub>3</sub> and T<sub>4</sub> oscillated periodically, much more than the normal fluctuation, likely due to unstable feeding. After noticing that the issue affected torrefaction tests, the smoothness of feeding hemlock particles was checked under room temperature and turned out to be satisfactory. It is postulated that the disturbance was caused by screw misalignment because of bending stemmed from aging and heat expansion. Moreover, the total length of the screw is  $\sim 1$  m and fastened at one end, which can shake/vibrate under too much stress. The hemlock feeding even completely stopped after 20 to 30 min of feeding during the oxidative torrefaction tests. As these tests were conducted at last, the severe feeding disturbance might be a result of continuous deterioration. The screw motor was turned off or set at speed doubled the target setting to restore the toque and overcome the friction. The duration of the run was recounted from the restart moment.


Figure C. 1 Bed temperature fluctuations caused by feeding disturbance.

## C.3 Thermal runaway in the oxidative torrefaction tests

The bed temperature in oxidative torrefaction tests could not be well-controlled with an oxygen concentration of 6% at a relatively high operating temperature due to the heat released from exothermic oxidation reactions. Figure C.2 illustrates the thermal runaway in the bed and the corresponding measures taken to lower the bed temperature. At ~20 min, the feeding stopped as described in the previous section and resumed subsequently. After several minutes, a rapid rise was seen in the bed temperatures. Air to the reactor was then turned off to stop the reactions, and temperatures dropped immediately. Once they fell back to the normal range at ~37 min, air was introduced again in the gas stream, and temperature at the discharge side increased immediately, and followed by  $T_3$  and  $T_4$ . After a second attempt to restart at a lower control temperature, the test was terminated due to the disturbance. Although in the presented data, the bed temperature seemed to be stabilized towards the end, a repeat test of this condition failed again due to the 156

thermal runaway. The substantial feeding fluctuation leading to large temperature fluctuations certainly aggravated the situation. The issue implied the existence of hot spots in the bed, probably in a relatively stagnant volume.



Figure C. 2 Thermal runaway during an oxidative torrefaction.

## Appendix D Continuous densification of torrefied materials in a pellet mill

Continuous densification of the torrefied sawdust produced in this work was carried out with a pellet mill (Model CL3, CPM, USA). According to Section 4.3, the effect of pulsation frequency on the product properties was not significant. Thus, torrefied sawdust from cases P1-F1.0 and P2-F1.0 were mixed to provide enough samples for continuously running the mill. Fine raw sawdust under the size of 250  $\mu$ m, which has been proven effective in binding torrefied materials in the densification process, was used as the binder (Peng et al., 2015). Different compositions of four tested samples are listed in Table D.1. All samples were conditioned to have a moisture content of 10% prior to pelletization.

#	Torrefied sawdust, %	Fine sawdust, %	Water, %
1	90	0	10
2	80	10	10
3	70	20	10
4	45	45	10

Table D. 1 Compositions of the samples to be pelletized.

Torrefied pellets and their densities are shown in Table D.2. The diameter and length of five pellets of each sample were measured with a caliper to calculate the pellet volume. The density was subsequently determined by the single pellet weight divided by the single pellet volume. The average of 5 repeated tests for each sample is reported in Table D.2. As the fine sawdust content increased, the color of the torrefied pellets became lighter, and the trend of pellet density was not clear with the errors in samples 2 and 3. Sample 4 with 45% fine sawdust had a slightly higher density, possibly due to the higher particle density of raw sawdust than that of the torrefied sawdust. However, pellets made from sample 3 were generally longer than other pellets,

suggesting an optimal binder content. More tests on energy consumption during the palletization and pellets durability need to be examined to identify the optimal amount of binders to be used.

#	1	2	3	4
Appearance				
Pellet density, kg/m <sup>3</sup>	<b>1087.2</b> ±8.4	1075.1±68.5	1141.2±54.3	1175.8±6.9

Table D. 2 Torrefied sawdust pellets.