

Study of gasification behavior for a biorefinery lignin waste in a fluidized bed gasification reactor

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Abstract

Lignocellulosic biorefineries, paper and pulp industries across the globe can convert cellulose and hemicellulose parts of the biomass into higher valued products. However, lignin from biomass is an underutilized biorefinery waste. Value-added applications of lignin waste should be investigated to produce low-molecular-weight compounds as an alternative to petrochemical compounds. Valorization and lignin recovery play an important role in 'green shifts' for such industries. In this work, the authors performed gasification of lignin pellets obtained from one biorefinery located in Finland. A 20-kW pilot-scale bubbling fluidized bed gasifier was used for the experiments. A computational particle fluid dynamics model based on a multi-phase particle in cell approach was developed for the same process. The developed model was validated against the experimental results.

The experimental results showed good conversion of lignin pellets into permanent light gases such as carbon monoxide, hydrogen, methane, etc. The average production of product gas and the lower heating value were 5.74 Nm³/hr and 4.95 MJ/Nm³, respectively. The average molar gas compositions obtained from the experimental study were 0.04 for CH₄, 0.16 for CO, 0.15 for CO₂, 0.13 for H₂ and 0.51 for N₂.

Keywords: lignin waste, lignin gasification, fluidized bed, CPFD

1. Introduction

Biomass is the world's fourth largest primary energy source, after coal, petroleum, and natural gas. Biomass now accounts for around 15% of total global energy use (Ankolekar and Kulkarni, 2018). The carbon present in the biomass is non-fossil in origin and considered as a renewable energy source. Biomass is a biobased material with a wide variety of sources such as lignocellulosic biomass, food waste, fish ensilage, etc. Lignocellulosic biomass primarily consists of cellulose, hemicellulose, and lignin. Lignin forms a part of the secondary cell walls of plants that helps to maintain the structural integrity of the plants. Lignin is the most abundant natural aromatic polymer which is found in most of the terrestrial plants on earth. Lignin accounts for 15–40% of dry weight (Ragauskas *et al.*, 2014) in terrestrial plants. The exact composition and the content of lignin in plants depend upon the botanical species. Millions of tonnes of lignin are produced as a byproduct from the paper, pulp industries, biorefineries, etc every year. The majority of lignin produced from such processes is currently used as a low-cost fuel to balance energy needs for the same process. Annually around 5–36×10⁸ tonnes of lignin are produced across the globe (Gellerstedt and Henriksson, 2008). Lignin residue can be used directly as fuel in combustion furnaces, boilers, etc. or can be used as binders, emulsifiers, concrete

additives, dyestuff dispersants, components for composites and copolymers, etc after chemical transformation (Calvo-Flores and Dobado, 2010). The major challenge for lignin conversion is its heterogeneous properties, such as molecular weight, functionality, and thermal properties depending upon the different sources and processing methods (Saito *et al.*, 2014). There are no significant applications of lignin waste on a commercial scale other than burning it as low-grade boiler fuel for an energy source.

Different alternatives for lignin conversion include biochemical conversion such as hydrolysis, fermentation, etc, and thermochemical conversions such as pyrolysis and gasification. Gasification converts lignin waste into low molecular weight gases that can be used for various applications. Gasification is primarily a thermochemical conversion process where thermal heat breaks down the solid materials into product gases such as carbon monoxide (CO), hydrogen (H₂), methane (CH₄), carbon dioxide (CO₂), etc. The mixture of CO and H₂ is also known as syngas, a common valuable product for the production of alternative fuels. Gasification of lignin waste can handle a certain variation of lignin properties.

A bubbling fluidized bed (BFB) gasifier uses solid bed materials to heat up the solid fuels in the presence of a limited supply of oxidizing agents (air,

oxygen, or steam) to produce light hydrocarbon gases such as CO, H₂, CH₄, CO₂, etc. The BFB gasifiers operate in the temperature range of 700-1100°C (Franco *et al.*, 2003). Good mixing and high heat capacity of the bed material result in a higher heat transfer rate to the biomass particles, thus resulting in higher conversion. The operation of the BFB gasifiers involves multiphase flow, different chemical reactions, and heat transfer. The effect of different parameters and designs of gasifiers can be studied with the help of modelling and simulation in a relatively short time period. Computational fluid dynamics (CFD) is used to model the systems handling the fluid flow. Conventional CFD is a well-accepted technique for single-phase systems while multiphase CFD is used to investigate the systems handling both solids and fluids.

Multiphase CFD models are based on either Eulerian-Eulerian (EE) or Eulerian-Lagrangian (EL) approaches. Both the solid and gas phases are treated as a continuous phases in the EE approach where two phases are differentiated by their volume fraction. This method lacks the discrete nature of solid particles and the transient information of the two-phase interactions (Bin *et al.*, 2009). EL approach preserves the discrete nature of solid particles where the solid phase can exchange the mass, momentum, and energy with the fluid phase, i.e., strong coupling between the phases. The EL approach gives high loading to a computer central processing unit due to the need of tracking individual particles in the system and the requirement of the small-time steps for solving the particle collisions (Ku *et al.*, 2015). The EL approach is computationally expensive and is limited to only 2×10^5 particles (Gidaspow *et al.*, 2004).

Multi-Phase Particle-In-Cell (MP-PIC) modelling incorporates the EL approach that eliminates the need for tracking individual particles. Computational particles for MP-PIC modelling are a group of particles (called parcels) with similar properties such as size, density, residence time, velocity, etc. The parcels are modelled in a discrete frame and the particle interactions are modelled in the Eulerian frame. Barracuda Virtual Reactor (VR) is a commercial software based on the MP-PIC approach, also known as a computational particle fluid dynamics (CPFD) approach. The rapid development of the graphic process unit in computers has made the CPFD simulation capable of simulating the real process in a short time. The major advantage of CPFD is that it can downsize the billions of particles in a large commercial plant to millions of computational particles (Chen *et al.*, 2013). The governing equations for the CPFD approach can be found in the study of Snider *et al.* (Snider *et al.*, 2011).

1.1 Background

Lignin is a major byproduct of biomass-based biorefinery, paper and pulp industries. This led to different researchers focusing on lignin as a low-cost renewable raw material. Biochemical conversion of lignin waste is widely studied over time (Boerjan *et al.*, 2003; Bugg and Rahmanpour, 2015). However, heterogeneity and higher resistance to chemical and physical actions (Ralph *et al.*, 2019; Strassberger *et al.*, 2014) limit the lignin valorization via biochemical conversion pathways. The biochemical conversion technologies still depend upon improvements and innovation in terms of product separation and catalysts (Strassberger *et al.*, 2014). Therefore, in this study, the authors attempt to convert pelletized lignin waste into syngas via gasification, a thermochemical conversion technology. Different technological hurdles must be overcome to make lignin processing feasible, and the authors believe this study gives a step forward toward lignin waste valorization. The present study aims to gasify lignin pellets obtained from a biorefinery in a pilot scale bubbling fluidized bed gasifier located at the University of South-Eastern Norway (USN).

Literature study in the field of lignin gasification gave a limited number of recent studies, suggesting that lignin gasification is an emerging topic. Liakakou *et al.* (Liakakou *et al.*, 2019) have studied the gasification of lignin rich residues in an updraft, bubbling fluidized bed, and MILENA indirect gasifier. The study shows the feasibility of gasification for lignin extracted from a steam explosion (lignin A) and enzymatic hydrolysis (lignin B). Fluidized bed gasification gave the lowest tar content compared to the other two technologies. Different combinations of lignin and gasification technologies were able to give the H₂/CO ratio of 0.6 to 1.0 and the tar yield was between 18-108 g/Nm³. The gasification behaviour of the three technologies was quite different and was able to give the overall gasification behaviour of lignin residue (Liakakou *et al.*, 2019).

Liakakou *et al.* (Liakakou *et al.*, 2021). performed gasification of lignin rich residue obtained after a steam explosion and enzymatic hydrolysis of wheat straw in the MILENA gasifier. The authors compared the results with the gasification of beechwood chips from the same gasifier. The average volume percentage of the major product gas is presented Tab 1.

Table 1: Product gas composition from Liakakou *et al.*

	CO	H ₂	CO ₂	CH ₄	N ₂
Beech wood	28.8	32.1	25.2	8.8	1.2
Lignin	19.8	35.5	24.4	11.4	1.4

The study showed reasonable product gas composition from the gasification of lignin waste

(Liakakou *et al.*, 2021). The basic background on the discussed topic is presented in the beginning. The relevant literature, theory, research gap, and the nobility of this study are presented in Chapter 1. The detailed methods and the properties of the materials are presented in Chapter 2. The results and discussions of this study are described in detail in Chapter 3, and the conclusion is drawn at last.

2. Methodology

2.1. Experimental setup

Experiments were performed at the pilot scale BFB gasifier located at USN Porsgrunn. Fig. 1 shows the isometric view of the BFB gasifier, and the gasifier is designed to operate at atmospheric pressure.

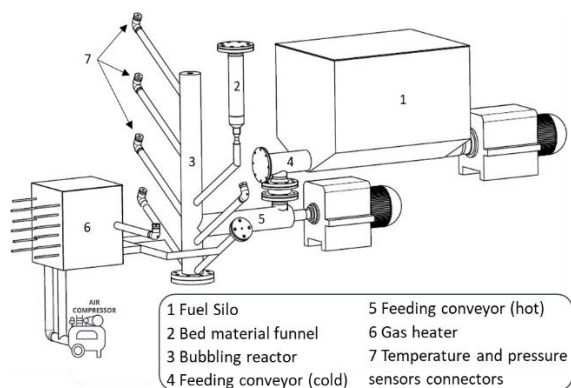


Figure 1: BFB gasifier with auxiliary connections at USN.

The setup consists of a reactor (3), biomass storage silo (1), biomass feeding screws (4 and 5), bed material funnel (2) and a chimney with flare. The setup is facilitated by three electrical heaters with 3kW each, one is the gas heater (6) used to heat the

gasifying agent, and the other two are used to heat up the reactor. The reactor has an inner diameter and height of 0.1 m and 1 m, respectively. An air preheater heats the gasifying agent (compressed air) before entering the reactor to a temperature of around 450°C. Fuel is stored in a sealed silo and is conveyed to the reactor using two screw conveyors. Biomass is fed at a height of 250 mm from the distributor. The gasifier has different pressure and temperature measurement sensors at different positions. The facility also has sensors for the detection of H₂, CO and N₂ to identify any gas leakage. Silica sand in the range of 850 – 1000 μm and density of 2650 kg/m³ was used as bed materials. The initial bed height was 0.22 m. Characterization of the lignin pellets was performed at the Eurofins testing facility to know the elemental composition. The proximate and ultimate analyses are presented in Tab. 2.

Table 2: Characterization of lignin pellets.

Proximate analysis (wt.%, wet basis)	
Fixed carbon	25.68
Volatiles	65.8
Moisture	8.2
Ash	0.32
Ultimate analysis (wt.%, dry basis)	
Ash	0.32
C	54.8
H	6.3
N	0.78
S	0.11
Cl	0.01
O (by difference)	37.68
LHV (MJ/kg, dry)	23.51

Table 3: Reaction kinetics for the CPFDF model.

Reactions/reactions name	Reaction rate kinetics (mol m ⁻³ S ⁻¹)	Enthalpy (kJ/mol)
2C + O ₂ ↔ 2CO		
Char partial oxidation (Snider <i>et al.</i> , 2011)	$4.34 \times 10^7 m_s \text{Texp}\left(\frac{-13590}{T}\right)[\text{O}_2]$	+221.0
C + O ₂ ↔ CO ₂		
Char oxidation (Sreejith <i>et al.</i> , 2015)	$5.7 \times 10^{12} m_s \text{exp}\left(\frac{-4595}{T}\right)[\text{O}_2]^{0.78}$	-394.0
C + CO ₂ ↔ 2CO		
CO ₂ gasification (Thapa <i>et al.</i> , 2014)	$1.12 \times 10^8 m_s P^{0.31} \theta_r \text{exp}\left(\frac{-29518}{T}\right)[\text{CO}_2]$	+172.0
H ₂ + 0.5O ₂ ↔ H ₂ O		
H ₂ oxidation (Bates <i>et al.</i> , 2017)	$5.69 \times 10^{11} \text{exp}\left(\frac{-17610}{T}\right)[\text{H}_2][\text{O}_2]^{0.5}$	-241.9
CO + 0.5O ₂ ↔ CO ₂		
CO oxidation (Xie <i>et al.</i> , 2013)	$5.62 \times 10^{12} \text{exp}\left(\frac{-16000}{T}\right)[\text{CO}][\text{O}_2]^{0.5}$	-283.0
CH ₄ + 1.5O ₂ ↔ CO + 2H ₂ O		
CH ₄ oxidation (Bates <i>et al.</i> , 2017)	$5.0118 \times 10^{11} \text{exp}\left(\frac{-24357}{T}\right)[\text{CH}_4]^{0.7}[\text{O}_2]^{0.8}$	-519.4
CO + H ₂ O ↔ CO ₂ + H ₂		
WGS (Xie <i>et al.</i> , 2013)	$7.68 \times 10^{10} \text{Texp}\left(\frac{-36640}{T}\right)[\text{CO}]^{0.5}[\text{H}_2\text{O}]$	-41.0
CH ₄ + H ₂ O ↔ CO + 3H ₂		
Methane reforming (Solli <i>et al.</i> , 2018)	$3 \times 10^5 \text{exp}\left(\frac{-15042}{T}\right)[\text{CH}_4][\text{H}_2\text{O}]$	+201.9

Lignin pellets were gasified in a bubbling fluidized bed reactor with air as a fluidizing agent. Experiments were performed at different airflow rates (thus giving a different equivalence ratio) and the product gas was analyzed in a gas chromatograph.

2.2 Computational model

A simulation model was developed using Barracuda VR 21.1.1. WenYu-Ergun drag model was used with 40% momentum retention after a particle-particle collision. The reactor was modelled as an open cylinder with a diameter of 0.1 m and a height of 1 m. The developed geometry was divided into 10000 cells with the help of a built-in mesh generator. Fig. 2 shows the boundary conditions, computational cells, and initial bed materials of the developed model.

The formation of the tar and the higher hydrocarbons were neglected during this study. Arrhenius reaction rate model was used to model the chemistry in the reactor. The reaction kinetics is presented in Tab. 3. Major reactions were considered for the model, and their reaction rate kinetics were taken from the literature.

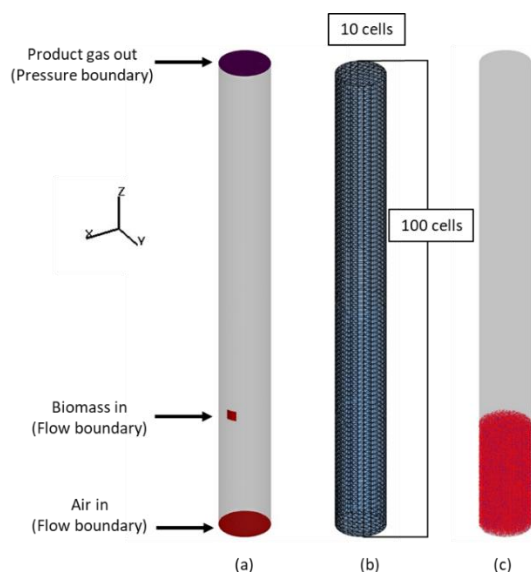


Figure 2. (a) boundary conditions, (b) computational cells and (c) initial bed materials.

3. Results and discussions

The reactor temperature and pressure for a window of 30 minutes during the experiments are presented in Fig. 3.

The results show a certain variation in the bed temperature during the experiments. Biomass was fed via a screw conveyor which gives some degree of variation from the mean feeding rate. Reactor temperature decreases with higher biomass feed rate and increases with lower biomass feed rate. This is because biomass gasification is an endothermic

process, which takes heat from the heated bed materials.

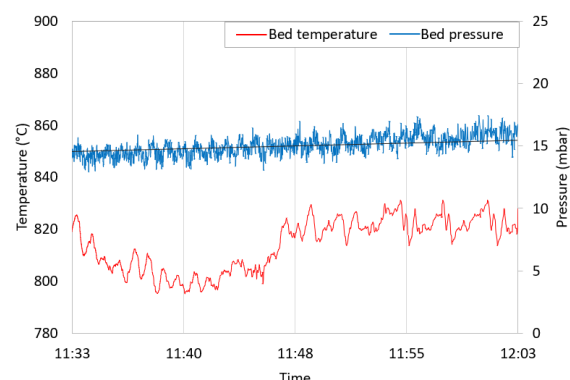


Figure 3: Temperature and pressure variation during the experiment.

A higher biomass feed rate consumes larger amounts of reactor heat, thus lowering the reactor temperature. Lower biomass feeding could lead to a higher reaction rate which in turn increases the reactor temperature. Therefore, variation in reactor temperature gave a certain level of variation in the product gas composition for different samples. Average gas composition was taken for further analysis which minimizes a certain level of measurement uncertainties during the experimental process. The reactor's bed pressure increased slightly with time due to the accumulation of char and ash during the gasification process. As the ash composition was relatively low for the lignin pellets, char gasification is an important aspect of these types of reactors. Char gasification is relatively slow compared to the devolatilization and homogeneous phase reactions (Bermudez and Fidalgo, 2016). Therefore, an alternative configuration such as a dual fluidized bed could be the better alternative to fully utilize the char generated during the gasification process. Char is combusted in a combustor reactor to heat the bed materials which are recirculated back into the bubbling fluidized bed reactor to optimize the biomass conversion.

A simulation model was developed using Barracuda V 21.1.1 (Software[©], 2022) and the simulation results were compared with the experimental results. The average gas compositions for the experimental study and simulation study are presented in Fig. 4. Average molar composition is taken from 40 to 80 seconds for the simulation. The model predicts the fractions of product gases comparatively well and there is a good agreement between the experimental and simulation results regarding the major gas species such as carbon monoxide (CO), carbon dioxide (CO₂), hydrogen (H₂) and nitrogen (N₂). The difference observed between the simulation results and the experimental results could be due to the limited number of reactions considered for the

simulation model, a significant number of reactions occurs during an experimental process. However, the difference is not significant and therefore the developed model was used to study the lignin pellets gasification in depth.

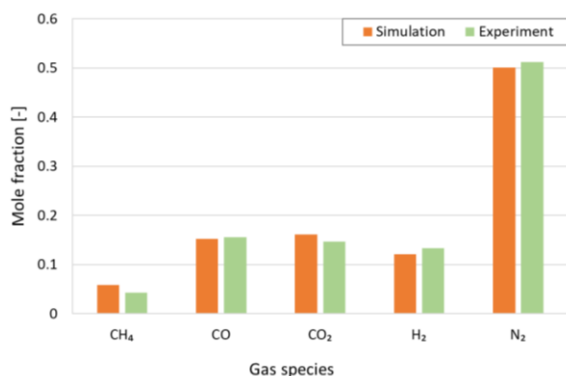


Figure 4. Comparison of average gas species at ER 0.165.

A number of chemical reactions occur during the biomass gasification process. However, only a certain number of major chemical reactions were included in the simulation model for reasonable accuracy and simulation time. The average oxygen concentration during the simulations was zero whereas around 1% of oxygen was present during the experimental analysis. This shows some degree of air contamination during the sampling process. The CFPD model shows a comprehensive result regarding the molar concentration and reactor hydrodynamics during a gasification process. The product gas compositions were monitored during the simulations with respect to time at the reactor outer boundary. Fig. 5 shows the composition of the product gases over simulation time. The reactor reaches steady state conditions after around 10 seconds of simulation time. The CO₂ production started abruptly after around 3 seconds, which counterbalances the O₂ inside the reactor. The high fraction of carbon dioxide at the start represents the combustion process due to the presence of an excess amount of oxygen.

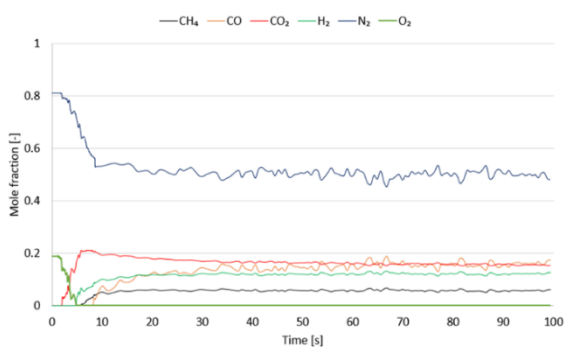


Figure 5. Mole fraction of product gases with respect to time.

The start of the production of CO, H₂ and CH₄ after around 10 seconds of simulation time represents the start of the gasification process. The process gradually shifted from combustion to gasification process. The average molar composition fluctuates around their mean value at steady state conditions. A certain level of variation in the composition illustrates different physical and chemical transformations inside the reactor.

The product gas composition during the simulations was monitored in the reactor. Fig. 6 shows the mole fraction for methane, carbon monoxide and hydrogen at the center plane of the reactor.

Non-distinguishable gas variation inside the bed represents that the heterogeneous reactions are less significant as compared to the homogeneous phase reactions. This suggests that the heterogeneous reactions in the gasification process are slow and are often considered as a rate-limiting process compared to the homogeneous phase reactions. Certain abrupt changes in gas composition can be seen in the biomass feeding regions. This is the region where biomass first enters the reactor, thus altering the flow behaviour. A number of phenomena occur simultaneously giving distinct chemical variations in this region.

Additionally, devolatilization and char mixing with bed materials give different dynamics and properties variation in this region. A certain level of variation in gas composition can be seen in the freeboard regions. The carbon monoxide concentration increased slightly along with the reactor height that indicating the dominance of methane reforming reaction and backward water gas shift reaction.

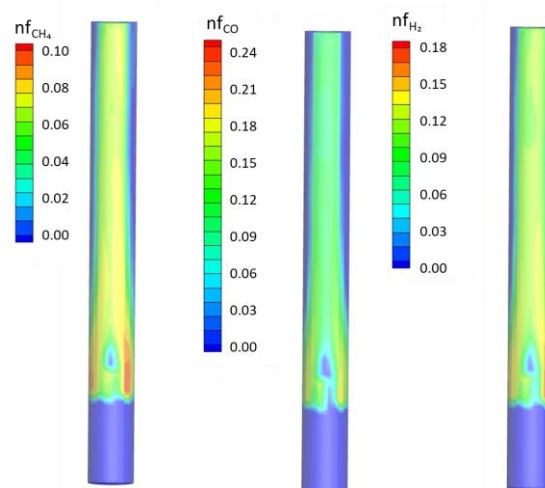


Figure 6. Product gas composition (mole fraction) along xz plane (at y=0.05m).

Gasification of lignin is an emerging aspect of lignin valorization. The literature study gave only certain studies on this topic. It is important to compare the quality of the product from this study to the results

published in the literature. The mole percentage of major gas species excluding N₂, C₂H₂, C₂H₄, C₂H₆ and other minor gas components is compared in Tab. 4.

Table 4: Comparison of molar percentage.

	CH ₄	CO	CO ₂	H ₂
Current study	9.05	32.51	30.59	27.85
Liakakou et al	12.51	21.73	26.78	38.97

As seen from the table, there is considerable variation between the two results. Liakakou et al have used the MILENA gasifier, a dual fluidized bed gasifier operated with steam as a fluidizing agent. However, the air was used as the gasifying agent in the current study which gives lower hydrogen production compared to the MILENA gasifier. Further, variation in operating conditions, bed materials, equivalence ratio, etc would normally give the variation in the product composition. Therefore, further experiments in different gasifiers at different conditions would give a broader perspective of the generated product gas applications.

4. Summary and conclusions

This study gave a comprehensive result for the air gasification of lignin pellets. Experiments were performed in a pilot scale BFB gasification reactor. A simulation model based on MP-PIC modelling approach was developed to study the reaction chemistry and the bed hydrodynamics inside the reactor. The average product gas obtained from the model was validated against the experimental results. The product gas varied with a variation of equivalence ratio in the gasifier. The simulation study showed a higher fraction of CO₂ at the start, suggesting combustion has taken place. The conversion shifted towards gasification at steady-state conditions. The average gas compositions from the experimental study were 0.04 for CH₄, 0.16 for CO, 0.15 for CO₂, 0.13 for H₂ and 0.51 for N₂. The lower heating value and the carbon conversion efficiency were 4.95 MJ/Nm³ and 40.3% respectively. Different sets of experiments are needed to generalize the product gas quality and quantity from the gasification of lignin waste. The developed CPFD model can be used to optimize the model in terms of gasifier performance, fuel flexibility, and reduced emissions. The model can be used to scale up the reactor to an industrial scale reactor to investigate the behaviour at a commercial scale.

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