Model Validation of Biomass-Coal Blends Co-Pyrolysis to Produce Hybrid Coal

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Abstract. Co-pyrolysis of coal and biomass blend to produce hybrid coal has recently been experimentally studied by some previous researchers. For similar generated energy, a newly developed hybrid coal is claimed to be more environmentally friendly compared to the coal only due to the release of neutral CO2. To acquire a better understanding of co-pyrolysis of coal and biomass blend, an experiment had been carried out in a tubular furnace reactor. For this purpose, the blends of constant mass composition of 20 wt% sawdust and 80 wt% low-rank coal were used throughout the study. It was found from the experiment that approximately 42.1% carbon, and 1.6% of ash were produced from the co-pyrolysis blend. Then, a steady state simulation of co-pyrolysis was developed using Aspen Plus v8.8 to predict the hybrid coal carbon content and required heat to perform the co-pyrolysis. The model simulation showed that hybrid coal yielded 44.0% carbon, which was at 4.5% deviation from the experimental study. The model had also been successfully used to estimate heat required to produce hybrid coal. It predicted that the equivalent heat of 336.2 kW was required to produce hybrid coal from 1,000 kg/h blend feed. The heat generated by the modeling of sawdust biomass combustion for fuel purposes was also estimated to supply heat for endothermic co-pyrolysis. It was found that 1,000 kg/h sawdust was predicted to be equivalent to 371.4 kW. This suggests that for scaling up purpose, ratio of sawdust fuel to blend feed of 1:1.1 is sufficient for this process.

Keywords: co-pyrolysis, hybrid coal, low-rank coal, sawdust, Aspen Plus

1. Introduction

Indonesia has abundant coal resources and reserves, which are 166 billion tons in resources and reserves of 37 billion tons or around 22% of its resources (ESDM RI, 2018). However, around 70% of them are low-rank coal with a calorific value less than 21.35 kJ/g and moisture content (up to 30%) (DEN, 2016). The low-rank coal is not efficient for direct combustion purposes and not environmentally friendly, as the coal combustion produces a high amount of carbon dioxide (CO2). The CO2 released by coal processing is a matter of great concern because of its contribution to the global warming. Carbon footprint is an indicator used for quantifying how much CO2 released during the process. Coal has a positive carbon footprint, especially when it was utilized by combustion. Meanwhile, biomass is known as a fuel that is environmentally friendly because it has a zero-carbon footprint. For some biomass, the amount of carbon dioxide released during thermal processing will be equal with the amount of CO2 removed from atmosphere by photosynthesis throughout the life of the biomass so the carbon footprint for biomass can be assumed zero (Sami et al., 2001). One alternative of those matters is by using both coal and biomass, blended and co-pyrolyzed. Co-pyrolysis is basically a process of decomposition of thermally organic matter without or minimum amount of oxygen with the raw material in the form of two or more fuels, resulting a solid fuel known as hybrid coal (Park et al., 2010; Lee et al., 2013; Jeong et al., 2015). Hybrid coal has some advantages: (a) calorific value of coal expected to be increased; (b) moisture content would be decreased; (c) hydrophobicity of biomass

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increased so that decaying is inhibited; (d) reducing CO₂, NOₓ, and SOₓ pollutants compared to conventional coal combustion; (e) increasing the economic value of utilizing biomass waste into more valuable products (Sami et al., 2001; Collot et al., 1999; Guo et al., 2015).

Therefore, research related to the co-pyrolysis of low-rank coal with biomass waste needs to be carried out. Recent modeling and simulation of pyrolysis of coal and biomass blend were conducted using raw material beech wood in temperature 450°C (Idriss et al., 2018). Municipal green waste pyrolysis simulation also had been carried out at temperature 550°C (Kabir et al., 2015). This paper provides an experiment of a hybrid coal production in laboratory scale and its simulation model. The steady state simulation of co-pyrolysis was developed using Aspen Plus v8.8 to predict the hybrid coal carbon content and required heat to perform the co-pyrolysis. Modeling and simulation of co-pyrolysis will provide another basis for its understanding and analysis to help in explaining the phenomena occurring in the process. It is also served as a basis for further scaling up and development of co-pyrolysis plant.

2. Experimental Method

In the previous study (Rizkiana et al., 2018), co-pyrolysis of coal-biomass blends experiment was conducted using a tubular furnace with 5 cm in diameters and 50 cm in height. Nitrogen gas from the tank flowed into the furnace which rate was controlled by the rotameter. Indonesian sub-bituminous coal and mahogany sawdust obtained from a furniture factory in Ciamis, Indonesia were used as hybrid coal raw materials. Both of feeds were reduced to smaller particles, 30-100 mesh, then mixed with the composition of coal and biomass 80% and 20% in mass fraction. Rounded shaped granules were made from the mixture with 1 cm in diameters. Dried granules were packed and later placed inside the furnace to do the co-pyrolysis stage. In the co-pyrolysis stage, the granules were heated in temperature 300°C, and nitrogen gas was flowed to maintain inert condition for an hour. Hybrid coal as a product then analyzed by proximate and ultimate analysis (Rizkiana et al., 2018; Sasongko et al., 2017). Table 1 shows the proximate and ultimate analysis of the lignite coal and mahogany sawdust (Rizkiana et al., 2018; Sasongko et al., 2017).

Table 1. Proximate and ultimate analysis data of biomass, coal, and feed (Rizkiana et al., 2018; Sasongko et al., 2017).

| Proximate (%, adb) | Source | MC | VM | FC | Ash |
|-------------------|--------|----|----|----|-----|
| Coal              | 18.02  | 42.2 | 37.71 | 2.07 |
| Sawdust           | 9.9    | 70.23 | 18.77 | 1.10 |
| Feed              | 16.40  | 47.81 | 33.92 | 1.88 |

| Ultimate (%, adb) | Source | C   | H   | O   | N   | S   | Ash |
|-------------------|--------|-----|-----|-----|-----|-----|-----|
| Coal              | 54.81  | 5.46 | 36.78 | 0.71 | 0.08 | 2.07 |
| Sawdust           | 45.88  | 6.03 | 46.77 | 0.19 | 0.03 | 1.10 |
| Feed              | 53.02  | 5.57 | 38.78 | 0.61 | 0.07 | 1.88 |

MC: moisture content
VM: volatile matter
FC: fixed carbon
3. Modelling and Simulation

3.1 Simulation Model Development

The simulation model was carried out using an equilibrium model approach in Aspen Plus® v.8.8 to present the pyrolysis process. In this simulation, coal and sawdust components were defined as unconventional component based on their ultimate analysis, including C, H, O, N, S, and ash elements, also its proximate analysis as shown in Table 1. The feed defined as a wet mixture of biomass and coal, which its proximate and ultimate was approximated by using 20% composition of biomass and 80% of coal. The use of an equilibrium approach used to estimate the yield and composition of pyrolysis products based on reactor conditions. Property methods IDEAL was used as a base property method for the whole system, since the simulation involved conventional components such as H₂O, N₂, and O₂ at low pressure.

The simulation divided into three main processes. The first process was feed drying in RSTOIC reactor (COALDRYN) at temperature 108°C and in atmospheric pressure condition. The dry feed then separated from the moisture in FLASH separator (DRYFLAS2). Since the input feed in Aspen Plus is unconventional component, the dry feed needed to be converted into known species by Aspen Plus. Dry feed was degraded to its component in a RYIELD (DECOMPL) reactor to C, H₂, N₂, O₂, S₂. After unconventional components converted, it was mixed with nitrogen from the outside, which nitrogen acts as a carrier gas in co-pyrolysis that occurred in RGIBBS (PYROLYZ). The operation parameter in the pyrolysis process is shown in Table 2. Then, the third process, gas and solid products of pyrolysis (ash and carbon) separated at separator (SPLIT). Models schematic of co-pyrolysis in Aspen Plus showed at Figure 1.

Table 2. PYROLYZ Input Operation Condition.

| Feed flow rate | 1.000 kg/hr |
|----------------|-------------|
| Pyrolysis temperature | 300°C       |
| Pyrolysis pressure | Atmospheric |

3.2 Model Assumptions

There are some assumptions that were used at this modeling:
- a) all the processes were at a steady-state condition in atmospheric pressure;
- b) feed (coal and biomass blends) decomposition happened spontaneously;
- c) pyrolysis products consist of C and ash as solid products and CO, CO₂, H₂, H₂O, CH₄, NO, NO₂, S, SO₂, H₂S, SO₃ as gas products and tar formation was ignored. The feed got decomposed as shown in Equation (1)

\[ \text{Feed} \rightarrow \text{Char (carbon+ash)} + \text{Liquid (Tar)} + \text{Gas} \quad (1) \]

Figure 1. Aspen Plus simulation process scheme.
4. Results and Discussion

4.1 Simulation Results and Model Validation

Hybrid coal production was carried out in a series of trials by varying the composition of biomass in the mix respectively 20, 30, and 40% (Rizkiana et al., 2018; Sasonko et al., 2017). In this simulation study, variation with 20% biomass composition in the feed was investigated. The simulation model basic result data in mass balance form were given in Table 3. Meanwhile, the comparison between experimental results with simulation model presented in Table 4.

Simulation of co-pyrolysis resulting in hybrid coal with carbon yield 440 kg or 44.00% of its original feeds. Meanwhile, from the experiment itself (Rizkiana et al., 2018), the carbon component yield at the end of pyrolysis is 42.10%. For the ash component, in the simulation, ash found about 1.59% of its original feed. Meanwhile, the experiment showed that the ash is about 1.68%. So, from the simulation itself, from 1,000 kg/h fresh feed, hybrid coal produced is 455.6 kg/h with the assumption that hybrid coal only consists of carbon and ash.

If we compare the simulation result to the experiment result, carbon content from the simulation result only had a deviation of 4.5% from the experimental result. Meanwhile, the ash content of simulation was also not differed too much from its experimental result, only 5.4%. The deviation between experimental and simulation data might be happened due to the simulation data considered that the process was almost ideal. Meanwhile, in the experiment, there is no such thing as an ideal condition. Despite having small differences, the model can be used to simulate the experiment for predicting the amount of carbon yield on hybrid coal product.

Table 3. Mass balance of pyrolysis simulation in kg/h.

| Comp | Feed | Gas carrier | Gas product | Solid product |
|------|------|-------------|-------------|--------------|
| H₂O  | 351.94 |             |             |              |
| N₂   | 1,000 | 1,005.16    |             |              |
| O₂   | 0     |             |             |              |
| NO₂  | 0     |             |             |              |
| NO   | 0     |             |             |              |
| S    | 0     |             |             |              |
| H₂S  | 0.72  |             |             |              |
| CO₂  | 31.31 |             |             |              |
| CO   | 0.11  |             |             |              |
| C    | 439.71|             |             |              |
| H₂   | 8.69  |             |             |              |
| SO₃  | 0     |             |             |              |
| SO₂  | 0     |             |             |              |
| FEED | 1,000 |             |             |              |
| ASH  | 15.89 |             |             |              |

Table 4. Comparison between component yield of hybrid coal experimental result and model prediction.

| Component | Experiment | Simulation Model | Deviation |
|-----------|------------|------------------|-----------|
| Carbon    | 42.10%     | 44.00%           | 4.5%      |
| Ash       | 1.68%      | 1.59%            | 5.4%      |
4.2 Heat Required for Pyrolysis

To know how much heat required at the unit where the pyrolysis, a simulation was done with following operating condition: feed drying at 108°C (Wulandari et al., 2019), decomposition at 200°C, and pyrolysis took action at 300°C, whereas the pressure is atmospheric. Mass balance of input and output of the simulation showed in Table 2 that concludes from 1,000 kg/h mixture of coal and biomass with a ratio of 0.8/0.2.

From the simulation, the heat needed to do pyrolysis is 336.2 kW per 1,000 kg/h feed or 336.2 W/kg feed. If the co-pyrolysis of the feed was conducted in 1 hour, then the heat required as equivalent to 1210.32 kJ/kg. This much of heat were initially supplied by an electrical source. It is still possible to do since the experiment was done on a laboratory scale. But when do scaling up, using this much amount of heat by electrical source can be a waste of money. Instead, heat can be supplied by another form of energy, such as from biomass combustion. So, a simulation was done to estimate how much biomass needed. Scheme of simulation still the same as above, but there were two units, the combustion unit, and pyrolysis unit. The heat produced in the combustion unit was utilized for the pyrolysis unit.

In an ideal system, all biomass combustion heat will be absorbed by coal-biomass blends until it can form hybrid coal, without any heat being released into the environment or lost with the flue gas. So, the amount of combusted biomass needed will be as low as possible. Biomass heat of combustion was calculated using Dulong equation, as shown in Equation (2) below (Mason & Gandhi, 1980).

\[ Q \ [\text{Btu} / \text{lb}] = 145.44C + 620.28H + 40.5S - 77.54 \]  \hspace{1cm} (2)

From this equation, it was found that the combustion heat of biomass was 6,787.74 Btu/lb or equivalent to 15,788 kJ/kg. Whereas the heat needed for pyrolysis is defined as the difference between HHV products (including gas products) with HHV reactants or raw fuels (Kodera & Mamoru, 2016). The energy balance scheme for pyrolysis is presented in Figure 2 below.

**Figure 2.** Heat balance scheme for pyrolysis process (Kodera & Mamoru, 2016).

Pyrolysis heat is used for sensible heat and latent heat of the product, also heat loss to the environment (Kodera & Mamoru, 2016). In ideal processing, heat loss to the environment is not considered, so the heat of pyrolysis will only be consumed as sensible heat and latent heat. In hybrid coal, sensible heat is heat used to increase the temperature of the feed and the product to reach the pyrolysis operating temperature. While latent heat is used to evaporate water components (moisture content) in the feed.

Various studies have been conducted to calculate the heat of pyrolysis. The heat needed for coal to undergo pyrolysis process in a fixed bed type reactor is 418.6 kJ/kg, while sawdust biomass is 434
kJ/kg with pyrolysis temperature of 300-500°C under atmospheric pressure conditions (Genetti, 1999; Wan et al., 2015; Velden et al., 2010). Taking the composition of 80% coal and 20% biomass in the mixture, the approximated heat required for co-pyrolysis is 421.7 kJ/kg. If all heat produced from biomass combustion is used to make hybrid coal, then 1 kg of complete combustion of sawdust biomass can produce 37 kg of hybrid coal under operating conditions pyrolysis temperature 300-500°C at atmospheric pressure, or at fuel: feed ratio of 1:37.

Back into the simulation, the amount of biomass combusted was the same as coal-biomass blends in a co-pyrolysis unit, which is 1,000 kg/h biomass. This simulation considered as an adiabatic system, where there is no heat supplied in the process. All heat from combustion of biomass was transferred to pyrolysis zone. Composition of biomass (proximate and ultimate) was shown in Table 1 that is from sawdust. Combustion of biomass will be done in an excess air environment. From the simulation, the heat that can be obtained from combustion is 371.4 kW/1,000 kg/h sawdust.

From the simulation, a ratio of fuel: feed that will be needed for pyrolysis process is approximately in 1:1.1 ratio. There was a big difference in heat calculated with equation and simulation. This was because the simulation process considering heat loss in the process, such as heat that was loss with flue gas or heat that was used for heating the reactor. The simulation can be used to design a scale-up reactor from the previous experiment. The process in the simulation was also a continuous process. Therefore, in scaling up an experiment, the reactor also should operate in a continuous operation. Type reactor that is suitable for this is a double chamber rotary kiln reactor, where the feed was co-pyrolyzed in the inner chamber of the reactor, and the combustion happened at the outside chamber.

5. Conclusion

In this study, co-pyrolysis the mixture of lignite and sawdust was investigated by simulation and validated using experimental result. Blend of constant mass composition of 20% sawdust and 80% low-rank coal were used throughout the simulation and experiment. Three main stage of co-pyrolysis process simulation was modeled using Aspen Plus V.8.8. The feed, model assumption and Aspen Plus simulation blocks were discussed. To simplify the simulation, the model ignored the possibility of gas and liquid (tar) formation in the hybrid coal product, only carbon and ash component were considered. It was found that from the simulation, co-pyrolysis of lignite and sawdust blends with mass composition of 80% and 20% resulting product which further called as hybrid coal with carbon mass yield of 44%. Compared with experimental result of co-pyrolysis, the carbon content from the hybrid coal is 42.1%. The simulation model can be used to approximate the experiment, because the deviation of simulation and experiment was only 4.5%. The heat of co-pyrolysis required was also discussed. From the simulation, heat needed to do co-pyrolysis is 336.2 kW per 1,000 kg feed or 336.2 W/kg feed. For the scaling up of the co-pyrolysis process, the energy required for co-pyrolysis will be supplied from the biomass fuel combustion energy instead from electrical source. From the simulation, heat that can be obtained from combustion is 371.4 kW/1,000 kg sawdust. With this condition, the ratio of fuel with the feed will be 1:1.1. This simplified model of co-pyrolysis lignite and sawdust using Aspen Plus will provide some information for further development of co-pyrolysis process to become industrial scale.

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