Effects of coal and biomass types towards the quality of hybrid coal produced via co-torrefaction

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Abstract. Coal is an abundant source of energy in Indonesia, but the reserves are dominated by low-rank coal, which is less favorable to be utilized. Indonesia also has various biomass that has potential to be used as an alternative energy source. Combining low-rank coal and biomass in a co-torrefaction process resulted in hybrid coal: an upgraded solid fuel that has higher calorific value than its original coal. The aims of this research are to analyze the effect of different biomass and coal blends on hybrid coal yield, calorific value, energy yield, and non-neutral CO\textsubscript{2} emission reduction. This study utilized two types of coal that are X and Y, and three different types of biomass: (a) sugarcane bagasse, (b) rubberwood, and (c) empty palm fruit bunch that are being most widely found in Indonesia. The blends consist of 30\%wt biomass were co-torrefied in a vertical tubular furnace reactor for 60 minutes with temperature 300°C in an inert environment and ambient pressure. Solid yields of hybrid coal founded in the range of 57.0 to 63.8\% which different types of biomass gave significant effect. Calorific value was increased 37.6-44.1\% to 5681-6288 kcal/kg from its original coal. The energy yield ranges from 77.0 to 89.0\%. The product reduced non-neutral CO\textsubscript{2} emission within the range of 18.1-22.2\%.

1. Introduction
Indonesia is one of the largest coal-producing countries in the world. Based on the Performance Report of the Directorate General of Mineral and Coal, the Indonesian Ministry of Energy and Mineral Resources, coal reserves and resources until September 2018 are estimated to be 37 and 166 billion tons, respectively [1]. However, Indonesia’s coal reserves and resources are dominated by low-rank coal by around 70\%. The common problems of low-rank coal which less favorable for direct combustion purposes are its high moisture content resulting in low calorific values, low efficiency, and high transportation costs [2]. The low-rank coal also not environmentally friendly because it emits a huge amount of CO\textsubscript{2} gas during the process, which considered the main contributor to global warming.

Indonesia also has an abundant resource of biomass waste but has not been utilized properly. Biomass comes from various sources of waste, such as from forestry, agriculture, livestock, industry, and settlements and the aquatic environment [3]. Biomass is a potential and new renewable energy source because it is more environmentally friendly, considering that CO\textsubscript{2} emissions from biomass are neutral and low in sulfur content [4]. However, biomass has a disadvantage at its low heating values due to its high water and oxygen content.
A method that can be used to improve the quality of low-rank coal is by co-torrefaction with biomass, producing new solid fuel called hybrid coal. The co-torrefaction process is expected to provide several benefits such as (i) an increase in coal calorific value, (ii) decreased coal-water content, (iii) increased biomass hydrophobicity which prevents decay, (iv) decreased non-neutral CO\textsubscript{2} emission through the utilization of biomass waste for energy generation, (v) increased utilization of low-rank coal and biomass waste [4][5][6]. Several previous studies have been carried out to obtain hybrid coal by mixing and processing coal and various types of biomass, such as Larix leptolepis wood sawdust [4], yellow poplar wood [6], sugarcane juice, and molasses [5]. The research on using various types of coal and native Indonesian biomass waste has not been done much.

This study aims to determine the effect of coal and biomass types towards hybrid coal quality based on solid yield, proximate components, constituent elements, calorific value, energy yield, and non-neutral CO\textsubscript{2} emissions. In this study, the two types of lignite coal used, namely X and Y which are obtained from the Mineral and Coal Technology Research and Development Center (Puslitbang tekMIRA). Biomass waste used are empty palm fruit bunch obtained from Mulawarman University, rubberwood sawdust obtained from one of the furniture industries in Pati and sugarcane bagasse obtained from the Trangkil Sugar Factory, Pati. Torrefaction is done in a vertical tubular furnace reactor with a 1.6 L/minute nitrogen rate, at ambient pressure and temperature of 300°C for 60 minutes. The composition of biomass in the mixture of coal and biomass is 30%-w/w. The results of these experiments can be used in determining which type of coal and biomass used as raw materials that produce the best quality of hybrid coal. It also serves as a reference for a further scaled-up co-torrefaction reactor.

2. Methodology
The two types of coal used named X and Y. Coal was reduced to 28-100 mesh in diameter, while the three types of biomass used, which were sugarcane bagasse (BG), rubberwood sawdust (K), and empty palm fruit bunches (TKS), were ground to 48-100 mesh. Binder made from a starch solution was used to make coal-biomass granules sized 1-1.5 cm in diameter. Coal and biomass type variation used were shown in Table 1.

| Variation | Coal Type | Biomass Type          | Code  |
|-----------|-----------|-----------------------|-------|
| 1         |           | Empty palm fruit bunch | X-TKS |
| 2         | X         | Rubberwood sawdust     | X-K   |
| 3         |           | Sugarcane bagasse      | X-BG  |
| 4         |           | Empty palm fruit bunch | Y-TKS |
| 5         | Y         | Rubberwood sawdust     | Y-K   |
| 6         |           | Sugarcane bagasse      | Y-BG  |

The experiment was carried out using a vertical tubular fixed bed reactor with a diameter of 5 cm and a length of 50 cm which was equipped with a temperature controller to maintain the co-torrefaction temperature. The reactor was connected to a nitrogen tank with a flow rate regulator to measure the nitrogen gas flow rate. The reactor was heated and maintained under conditions of low-temperature co-torrefaction (300°C) and ambient pressure for one hour. Raw samples, which have been weighed and characterized early, were placed in stainless basketball sieves and inserted into the reactor. Updraft nitrogen gas flowed to the furnace so that co-torrefaction done in an inert environment and nitrogen also acts as a carrier gas that prevents volatile matters build up during the process. After co-torrefaction for 1 hour at a temperature of 300°C, the furnace was cooled down into room temperature. Hybrid coal product was removed from the furnace, weighed, and characterized. The characterization of raw materials included proximate analysis, ultimate analysis, analysis of calorific value, and analysis of lignocellulose content and characterization of hybrid coal products included proximate analysis, ultimate analysis, and analysis of calorific value. Proximate analysis including quantifying moisture content, volatile matter, carbon, and ash using ASTM D.3173, D.3175, D.3172 and D.3174 respectively.
Meanwhile, characterization using ultimate analysis measures carbon, hydrogen, nitrogen using ASTM D.5573 as a basis and also oxygen and sulfur using ASTM D.3176 and D.4239. ASTM D.5865 used to analyze the calorific value of each material. All characterizations were carried out at the Mineral and Coal Technology Research and Development Center (tekMIRA), Bandung except for the analysis of lignocellulose content, which is carried out at the Pulp and Paper Center (BBPK), Bandung. Other than that, the actual yield of hybrid coal products obtained from the weight ratio of solid products vs. the blends of coal and biomass inserted into the reactor.

![Figure 1. The scheme of hybrid coal co-torrefaction experimental equipment.](image)

3. Results and Discussions

3.1. Effects of coal and biomass types to solid yields

Solid yield is defined as the mass ratio between the co-torrefied solid product and the raw material. Hybrid coal solid yield for various coal and biomass samples are shown in Figure 2. Based on Figure 2, the hybrid coal solid yield obtained was in the range of 57.04-63.8% with errors between the experimental and theoretical results in the range of -9.85-12.76%. The theoretical solid yield was calculated based on the solid yield data obtained through torrefaction of each raw material, which shown in Figure 3.

Based on Figure 2, the type of coal affects the synergistic effect on hybrid coal solid yield. A positive synergistic effect occurs on all hybrid coal with Y coal raw material, while a negative synergistic effect occurs on all hybrid coal with coal raw material X. The negative synergistic effect on solid yield is likely to occur because of the H and OH radicals released by biomass increase the coal's aromatic ring breakdown, considering the high H content of biomass [7]. Another possibility is the presence of a catalytic effect of inorganic components on biomass ash [8]. The positive synergistic effect on solid yield is likely to occur because the residual thermal decomposition of biomass accumulates on the surface of coal molecules then undergoes polymerization and condensation reactions to produce tar which clogs the pores of coal molecules [9]. This event causes blockage in coal volatile matters path so that the thermal decomposition of coal is inhibited [10]. The results of the analysis of variance (ANOVA) on solid yield data indicate that there is a significant effect on the types of biomass and there is no significant effect on the types of coal. The insignificant interaction between coal and biomass on the hybrid coal solid yield is also identified based on the interaction plot.
Hybrid coal solid yield can be influenced by the content of lignin, hemicellulose, and cellulose in biomass. At a temperature of 300°C, the largest decomposition occurs in the hemicellulose component, while cellulose and lignin components decompose in small amounts [11]. This indicates that the hemicellulose component becomes an important parameter in biomass torrefaction at a temperature of 300°C. The results of the analysis of the biomass lignocellulose component are shown in Table 2. The solid yield from coal and biomass raw material results are shown in Figure 3. Based on Table 2, the empty palm fruit bunch should provide the least solid yield because it has the highest hemicellulose content. However, the experimental results in Figure 2 show that the least solid yield is achieved by the coal X-rubberwood mixture, which is different from the estimation based on Table 2. The experimental results in Figure 3 show that the least solid yield is found on hybrid coal made from empty palm fruit bunch, which is the same as estimation made based on Table 2. This happens because lignocellulose constituents in biomass consist of several complex compounds.

**Table 2.** Biomass lignocellulose composition data

| No | Parameter    | Unit     | Sample |
|----|--------------|----------|--------|
|    |              |          | K      | TKS | BG   |
| 1  | Lignin       | %        | 27.82  | 20.78 | 21.52 |
| 2  | Hemicellulose| %-%w/w   | 23.64  | 31.65 | 31.26 |
| 3  | Cellulose    |          | 48.54  | 47.57 | 47.21 |
In the torrefaction mechanism, the reactions that occur in biomass vary and can produce different substances. Hardwood and softwood biomass with similar hemicellulose compositions under identical torrefaction conditions may give different solid yield [11]. Biomass components, especially hemicellulose, contain xylan in varying amounts. Xylan (4-O-methylglucuronoxylan) is the most reactive component in the torrefaction process, and its decomposition rate is faster than other biomass components so that mass loss in biomass is more influenced by xylan content in biomass than overall hemicellulose content [11]. To validate the loss of mass obtained, an analysis of xylan content needs to be conducted.

3.2. Effects of coal and biomass types towards proximate components

Proximate components describe general characteristics of the distribution of coal, biomass, and hybrid coal components[12]. Proximate components consist of moisture, volatile matter, fixed carbon, and ash. The proximate analysis results of raw materials and hybrid coal products are shown in Figure 4. The moisture contents of hybrid coal were low and in a narrow range, which is 0.6-2.1%-w/w. When biomass undergoes torrefaction, some hydroxyl groups are broken through dehydration [13]. It prevented the formation of hydrogen bonds so that hybrid coal becomes hydrophobic. Moreover, the heat absorbed by the feed made the moisture contents to vaporized at temperature 108°C and continued by the vaporization of volatile matter at temperature more than 200°C [14].

![Figure 4. Proximate analysis results](image)

The volatile matter of hybrid coal varies and is in the range of 39.2-47.1%-w/w, while the fixed carbon ranges from 45.3-47.7%-w/w. The biomass component, mainly hemicellulose, decomposes, and is released as volatile matter during torrefaction [11]. It causes a decrease in volatile matter accompanied by an increase in fixed carbon of hybrid coal. In raw X coal, sugarcane bagasse biomass causes the largest decrease in volatile matter content of hybrid coal, followed by rubberwood and empty palm fruit bunch. In raw Y coal, sugarcane bagasse biomass causes the largest decrease in volatile matter content of hybrid coal, followed by empty palm fruit bunch and rubberwood. This difference is caused by synergistic effect differences between each type of coal and biomass, causing different thermal decomposition processes [8].

Hybrid coals containing raw Y coal have higher volatile matter content than hybrid coals containing raw X coal, which are similar to volatile matter content trends in raw coal. Hybrid coals containing raw Y coal have higher carbon content than hybrid coals containing raw X coal, which are similar to fixed carbon content trends in raw coal. The lignocellulose component has decomposed naturally in coal so that only a small portion of the volatile matter of coal is released during torrefaction. It is proven by
solid yields of raw coal torrefaction are higher than solid yields of raw biomass torrefaction, based on Figure 3.

Hybrid coals containing empty palm fruit bunch have the highest ash content, followed by sugarcane bagasse and rubberwood. Hybrid coals containing raw X coal have higher ash content than hybrid coals containing raw Y coal. They are similar to ash content trends of the raw materials. This happens because the mass of ash in the sample does not change during torrefaction [10].

3.3. Effects of coal and biomass types towards constituent elements

The constituent elements of coal, biomass, and hybrid coal are obtained from the ultimate analysis results shown in Figure 5. An increase in C content followed by a decrease in O and H content of hybrid coal products compared to the raw materials occurs. Decreases in the moisture content and volatile matter content followed by an increase in the fixed carbon content during the torrefaction change the elemental composition of hybrid coal because water and volatile matter contain many O and H, while fixed carbon contains many C [13].

Van Krevelen diagram of raw materials and hybrid coal products in Figure 6 shows a decrease in the ratio of atomic O/C and H/C in hybrid coal products when compared to its constituent coal. It indicates an increase in coal rank after being processed into hybrid coal. Hybrid coals containing empty palm fruit bunch have the smallest O/C and H/C values. It indicates that the types of empty palm fruit bunch's volatile matter are the most easily decomposed, as proven by the data of solid yield of raw empty palm fruit bunch torrefaction that have the lowest value. Raw X coal has a smaller O/C and H/C atomic ratio than raw Y coal so that it can be said that X coal rank is better than Y coal. However, the rank of hybrid coal produced does not follow the trend of its constituent raw coal. Hybrid coals containing raw X coal have a smaller H/C atom ratio than hybrid coals containing raw Y coal, while the reverse result is obtained for O/C atomic ratio value. It shows that X coal is more difficult to be volatilized than Y coal, as proven by the data of solid yield of raw X coal torrefaction X is higher than the solid yield of raw Y coal torrefaction.
3.4. Effects of coal and biomass types towards calorific value

The calorific value of raw materials, hybrid coal products, and several coal standards in the form of gross calorific value (GCV) are shown in Figure 7. It shows increases in the calorific values of hybrid coals in the range of 37.6-44.1% compared to their constituent raw materials. Decreases in the moisture and volatile matter contents followed by an increase in the fixed carbon content during torrefaction cause an increase in calorific value because the carbon content is the main source of heat energy released when burning a fuel [13]. Hybrid coal products have calorific values in the range of 5681-6288 kcal/kg. Hybrid coals containing raw Y coal have higher calorific value than hybrid coals containing raw X coal, which are similar to calorific value trends in raw coal. It indicates that the calorific value of hybrid coal is mainly affected by the calorific value contributed by raw coal. Meanwhile, biomass types do not show a trend towards the calorific values of hybrid coal because the calorific values of the raw biomass used are in a narrow range, which is 3925-3979 kcal/kg.
3.5. Effects of coal and biomass types towards energy yield

Energy yield is a ratio of energy amount of torrefied solid product to the energy amount of its raw materials. Hybrid coal energy yield in Figure 8 varies and ranges from 77.0-89.0%-w/w. The sequence of hybrid coal energy yield is similar to the sequence of hybrid coal solid yield. It indicates that solid yield is a more dominant factor than calorific value, so that the effects of biomass and coal on energy yield are similar to the effects of biomass and coal on solid yield [13].

![Figure 8. Hybrid coal energy yield](image)

3.6. Effects of coal and biomass types towards CO₂ emissions reduction

Non-neutral CO₂ emissions reductions of hybrid coal when they are used as fuel are shown in Figure 9. The reduction values vary and are in a narrow range, which is 18.1-22.2%. These results indicate that hybrid coal is more environmentally friendly than raw coal when used as fuel. The highest reduction value is achieved by hybrid coal from rubberwood mixtures. Hybrid coals containing raw Y coal have higher reduction values than hybrid coals containing raw X coal do. CO₂ emissions reduction of hybrid coal is proportional to the calorific value of hybrid coal and inversely proportional to the composition of non-neutral C in the product. A high calorific value reduces the mass of hybrid coal needed. A low composition of non-neutral C lowers the non-neutral CO₂ emitted.

![Figure 9. Hybrid coal non-neutral CO₂ emissions reduction](image)
4. Conclusion
From this study, it can be concluded that hybrid coal solid yield is in the range of 57.0-63.8%. Coal types do not have a significant effect on solid yield while biomass types have a significant effect on solid yield. In addition, it is found that there are no significant interactions of coal and biomass on solid yield.

From the result of proximate and ultimate analysis, there is an increase in fixed carbon, a decrease in moisture content, a decrease in the volatile matter and a decrease in the atomic ratio O/C and H/C in hybrid coal. It causes an increase in hybrid coal calorific value by 37.6-44.1% to be 5681-6288 kcal/kg. Y hybrid coals have higher calorific value than X hybrid coals do. Meanwhile, the energy yield of hybrid coal was in the range of 77.0-89.0%. Coal and biomass types give varied effects on hybrid coal energy yield with the same sequence as hybrid coal solid yield.

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