Simultaneous sulfur dioxide and mercury removal during low-rank coal combustion by natural zeolite

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ABSTRACT

Sulfur dioxide (SO2) and trace metal such as mercury emission during combustion of low-rank coal cause a significant impact on the environment and health. Flue gas at coal-fired power stations is one of the main sources for the emissions of SO2 and mercury metal. Low-cost and sustainable technologies for the removal of SO2 and mercury from flue gas have become increasingly important nowadays. This paper presents the study of simultaneous removal of SO2 and mercury over natural zeolite as an adsorbent in briquette and pulverized. The research is focused on evaluating adsorbent to coal optimum ratio towards adsorption performance on SO2 and mercury removal. The experiments on the removal of SO2 and mercury were carried out using horizontal electric furnace with different combustion temperature and adsorbent ratio. SO2 in the flue gas as the result of the combustion process which exits from the outlet was analyzed using Gas Combustion and Emission Analyzer (E4400, E-Instrument). Mercury metal residues in the bottom ash were analyzed using NIC Mercury SP Analyzer. An increasing zeolite adsorbent SO2 content in flue gas decreased. The optimum SO2 removal was determined on 4% zeolite adsorbent ratio. It also has been found that 8% natural zeolite ratio to low-rank coal show optimum condition to adsorb mercury for all temperature condition for both briquette and pulverized conditions.

1. Introduction

Coal is a majority source of energy for industry and becomes the preference for many investors because it can lower the operational cost and is highly available in Indonesia. The Indonesian coal deposit of 70% is considered low-rank coal. Coal as an energy alternative is widely used as the electric source in the industries producing cement and fertilizer. Possible future rises in the price of natural gas and the demand for ammonia and its derivatives especially for the fertilizer industry motivate the consideration of coal as a feedstock [1, 2, 3]. Indonesia’s low-rank coal is a source of energy with low economic value because it has high water content, low calorie, and difficult to be exported. Coal is considered low rank due to its high moisture, ash, sulfur, nitrogen, trace metals, and low carbon content, especially exhibiting high volatile and low heating value [4, 5, 6, 7, 8]. The utilization of low-rank coal in technology can be generally found in combustion, liquidation, gasification, and pyrolysis. Most of the power plants utilize low-rank coal with direct combustion technology.

Sulfur dioxide (SO2) emission during coal combustion to the environment generally comes from power plants as the primary energy sector. Power generator using coal and industrial activities are the major source of SO2 emissions [9, 10, 11]. Coal combustion also generates nitrogen oxide (NO) which will produce nitrogen dioxide (NO2) in the atmosphere as another air pollutant, and pollution control was studied [12, 13, 14]. SO2 and NO2 emission produced during coal combustion causes acid rain and leads to problems such as corrosion. Several health problems and diseases such as throat disease, eyes, nose irritation, cough, and headache appear as the result of short exposure to this polluted air due to sulfur and nitrogen oxides content especially for people with asthma [15, 16, 17]. Coal combustion also produces ash contains trace metals that are harmful and has a carcinogenic effect during combustion. Several kinds of research show that zeolite can be used in controlling SO2
and tracing metals emission [18, 19, 20, 21, 22, 23]. The use of zeolite both natural and synthetic as an adsorbent is more advantageous compared to carbon-based adsorbent due to its thermal tolerance and resistance to acid and easily regenerated and low cost.

Power plant using coal as fuel is the most important mercury emission contributor to the air because of its high volatile and corrosion potential by SO2 and NO2 [24, 25, 26]. Sub-micron mercury particles are emitted during coal combustion [9, 10, 11] and there are few research related to trace element especially mercury. The study on mercury classification modelling and its trace element characteristic has been studied by [27, 28, 29, 30, 31].

Some of the heavy metals (trace element) found in coal such as Hg, Pb, As, Ba, Be, B, Cd, Cr, Ta, Se, and Mo emitted are not dependent only on the amount of its content in the coal [27]. The emission may be an advanced combination of (a) the period of bed use and fatigue condition of the metal, (b) combustion temperature, (c) ash content and its composition, (d) fuel density, and (e) chlorine and sulfur content concerning trace metal volatility. This study stressed mercury released and captured during coal combustion in briquette and pulverized condition. During coal combustion, the mercury element in coal is totally vaporized in the vapor phase, since the species in high vapor pressure at typical boiler exit temperature. Although, mercury (Hg) concentration in coal is typically very low, important attention is stressed on its emission since it is highly toxic and harmful to human health. Several previous works have been done to capture SO2 and trace metals during low-rank coal combustion and the effect of other species such as chlorine on its capture mechanism [32, 33, 34, 35, 36, 37, 38]. Although several studies have been conducted to see desulfurization and trace metal capture of low-rank coal, the simultaneous desulfurization, and mercury adsorption during combustion of low-rank coal using zeolite is still very limited. The advantage using of natural zeolite adsorbent is due to its low cost and wide availability.

Zeolite which is a polar adsorbent in general can be distinguished into two, namely natural zeolite and synthetic zeolite where zeolite is generally a three-dimensional structured silica-alumina crystal, formed from tetrahedral alumina and silica with cavities inside containing metal ions, usually alkaline or alkaline soil and water molecules that can move freely. Empirically, the zeolite molecular formula is Mx/n (AlO2)x (SiO2)y. Zeolite structure is so far known to vary. Natural zeolite usually contains cations K+, Na+, Ca2+, or Mg2+ whereas synthetic zeolite usually contains only K+ or Na+ cations. In natural zeolite, the presence of water molecules in pores and free oxides on the surface such as Al2O3, SiO2, CaO, MgO, Na2O, K2O can cover the pores or active sites of the zeolite to decrease the adsorption capacity and catalytic properties of the zeolite [39, 40]. Under normal circumstances, the vacuum in the zeolite crystals is filled with free water molecules that form a spatial around the cation. When the crystal is heated for several hours, usually at a temperature of 250–900 °C, then the zeolite crystals have the function of absorbing gas or liquid. The absorbance of zeolite depends on the amount of vacuum and surface area. Adsorption on the zeolite itself occurs on the surface of the adsorbent pore. Zeolite particles have three types of pores, macropore and micropore (with sizes of >50nm and <2nm, respectively). Between them is mesopore. Macropore is the entrance into the particles to the micropore. Macropore does not contribute to the large surface area of zeolite adsorbents. In contrast, micropore is the cause of the large surface area of zeolite adsorbents. Such micropores are mostly formed during the activation process. It is at this micropore that most adsorption events occur. To analyze the microstructure (microstructure) of adsorbents using the method of scanning electron microscope (SEM) micrometer scale to nanometer, and method Thin Slice (thin incision) millimeter scale to get a visual picture of pore structure and percentage of mineral content in rocks with petrography test.

Bentonite is a term on clay containing montmorillonite, in the world of trade and belongs to the dioctahedral group. Bentonite can be divided into 2 groups based on the content of aluminum silicate hydros, namely activated clay and fuller's earth. Activated clay is clay that lacks the power

of paint, but its paint ability can be increased through certain processing. Meanwhile, fuller's earth is used in filling or cleaning wooden materials from fat. While based on the type, bentonite is divided into two, namely:

1. **Wyoming Type (Na-bentonite – Swelling bentonite)**

Na-bentonite has the power to expand up to eight times when dipped in water and remains dispersed for some time in the water. In a dry state white or cream, in wet and exposed to sunlight will be shiny. Comparison of soda and lime is high, the colloidal suspension has a pH: 8.5–9.8, cannot be activated, the exchange position is occupied by sodium ions (Na+).

2. **Mg. (Ca-bentonite – non-swelling bentonite)**

This type of bentonite is less inflated when dipped in water, and remains dispersed in water, but naturally or after activation has good sucking properties. The comparison of Na and Ca content is low, the colloidal suspension has a pH: 4–7. The ion exchange position is more occupied by calcium and magnesium ions. In dry conditions are rapid slaking, gray, blue, yellow, red, and brown. The use of bentonite in the refining process of cooking oil needs to be activated first [41]. Bentonite clay is very interesting to research because this clay has a layered structure with the ability to expand (swelling)and has cations that can be exchanged [42]. Although bentonite clay is very useful for adsorption, its adsorption capabilities are limited [43]. The use of clay as an adsorbent has several advantages because clay especially bentonite type has an inter-layer structure that can be modified to improve its properties. Besides, the use of clay as adsorbents can be regenerated [44].

Kaolin is a clay mineral with a 1:1 layer structure with a base unit consisting of a Tetrahedral Sheet SiO4 and an octahedral sheet with Al3 as an octahedral. Kaolin is usually located as a pure kaolinite mineral or related mineral e.g., halloysite, nacrite, and dickrit which are joined by other minerals such as smectic, mica, quartz, and feldspar as impactors. Kaolin impregnator materials especially quartz have a composition of SiO2 and are about the same size as kaolin in an aggregated state so that the purification process using sifting is less effective. Kaolin minerals have a multi-layered pseudo-hexagonal structure with a size of 1–10 μm. In general, individuals of kaolin are usually composed of 10–50 layers of alumina-silica with a layer thickness of several tens of nanometers to several micrometers [45].

This research aims to reduce SO2 and trace metal emission especially mercury during coal combustion for both pulverized and briquette form by adding natural zeolite adsorbent. The adsorbent will capture the sulfur and trace metal remain in ash instead of flying into the air. The use of non-carbon adsorbent such as zeolite, bentonite, kaolin, and lime powder is also capable to absorb Hg, Pb, Cd, Na as well as several volatile and non-volatile metal [46]. Natural zeolite has water molecules in the pores and free oxides on the surfaces such as Al2O3, SiO2, CaO, MgO, Na2O, K2O. The free oxides can cover the pores or active sites of the zeolite and can reduce the adsorption capacity.

2. **Experiment procedure**

The experiment used coal from Kaway XVI of West Aceh Sub-District in Aceh Province, Indonesia, and the natural adsorbent (zeolite) was obtained from West Java Province, Indonesia. Coal has low rank if it has high moisture content of 8.83%, sulfur content of 0.38%, and ash of 5.4% ash (db) with calorific value of 5904 cal/gr [47]. The size of coal and zeolite was crushed and screened to 60 mesh using a crusher and ball mill, then was briquetted using a briquette holding machine without binder, while the rest of the sample was left in pulverized condition. The mercury content in the ash and sulfur were analyzed. The adsorbent was then added with various concentration ratios from 4 to 12% of the weight and formed into briquette and pulverized form. The briquette was molded in coal briquette molding equipment with pressure level of 10 ton/cm² (SNI 047, 2006).
3. Result and discussion

3.1. Effect of temperature and adsorbent on SO2 emission

SO2 emissions from coal combustion mainly depend on the sulfur content in the coal. Effect of operating condition shows the profile of SO2 generated during combustion as shown in Figure 2. The oxidation process occurred on surface of coal particle, i.e., a coal which was finely crushed, and consequently exposing more surfaces for a given weight. It reacted more rapidly with oxygen than with a similar sample for both in briquette and pulverized forms. Figure 2 shows SO2 profile during coal combustion at different temperatures without adsorbent for pulverized and briquette cases. The figure shows SO2 emissions in ppm increased with the increasing of temperature, and then appeared to decrease after 4 min and 3 min for briquette and pulverized forms, respectively. The decrease of SO2 emission after 4 and 3 min might be caused by the decrease of sulfur content in coal.

The experiment shows the increase of SO2 which was burned earlier as volatile matter combustion. The SO2 profile after 4 min tended to decrease due to fixed carbon combustion and the sulfur content in fixed carbon was low. SO2 emission for pulverized case was higher than briquette case for all temperatures. Higher SO2 emission for pulverized due to particles was smaller than briquette form. In briquette conditions, it took time to reach the combustion temperature (flash point), so it was not fast and the oxygen diffuse factor into the briquette was slower than the pulverized condition because the surface area was smaller. The small particle gave large surface area and easily made oxygen diffusion. The result of pulverized form gave more complete combustion compared to briquette form. Complete combustion gave more SO2 emission compared to uncomplete combustion. The smaller sorbent particles gave higher SO2 reduction and CaO conversion, even it appeared in the pulverized coal combustion [37]. In pulverized conditions, the diffuse rate of SO2 gas into adsorbsent was easier because the surface area was larger.

The effect of adsorbent on SO2 adsorption during coal combustion at different temperature for briquette and pulvzerized case at 4 min is shown in Figure 3. The figure shows effect adsorption of SO2 by adsorbent decreased SO2 emission as the adsorbent concentration increased. Similar tendency is shown on higher SO2 concentration for pulverized form compared to briquette form. The result also suggests smaller particle will give higher SO2 concentration due to complete combustion. The same phenomena are also shown higher temperature will give a higher concentration of SO2 due to more complete combustion process.
Optimum adsorption condition is shown on 4% adsorbent concentration at 700 °C combustion for pulverized condition.

The result also shows the ability adsorbent on SO₂ adsorption in briquette form was better than pulverized form. Since volatilized and emitted in vapor phase, the increasing temperature will increase the SO₂ concentration. Coal in pulverized form is easily burned due to higher surface area than briquette form, resulting in higher SO₂ concentration. The optimum adsorption capacity was determined at 6% for briquette and pulverized 8% form, respectively.

3.2. Effect of temperature and adsorbent on mercury adsorption

Figure 4 shows the influence of adsorbent ratio toward mercury content in bottom ash at various combustion temperatures. The result shows that zeolite adsorbent had a proportional effect on adsorption capability of mercury. The concentration of mercury remained in bottom ash (sampling boat) as the zeolite adsorbent ratio increased. The result suggests zeolite adsorbent had significant effect on mercury adsorption, and optimum adsorbent ability to adsorb mercury was observed at 8% ratio for both briquette and pulverized forms. After 10% of optimum adsorbent ratio, adsorption capability of adsorbent to adsorb mercury decreased for all temperatures and started to befall beyond 10% ratio.

Effect of temperature had inverse influence over mercury content in bottom ash. The concentration of mercury in bottom ash decreased as the combustion temperature increased due to more volatile property of mercury in higher temperature. This is because the increase of temperature during combustion released mercury in the more volatile form and rate of diffusion volatile mercury to adsorbent was slower than devolatilization rate of mercury from coal. In briquette conditions, some briquettes were not perfectly burned; thus, some mercuries were left in the ash and unburned carbon. In pulverized conditions, the carbon was perfectly burned; thus, some mercuries evaporated due to the volatile form of the mercury. Mercury measurements were performed on ash left behind after burning, as it is considered an adsorbent. Volatile mercury can pollute the air and easily disbursed and is very hard to capture by increasing of combustion temperature. The use of natural zeolite as an adsorbent is expected to control release to the air and control mercury content remain in bottom ash. The effective temperature of zeolite to capture mercury emission during combustion is 600 °C–700 °C. At temperature 800 °C, the effectiveness of zeolite to adsorb mercury decreases for both case briquette and pulverized forms. The optimum ratio of adsorbent in mercury adsorption for briquette was achieved at 8% adsorbent ratio for both 600 °C and 700 °C combustion temperatures. While in pulverized conditions, the optimum condition was obtained at 10% adsorbent at temperatures of 600 °C and 700 °C.

3.3. Adsorption capacity of zeolite

SO₂ and mercury adsorption during coal combustion occurred simultaneously, requiring observation on the adsorption capacity. Adsorption capacity is the ability of one gram adsorbent to adsorb adsorbate by the following equation:
The adsorption capacity of mercury by zeolite for briquette and pulverized cases at different temperature is shown in Figure 5. The result shows adsorption capacity optimum at 6% and 8% for briquette and pulverized forms, respectively. In general, the adsorption capacity or efficiency will increase as the sorbent ratio increases. The results show that adsorption capacity of mercury in briquette form was better than pulverized form. Adsorption efficiency of sorbent showed significant effect of temperature change. The adsorption capacity decreased as the temperature increased. Since mercury is easily evolved with increasing temperature, adsorption capacity of zeolite decreases as the temperature increases. Figure 5 suggests that when combustion temperature was low, some mercuries did not volatilize but stayed in bottom ash, then were captured by the sorbent or deposited on it. As the result, adsorption capacity of zeolite was high at low combustion temperature. When some volatile mercuries existed in the gas phase, some formed particles were captured by sorbents. Another mechanism is that mercury particles were captured by sorbents through nucleation and coagulation.

4. Conclusion

Simultaneous adsorption SO2 and mercury during coal combustion using natural zeolite was observed. SO2 emission during combustion tended to increase during at the earlier combustion process. The adsorption capacity of mercury was influenced by its high volatility and tended to decrease as the temperature increased. The optimum adsorption capacity was determined at 6% and 8% for briquette and pulverized forms, respectively. The adsorption capacity of zeolite to adsorb SO2 and mercury was controlled by diffusion rate of the gases to zeolite particles. The performance of zeolite to adsorb SO2 and mercury tended to decrease at 800 °C because it started to desorb.

Figures 5. Adsorption capacity of mercury by zeolite for (a) briquette and (b) pulverized cases at different temperature.

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Q = \frac{C_{A} - C_{A0}}{M_{Absorbent}}
\]  

(1)

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