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# PROCEEDING



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## Development of Pre-treatment Process for Aceh Low-rank Coal by Simultaneously Capture of Sulfur and Trace Element

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### Abstract

Low-rank coal is a potential fuel for power plant, both for gas and steam turbine systems. Since the low-rank coal contain high fraction of sulfur and some trace metals, therefore burning of that coal will release SO<sub>2</sub> emission in flue gas and trace element particulates in fly ash. In order to suppress the SO<sub>2</sub> emission in flue and trace element (especially Hg) in fly ash, pulverized coal was mixed with bentonite (as desulfurizer and adsorbent) before subjected to furnace or burner. Introduce of bentonite hopefully can capture the sulfur compound (both in form of SO<sub>2</sub> and S) and trace element particularly mercury (Hg). Experimental was performed at desulfurizer/adsorbent content of 0, 2, 4, 6, 8, 10, 12, 14 and 16% (by weight) and burning temperature of 700, 800 and 900 °C. The 20 gr sample of coal-bentonite mixture with particle sizes -60/pan was put in the ceramic boat then placed in the Electric Tube Furnace. Air flow rate was fixed at 1.5λ (λ = stoichiometric air consumption) and reaction was run for 30 minutes. Sulfur and Hg retentions in bottom ash were analyzed using Spectrophotometer and AAS, respectively. It was found that the addition of bentonite can increase S and Hg retention in bottom ash. Maximum adsorption capacity of S and Hg was observed at bentonite dosage of 16% and combustion temperature of 700 °C. Adsorption efficiency for S reached up to 63.5%, while for Hg was 57.2%.

**Keywords:** *Aceh low-rank coal; adsorbent; bentonite; bottom ash; desulfurizer; fly ash; SO<sub>2</sub> emission; traces element.*

### 1 Introduction

Use of low-rank coal as a fuel in power plant restricted by some factors such as release the GHGs emissions, particulates and trace elements that bring serious environmental problems. Many technologies have been developed and many researches have been attempted in order to reduce the problems. However, nowadays the research and development (R&D) on the technologies are generally focused on those issues done separately, not simultaneously. For example, flue gas desulfurization (FGD) system can only be used to capture the SO<sub>x</sub> emission or flue gas re-burning technologies that only possible to handle NO<sub>x</sub> emission or co-combustion of coal-biomass technologies, which are addressed to solve the problems triggered by CO, CO<sub>2</sub> and N<sub>2</sub>O emissions. First generation for the calcium-based scrubber (a type of FGD) was made in Great Britain in 1920.

Only a few literature can be found study about simultaneous reduction of the SO<sub>x</sub> emission (or other sulfur compounds) and trace elements (or trace metals), especially mercury (Hg) [Ma et al., 2014; Wang et al., 2013]. They applied iron-based sorbents in their investigations. On the other hand, the R&D on the desulfurization technologies using several adsorbents or desulfurizers can be found anywhere [Cheng et al., 2003; Zhou et al., 2001]. Moreover, similar fact might be faced for R&D on the mercury capture by some adsorbents, including bentonite (both natural and treated/modified) [Li et al., 2014]. There is no, at the moment, observation on the simultaneous adsorption of the SO<sub>x</sub> emission and trace mercury using bentonite adsorbent.

Bentonite is an absorbent aluminum phyllosilicate, impure clay consisting mostly of montmorillonite, a clay mineral of the smectite group [Bergaya & Lagaly, 2013; Williams et al., 2009]. The absorbent clay was given the name bentonite by Wilbur C. Knight in 1898. There are different types of bentonite,

each named are respective to dominant element, such as potassium (K), sodium (Na), calcium (Ca), and aluminum (Al) [Borrelli et al., 2013]. Ca-bentonite is the most prevalent of the smectite and is found in many geographical regions in the world. Na-bentonite is relatively rare compared to Ca-bentonite, and it swells more in water than calcium bentonite [Murray, 2006; Fu & Chung, 2011].

Bentonite usually forms from weathering of volcanic ash, most often in the presence of water. For industrial purposes, two main classes of bentonite exist: Na- and Ca-bentonite, which is widely used as a thickener and extender for paints, as an additive in ceramics. The material also applied in producing the health products, cosmetics, foods and pharmaceuticals. However, each of these applications requires specific properties and characteristics of the material [Viseras et al., 2010; Allo & Murray, 2004]. The main uses of bentonite are for drilling mud, binder (e.g. foundry-sand bond, iron ore pelletizer), purifier, absorbent (e.g. pet litter), and as a groundwater barrier.

Chemical contents of natural bentonite mostly comprises of SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, MgO, Fe<sub>2</sub>O<sub>2</sub>, CaO, Na<sub>2</sub>O, K<sub>2</sub>O, TiO<sub>2</sub>, P<sub>2</sub>O<sub>5</sub>, MnO, etc. What compound is dominant depend on region from where the bentonite come from [Arbaoui & Boucherit, 2014; Guerra et al., 2013]. The characteristics of bentonite are influenced by the number of smectite and by the exchangeable cations present in the interlayer space [Hanuláková et al., 2013]. Cation exchange (such as Na in, Ca out) increases the distance between the layers structures of bentonite, this allowing for entrance of the water [Barbanti et al., 1997]. High content of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>, with small contents of other oxides, was observed in the natural bentonite sample [Akpomie & Dawodu, 2015; Li et al., 2014]. Scanning electron microscopy (SEM) of bentonite shows individual particles that are irregular platelets and tend to form thick and large agglomerates [Nones et al., 2015].

The paper discussed the possibility utilize of natural bentonite for an adsorbent in the low-rank coal combustion for capturing the sulfur and mercury simultaneously. Effect of adsorbent content in coal sample at several burning temperatures is presented here in order to give an evident role of the adsorbent in the proposed work.

## 2 Materials and Method

Materials used in the study were coal (low-rank coal from West Aceh), natural bentonite (from Lampung) and strong acid such as HCl and H<sub>2</sub>SO<sub>4</sub>. Sulfur and mercury contents in the raw coal were 87 and 320 ppm, respectively, whereas chemical composition of the bentonite in this research are (by weight): SiO<sub>2</sub> 47.78%, Fe<sub>2</sub>O<sub>3</sub> 2.49%, Al<sub>2</sub>O<sub>3</sub> 9.11%, CaO 5.09% and the rest are some others compounds such as MgO, Na<sub>2</sub>O, K<sub>2</sub>O, TiO<sub>2</sub>, P<sub>2</sub>O<sub>5</sub>, MnO, and so forth with the contents <1%. Those all chemical contents were slightly lower compared to the Na-bentonite used by Li et al. [13] and Akpomie & Dawodu [1] in their investigation. Moreover, equipment needed were crusher, ball mill, vibrating screen, compressor, electric tube furnace, ceramic boat, clay boat, Atomic Absorption Spectrophotometer (AAS), Spectrophotometer, desiccator, flow meter, electric balance, etc.

The coal and bentonite at first separately crushed and pulverized to have a particle size -60/pan mesh. Those materials then homogenously mixed before burned at several contents of bentonite (i.e. 0, 2, 4, 6, 8, 10, 12, 14 and 16% by weight, respectively). After that, 20 gram of the coal and bentonite mixture sample put in the ceramic boat and further placed in the electric tube furnace. The samples therefore were burned at temperature of 700, 800 and 900 °C, respectively for 30 minutes with air flow rate 1.5λ liter/minute (λ is stoichiometric air flow rate) [Yao & Naruse, 2005]. Since the combustion temperature relatively higher, there was no calcination treatment applied to the bentonite before utilized. This idea was also supported by the fact that bentonite might be calcined at lower temperature up to 300 °C [Li et al., 2014].

Sulfur and mercury contents in the raw coal and bottom ash samples were analyzed by Spectrophotometer and AAS, respectively. Pulverized raw coal and bottom ash stored in desiccator for analysis purpose. It should be note that the bentonite-captured S and Hg were retained in bottom ash, while the un-captured S and Hg were emitted in the fly ash. The measured data were used to evaluate the adsorption efficiency both for mercury and sulfur by the following equation:

$$\eta = \frac{C_i(a = n) - C_i(a = 0)}{C_i(\text{in coal})} \times 100\% \quad (1)$$

### 3 Result and Discussion

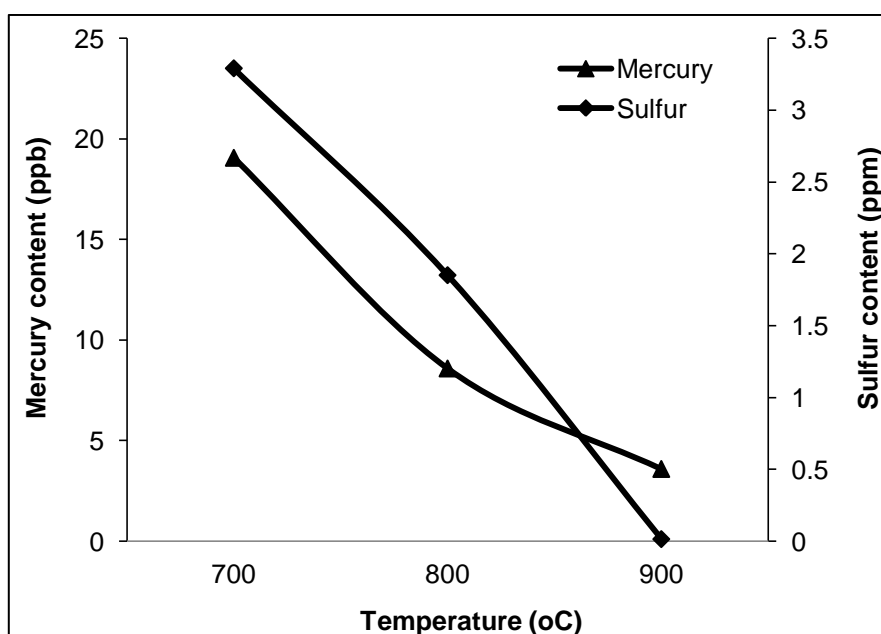
In this section the data and discussions are provided in three sub-section on preliminary test, effect of bentonite addition on the mercury and sulfur contents in the bottom ash and adsorption efficiency, respectively.

#### 3.1 Preliminary observation

Coal combustion generally produced fly and bottom ashes as by product or solid waste. Vaporized trace metals in the coal combustion will partly be suspended in the fly ash and some of un-vaporized will be retained in the bottom ash. Meanwhile, most organic sulfur and pyrite were oxidized and released together in the flue gas, and small part of the sulfur might be captured by the alkaline components of coal ash such as CaO, MgO, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>2</sub>, Na<sub>2</sub>O, K<sub>2</sub>O and remained in the bottom ash. It was found that the alkaline sulfates were dominant at lower temperature under oxidizing conditions [Yan et al., 1999]. Therefore, based on those finding it is expected that those trace elements and sulfur have high tendency to go down to the bottom ash. It is well known that the bottom ash is more easy to handle compared to the fly ash. In order to increase the possibility of those elements or compounds to be retained in the bottom ash, introduce of bentonite in the coal combustion was conducted in this examination.

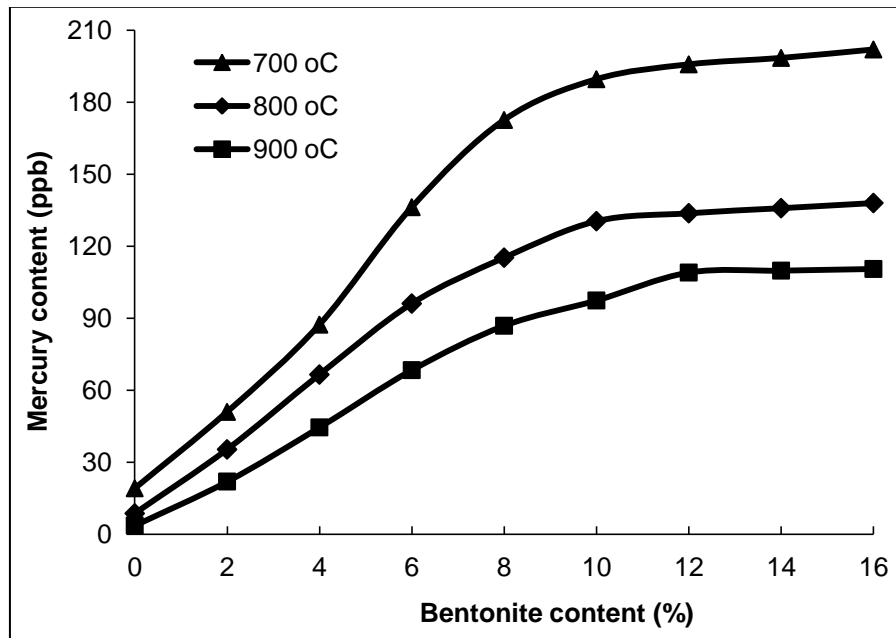
Figure 1 shows the effect of combustion temperature on the Hg and S contents in the bottom ash. The Hg and S contents sharply decreased in regards the increase of temperature. Experimental temperature in this study was fixed in the range of 700-900 °C that was almost same to the Fluidized Bed Combustion (FBB) operating condition [Manninen et al., 1996]. Since the boiling points of Hg and S (i.e. 356.7 and 444.6 °C, respectively) were much lower than operating conditions, that the reason why those elements greatly tended to vaporize in higher temperature. Even though mercury has high volatility, it was still existed in the bottom ash at temperature of 900 °C. That phenomenon might be exhibited by the particulate-bounded mercury (Hgp) from the un-burned carbon in the bottom ash [Wilcox et al., 2012].

Thermodynamic calculations have shown that Hg completely volatilized in the range of FBC operating temperature [Manninen et al., 1996]. However, in this investigation the fact departed from the calculation. The interesting phenomenon was reflected at the interval temperature of 850 to 900 °C; the S curve crossed the Hg curve, it described that the S vaporization rate much higher than that of the Hg vaporization rate in that range of temperature. The different evidence was presented by S element, in which totally vaporized at the higher temperature of 900 °C (or no S content in the bottom ash). Since the existence of S in the bottom ash significantly affected by temperature, it might be said that the Aceh low-rank coal mostly contains inorganic S [Xu & Kumagai, 2003].

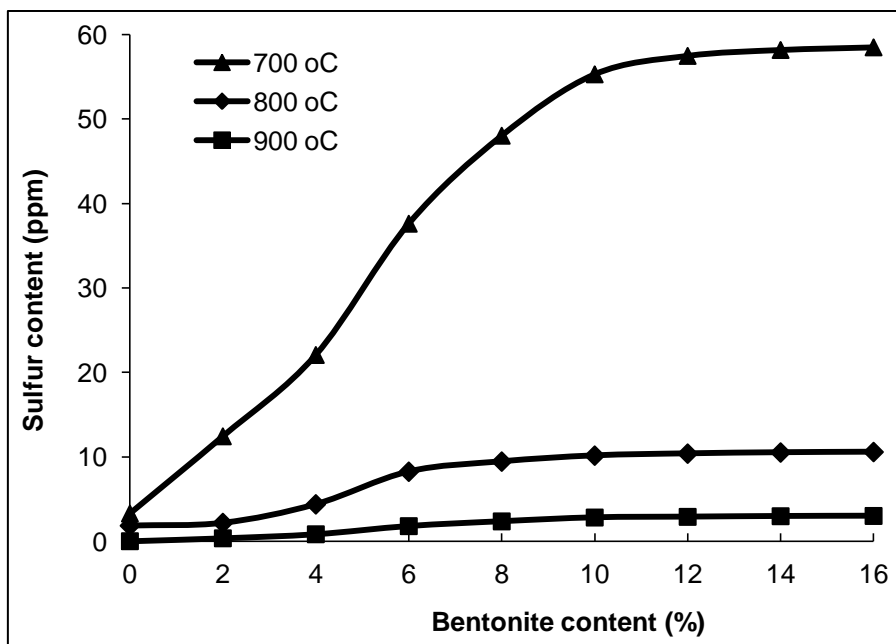


**Figure 1** Mercury and sulfur contents in the bootom ash before mixed with bentonite  
**3.2 Effect of bentonite addition on the mercury and sulfur contents in the bottom ash**

As was mentioned above, addition of the bentonite was addressed to reduce Hg and S contents in the fly ash and flue gas. It is expected by introduce the bentonite as an adsorbent might promoted the affinity or bounding force between the trace metals (Hg) and mineral in the bentonite through the adsorption mechanism and simultaneously lowered the affinity of Hg on S or SO<sub>2</sub> [Huang et al., 2004]. In coal combustion, Hg is possible to react with S or SO<sub>2</sub> to form HgS, and then suspended in the flue gas. In case of S, that attempting enhanced the possibility for inorganic S such as pyrite, marcasite, sulfates and also organic S such as mercaptans, aliphatic and aryl sulfides (even exist in small amount) to be bounded by the alkaline components of bentonite and coal ash, henceforth settled down to the bottom ash. As was proved by Yan et al. [23] that the alkaline sulfates were dominant at lower temperature, it was the reason why S content in the bottom ash higher at temperature of 700 °C because this is the lowest temperature in the examination.



**Figure 2** Mercury content in the bootom ash after introducing the bentonite



**Figure 3** Sulfur content in the bottom ash after introducing the bentonite

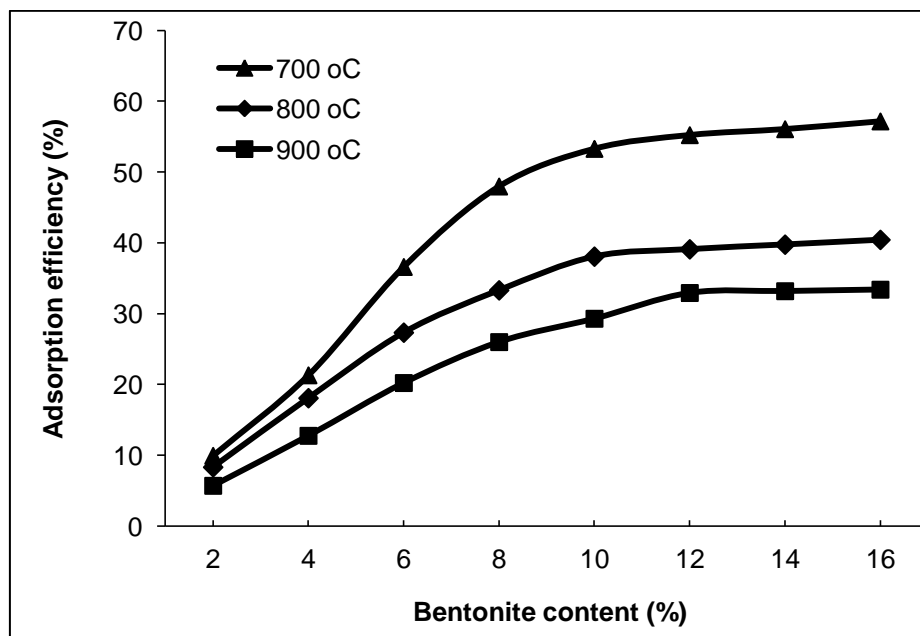
Profiles of Hg and S concentration in the bottom ash are presented in Fig. 2 and 3. It is seen from the figures that the Hg and S concentrations in the bottom ash rise by enhance the bentonite content in the fuel mixture, but it decreased by improvement the burning temperature. The greater concentrations were obtained at the bentonite dosage of 16% and temperature of 700 °C, and the sharp increase also appeared at the same temperature but in the range of bentonite content 0 to 10%. At higher temperature (in this investigation  $\geq 800$  °C) Hg and S concentrations in the bottom ash were small, due to desorption process might taken place at elevated temperature. Moreover, It was observed S was mostly turned into SO<sub>2</sub> gas, less than 10% was retained in the fly ash and less than 1% was held in the bottom ash. On the other hand, about 60% of the calcium was retained in the fly ash and less than 10% was found in the bottom ash [Cheng et al., 2003].

Since the alkaline compounds in this applied bentonite were minor, the effectiveness in the SO<sub>2</sub> capturing was low. Those two facts (desorption phenomenon and low alkalines contents in the adsorbent) influenced the presence of S in the bottom ash in double especially at higher temperature, as can be seen in Fig. 2 and 3 where the curves for S at 800 and 900 °C, respectively much flat compared to the Hg curves ones. Based on the finding data, it might be claimed that the retention of S in any form remarkably controlled by physical adsorption.

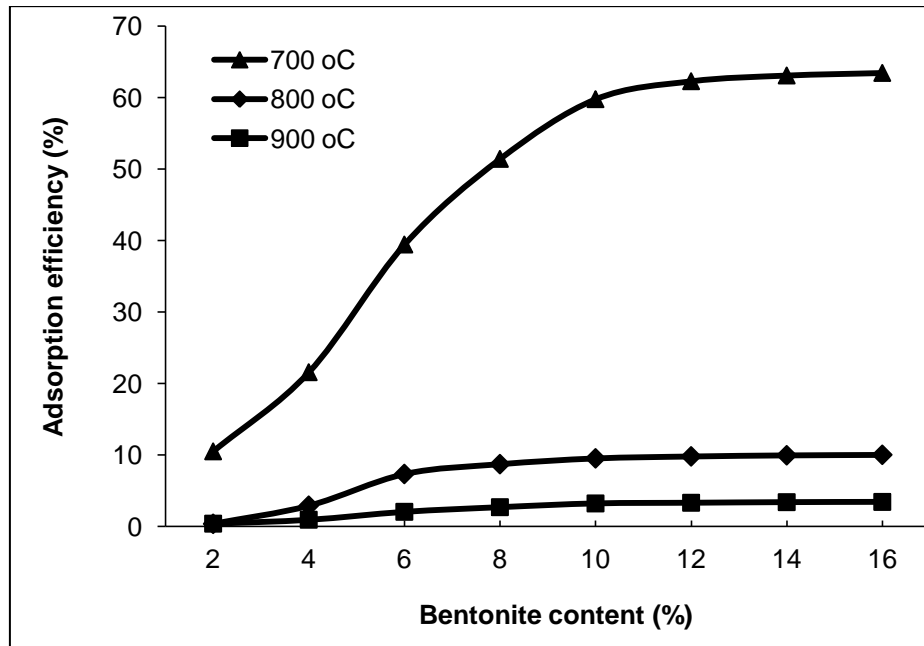
### 3.3 Adsorption efficiency

Although the general trend can be found that the S retention efficiency in the bottom ash was promoted by an increase in molar ratio of Ca/S, as shown in Fig. 4 and 5, however effect of temperature more dominant. Fig. 5 described the adsorption efficiencies for temperatures of 800 and 900 °C only up to more and less 10%. The highest efficiency was appearing at temperature of 700 °C and the bentonite content of 16%, i.e. 63.5%. In despite, the Hg adsorption efficiency was equally affected by both parameters (i.e. Ca/S ratio and combustion temperature) as exhibited by the curves in Fig. 4. The maximum efficiency was read at the same conditions to those observed for the S, with the efficiency as much as 57.2%.

It was discussed that silicon compounds are beneficial to SO<sub>2</sub> reduction from the flue gas. On the other hand, it should be noted that SiO<sub>2</sub> cannot only form silicates that enwrap the sulfation product CaSO<sub>4</sub>, but also form a CaO–SiO<sub>2</sub> phase that has a low capacity for sulfur retention. Whether the Fe–Si–Ca sorbents are beneficial or harmful during coal combustion depend upon factors such as heating rate, furnace temperature and reaction activity of the silicates [Cheng et al., 2003]. It is reported that simultaneously adding Fe<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> into CaCO<sub>3</sub> with the same weight ratio of 0.1% promotes the sulfur removal efficiency from 41.2 to 53.8% at a constant heating rate from room temperature to 1080 °C [Chang et al., 1998].



**Figure 4** Adsorption efficiency for mercury



**Figure 5** Adsorption efficiency for sulfur

## 4 Conclusions

Based on the results and discussion, some conclusions listed here.

1. Raw or natural bentonite can be used as an adsorbent for simultaneously capture the sulfur and mercury on the coal combustion.
2. That bentonite has showed a good performance as an adsorbent for that propose.

The influence of bentonite for the sulfur adsorption was not significant, however for mercury provided more important role.

## 5 Nomenclature

$\eta$	=	adsorption efficiency (%)
$C_i (a = 0)$	=	mercury or sulfur concentration in the bottom ash without adsorbent (ppm)
$C_i (a = n)$	=	mercury or sulfur concentration in the bottom ash with adsorbent (ppm)
$C_i$ (in coal)	=	mercury or sulfur concentration in the raw coal (ppm)

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