Journal of Environmental Engineering and Landscape Management
ISSN 1648-6897 / eISSN 1822-4199
2022 Volume 30 Issue 3: 342–349
https://doi.org/10.3846/jeelm.2022.16294

COMPARISON OF REMAINING COAL-BURNING ASH-BASED ON CD, PB, AND HG CONCENTRATION AT DIFFERENT TEMPERATURES: A CASE STUDY IN ACEH PROVINCE

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Received 09 May 2021; accepted 09 November 2021

Highlights

- The use of natural adsorbents applied in this work serves to capture heavy metals during the combustion process.
- Heavy metal Pb can produce the highest absorption capacity of 39.85 ppm/gr compared to heavy metals Cd and Hg.
- The most optimum absorption efficiency was recorded for heavy metal Cd by 22.96% compared to heavy metal Pb and Hg.
- The ratio of zeolite adsorbent used for coal binder is 2%, 4%, 6%, 8%, and 10% and temperature 600 °C, 700 °C and 800 °C.

Abstract. This study aims to investigate the efficiency level of absorption of heavy metals Cd, Pb, and Hg. Combustion is carried out using coal with the addition of absorbent ratios of 2%, 4%, 6%, 8%, and 10%. The adsorbent used is natural zeolite which is widely available and inexpensive. This study provides practical implications for the easy and inexpensive removal of heavy metal emissions during combustion. The results show that the maximum efficiency level for Cd metal reached 22.96% which was recorded at a temperature of 600 °C for an adsorbent ratio of 10%. The maximum efficiency level of Pb metal from the experimental results was obtained at a temperature of 600 °C with an adsorbent ratio of 10% to 10.83%. Meanwhile, the efficiency level for Hg metal produced was 0.05% which was recorded at the adsorbent ratio of 10% at 800 °C. The maximum total capacity of Pb metal for each tested combustion temperature was 600 °C 39.85 mg/kg, 700 °C 25.43 mg/kg, and 800 °C 7.21 mg/kg. On the other words, the higher the combustion temperature tested, the lower the absorption efficiency rate obtained.

Keywords: briquets, coal-burning, metal, combustion, efficiency.

Introduction

In recent years, haze resulting from burning has become increasingly common, especially in the largest cities around the world. The result of combustion using fossil fuels becomes a major source of emission production as described in several previous publications (Bi et al., 2007; Wei et al., 2018; Xie et al., 2008). The absorption of particulate pollution in most areas of Mainland China has also been investigated by (Wei et al., 2018). Combustion of biomass by municipal incineration can account for 9.8% of the annual mean mass concentration of PM10 as reported by (Xie et al., 2008). Coal is the largest source of electricity generation in the world amid global efforts to reduce the use of coal. The International Energy Agency or IEA stated in 2019 that coal holds 38% of the total share of power generation sources globally (Erdiwansyah et al., 2019b, 2021). The use of coal energy, especially in

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Indonesia, until 2025 is predictably high as reported in a study by (Erdiwansyah et al., 2019a). Since 2016, global coal production from year to year has continued to increase. Meanwhile, global coal reserves are recorded at more than one trillion tons. The IEA data report shows that total world coal production managed to reach 7.9 billion tons in 2019, higher than in 2014. Thermal and lignite coal account for about 86% of this production and the remainder is metallurgical coal.

Indonesia is the fourth largest country in coal production after China, India, and the United States. In 2019, coal production in Indonesia reached 616 million tons, China had 3.7 billion tons, India had 783 million tons, and the United States had 640 million tons (Kementerian Energi dan Sumber Daya Mineral 2016, 2019). Meanwhile, the Chinese Department of Statistics in 2018 reported that China contributed the most to coal production which reached 69.3% and contributed 59% to national energy production (Cheng et al., 2019). The working substances of coal are potentially very dangerous (Shao et al., 2015). Significantly, particulate pollutants have a major impact on atmospheric chemistry and climate change, especially for human health (Jones et al., 2009; Kan et al., 2012; Pian et al., 2016). Emissions resulting from burning coal in households used for heating and cooking are closely related to burning coal as in China (Li et al., 2012). Coal consumption processing and control systems must be a priority, especially for PM2.5 pollution (Xie et al., 2020).

Investigations regarding the composition of the elements of coal are very important to know geological information related to the environment. Toxic metals and metalloids found in coal can be released to the surface, groundwater, air, and soil during the combustion process (Tang et al., 2009). The toxic elements from coal can evaporate and condense on the ash particles for the energy production process (Yan et al., 2000). In general, cadmium (Cd) can be found in organs, pyrite, clay, and coal carbonate minerals (Yang, 2010; Shi et al., 2018). The Cd contained in coal is up to 80% lower than monosulfide, especially sphalerite. Meanwhile, 20% of Cd is bound to be the same as silicate and pyrite (Finkelman et al., 2018). Fly-ash particles containing Cd vapor obtained during off-gas cooling can be removed by electrostatic deposition (Goodarzi & Huggins, 2001). Mercury (Hg) found in coal can be associated with pyrite, calcite, marcasite, and cinnabar (Diehl et al., 2004; Hower et al., 2005). The toxic element in Hg cannot be broken down and accumulates in the human body so that it can cause the body's organs and nervous system (Tang et al., 2009). Power generation using coal can increase the Hg content of flying ash which can hurt the environment (Dittert et al., 2007). Bottom ash that accumulates from various toxic elements can create consequences for the health of people who live around the power plant area for a long time (Dai et al., 2012).

The purpose of this research was to study the residual ash from burning coal based on metal concentrations with different adsorbent ratios and temperatures. The mini-style zeolite adsorbent was chosen because it is widely available in Indonesia and is cheap and resistant to high temperatures. Selection of the three heavy metals Hg, Cd, and Pb because they are found in coal and are toxic. The adsorbent used is natural zeolite which is widely available and inexpensive. This study provides practical implications for the easy and inexpensive removal of heavy metal emissions during combustion. The experiments in this work were carried out three times with each combustion temperature of 600 °C, 700 °C, and 800 °C. Each experiment used five adsorbent ratios of 2%, 4%, 6%, 8% and 10%. The aim of this test is to determine the level of absorption, absorption, and absorption efficiency of the ratio of the adsorbent with different combustion temperatures. The results of this work can provide a scientific reference in the briquette production process by adjusting pollutants to the atmosphere. However, it also provides a huge potential for insights into the production that is obtained. The results of the comparison of each treatment with different adsorbent ratios and combustion temperatures are a special discussion in this work. The novelty of this research is that the natural adsorbent used can capture heavy metals during the combustion process. In addition, the weight of the natural adsorbent applied for combustion temperatures of 600 °C, 700 °C, and 800 °C can adjust to the given treatment.

1. Experimental setup and material

1.1. Experimental setup

The experiment used coal from Kaway XVI of West Aceh Sub-District in Aceh Province, Indonesia, while 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 (Mahidin, 2009). 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 percent 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/cm^2 (SNI 047, 2006). Coal was crushed using a crusher until the particle size was homogenized up to 60 mesh. Then it was printed using a coal briquette printer with a pressure of 10 tons/cm^2. The size of the coal briquettes was printed with variations in cylinder shape up to 6 in diameter and the same cylinder briquette height was 10 cm. The purpose of milling was to homogenize the size (60 mesh) for better mixing and a more perfect briquetting process.

The furnace for the experiment in this study is shown in Figure 1. The specifications of the tools used in this study are shown in Table 1. The test experiments carried out in this work are shown in Table 2.

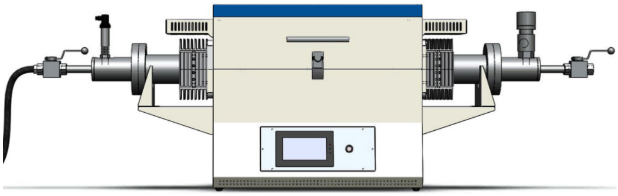

Figure 1. Combustion Chamber

Table 1. Specifications of tube furnace

No	Properties	Range
1	Tube material	Ultrahigh temperature alloy
2	Tube diameter	Φ85 mm
3	Tube length	1000 mm
4	Heating length	440 mm
5	Constant heating zone	200 mm
6	Sealing method	Pressure flange
7	Pressure measured	Semiconductor pressure gauge
8	Over-pressure protective	Safety relief valve
9	Pressure adjusts	Manual and auto adjustable
10	Over temperature alarm	Over-temperature power off
11	Working temperature	≦1100 °C
12	Working pressure	≦20 MPa @800 °C ≦12 MPa @900 °C ≦6 MPa @1000 °C ≦4 MPa @1100 °C
13	Temper accuracy	±1 °C
14	Temper control mode	AI-PID 30 Programmable control
15	Display	7” LCD Touch Panel
16	Working gas	N2, Ar2, O2 and other non-corrosive gas
17	Power supply	AC110V/AC220V 50Hz/60Hz

Table 2. Treatment and combustion temperature

No	Briquet Batubara	Temperature (600, 700, and 800 °C)
1	2% adsorbent	Treatment-I
2	4% adsorbent	Treatment-II
3	6% adsorbent	Treatment-III
4	8% adsorbent	Treatment-IV
5	10% adsorbent	Treatment-V

From each of these combustions, there is residual ash that remains in the ceramics boat (bottom product) which is then temporarily stored in a desiccator to condition the temperature of the combustion in the furnace with the ambient temperature.

1.2. Material

The materials used in this research are coal, synthetic zeolite, and a strong acid reagent solution that functions for Hg, Pb, and Cd metals. In addition, the equipment used includes crusher, ball mill, vibration screen, coal briquette printer, compressor, tube furnace, electrically stainless-steel reaction tube, industrial gas combustion, and emission analyzer (E4400), desiccator, ceramics boat, multi ware, atomic absorption spectroscopy (AAS) 600, wet gas meter, chemical glass, Erlenmeyer, and flow meter. The uses and functions of each tool used in this study are described in Table 3. The structure of the process of materials used in this study is detailed in Figure 2. The process is carried out from the beginning of the preparation process to the final test.

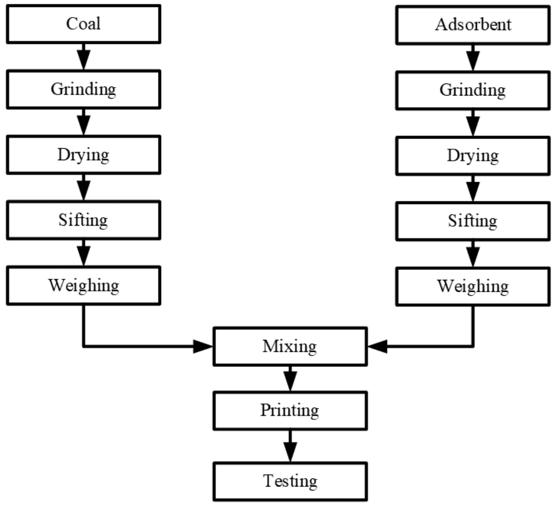

Figure 2. Diagram for material process

Table 3. Types of research tools and their respective functions

No	Categories	Using
1	Crusher	Crushing of coal and zeolite
2	Ball Mill	Refine coal and zeolite
3	Vibration Screen	Coal and zeolite sieving
4	Coal Briquette Printer	To print briquettes
5	Compressor	Air combustion
6	Tube Furnace	Combustion Chamber
7	Electrically Stainless-Steel Reaction Tube	Pipe of combustion
8	Gas Combustion, and Emission Analyzer (E4400, E Instrument)	SO_2 and CO
9	Desiccator	Saver of sample
10	Ceramics Boat	Place the briquettes in the combustion chamber
11	Multi ware	Sample and briquette storage containers
12	Atomic absorption spectroscopy (AAS)	Cd, Pb, and Hg
13	Wet Gas Meter	Flow rate calibration
14	Chemical Glass	Material analysis of heavy metals
15	Erlenmeyer	Container for analysis process
16	Flow meter	Flow meter

There are two variables carried out in this study, namely fixed variables and random variables. Fixed variables include briquette weight, cylindrical briquette, coal particle size pressing pressure, zeolite, airflow rate, and combustion time. Meanwhile, the random variable include briquette combustion temperature and adsorbent ratio.

2. Result and discussion

The research in this paper was conducted to investigate the maximum point of absorption efficiency levels for cadmium (Cd), lead (Pb), and mercury (Hg) with the addition of adsorbents of 2%, 4%, 6%, 8%, and 10%. The test was carried out at a combustion temperature of 600 °C, 700 °C, and 800 °C for each treatment. The main fuel used is coal and adsorbents as pollutants. Residual samples from combustion products such as heavy metal content were analyzed in the laboratory of atomic analyzer spectrometer. Based on the results of the experiments carried out, it can be explained the comparison between each adsorbent ratio and the treatment is described below.

2.1. Comparison of bottom ash weight

The experimental results with combustion testing at temperatures of 600 °C, 700 °C, and 800 °C for different adsorbent ratios are shown in Figure 3a. It is shown that the higher the adsorbent ratio given, the higher the remaining combustion ash obtained. The highest remaining burning ash was recorded at the burning temperature of 600 °C. The results of the investigation show that when the combustion temperature increases, the remaining ash obtained will also be lower. This is because the fuel that is put at low temperature does not burn properly and vice versa; while if the combustion temperature is higher, then the fuel can burn optimally. The highest remaining burning ash at a temperature of 600 °C reached 2.01 grams. Meanwhile, the remaining ash produced when the temperature reached 800 °C was 1.62 grams, 0.39 grams lower than at 600 °C, and 0.3 grams lower than 700 °C. The higher the temperature, the more complete the combustion, and the more heavy metals emitted.

The results of experimental measurements for mercury are shown in Figure 3b. It can be explained that the highest residual ash weight recorded at 600 °C reached 2.01 grams for an adsorbent ratio of 10%. The results of measurements carried out with temperatures of 700 °C and 800 °C of lower ash residue were 1.92 grams and 1.62 grams, respectively. Meanwhile, the results of measurements of the remaining metal ash of Pb and Cd with an experimental combustion temperature of 700 °C and 800 °C were the same as Hg shown in Figure 3a and 3c. However, the higher the temperature given, the more perfect the combustion rate will be. This is because the high combustion temperature can burn fuel optimally so that the resulting efficiency is better. While testing the combustion at a low temperature, the fuel that was entered did not burn out. However, the experiments carried out in this test show that the remaining combustion ash produced was almost the same for testing the entire temperature. However, the resulting levels of efficiency and absorption show a difference. Meanwhile, investigations for the recovery of coal fly ash (CFA), coal bottom ash (CBA), and rice husk ash (RHA) for a partial replacement of ordinary Portland cement in translucent concrete have recently been discussed (Lo et al., 2021). The results of the investigation show that the carbon footprint from shifting ash can be reduced by 9.9–20.6%/m^3 of translucent concrete. However, the substitution of the ash material carried out in their study aims to reduce the carbon footprint that can be found in translucent concrete. The use of adsorption for maximum metal absorption capacity for Pb (II) and Hg (II) ions can be generated up to 270.3 and 400.0 mg/g (Fu et al., 2019). These results were obtained from the application of XPS spectrum with high resolution. The results show that the investigation of the residual weight of ash against heavy metals Cd, Pb, and Hg with the addition of an adsorbent ratio of 2–10% as the results of research in this work have not been found.

2.2. Comparison of the total bottom metal

The results of the total metal comparison analysis based on the experiments conducted show that the phenomena that occur are almost the same as the results of the analysis for the remaining burning ash. The total heavy metal collected from the highest combustion product was recorded in Pb metal at 51.10 mg for an adsorbent ratio of 10% at 600 °C as shown in Figure 4c. While the total weight of Cd and Hg were 2.79 mg and 0.32 mg, respectively, with

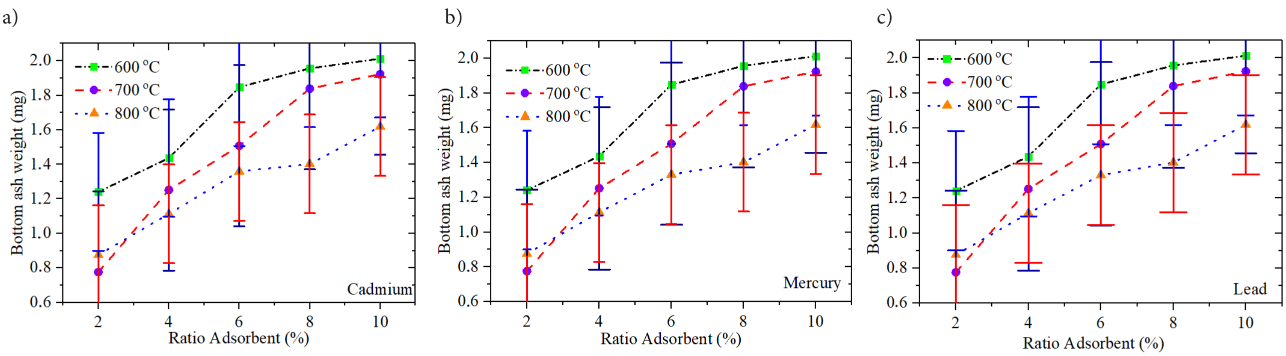

Figure 3. Remaining Cd, Hg, and Pb ash on the adsorbent ratio for different temperatures

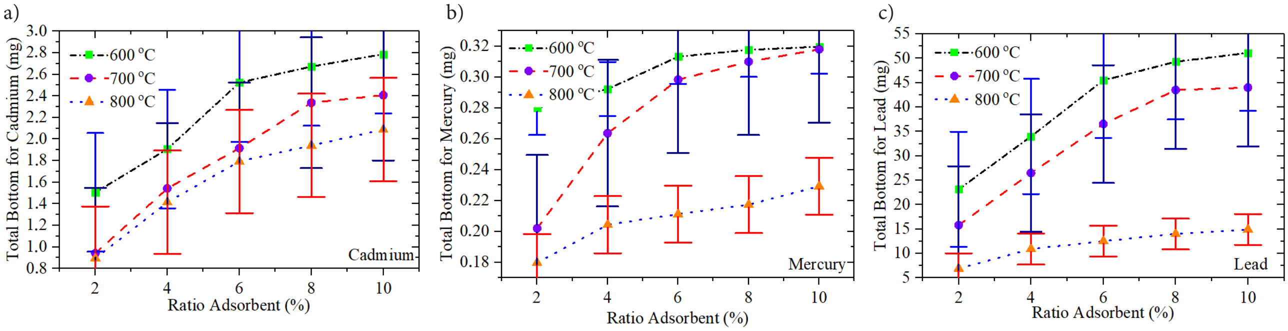

Figure 4. Total Cd, Hg, and Pb metal based on the adsorbent ratio for different temperatures

an adsorbent ratio of 10% as presented in Figure 4a and Figure 4b. A good and recommended standard adsorbent ratio for coal combustion binders is between 6–10%. If the adsorbent ratio is used too much, it is not optimal because the adsorbent system evaporates very quickly. Research using samples of fly ash, bottom ash, and surface soil from a waste incineration plant with the aim at investigating heavy metals in microplastic content has been studied (Shen et al., 2021). Whereas heavy metals such as Cr, Cu, Zn, Pb can be automatically adsorbed on the microplastic surface. The resulting Cr and Pb content reached 8.69, 2.15, 3.67 mg/kg and 9.67, 92.58, 7.27 mg/kg on bottom ash, fly ash, and soil. Their results were inferior to the results carried out in this work. However, the experimental treatment carried out showed a fundamental difference. The most widely used heavy metal investigation is to remove Cd2 +, Pb2 +, and Zn2 + ions as reported in a review study (Fouda-Mbanga et al., 2021). Meanwhile, investigations regarding the total metals Cd, Hg, and Pb based on the ratio of adsorbent and temperature are still lacking in various publications.

Particle size is very influential on combustion characteristics. Fine coal particles will make the coal burn faster and the process of releasing heavy metals is easier and faster so that more metals are emitted. Meanwhile, for adsorbents, the smaller the particle size, the larger the contact area, so that more heavy metal emissions can be captured. The importance of this research is to be able to absorb heavy metals during combustion in the presence of natural adsorbents used.

2.3. Comparison of absorbed metal

Five experimental captions based on the ratio of the adsorbent to each combustion temperature were carried out to investigate each metal that was absorbed for each combustion temperature tested. The results of the analysis show that the highest metal absorption was recorded for the Pb metal of 43.89 mg, which was obtained at the adsorbent ratio of 10% to 600 °C as shown in Figure 5c. Meanwhile, the absorption rate of Hg resulting from testing with different adsorbent ratios only reached 0.19 mg, much lower than heavy metal Pb as presented in Figure 5b. Meanwhile, the total Cd metal absorbed from the experiments with the highest different adsorbent ratios only reached 1.88 mg, which was obtained from the ratio of 10% for the combustion temperature of 600 °C as shown in Figure 5a. The results of the tests show that the higher the combustion temperature can reduce the absorption rate of heavy metals. This is because higher temperatures can burn coal completely. Meanwhile, the adsorbent ratio as a treatment which is a coal binder also looks better if the given ratio is higher. Meanwhile, the high efficiency produced, both absorption processes with higher adsorbent ratios and temperatures, showed a better level of efficiency. A better absorption efficiency level was obtained for heavy metal Cd than for Pb and Hg. The highest absorption efficiency level in heavy metal Cd reached 22.96% which was recorded at the adsorbent ratio of 10% to a combustion temperature of 600 °C as shown in Figure 6a.

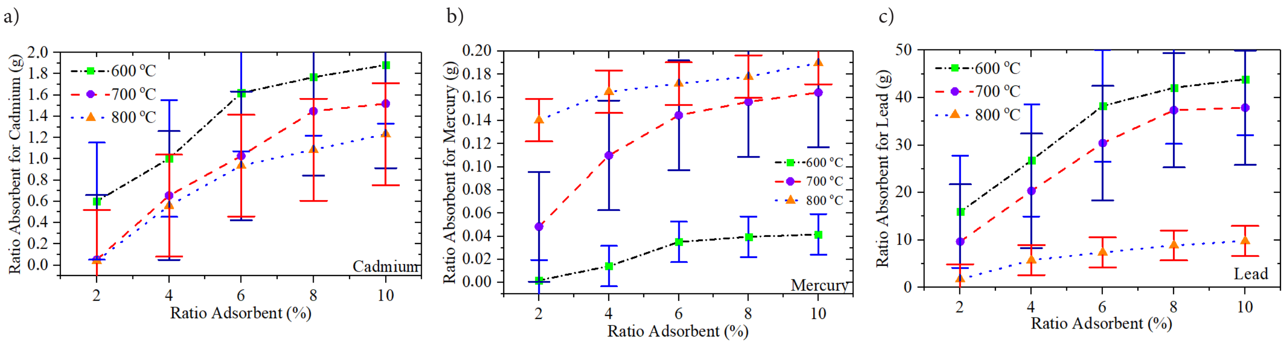

Figure 5. Total metal Cd, Pb, and Hg absorbed based on adsorbent ratio at different temperatures

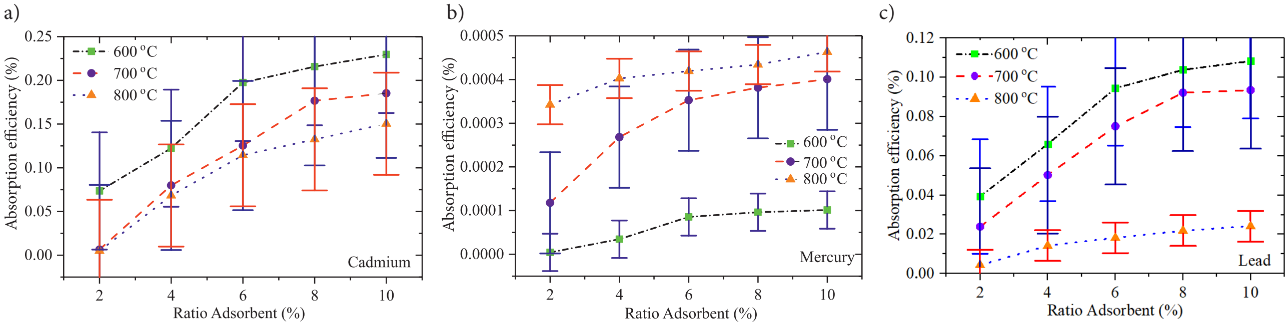

Figure 6. Efficiency absorption Cd, Hg, and Pb metal for ratio at different temperatures

2.4. Comparison of absorption efficiency

Furthermore, the analysis of the highest Pb absorption efficiency level of 10.83% was recorded at the adsorbent ratio of 10% for the combustion temperature of 600 °C. The absorption efficiency level of Pb metal was much lower than that of heavy metal Cd as shown in Figure 6c. Meanwhile, the absorption efficiency level resulting from the testing of heavy metal Hg was significantly lower than the heavy metal Cd and Pb. The absorption efficiency level of heavy metal Hg reaches 0.10% as shown in Figure 6b. This shows that the efficiency level resulting from the testing process for heavy metal Hg is not good. Investigations regarding the mass absorption efficiency (MAE) of elemental carbon (EC) which were sampled as a measurement of emission sources through solid fuel combustion using thermal optical carbon analysis tools have also been carried out (Shen et al., 2013). The results of the measurements carried out show that the absorption efficiency of combustion reached 7.9 (4.8–11) mg/kg. Meanwhile, the use of four types of adsorbent material used has a significant removal efficiency for the treatment of sponges, especially for heavy metal Cr with each of (19.09%) and cotton (26.36%). Meanwhile, in this research, the level of absorption efficiency studied was the addition of the adsorbent ratio for the absorption efficiency of heavy metals Cd, Pb, and Hg.

2.5. Comparison of absorption capacity

The highest absorption efficiency level of mercury was obtained at an adsorbent ratio of 10% when the temperature reaches 800 °C of 0.46%. Meanwhile, the absorption efficiency level at 600 °C and 700 °C reach 0.10% and 0.40% as shown in Figure 7a. The absorption capacity obtained when the combustion temperature reaches 800 °C was 0.35 mg/kg, while when the combustion temperature of 600 °C and 700 °C were 0.03 mg/kg and 0.15 mg/kg, respectively. Meanwhile, previous studies show that the addition of the maximum adsorption capacity of Cu (II) and Pb (II) reached 61.96 and 138.11 mg/kg, respectively (Jiang et al., 2020). However, the treatments and materials used in their study were significantly different from this study. There have been many studies on the use of coal fuel for various analyses, but investigations in particular for the addition of adsorbents to absorb heavy metals such as Cd, Pb, and Hg are still very rare in previous publications. Thus, this research can provide new references in terms of heavy metal absorption by adding different adsorbent ratios. This research is a continuation of our previous work which has been published recently (Gani et al., 2021).

The highest absorption capacity of Pb metal with different adsorbent ratios and combustion temperatures of 1.51 mg/kg was recorded at the adsorbent ratio of 2% to 600 °C. The absorption rate of Pb metal was higher than the heavy metal Hg as shown in Figure 6b. Overall, the Cd absorption rate for the three combustion temperatures tested was higher than the Hg variety. Meanwhile, the absorption rate for Pb metal was slightly better than Hg metal, especially at the combustion temperature of 600 °C and 700 °C. The maximum absorption capacity rate for Pb of 39.85 mg/kg is shown in Figure 8. This result is the

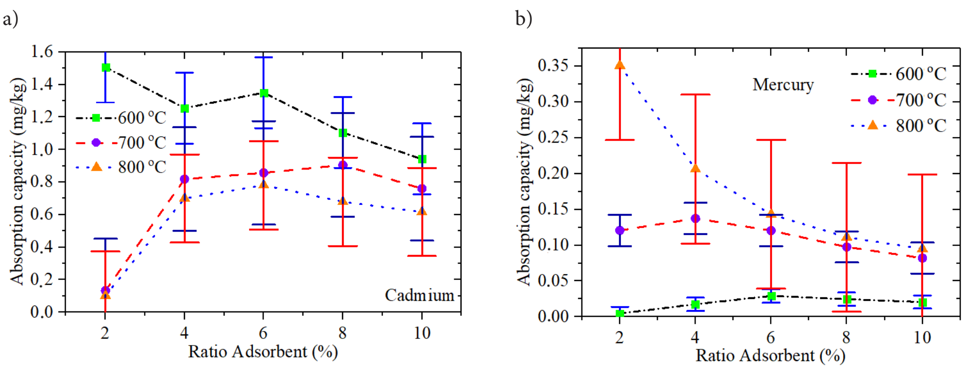

Figure 7. Total absorption capacity Hg and Cd with ratio and different temperatures

highest compared to Cd and Hg. The overall absorption capacity of Pb showed higher yields for all the different treatments and temperatures.

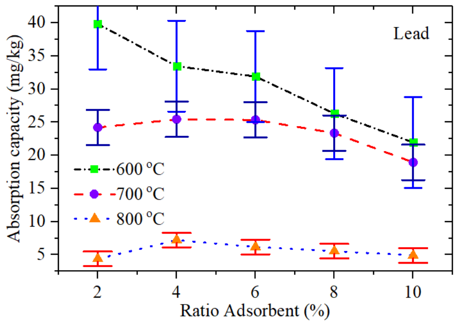

Figure 8. Total absorption capacity Pb with ratio and different temperatures

Conclusions

Investigations on the experiments in this study were conducted to compare and find the most optimum results from different ratios and combustion temperatures. Heavy metals analyzed in this test include Pb, Hg and Cu. Heavy metal Pb produced the highest absorption capacity of 39.85 mg/kg at a combustion temperature of 600 °C with an adsorbent ratio of 2%. Heavy metal Pb also produced the highest absorption rate of 43.89 mg when the combustion temperature was 600 °C with an adsorbent ratio of 10%. Meanwhile, the best absorption efficiency was recorded for heavy metal Cd by 22.96% compared to heavy metal Pb and Hg. Based on several experiments, the results show that heavy metal Pb was more dominant in terms of absorption and absorption compared to heavy metals Cd and Hg. Meanwhile, the remaining amount of ash from the three tested combustion temperatures produced the same value.

Acknowledgements

This research supported by Universitas Syiah Kuala, Research Institutions, and Community Service.

Conflict of interest

There are no conflicts to declare.

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