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Iron bearing oxide minerals separation from rare earth elements (REE) rich coal fly ash

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Abstract. Recovery of Rare Earth Elements (REE) from waste is an important alternative to solve global REE crisis. Therefore, coal fly ash containing REE becomes a potential candidate as an alternative source of REE. Indonesia has a large potential of coal resources, however unlike other type of coals which is rich in SiO₂ and Al₂O₃, Indonesian coal contains significant amount of Fe₂O₃. As a consequence, fly ash resulted from burning Indonesia coal is also rich in iron oxides. The presence of high concentration of iron oxides will however reduce effectiveness of REE recovery from coal ash through hydrometallurgy process. Therefore, iron oxides in the coal fly ash needs, firstly, to separate preceding further REE separation. This study determined the optimal process conditions in the iron recovery process from coal fly ash. The variables studied were fly ash grain size, magnetic field strength and separation techniques. The iron recovery was determined by mean of the ratio of iron concentration after and before magnetic separation. Experimental results showed that increasing magnetic field strength and decreasing coal fly ash grain size in a wet separation will increase the recovery of magnetic iron component. Magnetic field strength of 4300 Gauss and grain size of -400 mesh were the optimal values in this study with iron recovery of 25.89%.

1. Introduction

Rare Earth Elements (REE) are elements of lanthanide group widely used in industries such as petroleum industry, electronics, information, and communication devices, even in green technology. REE play a key role in improving performance products, especially in clean energy technology and consumer goods [1][2][3]. Along with the extensive use of green technology to reduce global warming and improve energy efficiency, utilization of REE in various products is predicted to increase [4], between 2006 and 2035 with annual growth rate between 3.7% -8.6% [1]. Despite of its high annual demand rate the availability of REE is not enough, this condition is known as REE crisis [4][5][6]. One scenario to overcome this scarcity is to extract REE from waste containing REE including from coal fly ash.

Coal fly ash is known to contain REE and it becomes a candidate for REE alternative source. Several studies show that the amount of REE in coal fly ash (CFA) is relatively high so that several countries such as China, USA and South Africa consider coal fly ash as an alternative source of REE.

In 2015, Indonesian steam power plants had produced 4.38x10⁶ tons coal fly ash and bottom ash and it is predicted that the production will reach by 8.31x10⁶ tons in 2019 [7]. This information shows



that Indonesia has an abundance resource of coal fly ash and coal bottom ash. During coal combustion process, concentration of trace elements in the coal fly ash will increase up to 30 times [8]. Therefore, utilization of coal fly ash as a source of REE might become more attractive in the near future. In addition, the recovery of REE from fly ash will become an alternative solution to reduce environmental pollution.

In recent years, there have been many studies in several countries related to REE recovery from coal ash. Acid leaching is usually used to extract REE element in coal fly ash. The results show that the dissolution process was highly depending on fly ash particle size, solution concentration and dissolution temperature [9][10].

In Indonesia, Anggara *et al.* (2018) conducted analysis of REE contain in coal from Banko coalfield, South Sumatera Basin using Inductive Couple Plasma Mass Spectroscopy (ICP-MS). It was found that the REE concentration in Banko coal is between 2.4 and 118.4 mg/(kg coal). When the coal was burned, the REE concentration in fly ash was predicted higher than 1000 ppm. This value was higher than world global average value[11]. In addition, the result of petrographic analysis of fly ash from South Sumatera coal combustion showed that coal fly ash before sieving has 12.7% unburned carbon, 41.5% glass, 15.3% quartz, 11.8% Iron oxide, 6.0% spinel, 12.7 % mullite [12].

The presence of iron oxides in coal fly ash can reduce the efficiency of REE recovery through leaching process (hydrometallurgy). Iron oxides can compete with REE to react with acid in leaching process. Therefore, to increase the REE recovery, iron oxides in the coal fly ash must be firstly eliminated. Magnetic separation and froth flotation are some techniques for reducing iron oxides from iron rich material. However, there are limited reports in case of reducing iron oxides content from REE rich coal fly ash. Lin *et al.* (2017) performed separation of iron oxides from USA coal fly using dry magnetic separation. They found that iron contents in magnetic component tend to decrease when magnetic field strength increases. In addition, REE are associated with aluminosilicates groups and are enriched in nonmagnetic component [13]. This REE analysis agrees with previous finding reported by Zhang *et al.* (2015) for United Kingdom coal fly ash, but there was no information whether the iron oxides are magnetic or nonmagnetic component [14]. In case of Indonesian coal fly ash, there is no report related to magnetic separation for reducing iron oxide content in REE rich coal fly ash. Even though , it has been known that coal fly ash from Indonesia contains relatively high iron oxides [12].The aim of this study is to investigate magnetic separation variables in order to remove iron oxides from fly ash. This will eventually increase REE content in the ash and therefore help improve the efficiency of REE recovery process.

2. Experimental method

Fly ash was produced from a commercial steam power plant which uses coal from South Sumatra as a source of energy. The first step was to sieve the samples in order to get the sample size +200 mesh and -400 mesh. The iron concentration in the fly ash samples were analyzed using ICP-MS performed by ALS Canada Ltd. (Vancouver, Canada) using fused bead method prior to acid digestion, followed by mineralogical analysis using X-Ray Diffraction (XRD, PANalytical type X'pert3 Powder). Subsequently the iron containing material was separated using magnetic separators developed by Research and Development Centre for Minerals and Coal Technology, Bandung. There were two kinds of magnetic separator, i.e dry magnetic separator and wet magnetic separator. Both magnetic separation techniques were used in this work. In wet separation, water was added to fly ash sample with a certain L/S then stirred until the mixture was well mixed. The mixture was then poured into magnetic separator with a specified magnetic strength. Magnetic components would be attracted to magnetic ball inside separator. The non-magnetic component flew to the bottom of separator and was then collected. Magnetic field strengths used in the wet separation study were 4300 Gauss and 10800 Gauss. In dry separation, fly ash particle was spread on a belt conveyor and passed into a magnetic separator. The magnetic component would be attracted to magnetic disc inside separator and non-magnetic component stayed on conveyor belt which is moving out of the separator. Fly ash sample in dry magnetic separator was set at code 1 for vibration to adjust the feed thickness. Magnetic field

strength used in this study were 1100 Gauss, 1900 Gauss, 2650 Gauss, 4300 Gauss and 10800 Gauss. The amount of iron concentration in magnetic and non-magnetic components was analyzed using AAS (Perkin Elmer 3100).

The amount of iron collected as magnetic compound is calculated following this equation:

$$\alpha = \frac{m C_s}{M C_0} 100\%$$

where, α = percentage of iron recovery (%),

m = mass of magnetic compounds (grams),

C_s = iron concentration in magnetic compounds (wt%),

M = initial mass of fly ash sample before separation (grams),

C_0 = initial concentration of iron in fly ash before separation (wt%)

3. Results and discussion

3.1. Analysis of major compounds and mineralogy in coal fly ash

Table 1 shows major oxides in the coal fly ash before sieving using ICP-MS. The iron oxides content in this work is quite similar to result from petrology analysis previously reported by Besari (2018). As seen in the table, the concentration of iron oxides in the fly ash is 13.75%. This value is relatively higher compared to that of coal combustion from other countries, which is below 8% [15][16]. Therefore, reducing the iron oxides content from the fly ash is essential to increase efficiency of REE recovery through acid leaching process.

Table 1. The composition of major compounds in Coal Fly Ash before sieving

SiO ₂ (%)	Al ₂ O ₃ (%)	Fe ₂ O ₃ (%)	CaO (%)	MgO (%)	Na ₂ O (%)	K ₂ O (%)
42.6	20.4	13.75	9.62	6.61	0.67	0.89

Previous study conducted by Besari (2018) found that unburned carbon is still present in coal fly ash [12]. The unburned carbon must be reduced because it could potentially interfere acid leaching process for REE recovery. The presence is indicated with the appearance of coarser grains with black colour. In this work, the unburned carbon was removed mostly as fraction of +200 mesh grain size.

Table 2 shows the iron concentration related to grain size. It is shown that the smaller size fraction contains higher concentration of iron.

Table 2. Iron concentration in fly ash fractions after sieving

Fraction	Iron Concentration (ppm)
+200 mesh	75512.3
-400 mesh	83229

In Table 3, it can be seen that in the fine fly ash, the fraction of hematite and magnetite are respectively 4.4% and 9.5%. In term of iron oxides, the content of both was 13.9%. Table 3 also shows the presence of amorphous glass fraction which amount reaches to 46.6%. In comparison to petrographic data from previous study [12], the concentration in this work is relatively higher. Experimental result in this work shows that iron oxides and glass fraction can be concentrated in the finest fraction (-400 mesh).

Table 3. Mineralogical analysis on -400 mesh fraction using XRD

Quartz (%)	Hematite (%)	Magnetite (%)	Mullite (%)	Amorphous (%)
19.9	4.4	9.5	19.5	46.6

The increase of hematite, magnetite and glass fractions was also reported for coal fly ash from Poland and Kentucky [6][17]. Lower hematite and magnetite concentration in the coarser grain size is due to the presence of high concentration of organic compounds as well a slow pyrite and clay mineral concentration in the initial coal [6]. During combustion process, organic content partly becomes unburned carbon whereas pyrite and clay minerals become hematite, magnetite and glass fraction[18]. The formation of hematite indicated that boiler temperature possibly reached as high as 1400°C [19].

3.2. Iron oxide recovery from coal fly ash

The iron content in the fly ash within fraction of -400 mesh was then separated based on its magnetic properties using magnetic separator. This separation process is needed in order to reduce the iron content in the nonmagnetic components. The REE was predicted to concentrate in the glass fraction which is classified as nonmagnetic components of coal fly ash. Therefore, the iron content in nonmagnetic components is reduced in order to increase the efficiency of further REE recovery through acid leaching process. In this work, the influence of magnetic field strength, grain size and separation method were investigated.

3.2.1. Effect of magnetic field strength.

Hematite and magnetite have different magnetic properties. Hematite is paramagnetic while magnetite is ferromagnetic. Therefore, a greater magnetic strength is needed to separate hematite compared with the magnetite.

Table 4. Effect of Magnetic Field Strength on Iron concentration

Method	Grain Size, (mesh)	Magnetic Field Strength (Gauss)	Mass of Magnetic Components (gram)	Mass of Magnetic Components Percentage (wt%)	Iron Concentration in Magnetic Components (wt%)
Dry separation	+200	1100	57.68	59.35	7.47
		1900	70.98	72.77	7.82
		2650	75.65	77.35	9.63
		4300	80.01	82.37	9.71
Wet separation	-400	4300	9.49	18.99	13.78
		10800	17.98	35.83	12.40

Table 4 shows that increasing magnetic strength can increase iron concentration in the magnetic component and it also increases the amount of magnetic mass that is collected. Higher magnetic strength can attract more iron oxides as they are collected as magnetic component. However, fly ash also contains other minerals that might stick each other. This will cause that separation within magnetic field will also attracts others minerals. This is indicated by petrographic analysis data from Besari[12], as also shown in Figure 1, that some iron oxides were trapped in glass or quartz fraction. Therefore, an increase of magnetic strength is needed to attract iron oxide minerals as well as other minerals to magnetic fraction. However, experimental work in this study showed that fine fly ash



fraction is more hygroscopic and leads to entrainment among particles. The entrainment causes that non-iron components are likely to be also attracted to the magnetic components.

Figure 1. Petrographic analysis for iron oxide after Besari, 2018 [12]

3.2.2. Effect of coal fly ash grain size

Table 5 shows the effect of fly ash grain size on iron concentration in different separation techniques.

Table 5. Effect of Fly Ash Grain Size on Iron Concentration

Method	Magnetic field strength (Gauss)	Particle Size (mesh)	Iron Concentration in magnetic components (wt%)	Mass of Magnetic Component Percentage (wt%)
Dry separation	1100	+200	7.47	59.35
		+325	8.94	55.04
		-400	Cannot be separated using this method	
		+200	9.71	82.37
Wet separation	4300	+200	9.07	29.48
		-400	13.78	18.99

Table 5 shows that for both separation techniques decreasing particle size results the increase of iron concentration in the magnetic components. However, iron in the grain size fraction of -400 mesh cannot be separated using dry separation technique. This was due to entrainment phenomena that is more likely to occur for fly ash with particle size of -400 mesh so that non-iron components is also be attracted to the magnetic component.

As shown in Table 5, coarser grain size fraction contains lower iron concentration compared with the finer one. This is because the coarse coal size contains more unburned carbon resulted from coal combustion process. This result is in accordance with previous study which stated that separation using dry magnetic separator is limited for any material not finer than 75 μ m. For finer material, the effectiveness of separation would significantly reduce due to the effect of air current, particle-particle entrainment, and particle-device adhesion[20].

Based on results from both separation techniques, it is shown that the amount of separated magnetic components from wet separation was lower than that from dry separation. In the wet technique, particles are separated off in water so that the occurrence of entrainment can be avoided. Therefore, the iron separation process can be done more effectively and the concentration of iron is higher than that of dry separation technique.

Table 5 also shows the effect of separation techniques on the iron recovery with the same particle size fraction and magnetic field strength. Although, the iron concentration in magnetic components has slight differences, there is a significant difference in the total amount of magnetic components that

can be separated. With dry separation technique, the collected magnetic components are 82.37% of coal fly ash. However, this result is not in a good agreement with the result of the mineralogy analysis, as reported in previous study [12]. This indicates that some non-iron components are likely being collected into magnetic components due to particle-particle entrainment. On the other hand, REE was predicted to present on the glass phase surfaces which are a non-magnetic component. The presence of non-iron components in the magnetic components will eventually decrease REE recovery. As a consequence, if dry separation technique is used, there will be significant amount of REE in the magnetic components and still mixed with iron components. This will cause further acid leaching process to recovery REE becoming more difficult and inefficient.

In this study, the most effective method to separate iron oxide from coal fly ash is using wet separation techniques with particle size of -400 mesh and 4300 Gauss magnetic field strength. In this condition the iron concentration is 13.78% with mass of magnetic components is 18.99%. So, the iron recovery will be 25.89%.

4. Conclusions

To separate iron components from fly ash, dry separation techniques should be used for fly ash with particle sizes larger than fraction of +200 mesh, while wet separation is for finer particles. The higher magnetic strength can increase iron concentration, glass, and mullite fraction in the magnetic component. The optimum value of magnetic field strength in this study is 4300 Gauss. Fly ash with fine particle size fraction has higher iron concentration in the magnetic component. The highest iron concentration can be achieved from fly ash having particle size fraction of - 400 mesh with an iron recovery of 25.89%.

5. Suggestion

It is recommended to investigate the effect of magnetic field strength between 4300 Gauss and 10800 Gauss in wet separation techniques for iron recovery in magnetic components.

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