Extraction of Alumina from the Coal Fly Ash by Hydrochloric Acid

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Abstract

Fly ash landfills that accumulate a by-product of coal combustion and gasification represent a permanent threat to the surrounding environment due to many factors (air and water pollution, soil contamination, wildlife poisoning, etc). Moreover, disposed coal fly ash may contain significant amounts of valuable elements that are not extracted. To improve the above situation, a combined ash treatment process was developed for utilisation of the coal fly ash waste from coal-fired power stations. The ash treatment includes three stages: 1) magnetic separation of an iron-containing fraction, 2) carbon separation by floatation, and 3) extraction of aluminum by the autoclave hydrochloric acid leaching. The lab-scale results of the ash treatment applied to the Ekibastuz brown coal fly ash from the Omsk power stations (Russia) were presented and discussed. The XRD analysis showed that the fly ash consists primarily of quartz, mullite and magnetite. It was found that the magnetic fraction separated at the first stage is enriched in magnetite (over 20 wt %), the carbon content in the concentrate after flotation increases to 27 wt %, and 90-95 % of aluminum can be extracted during the autoclave acid leaching. The SEM analysis showed that the magnetite phase is grown on the surface of aluminosilicate spheres as ~ 1 mm cubic crystals. The effect of the autoclave temperature and exposure time on the Al extraction efficiency was also investigated and analysed in the present paper. The optimal autoclave temperature and exposure time were found to achieve the maximum Al extraction efficiency. It was also found by the SEM microanalysis that further extraction of aluminum is not economically feasible since the remaining Al is evenly surrounded by SiO_2 in the fly ash particles.

Keywords: coal fly ash, magnetic separation, flotation, leaching, hydrochloric acid.

1. Introduction

A lot of power stations world-wide still use coal combustion as a main process to generate electricity and heat. In the city of Omsk thermal power stations utilize brown coal from the Ekibastuz coal field (Kazakhstan), which is characterised by high ash content (up to 40 %) [1]. Electricity and heat for the Omsk city is produced mainly by two coal-fired power stations TPS-4 and TPS-5 that generate 450 000 and 1 150 000 tons of coal fly ash (CFA) per year, respectively. In total, ash disposal sites near Omsk accumulate more than 75 billion tons of CFA [2].

CFA usually contains from 20 to 35 wt % of Al_2O_3, which can partially be extracted by different methods [3,4]. Recent studies on the Al extraction from CFA using acid leaching show that hydrochloric acid is the most reactive in regard to Al compared to sulphuric, nitric and hydrofluoric acids [5]. However, if leaching is carried out at atmospheric pressure (e.g. 6M HCl, T = 107 °C) the extent of Al extraction does not exceed 70 % [6,7] One of the most perspective methods to intensify leaching of a high-silica aluminum-containing feedstock is to use autoclaves [8,9]. This method allows to exclude preliminary annealing and two-stage leaching. As silicon dioxide does not react with acids a preliminary desilication is not necessary. Besides aluminum iron will also go into the solution, which makes it necessary first to remove magnetite by magnetic separation.

In the present work a combined treatment of the raw CFA collected from the Omsk TPS-4 was carried out in three stages: 1) iron separation by magnetic separation, 2) carbon separation by floatation, and 3) aluminum extraction by autoclave acid leaching.

2. Experimental

The raw CFA collected from the Omsk TPS-4 ash disposal site (that contains ~37 billion ton of fly ash) was used as an initial waste material. The CFA is produced from combustion of the Ekibastuz brown coal (Kazakhstan) on the Omsk TPS-4. The average chemical composition of the raw CFA is given in Table 1.

Table 1. The average chemical composition [wt. %] of CFA from TPS-4 (Omsk, Russia).

Component	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	K_2O	CaO	MgO	MnO	Na_2O	P_2O_5	LOI*
Contents	59.66	24.26	5.42	1.15	0.74	0.88	0.40	0.11	0.44	0.45	6.49

*LOI – loss on ignition at 1000 °C

A roll-type magnetic separator MBS 150x125 (NPO “Erga”, Russia) was used to separate the magnetic and non-magnetic fractions of CFA. Initial CFA samples of ~500 g were mixed with water and loaded into the separator. The magnetic field strength applied was 110 mT.

Carbon flotation tests were carried out in a flotation machine 189FL (REC “Mekhanobr-Tekhnika”, Russia) using methyl isobutyl carbinol (MIBC) as a foamer (frother) and kerosene KO-25 as a collector. Typical CFA amount and water volume were 20 g and 100 ml, respectively.

The flotation efficiency index commonly used in mineral processing was employed as an efficiency criterion:

$$E = [(\varepsilon - \gamma) / (100 - \alpha)] \times \cdot 100\% \quad (1)$$

where ε is the percentage of carbon/sulphur extraction from the non-magnetic fraction; γ is the amount of the carbon/sulphur concentrate relative to the initial mass of the non-magnetic fraction; α is the carbon/sulphur content in the CFA non-magnetic fraction.

Leaching of coal fly ash by hydrochloric acid to extract aluminum was carried out in a laboratory autoclave using stainless steel capsules with 50 ml Teflon inserts (Deschem, China). The autoclave temperature was governed by a PID controller with ±1°C accuracy. The heating time from room temperature up to a particular temperature was approximately 1 hour. The exposure times at different temperatures (160, 170, 180, 190, 200°C) were 1, 2, 3, 4 hours, respectively. The HCl concentration was 345 g/l (30 %); the ratio S:L = 1:5.

3. Results and Discussion

Figure 1 showed the XRD results of the initial CFA (TPS-4, Omsk, Russia), and the magnetic & non-magnetic fractions after the wet magnetic separation. It can be seen that the initial CFA consists mainly of mullite, quartz and magnetite. In general, the mullite phase is a non-stoichiometric solid solution between SiO_2 and Al_2O_3 with an approximate formula $Al_6Si_2O_{13}$. The quartz and magnetite phases have stoichiometric formulae SiO_2 and Fe_3O_4, respectively; however, while quartz is a SiO_2 polymorph, magnetite generally represents a solid solution between FeO and Fe_2O_3 with a spinel-type structure that can accommodate other atoms like Ca, Mg and Mn.

The effect of the autoclave temperature and exposure time on the aluminum extraction efficiency was investigated for the autoclave hydrochloric acid leaching process. The Al extraction efficiency can reach ~95 % at the following optimal process parameters: T = 200 °C, C_{HCl} = 345 g/l, τ = 3 h, S:L = 1:5. The activation energy of the leaching process determined from the experimental data was 56.5 kJ/mol, which means that the process limiting stage is probably surface chemical reaction.

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6. References

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