Conference Paper

Alkali Fusion-Leaching Method for Comprehensive Processing of Fly Ash

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Abstract

Fly ash, composed of mullite, hematite, amorphous silica and quartz, is a promising source for the recovery of alumina and silica. Desilication with help of NaOH and alkali fusion-leaching method and utilization of alumina and silica in the fly ash for preparation of sodalite and silica white were explored in this research. The samples were characterized by using wet chemical analysis and X-ray diffraction. The optimal extraction of SiO_2 from Reftinskaya power plant fly ash was 46.2% with leaching at 95°C for 3 h. Sodalite was synthesized at 200°C for 1 h followed water leaching at 95°C for 1 h. Silica white with specific surface area 180-220 m2/g was prepared by carbonation of the Na_2SiO_3 solution at 40°C for 90-120 min. The as-prepared silica has a purity of 98,8%. The proposed method is suitable for the comprehensive utilization of the fly ash.

Keywords: fly ash, alkali fusion, desilication, sodalite, silica white, carbonization, comprehensive utilization

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1. Introduction

The combustion of coal in thermal power plants annually produces a large quantity of fly ash and slag waste, which are anthropogenic source of environmental pollution. The damage caused by fly ash to the environment is well known. A large part of the methods of disposal of ash and slag waste eventually leads to their proliferation on the exposed areas of the territory. The steady accumulation and improperly storage of fly ash leads to an increase of the occupied area, soil degradation and hazards for people and the environment.

Meanwhile, ash and slag waste contains a large amount of valuable components and their recycling can be economically and environmentally beneficial alternative to disposal. In recent years, especially in China, has made great efforts to develop effective methods for the processing of fly ash [1–4]. For example, in this country, about 20% of the generated fly ash being used in concrete production. Other areas of utilization include: land reclamation, ceramic industry, production of catalysts and

Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	K_2O	CaO	TiO_2	Fe_2O_3	LOI	S	C
0,75	0,51	23,4	65,0	0,41	0,59	1,85	1,170	4,85	1,4	0,25	1,5

Table 1: Chemical composition of the fly ash (wt.%).

carriers of catalysts, sorbents, deep separation of useful components, production of zeolites and extraction of valuable metals.

Experience from numerous studies on processing of fly ash shows that the most difficult step is the opening of the mineral called mullite (it can form several stoichiometric forms from $Al_6Si_2O_{13}$ to Al_4SiO_8), since it is not soluble at atmospheric pressure either in acids or in alkalis even at high concentrations. Therefore, in order to open the mullite its activation by sintering at temperatures above 800-1000°C or leaching under high pressure in autoclaves at temperatures above 250°C and the concentration of caustic alkali of over 400 g/L is proposed.

We have conducted earlier research on the processing of aluminum raw materials [5, 6] showed that the fusing aluminum containing minerals with alkali eliminates the need of autoclave or high-temperature processing. To explore the possibility of applying this method for the processing of fly ash studies have been conducted on the fusing of pre-desilicated fly ash with caustic alkali at different temperatures from 100 to 300°C. Preliminary desilication in hot caustic liquor allows to reduce material and energy costs due to the enrichment of raw material for alumina, as it helps to extract amorphous silica-containing component.

2. Experimental

Fly ash, derived from Reftinskaya power plant, Sverdlovsk region, Russia, was used as the raw material. The chemical composition of the fly ash measured by wet chemical analysis is listed in Table 1. It contains 23.4% Al_2O_3 and 65.0% SiO_2 as the major valuable components. The X-ray diffraction pattern of the fly ash is shown in Figure 1. Other chemicals used in this research were analytical grade.

The fly ash can be desilicated in NaOH solution through alkali leaching treatment [7, 8]. About 1/2 silica in the fly ash was leached out and transformed into the Na_2SiO_3 solution. The solid product with higher Al/Si mole ratio was obtained.

The Na_2SiO_3 solution was reacted with CO_2 gas to produce silica white and Na_2CO_3 solution. The Na_2CO_3 solution was reacted with lime milk to produce calcium carbonate that is a material for concrete or alumina production by sintering method and NaOH solution which can be used for fly ash desilication.

The solid product with higher Al/Si ratio, mixing with sodium hydroxide was fused at 100-300°C for 2 h. The fused clinker was dissolved in water to produce the Na_2SiO_3

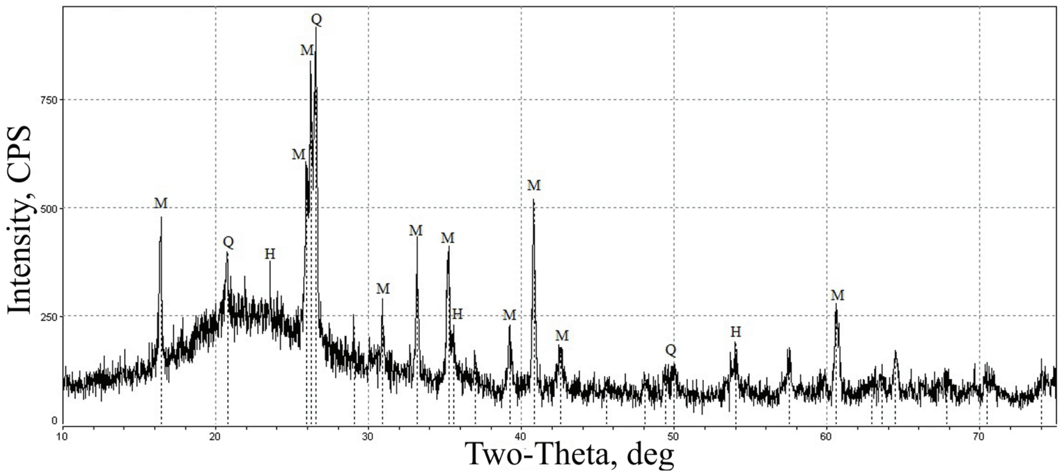

Figure 1: XRD pattern of the fly ash (M, mullite; Q, quartz; H, hematite).

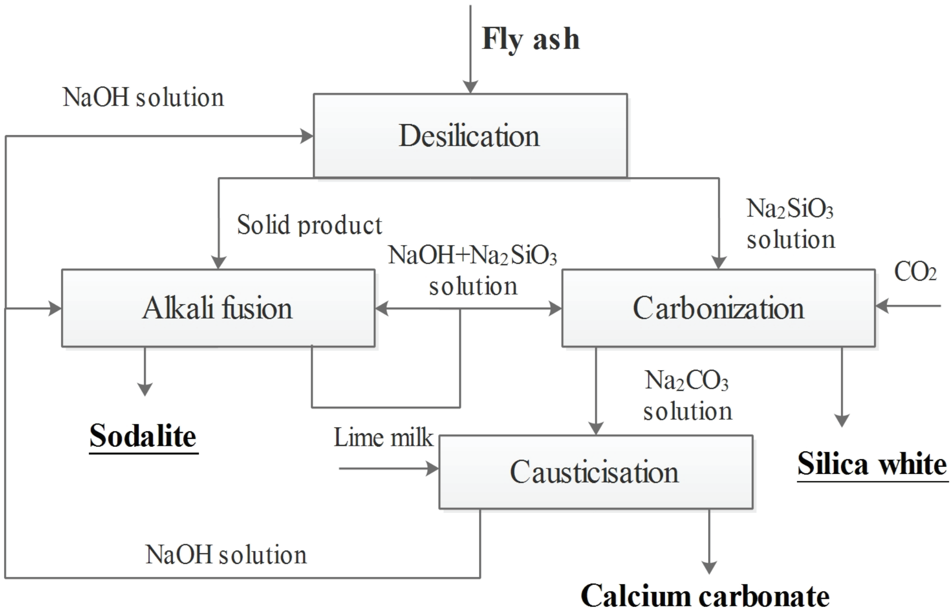

Figure 2: Flowchart for the processing of the fly ash and its comprehensive utilization.

solution and the solid product sodalite. Sodalite is a product which is used as sorbent or raw material for alumina production by sintering method. The whole processing flowchart for the processing of the fly ash and its comprehensive utilization is shown in Figure 2.

3. Results and Discussion

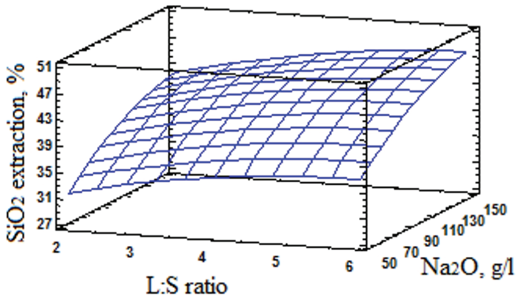

Figure 3: Estimated response surface of multifactor experiment at 3h of leaching time and 95°C.

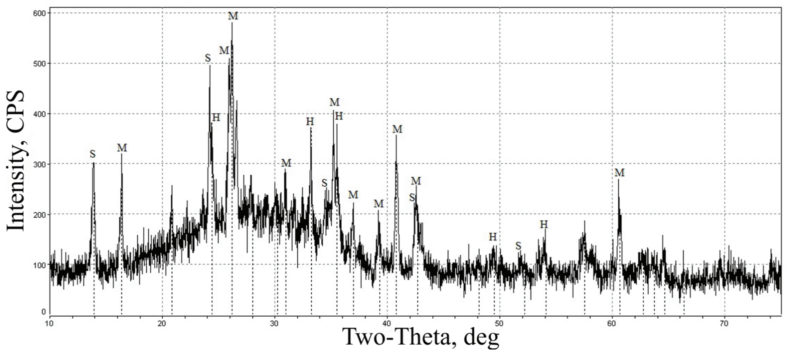

Figure 4: XRD pattern of the solid product obtained after alkali fusing at 115°C (M, mullite; H, hematite; S, sodalite).

3.1. SiO_2 Leaching from the Fly Ash

It is seen (Table 1) that the studied ash rich in silica (65%), and the content of Al_2O_3 is only 23.4 per cent. This indicates that almost all the alumina is contained in the mullite and an amorphous phase is represented mainly by silica. This fact allows to assume that at the initial stage we can try to enrich the ash for alumina and selectively removing a portion of silica in an alkaline solution using prior desilication by leaching at atmospheric pressure.

The temperature, L:S ration, Na_2O concentration in leaching solution and time of the process has a significant effect on the results of the fly ash desilication process. The temperature of atmospheric leaching was taken as maximum for our geographical conditions. In the result of multifactor experiment (Fig. 3) it was found that under optimal parameters (temperature = 95°C, leaching time = 3 h, L:S ratio = 1:4, Na_2O = 130 g/L) maximal extraction of SiO_2 in the solution was 46.2%. The solution may further be used to produce silica white, calcium silicate and liquid glass. Desilicated fly ash after washing from the silicate solution can be used for sodalite synthesis.

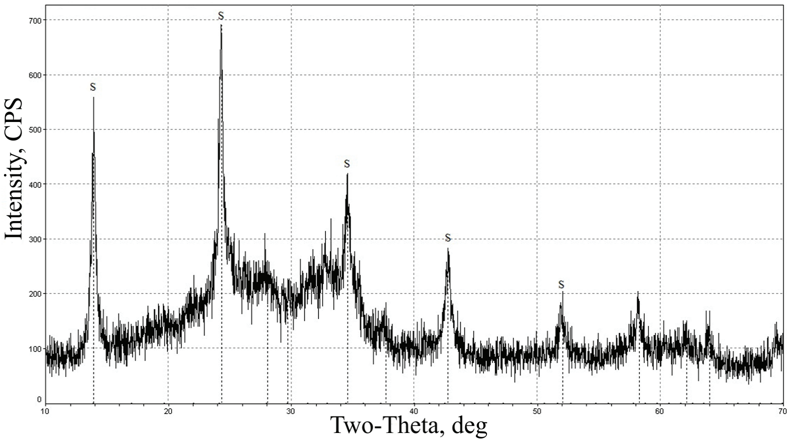

Figure 5: XRD pattern of the solid product obtained after alkali fusing at 200°C (S, sodalite).

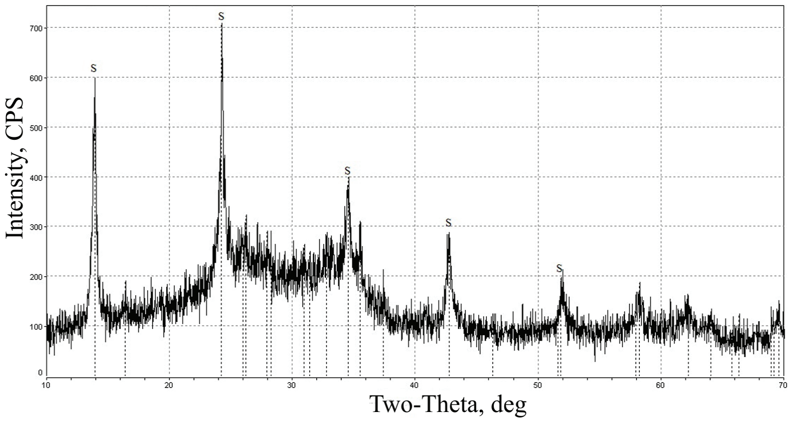

Figure 6: XRD pattern of the solid product obtained after fusing at 300°C (S, sodalite).

3.2. Preparation of Sodalite

The solid product obtained after desilication of the fly ash was reacted with NaOH at alkali/solid product mass ratio = 1.5 at different temperatures (115, 200 and 300°C) for 1 hour. The fused clinker was leached in water at 95°C for 1 hour to produce the NaOH-Na_2SiO_3 solution and the solid product which father was characterized by XRD analysis (Figure 4, Figure 5 and Figure 6).

It is obvious that already at 115°C, the peaks intensity of mullite begins to decrease and a new phase of sodalite appears. At 200°C the formation of sodalite is completely finished, i.e., all the mullite during solid-state reactions is converted into sodalite, which is very soluble even in weak acids and can be used to produce alumina by sintering. At 300°C is observed the same pattern as that at 200°C. Thus, there is a possibility of opening of mullite in the fly ash under milder conditions than currently existing technologies.

Samples	Reaction factors			Specific surface area of the product (BET), m^2/g
	Time, min	Na2O concentration, g/L	Temperature, °C	
1	50	126,5	40	329
2	70	126,5	40	293
3	90	126,5	40	226
4	120	126,5	40	180
5	70	126,5	25	464
6	70	126,5	60	475

Table 2: Experimental conditions and results for the carbonization of the Na_2SiO_3 solution.

Na_2O	MgO	Al_2O_3	SiO_2	CaO	Fe_2O_3	Total
0.61	0.01	0.05	98.84	0.2	0.06	99.8

Table 3: Chemical composition of the optimal silica white, (wt%).

3.3. Preparation of Silica White

One of the most promising products that can be obtained from the Na_2SiO_3 solution is a silica white which is widely used as filler for rubber and various sorbents.

In the solution according to the research after desilication of Reftinskaya power plant fly ash is extracted to 50% of the silica. The result is a Na_2SiO_3 solution. The composition of the solution obtained under optimum conditions has the following form: Na_2O = 126,5 g/L and SiO_2 = 87,73 g/L.

To determine the optimal parameters for extraction of the silica white, experiments were conducted using carbon dioxide as a neutralizing agent. The main variable parameters in the experiments were: temperature, concentration of the Na_2SiO_3 solution, and the duration of the carbonization (Table 2). The final pH in the all experiments was equal to 9.7-9.8.

Thus, the results of Table 2 shows that the optimal parameters for producing white silica from the Na2SiO3 solution is the temperature at 40°C and process duration 90-120 min with a final pH of 9.7 units, thus promoting the formation of silica with a specific surface area of 180-220 m^2/g. Table 3 presents the results of the chemical analysis of precipitated silica.

4. Summary

Preliminary leaching to extract silica, alkali fusion-leaching method and silica white deposition were explored using fly ash as the raw material. The optimal extraction of SiO_2 from Reftinskaya power plant fly ash was 46.2% with temperature = 95°C, leaching time = 3 h, L:S ratio = 1:4, Na_2O = 130 g/l. Sodalite was synthesized after alkali fusion-leaching method of predesilicated fly ash at 200°C for 1 h followed water leaching at 95°C for 1 h. Silica white with specific surface area 180-220 m2/g was prepared by carbonation of the Na_2SiO_3 solution at 40°C for 90-120 min. The as-prepared silica is in purity of 98,8%.

It can be concluded from the experimental results that the proposed method is efficient and energy-saving. Moreover, it appears to be suitable for the comprehensive utilization of the fly ash.

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