

The use of a modified fly ash as an adsorbent for lead

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Abstract

Coal fly ash was modified by hydrothermal treatment with NaOH solutions of varying concentrations. During the modification the zeolites, Na-P1 and hydroxysodalite, were synthesised. Increasing base concentration led to hydroxysodalite being the preferred product. Elemental analysis revealed that the modification treatment preferentially leached Si from the fly ash at low base concentrations. Elevated base concentrations, however, resulted in Al also being leached as well as the inclusion of Na in the zeolitic product. The modification activated the fly ash significantly with respect to specific surface area (SSA) and cation exchange capacity (CEC). SSA increased from $1.0 \text{ m}^2\cdot\text{g}^{-1}$ to a maximum of $62.7 \text{ m}^2\cdot\text{g}^{-1}$ while CEC rose from $25 \text{ mmol}/100 \text{ g}$ to a maximum of $300 \text{ mmol}/100 \text{ g}$. Metal sorption studies were performed with lead as the metal of choice. Adsorption experiments at $\text{pH} = 5$, revealed that all modified ash samples adsorbed significantly more Pb than the raw ash. The best adsorption was obtained for ash, modified with 3 M NaOH . 3 M NaOH -modified ash also proved the most effective sorbent when adsorption was determined as a function of pH.

Introduction

Large quantities of fly ash are produced during the combustion of coal in the production of electricity. Most of this ash is used in low level applications such as landfill. In South Africa, where high-ash content coal is used in power generation, $24 \times 10^6 \text{ t}$ of fly ash were produced in 1997. Only 5% of this ash was sold for reuse (Eskom, 1997). Increasing concerns about the environmental consequences of such disposal have led to investigations into other possible utilisation avenues. Furthermore, the conversion of a low-cost waste product into a higher level product would make the environmentally-friendly disposal of the remaining unused ash far more economically viable.

Fly ash is composed primarily of aluminosilicate glass, mullite ($\text{Al}_6\text{Si}_2\text{O}_{13}$) and quartz (SiO_2). These materials provide a ready source of Al and Si, which is necessary for the synthesis of zeolites. Low Si/Al ratio zeolites have been shown to be excellent sorbents for the adsorption of transition metals because of their high cation exchange capacities (CEC) and large pore volumes (Höller and Wirsching, 1985; Flanigen, 1991; Querol et al., 1997b). Specific applications include the use of zeolites in nuclear waste processing (McFarlane et al., 1997). Thus, the conversion of fly ash into zeolites of this type might prove to be effective for removing such metals from wastewater streams cheaply and effectively owing to the low cost of the ash starting material.

Several authors have reported the conversion of a sizeable fraction of fly ash into zeolites by the treatment of ash with concentrated NaOH solutions at elevated temperatures and pressures (Höller and Wirsching, 1985; Henmi, 1987; Mondragon et al., 1990; Catalfano et al., 1993; Lin and His, 1995; Park and Choi, 1995; Singer and Bergaut, 1995; Amrhein et al., 1996; Querol et al., 1997a; Garde et al., 1999). Recently more sophisticated treatments including the use of microwave radiation (Querol et al., 1997b) and fusion with NaOH followed by hydrothermal treatment have been reported (Shigemoto et al., 1993; Chang and Shih, 1998).

The objective of this study was to investigate the conversion of fly ash into zeolites. The specific aims were to analyse the zeolitic products for zeolite type and surface characteristics such as specific surface area (SSA) and CEC. Furthermore the efficiency of the

various products as far as the removing of lead was concerned, was also to be determined.

Most of the literature, detailing the synthesis of zeolites from fly ash, deals only with the actual synthesis and does not consider the applications thereof. Exceptions are the work of Singer and Bergaut (1995) and Lin and Hsi (1995) who looked at metal adsorption. It was thus an aim of the present work to study the adsorption of metals from solution.

Lead was chosen as the adsorbate because of its known toxicity. It has been classified as a list II material by the European Community Directive on Dangerous Substances (Council of European Communities, 1976). The effectiveness of the zeolites produced from fly ash for the removal of lead from aqueous solution would be monitored and compared to other adsorbents such as natural zeolites (Kesraoul-Oukl et al., 1993) and sphagnum moss peat (Ho et al., 1996). The ash could hopefully prove to be a low-cost alternative to existing technologies, ideal for a developing country such as South Africa.

Experimental

Zeolite preparation

Samples of fly ash were obtained from Ash Resources (Pty) Ltd. These were a beneficiated high-silica ash known as Plasfill 5. The zeolite products were produced using a modification of the procedure of Henmi (1987). 100 g of fly ash was added to $1\ 000 \text{ mL}$ of NaOH solution. The NaOH solutions varied in concentration from 1 M to 8 M . The slurries were then refluxed with overhead stirring under atmospheric pressure for 21 h . The product was collected by filtration, washed with hot distilled water and then dried at 90°C for 48 h .

X-ray diffraction

The modified ash samples were shaken in hot distilled water to remove any possible remaining NaOH prior to X-ray diffraction (XRD) studies. After drying, powder XRD patterns were obtained using a Philips PW 840 X-ray diffractometer, using CuK α radiation (1.541 \AA), set at 40 kV and 25 mA . The files shown in the Joint Committee on Powder Diffraction Standards (JCPDS) were used to identify various crystalline zeolite phases and other minerals present.

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