The synthesis of zeolite-P, Linde Type A, and hydroxysodalite zeolites from paper sludge ash at low temperature (80 °C): Optimal ash-leaching condition for zeolite synthesis

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ABSTRACT

Typically, the ash from incineration of paper sludge contains a high percentage of Ca in the form of anorthite (CaAl2Si2O8) and gehlenite (Ca2Al2SiO7). The Ca in the sludge originates from calcite that is included in paper as fillers. We applied acid leaching with HCl on the ash to reduce its Ca content. Zeolite was then synthesized from the leached ash through reaction with 2.5 M NaOH solution at 80 °C for 24 hours. The fraction of Ca and Al extracted from the ash correlates with the pH of the leachant. We determined the leachant pH (after 24 hours of leaching) associated with the Ca: Al:Si ratio in the leached ash that provided optimal production of zeolites with high cation-exchange capacity. During acid leaching, gehlenite dissolved out at higher pH than anorthite. In the case of pH > 5, both gehlenite and anorthite remained in the ash, and hydroxysodalite and LTA (Linde Type A) were synthesized in the product. In the case of pH = 1−5 in the leachant, gehlenite dissolved out but anorthite remained in the ash, and LTA and Na-P1 (zeolite-P) were produced. In the case of pH < 1, both gehlenite and anorthite dissolved out, and only Na-P1 was produced. The cation-exchange capacities of the products that contained hydroxysodalite, LTA, and Na-P1 were approximately 130, 200, and 120 cmol/kg, respectively. We conclude that acid leaching of paper sludge ash controls which of the three zeolite phases form, and that LTA and Na-P1 exhibit a higher cation-exchange capacity than hydroxysodalite. The most efficient production of zeolites with high cation-exchange capacity (about 220 cmol/kg) is obtained after leaching the sludge ash in solutions of around pH = 3. At this low pH, gehlenite has already dissolved out of the sludge ash; half the Ca content of the ash has been leached out, Si has not yet been leached, but Al has begun to be leached. After ash zeolitization, LTA coexists with Na-P1 in the product.

INTRODUCTION

During the manufacture of recycled paper, paper sludge is discharged as an industrial waste. The amount of sludge increases annually. To reduce its volume, this sludge is incinerated, which produces paper sludge ash. At present, although paper sludge ash is used as an additive to cement and as a material for artificial aggregates (Singh and Garg 1999; Kikuchi 2001), a large amount of the ash is disposed of by dumping. It has become more difficult to secure a sufficient amount of land for the waste disposal. It is, therefore, essential to develop new techniques of ash utilization for further recycling.

The conversion of the ash into zeolite has been investigated recently (Henmi 1989; Ishimoto and Yasuda 1997; Mun and Ahn 2001). In the conversion, aluminosilicate, which is a main component of the ash, is transformed into zeolites by reaction with alkalies. Because of their unique pore structures and ion-exchange properties, zeolites (including those synthesized from paper sludge ash) are applied to such tasks as water purification and soil improvement (Ishimoto et al. 2000, 2003).

Due to calcite included in paper as fillers, however, recent ash contains a high percentage of calcium in the form of anorthite and gehlenite. It is reported that ash with a high percentage of Ca can be converted into zeolites, but that the product has a low cation-exchange capacity (Catalfamo et al. 1994). Moreover, it also has been reported that Ca compounds in the ash prevent the separation of the production slurry into solid and liquid during the filtration and sedimentation process (Murayama et al. 2000). The reduction of Ca content, therefore, is an important issue for the production of desirable zeolite materials.

To control the Ca content for zeolite synthesis at low temperature (80 °C), we propose a Ca reduction process through pre-treatment of the paper-sludge ash by acid leaching. Many researchers have reported on production of zeolites from coal fly ash by means of different hydrothermal activation methods, such as classic alkali activation (Querol et al. 2001), alkali fusion (Shigemoto et al. 1993), and two-stage synthesis (Holman et al. 1999). Little information is available, however, on pre-treatment of the raw materials (ashes) by acid leaching. Additionally, more information is needed about zeolite synthesis at low temperature (<100 °C), which is less expensive than the production of zeolites under hydrothermal conditions (>100 °C). We have studied the leaching conditions of paper-sludge ash, the synthesis of zeolites from this ash at low temperature, and the cation-exchange capacity of the resultant product with the objectives of (1) demonstrating the relationship between starting materials for synthesis and the resulting zeolite phases, and (2) establishing the best ash-treatment conditions for production of zeolites.