SYNTHESIS AND CHARACTERIZATION OF HIGH SILICA ZEOLITES FROM COAL FLY ASH (CFA): TWO CASES OF ZEOLITE SYNTHESES FROM THE SAME WASTE MATERIAL.

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***Abstract*— Zeolites H/β and H/ZSM-11 were synthesized by the crystallization hydrothermal method (CH), using coal fly ash (CFA). Tetraethylammonium hydroxide (TEAOH), tetrapropylammonium bromide (TPAB) and tetrapropylammonium hydroxide (TPAOH) were used as structure-directing agents (SDA). The optimum conditions used in the experimentation were 140 °C and NaOH/CFA ratio=1.2 (wt/wt), during the crystallization hydrothermal method (CH), and microwave-assisted heating (MH) was used in some experiments. The structure and crystallinity of the synthesized materials were analyzed by X-ray diffraction, where the formation of the β and ZSM-11 zeolites was confirmed. The morphology of the obtained zeolites was observed by the scanning electron microscope technique. It was observed spheroidal particles for the ZSM-11 zeolite and ellipsoidal particles for the β zeolite, respectively. Both zeolites were ion exchange treated, in order to obtain the protonated form. Finally, the thermal stability and the loss of weight of the modified β zeolite were determined by using the termogravimetric and differential thermal analysis techniques. The modified zeolites were stable in the range of most gas chemical reactions (300-500 °C).**

***Keywords* — Zeolite H/β; zeolite H/ZSM-11; crystallization hydrothermal method; coal fly ash, structure-directing agents.**

I. INTRODUCTION

The amount of coal fly ash (CFA) generated by coal-based thermal power plants has been increased at an alarming rate throughout the world. The disposal of such a big quantity of fly ash has become an important topic. Thus, several new approaches have been adopted to utilize fly ashes not only to reduce the cost of the disposal but also to minimize environmental impact. Those elements other than calcium detected in the fly ashes exhibit a broad range of toxic effects to humans, terrestrial and aquatic life and plants. A number of these elements have the potential to bioaccumulate, including arsenic, chromium, lead, mercury, nickel and zinc (Bridgen and Santillo, 2002). Particulate pollution is implicated in the worsening of respiratory illnesses such as asthma, and in the increasing of premature mortality from respiratory and heart diseases (Bridgen and Santillo, 2002).

Recently, intensive efforts were made to promote the recycling of fly ash through zeolitization. Zeolites are microporous crystalline aluminosilicates with three-dimensional framework structures. Due to the high thermal and good dimensional stability, they have attracted a particular attention as catalysts in acid-mediated reactions (Maxwell and Stork, 1991; Corma, 1995). By virtue of their compositional and structural peculiarities, zeolites also have many other applications in various ion-exchange, adsorption, separation, and purification processes (Breck, 1974).

Several groups (Meshram *et al.*, 2000; Rayalu *et al.*, 2001; Miyake *et al.*, 2002; Querol *et al.*, 2002; Park *et al.*, 2000; Shigemoto *et* al., 1995; Singer and Berkgaut, 1995) around the world have studied the conversion of fly ash into various types of zeolites such as Y, X, A, P, F, KM, etc. However, there are no reports on the synthesis of commercially important high silica zeolite with three-dimensional 12-membered ring channels using fly ash as a source material. Using conventional source materials, high silica β (*BEA) zeolite, with a three-dimensional 12-membered ring channels, was synthesized for the first time in early 1967 (Wadlinger *et al.*, 1967), and its structure was discerned late in 1988 (Newsam *et al.*, 1988). Crystallization of zeolite β (*BEA) draws much attention because of its unique characteristics, in particular, its acidity and potential for acid catalysis (Kiricsi *et al.*, 1994; Jones *et al.*, 1999, Corma *et al.*, 2002; Prasad Rao *et al.*, 1998; Matsukata *et al.*, 2002).

Several factors have influence on the zeolitic formation when fly ash is used as a source of Al2O3 and SiO_2; Höller and Wirsching (1985), investigated zeolite formation after carrying out an alkaline activation of fly ash as a function of temperature, solution composition,