



Original Research Paper

Conversion of high silicon fly ash to Na-P1 zeolite: Alkaline fusion followed by hydrothermal crystallization

H. Kazemian^{a,b,*}, Z. Naghdali^a, T. Ghaffari Kashani^a, F. Farhadi^a

^a SPAG Zeolite R&D Group, Technology Incubation Centre, Science and Technology Park of Tehran University, Tehran, Iran

^b Department of Chemical and Biochemical Engineering, Faculty of Engineering, The University of Western Ontario, London, Ontario, Canada N6A 5B9

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ABSTRACT

In this research, we converted high silicon fly ash to a high ion-exchange capacity zeolite using a two stages conversion process. Alkaline fusion was applied to collapse the fly ash crystalline phases and release Si content. Then Si/Al ratio of the synthesis sol adjusted with appropriate amount of industrial grade materials. A synthesis solution with the molar ratio of 2.2 SiO₂:Al₂O₃:5.28 Na₂O:106 H₂O was hydrothermally crystallized to Na-P1 zeolite at 120 °C for 4 h. The as-synthesized zeolite characterized by means of X-ray diffraction, infrared spectroscopy, scanning electron microscopy and thermal analysis. Cation exchange capacity of the zeolite was determined using ammonium acetate method. The zeolitization remarkably improved the cation exchange capacity of the final product (e.g. 3.23 meq/g in comparison to the raw fly ash ~0.005–0.02 meq/g).

Due to the high CEC and sufficient whiteness of the final product, we suggest that the as-synthesized zeolitic powder is a potential candidate as a detergent builder.

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1. Introduction

Most of the world's produced fly ashes (FA) are solid wastes of coal combustion in the electricity generation processes. However, some other industries such as steel and ferrosilice industries also encounter hazards of this solid waste. Different industries produce FA with various chemical and mineralogical compositions. Disposal of fly ash that is a very fine submicron powder, has become an increasing economical and environmental burden. So far, the only realized utilization of FA is by low technology applications such as in construction materials and cements. Nevertheless, more than 500 million tones/year of fly ash is produced globally, of which only 20% effectively used [1]. This implies that finding techno-economically viable methods to treatment FA in a safe manner is an urgent need of the industries. In the recent decades, many efforts are being made to explore other potential applications such as using in cements and concretes, roadbed, gas absorbents and fertilizers. One possible new application for FA is the synthesis of value added zeolitic materials. Many researchers have investigated fly ash as starting material for hydrothermal synthesis of various zeolites. Fly ash has been used for direct hydrothermal synthesis [2–5],

hydrothermal synthesis from alkali-fused fly ash [6], and hydrothermal synthesis of zeolite using extracted silica [7]. Possible zeolitization of FA is a process analogous to the formation of natural zeolites of the volcanic bedrocks. Both volcanic ash and FA are fine-grained and contain large amounts of aluminates and silicates glasses [8]. In natural conditions, this glass fraction might be converted into zeolites by the influence of percolating hot groundwater, while the zeolites may subsequently be converted into other minerals.

This similarity of fly ash to some of volcanic minerals stimulated the idea of using fly ash solid wastes for conversion into the value added synthetic zeolites [9].

Since the results of preliminary studies on this issue were promising, more researchers have been dedicated their research life on this issue. As results of these activities, many articles related to manufacturing of various zeolitic materials from FA were published and many patents were registered. Most of the conversion procedures are based on alkaline hydrothermal crystallization, in which by applying different temperatures, pressures and crystallization duration times on various chemical compositions, varieties of zeolitic materials have produced with different potential applications [3].

While this conversion process may take tens to thousands of years in the nature, in a laboratory, the reaction duration can be reduced remarkably to some days or even to hours [10]. This implies that using of fly ash as a cheap and abundant source of Si and Al for making artificial zeolites can be considered as a promising method

* Corresponding author. Address: Department of Chemical and Biochemical Engineering, Faculty of Engineering, The University of Western Ontario, London, Ontario, Canada N6A 5B9, Tel.: +1 519 694 9453.

E-mail addresses: hossein.kazemian@uwo.ca, hosseinkazemian@gmail.com (H. Kazemian).

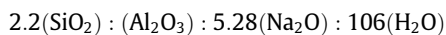
to treatment all type of FA based solid wastes. Usually, this method is an alkaline hydrothermal crystallization process, in which the fly ash is mixed with an alkali hydroxide such as NaOH and fuse at an elevated temperature to release the silica and alumina content in the form of water soluble sodium silicate and sodium aluminate. Then the chemical composition of resultant solution will be adjusted by adding sufficient amounts of Al and/or Si, etc. according to the desirable zeolitic phase. Such treatment processes can lead to the formation of various types of zeolite with different Si/Al ratios such as the zeolites P, A, X types, etc. The type of zeolite formed is a function of the temperature, pressure, chemical composition of synthesis gel/sol (e.g. concentrations of Al, Si and cation and type of the used salts), aging time, pH, and crystallization duration, etc. [11].

Following to the previous research of our group on the synthesis of various zeolites of NaA and FAU from high silicon fly ash [12–14], this paper reports the results of a new research work on the successful hydrothermal conversion of a high silicon fly ash from Iran Ferrosilice Company into zeolite P powders. Details of hydrothermal treatment of the fused fly ash, characterization results as well as ion-exchange capability of the converted product in comparison to the raw FA and some of other zeolitic materials will be compared and discussed.

2. Experimental

All of the chemicals used in this research were domestically produced industrial grade materials. High silicon fly ash was provided by Iran Ferrosilice Company, Semnan, Iran.

For fusion step, sufficient amount of the FA was mixed with appropriate amounts of NaOH in a FA/NaOH weight ratio of 1/1.2. The resultant mixture then fused in a muffle furnace at 550 °C for 1 h in air atmosphere. The heating rate of 5 °C/min was applied to elevate the temperature from ambient to 550 °C. The melted materials was then cooled to room temperature with cooling rate of 20 °C/min, well milled to fine particles using an agate mortar. Then 50 ml of 0.2 M solution of NaOH added to the mixture. The resultant slurry was then agitated vigorously by a mechanical stirrer for several hours at 70 °C. The resultant silica solution (Solution A) was cooled to room temperature and slowly added to the sodium aluminate solution. The aluminate solution was prepared by adding sufficient amounts of sodium aluminate to a NaOH solution (Solution B). Solution A was then added to solution B in several steps and the mixture was stirred to achieve a homogenous gel. Chemical composition of the resultant synthesis gel was as following:



To crystallize the zeolite, the prepared synthesis gel was transferred into a PTFE lined stainless steel reactor, sealed and heated in an electrical oven equipped with an “end over end” shaker, at different temperature of 100 °C and 120 °C under autogenously produced pressure with and without shaking. After 4 h of crystallization duration, in order to assure that the reaction is stopped, the reactor was quenched in cold water. The solid product then separated from supernatant by filtration using a filter paper, washed with excess amounts of de-ionized water to remove excess amount of sodium hydroxide and other none reacted materials and then dried overnight at 60 °C in an electrical oven. Chemical compositions of the utilized FA and the produced zeolite P was measured by X-ray fluorescence technique using an Oxford instrument (XRF; ED2000). The crystallinity of the final zeolitic product was evaluated by mean of X-ray diffraction technique using a Philips1130/90 instrument and furrier transform infra-red spectroscopy (FT-IR; Vector 22-Bruker). Morphology of

the crystalline zeolitic product was examined by mean of scanning electron microscopy using a Philips instrument (Model; XL30). Further characterization of the product was carried out by investigation of its thermal behavior by mean of thermal analyses techniques using a simultaneous thermal analyzer (STA-Rheometric Scientific). In order to evaluate the ion-exchange property of the as-synthesized zeolite, its cation exchange capacity (CEC) was measured by mean of ammonium acetate saturation method [15–16]. This method is based on saturation of all ion-exchange sites in ion-exchanger with ammonium ion at equilibrium condition. The time required for saturation (e.g. achieving to equilibrium) depends to the ion-exchange reaction kinetic.

In this research, due to relatively fast reaction kinetic of the as-synthetic zeolite P, the ammonium solution and the zeolite powders were in contact for 24 h in order to make sure that exchange reaction reaches to equilibrium. The zeolite to ammonium solution ratio was kept at 1/50. The Kjeldahl ammonia distillation technique was used to determine the ammonium concentration in the zeolitic phase. The CEC was calculated accordingly.

3. Results and discussion

The sodium hydroxide present in the fusion mixture, acts as an activator during fusion to form soluble silicate salt. Fusion of NaOH with fly ash produces sodium silicate and amorphous aluminosilicate, which can be easily dissolved in aqueous solution. The X-ray diffraction patterns of the used high silicon FA and the as-synthesized zeolitic material are shown in Fig. 1. The XRD pattern of zeolitized FA, exhibits several sharp diffraction peaks, which are different from those present in the untreated FA.

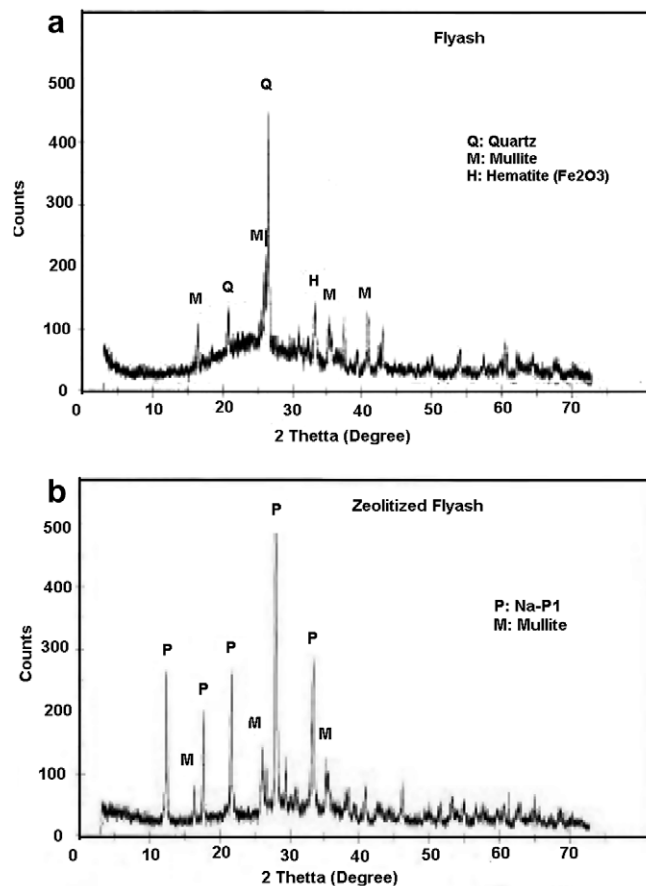


Fig. 1. X-ray diffraction patterns of the raw fly ash and as-synthesized zeolite Na-P1; Q = quartz, M = mullite, H = iron oxides, P = Na-P1 zeolite.

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