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Original Research Paper

Effect of microwave irradiation on crystal growth of zeolitized coal fly ash with different solid/liquid ratios

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ABSTRACT

In the present work, coal fly ash (CFA) was converted to zeolite (CFAZP) experimentally at atmospheric pressure via a conventional hydrothermal heating for 6 h at low temperature (90 ± 3 °C) followed by microwave irradiation for 30 min. The synthesized products were characterized using XRD, TGA/DTA, SEM, PSD, BET, and cation-exchange capacity (CEC) techniques. The effect of microwave on the crystal growth of nucleated CFAZP at different solid/liquid ratios (suspended CFA mass to NaOH solution volume, g/mL) was studied. A three-variable, three level central composite statistical experimental design was applied to investigate the effect of the independent variables on the response function defined as the ratio of the characteristic peak intensity at 2θ: 28° of a sample to that of the same peak of a sample run for 24 h with conventional heating. The relative peak intensity of CFAZP as high as 97% was achieved under optimum experimental conditions with 1 M of NaOH concentration, 6 h of conventional heating followed by 30 min microwave irradiation with a solid/liquid ratio of 0.40 g/mL. Under constant microwave energy, higher solid/liquid ratios led to higher relative peak intensity of the product.

1. Introduction

Coal fly ash (CFA), as a by-product of coal fired power plants, is considered as one of the biggest industrial waste streams in the world [1,2]. About 50% of the world's electricity energy is produced by burning coal [3]. Treatment of CFA is a worldwide priority due to several reasons, including but not limited to its harmful effects on the environment. In addition, disposing this solid waste, in landfills is a financial liability for the coal burning power plants [4]. In order to overcome this problem, numerous investigations have been conducted on the zeolitization of CFA as a source of silicon and aluminum. CFAZP is an interesting synthesized zeolite with high cation exchange capacity (CEC) [5–11].

Successful conversion of CFA to CFAZP was conducted using a conventional hydrothermal process at relatively high temperatures up to 200 °C for 8–100 h [12–15]. The synthesized product had a relatively high cation exchange capacity and crystallinity [16–18]. The characteristics of zeolites such as purity and CEC were strongly influenced by the hydrothermal heating methods and the synthesis parameters such as the reaction time, temperature, etc. [19–22]. Conventional hydrothermal treatment is an energy-intensive and time-consuming process which is difficult to commercialize. Using microwave (MW) and ultrasound (UST) as alternative energy sources could lead to homogenous nucleation and uniform distribution of thermal energy and decrease the reaction time remarkably from days down to hours, which can lead to an economically viable zeolitization process [23–28]. Inductive energies can strongly affect the synthesis of zeolites by accelerating the crystallization process [29–36].

Querol et al. [37] synthesized CFAZP from CFA by conventional and microwave-assisted hydrothermal alkaline activation experiments. Synthesis yields obtained from both methods were similar, but the activation time needed was drastically reduced from 24 to 48 h to 30 min by using microwaves. All experiments were carried out using 0.5–5.0 M of NaOH, CFA mass to NaOH solution volume ratios of 0.05–0.1 g/ml and the highest microwave power available (1000 W) with temperature control.

Inada et al. [38] investigated the effect of microwave irradiation on the yield of zeolite from CFA. The NaOH concentration was 2 M and the solid/liquid ratio was fixed at 0.125 g/ml for safe irradiation of microwave. The microwave heating was performed by using a household oven (500 W) refurbished to equip the vessel with a condenser under the ambient atmosphere. CFAZP was formed after

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the conventional treatment at 100 °C, but no zeolitic product was obtained by microwave heating alone after 2 h. In the combined experiment, however, zeolitization was achieved when the microwave was applied for 15 min in the course of the conventional heating. The initial MW irradiation was more effective. This is due to the stimulated dissolution of SiO_2 and Al_2O_3 from CFA. On the other hand, the microwave irradiation in the middle to later stages of the process retarded the crystallization of zeolite. Especially, the microwave heating at 45–60 min into the process influenced the zeolitization negatively.

Fukui et al. [39] synthesized phillipsite from CFA with a solid to liquid ratio of 0.04 g/ml at controlled temperature 100 °C (variable power) over 6 h of conventional heating and microwave heating. Microwave heating reduced the crystallization induction time compared to the conventional heating, but it retarded the crystal growth rate slightly leading to smaller particle size of the synthesized phillipsite. They showed that the crystal structure and chemical composition of the product powder did not depend on the heating method [15]. To the best of our knowledge, utilizing MW energy after a period of conventional heating for the crystallization of CFAZP from CFA has not been reported. In this study, CFA was converted into CFAZP by means of microwave irradiation of CFA after hydrothermal treatment in an alkali medium. Furthermore, the effectiveness of the hydrothermal and microwave-assisted hydrothermal method is compared.

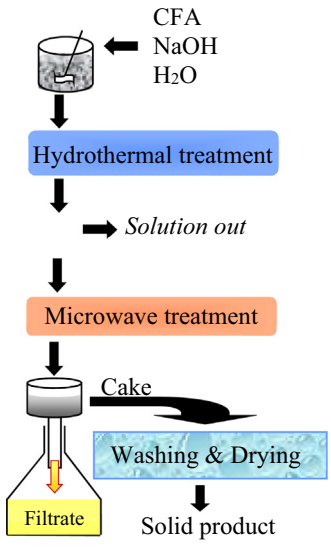
2. Materials and method

Coal fly ash, as a source of Al and Si, was supplied by a coal-fired power plant (OPG, Nanticoke) located in Ontario, Canada. Analytical grade sodium hydroxide (Alphachem, Canada) was used as received. Deionized water was used for the preparation of the solutions and washing.

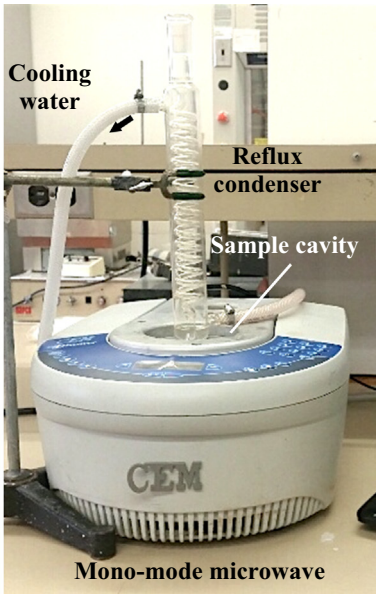
Two g of CFA and 0.8 g of sodium hydroxide granules were dissolved in 20 ml deionized water to form an alkaline slurry of 1 M (solid/liquid ratio of 0.10 g/ml). The mixture was subjected to conventional heating under atmospheric pressure at 90 °C for 6 h and stirred continuously using a heater stirrer (Thermo Scientific, China) at 170 rpm. The schematic of the synthesis procedure and microwave setup are illustrated in Fig. 1.

After hydrothermal treatment for 6 h in a 125-ml glass flask reactor in which Si and Al were extracted into the liquid, and achieving a desired solid/liquid ratio, a certain amount of the liquid phase was withdrawn by a syringe filter to adjust the solid/liquid ratio range. The latter was chosen in a way to allow the solids to form a wet bed for the safe irradiation of microwave and avoiding the hot spots issue. The residual mixture was poured into a cylindrical PTFE vessel (40 ml, 28 mm ID) attached to a reflux condenser. The suspension was subjected to microwave radiation for crystal growth at 105 °C for 15–30 min. Then, the filtered solid products were washed and dried overnight at room temperature. Microwave power was constant at 250 W during the experiments. The microwave treatment was performed in a self-adjusting microwave oven (single-mode microwave apparatus equipped with a magnetic stirrer, 2.45 GHz, CEM cooperation, Discover, USA), where the reaction temperature and power were monitored automatically.

Rigaku-MiniFlex powder diffractometer (Japan) was used to collect XRD diffraction pattern data of the synthesized samples using CuK_α (λ: 1.54059 Å) over the range of $5° < 2\theta < 40°$ with step width of 0.02°. The XRD patterns of the synthesized zeolite samples were compared with the characteristic peaks of CFAZP from the literature [40]. The peak intensities were calculated by the "peak fitting" algorithm in the MDI-Jade v 7.5 software. Intensities of the characteristic peaks were found for CFAZP main peaks at 2θ of 12.5°, 21.3°, and 28°. The relative peak intensity was defined as the ratio of the characteristic peak intensity at 2θ: 28° of a sample to that of the same peak of a sample run for 24 h with conventional heating. Crystal morphology of the produced zeolite was studied by scanning electron microscope (SEM) using a JSM 600F (Joel, Japan) operating at 10 keV. Thermal analysis was performed using a Mettler Toledo TGA/DTA 851e model (Switzerland) with version 6.1 Stare software by heating the samples from 25 °C to 600 °C with a heating rate of 10 °C/min under nitrogen purge of 40 mL/min. Master-sizer 2000 (Malvern Instruments Ltd., UK) was used for the particle size distribution (PSD) analysis. Brunauer-Emmett-Teller (BET) technique was used to measure the specific



(a) High Solid/Liquid (S/L) ratio procedure



(b) Microwave apparatus

Fig. 1. Schematic diagrams of experimental procedure and microwave set-up.