Conversion of Harmful Fly Ash Residue to Zeolites: Innovative Processes Focusing on Maximum Activation, Extraction, and Utilization of Aluminosilicate

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ABSTRACT: Reuse of the solid residue from coal fly ash alumina extraction (FAAE) by acid leaching is problematic. Conversion of this solid residue into aluminum-rich zeolite (13X) and silicon-rich zeolite (ZSM-5) was investigated in this research. The FAAE residue was activated by alkali roasting with Na₂CO₃ powder (110% mass fraction) at 890 °C for 60 min. Silicon and aluminum were mainly present as two mineral phases, Na₂SiO₃ and NaAlSiO₄, respectively, in the product obtained after roasting. The roasted product was dissolved in water (liquid/solid ratio of 2) after 20 min at 100 °C. The water-leaching liquor was investigated for total conversion to aluminosilicate zeolites without external aluminum or silicon addition. Hydrothermal synthesis of aluminum-rich zeolite 13X was successful after fine tuning of the conditions, although the filtrate had an unusually high SiO₂/Al₂O₃ molar ratio. Production of 13X consumed a large amount of aluminum, which increased the Si/Al ratio to a level suitable for synthesis of ZSM-5. The synthesis of ZSM-5 from the mother liquor of 13X was proved feasible. The FAAE residue was transformed into high-value zeolite products by nearly 100%. Additionally, the tail liquid of this process, mainly containing Na₂CO₃, was completely recycled. This process could be used to realize high-efficiency and high-value utilization of similar aluminosilicate solid wastes.

1. INTRODUCTION

Large-scale and efficient disposal of fly ash is problematic in China. To meet reduction targets for solid waste, the China Energy Group, which is the world’s largest coal and thermal power producer, has been investigating extraction of alumina from fly ash with a high alumina content at Zhungeer (Inner Mongolia, China).¹,² Leaching of this fly ash with hydrochloric acid produces metallurgical-grade alumina.³⁻⁵ By-products such as lithium, gallium, and other rare metals could also be recovered from the fly ash in the near future.⁶⁻⁹ However, the secondary solid waste created after alumina extraction from the fly ash is difficult to dispose of. According to relevant policies in China,¹⁰ alumina production plants that use fly ash as raw material should find uses for more than 96% of the waste. Insufficient use or disposal of the solid residue obtained after alumina extraction severely limits the development of alumina-rich fly ash as a resource. Thus, development of high-efficiency and high-value reuse technologies for the solid residue obtained after alumina extraction is of practical significance to relieve environmental pressures and promote industrial utilization of alumina-rich fly ash.

The solid residue obtained after alumina extraction is highly enriched in silicon (SiO₂ mass fraction of 60–80%, ash basis). Previous research on the solid residue obtained after alumina extraction has mainly focused on the production of silica-based materials such as autoclaved bricks, active calcium silicate, glass–ceramics, silicon carbide, and rubber fillers.¹¹⁻¹⁶
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The composition of the FAAE residue was SiO_2 78.7%, Al_2O_3 13.4%, and TiO_2 5.2% (Table 1). X-ray diffraction (XRD) analysis (Figure 1) indicated that the solid residue contained mullite (3Al_2O_3·SiO_2), quartz (SiO_2), anatase (TiO_2), and rutile (TiO_2). The main carrier minerals for Al_2O_3 and SiO_2 are mullite and quartz, and the main carrier minerals for TiO_2 are anatase and rutile. These minerals with quite stable crystal structures would exhibit fairly low reactivity in the process of activation and conversion of solid waste disposal or reuse. SiO_2 is found more in amorphous glass. The percentage of amorphous silica in the FAAE residue can be easily extracted from the solid residue obtained after alumina extraction, the original fly ash, and similar solid wastes.

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Benefiting from the acid-leaching treatment of FAAE, the solid residue of FAAE is more suitable for the synthesis of aluminosilicate zeolites than the original fly ash as iron, calcium, and other acid-soluble harmful components were removed along with aluminum in advance.

2.2. Methods. A four-step process was designed for simultaneous synthesis of zeolites 13X and ZSM-5 from the FAAE residue (Figure 2).

The first step was alkali roasting. In this step, the FAAE residue was mixed with Na$_2$CO$_3$ (110–130% mass fraction) and roasted at 830–910 °C for 60–90 min to effectively activate silicon and aluminum. Molten Na$_2$CO$_3$, which may contain some Na$_2$O (a decomposition product of Na$_2$CO$_3$), will destroy the structure of aluminosilicate crystals and form new water-soluble substances. This step is the key to the decomposition and reuse of the aluminosilicate solid waste. In future large-scale production, CO$_2$ in the roasting tail gas can be collected and used for the subsequent CO$_2$-bubbling process.

The second step was water leaching. In this step, the alkali roasting product was crushed to less than 200 mesh. This was followed by dry magnetic separation of the iron. A one-step water leaching process was then used to dissolve active silicon and aluminum. The leaching temperature ($T_1$) was 60–100 °C, the liquid to solid ratio (LSR) was 2–20 (mL/g), and the leaching time ($t$) was 20–120 min. The mixture was then filtered while still warm ($T_2$ = 20–75 °C) to separate the Si- and Al-rich aqueous phase from the solid residue. The aqueous phase was used as the raw material for zeolite synthesis. The solid residue from warm filtration was recycled back into the alkali roasting step, which minimized the production of secondary solid waste.

The third step was synthesis of zeolite 13X. For this step, 100 mL of water was added to approximately 400 mL of the Si- and Al-rich water-leaching liquor from the second step of preliminary hydrolysis. Then, CO$_2$ gas was bubbled into the system, and the mixture was hydrolyzed further to a certain pH value to prepare the precursor for synthesis of 13X. The zeolite was then synthesized in a polytetrafluoroethylene reactor at 90–110 °C for 15–30 h. The mother liquor obtained after removal of 13X by filtration was retained for subsequent synthesis of ZSM-5.

The fourth step was synthesis of zeolite ZSM-5. For this step, the mother liquor of 13X was further hydrolyzed by bubbling CO$_2$ gas again to a lower pH. Next, tetraropropylammonium hydroxide (TPOH) was introduced into the system as a template to prepare the precursor for ZSM-5. The ZSM-5 precursor was then transferred into a polytetrafluoroethylene reactor and crystallized under hydrothermal treatment (140–190 °C) for 24–72 h. The solid crystallization product was then filtered and washed using excess water to thoroughly remove Na$_2$CO$_3$ from the filter cake. After filtering and washing, the solid product was dried at 120 °C for more than 4 h and then roasted at 550 °C for 4 h to remove the organic template. The roasting product was pure sodium-type ZSM-5. The mother liquor of ZSM-5 contained mainly Na$_2$CO$_3$ and a small amount of silicon and aluminum. The Na$_2$CO$_3$ from the liquor was recycled back into steps 1 and 2 after evaporation, concentration, and crystallization. This minimized the volume of the waste liquid created in this step.

2.3. Analysis. X-ray fluorescence spectrometry (XRF; ZSX Primus, Rigaku, Tokyo, Japan) was used to quantitatively analyze the chemical elements in the solid residue obtained after alumina extraction, the intermediates, and the zeolite products. The instrument used contained a 4 kW X-ray tube with an ultrathin 30 μm Rh window and was equipped with an 8-crystal converter (LiF200, PET, RX-61/45/35/25, Ge, and LiF220). Data were obtained for the elements from boron to uranium, and the measurement accuracy was at the parts per million level.
XRD analysis of all samples was performed using a powder diffractometer (D8 Advance, Bruker, Germany) equipped with a ceramic X-ray tube (40 kV, 40 mA), a Cu Kα radiation source, and an energy resolution of <9%. All samples were scanned within a 2θ interval of 4–70° using a step size of 0.01°. EVA and TOPAS software (Bruker) were applied to the data to identify the mineral phases in the samples.

A field-emission scanning electron microscope (SEM; Nova NanoSEM, FEI, USA) was used in conjunction with an energy-dispersive X-ray spectrometer (X-man 50, Oxford Instruments, UK) to obtain information about the micromorphologies and elemental compositions of the zeolites synthesized in this study.

As for the determination of the chemical composition of the precursor liquor used for the zeolites syntheses, the content of Na, Si, Al, and other elements was determined by ICP-AES (SPECTRO ARCOS, Spectro, Germany), and the concentration of the CO₃²⁻ ion was determined using the ion-selective electrode (PXS-CO₂, Hangzhou Qiwei Instrument Co., LTD, China). The mass of H₂O was obtained by subtracting the total mass of measured ions from the total mass of the solution. Then, all the mass concentrations were converted to molarities to characterize the chemical composition of the precursor.

A laser particle size analyzer (Mastersizer 3000, Malvern Panalytical, UK) was used to analyze the particle size distribution of raw materials and products. The N₂ adsorption–desorption isothermal curves were obtained at 77 K (3-FLEX, Micromeritics, USA). The specific surface area was determined via the Brunauer–Emmett–Teller (BET) equation.

3. RESULTS AND DISCUSSION

3.1. Mineral Phase Transition during Alkali Roasting and Activating Effect.

Dv(X) was defined as the diameter at which X % (by volume) of the particles have a diameter less than that value. The particle size property of powder sample can be basically obtained commonly by fetching the values of Dv(10), Dv(50), and Dv(90). The FAAE residue particles have good fineness (Dv(90) = 44.7 μm, Figure 3), which is conducive to their subsequent activation or transformation reaction. No individual mineral particles (e.g. mullite, anatase, or quartz) were found under the SEM. Irregular particles adhering to each other (Figure 4) indicate that physical separation methods would be ineffective in extracting any single component.

Alkali (adding NaOH or Na₂CO₃) roasting is an effective method to decompose or activate aluminosilicate minerals. In the subsequent dissolution and extraction process, separating aluminum by acid-leaching and silicon by alkali-leaching, respectively, is a common approach. The natural consequences of using both acids and alkalis in the same system involve increased acid and alkali losses, more complex operation procedures, and an extra investment in acid- and alkali-proof equipment and pipelines. Silica and alumina could react with Na₂CO₃ and be transformed completely into water-soluble Na₂SiO₃ and alkali-soluble NaAl₂SiO₅ under proper roasting conditions. It seems feasible to complete roasting (alumino-silicate activation), leaching (Si & Al extraction), and utilization (zeolites hydrothermal syntheses) of aluminosilicate waste under a single alkali reaction system. Maximum conversion and use of the FAAE residue at the lowest cost of energy and materials would be achievable.

The effects of addition of sodium carbonate, the roasting temperature, and the roasting time on the composition of the alkali roasting product were investigated to optimize the alkali roasting conditions for subsequent Si and Al dissolution and zeolite syntheses. The XRD results (Figure 5) indicated that the alkali roasting products all contained Na₂SiO₃, sodium silicoaluminate, amorphous aluminosilicate glass, and Na₂CO₃ in different proportions.

As we all know, mineral phase content is positively correlated with the intensity of the diffraction peak. The original diffraction spectrums of alkali roasting products obtained under the same test conditions were stacked and compared in XRD analysis software (e.g., JADE). By observing the intensity (height) variation of the diffraction peaks of each mineral phase with different roasting conditions, their influence on the relative proportion of each phase (including the amorphous alumi-
Silicon and Aluminum. The FAAE residue was 156%.

The mass fraction of the alkali roasting product compared with min were selected as the optimum alkali roasting conditions. Calculated as follows

\[ LR = \left(1 - \frac{M_i}{M_o}\right) \times 100\% \]  

where \( M_i \) is the mass (dry basis) of the solid residue obtained after water leaching, and \( M_o \) is the mass (dry basis) of the original mass of the FAAE residue used for alkali roasting.

In a desired state, Si and Al were dissolved in NaOH solution and made to coexist in the aqueous phase as sodium silicate and sodium aluminate. Then, the extraction, separation, and reuse of Si and Al would be quite easy. However, actually, the alkali leaching conditions of Si and Al partially overlap with those of the hydrothermal synthesis of zeolites; Si and Al in the alkali hydrothermal environment tend to combine with Na to form sodalite (Na\(_8\)Al\(_6\)Si\(_6\)O\(_{24}\)(OH)\(_2\)) precipitation. Sodium silicoaluminate in nepheline and carnegeite, amorphous Si and Al, and Na\(_2\)CO\(_3\) in the nitrite phase showed poor solubility in the water leaching process and were components of the solid residue obtained after filtration. Therefore, the proportions of nepheline, carnegeite, natrite, and glass should be limited by controlling the roasting conditions. In consideration of the activation effect and the energy cost, a 110% mass fraction of Na\(_2\)CO\(_3\)-roasted FAAE residue under various conditions.

3.2. Optimization of Conditions for Water Leaching of Silicon and Aluminum. Obviously, the leaching rate (LR) of Si and Al is expected to be as high as possible. The LR was calculated as follows

\[ LR = \left(1 - \frac{M_i}{M_o}\right) \times 100\% \]

where \( M_i \) is the mass (dry basis) of the solid residue obtained after water leaching, and \( M_o \) is the mass (dry basis) of the original mass of the FAAE residue used for alkali roasting.

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Warm-keeping filtration was first found to be necessary. The dissolution product visibly became sticky when the temperature dropped during the filtration process, resulting in a significant decrease in the LR of Si and Al (entry 1 vs 2, Table 2). Considering the actual operating conditions, \( T_1 = 75 \, ^\circ C \) was selected as the feasible filtration temperature.

Table 2. Dissolution Conditions and the Leaching Rates of the Alkali-Roasted FAAE Residue

| test no. | liquid to solid ratio (mL/g) | leaching temp. \( T_1 \) (°C) | leaching time \( t \) (min) | filtration temp. \( T_2 \) (°C) | wash agent | leaching rate LR (%) |
|---------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|------------|------------------------|
| 1       | 20                          | 100                         | 30                          | 20                          | none       | 40.3                   |
| 2       | 20                          | 100                         | 30                          | 75                          | water      | 63.0                   |
| 3       | 20                          | 100                         | 20                          | 75                          | 67.6       |
| 4       | 10                          | 100                         | 20                          | 75                          | 62.4       |
| 5       | 10                          | 100                         | 30                          | 75                          | 59.5       |
| 6       | 10                          | 100                         | 20                          | 75                          | 75.2       |
| 7       | 5                           | 100                         | 20                          | 75                          | saturated Na\(_2\)CO\(_3\) solution | 89.7       |
| 8       | 2                           | 100                         | 20                          | 75                          | 93.2       |

By comparing test no. 4 and no. 5 in Table 2, increasing the leaching time would not increase the LR, which should be attributed to the excessive formation of zeolites (e.g., sodalite) under high alkalinity conditions.

The essence of water leaching is still alkali leaching. The alkalinity may come from the residual Na\(_2\)O or Na\(_2\)CO\(_3\) or the hydrolysis of sodium (alumino)silicate in the roasted product of the FAAE residue

\[ Na_2SiO_3 + H_2O \rightleftharpoons NaOH + SiO_2 - nH_2O\_1 ] (2)

\[ NaAlSiO_4 + H_2O \rightleftharpoons NaOH + Al(OH)_3\_2 \] (3)

The possible presence of hydrolysis products (SiO\(_2\)-nH\(_2\)O or/and Al(OH)_3) of sodium (alumino)silicate was believed to be another key factor causing filtration difficulty and a decrease in LR. In general, hydrolysis will be intensified by a temperature increase. A low liquid-solid ratio scheme was adopted to inhibit the hydrolysis reaction across the process of high-temperature \( (T_1 = 100 \, ^\circ C) \) dissolution. With less H\(_2\)O and more Na, the
2. Optimized parameters for water dissolution of the alkali-roasted product were finally determined as (T₁ = 100 °C, t = 20 min, T₂ = 75 °C, LSR = 2−5, using saturated Na₂CO₃ solution as the diluent and washing agent). Under this optimal condition, around 90 wt.% of the FAAE residue can be dissolved and separated for further use. The secondary residue was still dominant with silica and alumina and was sent back for the alkali roasting procedure.

3.3. Co-Production of Zeolites 13X and ZSM-5. The Si- and Al-rich filter liquor collected from Section 3.2 with a molar composition of SiO₂/Al₂O₃/Na₂O/CO₃²⁻/H₂O = 12:1:13:3:5:40 was taken as the only Si−Al source to synthesize zeolite 13X and ZSM-5.

The theoretical chemical composition of zeolite 13X is Na₂O·Al₂O₃·(2.8 ± 0.2)SiO₂·(6−7)H₂O. Zeolite ZSM-5 is a typical high-silica zeolite and commonly has a silica/alumina molar ratio (SAR) greater than 30. The SAR value of the filter liquor (SAR = 12) was between that of low-Si zeolite 13X (SAR = 2.6−3.0) and high-Si zeolite ZSM-5 (SAR >30). Adding an external Al-source or Si-source, respectively, to adjust the liquor SAR down or upward for synthesis of 13X or ZSM-5, is the most common thought. However, it is obviously unfavorable in terms of reducing the amount of solid waste.

Assuming that the low-Si zeolite 13X can be directly synthesized with the filter liquor, the mother liquid of 13X will have a higher SAR value suitable for the synthesis of ZSM-5 because most of the Al was consumed by 13X. In this way, the Si and Al in the filter liquor will be converted and utilized with maximum efficiency.

3.3.1. Hydrothermal Synthesis of Zeolite 13X. First, CO₂ gas was used to neutralize the too-high alkalinity of filter liquor to an appropriate pH to prepare the precursor gel for synthesis of 13X. The use of CO₂ gas instead of traditional alkalinity regulators, such as HCl or H₂SO₄ solutions, made recycling of CO₂ and Na₂CO₃ in the overall process possible and avoided corrosion of the equipment by strong acids. Additionally, production of the greenhouse gas CO₂ and highly saline waste water (NaCl or Na₂SO₄) was minimized. Thus, the process was environmentally friendly.

In conventional hydrothermal zeolite syntheses, alkalinity is usually characterized and controlled by two static parameters: Na₂O/SiO₂ and H₂O/SiO₂. The levels of these parameters are fixed at the point at which the materials are added. In this study, a single parameter (pH) was investigated for replacing Na₂O/SiO₂ and H₂O/SiO₂ in the 13X and ZSM-5 syntheses. The upper limit of the SiO₂/Al₂O₃ molar ratio for precursors of pure 13X syntheses ever reported has not exceeded 5.4. This was the first attempt to synthesize 13X in this ultra-high silicon environment (SAR = 12).

The experimental results indicated that the pH of the precursor gel had a direct effect on the hydrothermal synthesis of zeolite 13X in this ultra-high silicon system. Precise regulation of the gel pH between 12.9 and 13.6 gave pure 13X crystals (SAR = 2.32, energy dispersive X-ray spectroscopy data) with diameters of less than 15 μm (Figures 6 and 7A) under conventional hydrothermal crystallization conditions (90–110 °C for 15–30 h).

3.3.2. Hydrothermal Synthesis of Zeolite ZSM-5. Recovery and recycling of zeolite mother liquor for the next round of synthesis of the same zeolite are commonly used to induce crystallization and reduce the usage of the template. In this study, mother liquor of 13X was used as the material for another kind of zeolite, ZSM-5. This is the major novelty of this work.

Crystallization of 13X consumed excessive Al and Na as expected. The SAR of the mother liquor of 13X increased to 32, and the system alkalinity decreased to approximately pH 12. These values were closer to the ideal conditions for ZSM-5.
synthesis. However, even so, co-crystallization of ZSM-5 and mordenite was found to happen easily, with this SAR level still being relatively low. Impurities of zeolite NaP or analcime were detected when ZSM-5 was directly synthesized under this system alkalinity.

CO₂ was again used to adjust the system alkalinity and also to prepare the precursor of ZSM-5. When the pH was further adjusted to 10.5 by carbonation, pure ZSM-5 crystals (SAR = 24.3, energy dispersive X-ray spectroscopy data) with a diameter size of less than 6.5 μm (Figures 7B and 8) were synthesized in the presence of a template or a seed. The mother liquor of ZSM-5 that mainly contained Na₂CO₃ with trace quantities of silicon and aluminum was recovered to make wash agents (saturated Na₂CO₃ solution) and roasting aids (powder Na₂CO₃).

3.3.3. Further Characterization of Synthesized Zeolites from the FAAE Residue. As shown in Figure 9, particles of zeolite 13X have an ideal normal distribution and good fineness (Dₜ(90) = 10.1 μm). The particle size distribution of ZSM-5 is bimodal, indicating the presence of a certain proportion of submicron particles (Figure 10).

The specific surface area and pore characteristics of both two zeolites are at normal levels (Table 3). More in-depth characterization of the two zeolites synthesized will be displayed combined with the specific applications in the future.

Metal ions (Ca, Fe, Ti, etc.) other than Si, Al, and Na have positive or negative effects on the adsorption or catalytic performance of zeolites, sometimes significantly. The elemental composition of the synthesized zeolites was analyzed by XRF. The results showed that there was only a trace amount of TiO₂, Fe₂O₃, and others in the two zeolite products (Table 4). This means that the elements Ti or Fe, which may be activated synchronously in the process of alkali roasting, mostly enter into the filter residue of water leaching. After several “roasting-leaching” cycles, Ti with a high economic value will be enriched in the solid phase, from which the recovery of titanium resources may become an unanticipated earning created by this work.

Certain amounts of carbon were detected in both the zeolite products (Table 4), although the occurrence state of carbon is unknown for now. Carbon played an important role in the whole process of decomposition, transformation, and utilization of the solid residue. There have been few reports of the use of CO₂ alone for adjusting and controlling system alkalinity of the hydrothermal syntheses of zeolites in a carbonate (CO₃²⁻) environment. However, it has been shown that CO₃²⁻ can effectively improve the degree of silica condensation in alkaline media and promote the crystallization of zeolites with Mobil-5 structures (typically, ZSM-5). The detailed impact of the CO₃²⁻ environment on the zeolite pore structure and the zeolite acidic and catalytic performance deserves expanded research as this link is of importance for constructing the complete circulation of Na₂CO₃−CO₂−CO₃²⁻ and thus for the full conversion and use of aluminosilicate wastes. The focus on carbon cycling in inorganic chemical processes also has positive environmental implications.

4. CONCLUSIONS

The FAAE residue can be fully activated by alkali roasting with Na₂CO₃ (110% mass fraction) at 890 °C for 60 min. The product from roasting contains silicon and aluminum in the form of Na₂SiO₃ and NaAlSiO₄. These minerals dissolve in water after 20 min at 100 °C when the liquid/solid ratio is 2−5.

After optimization of the process, the LR of the alkali-roasted FAAE residue reached 90%.

A combined processing strategy was effective for full utilization of the silicon and aluminum in the water leaching liquor of the alkali-roasted FAAE residue. First, the Si- and Al-rich liquor was used to produce low-Si zeolite 13X. As expected, the Si- and Al-rich liquor was used to produce low-Si zeolite 13X. After optimization of the process, the LR of the alkali-roasted FAAE residue reached 90%.

A combined processing strategy was effective for full utilization of the silicon and aluminum in the water leaching liquor of the alkali-roasted FAAE residue. First, the Si- and Al-rich liquor was used to produce low-Si zeolite 13X. After optimization of the hydrothermal conditions for this process, the method was successful even though the SAR was much higher than that conventionally used in 13X synthesis. As expected,
production of 13X consumed a large amount of aluminum, which increased the silica/alumina ratio of the liquor to SAR = 32, suitable for the synthesis of ZSM-5.

Combined production of high-Si and low-Si zeolites maximized the utilization of both silicon and aluminum in the FAAE residue, reduced requirements for additional materials, and minimized the production of secondary waste. The strategies and practices developed in this study could be used to develop techniques for commercial disposal of fly ash in the future.

**Table 3. BET Surface Area and Pore Characterization of Zeolites Produced from the FAAE Residue**

|           | BET area (m²/g) | \(V_{\text{total}}\) (cm³/g) | \(V_{\text{micro}}\) (cm³/g) \(^a\) | \(V_{\text{meso}}\) (cm³/g) \(^b\) | \(d_{\text{pore size}}\) (nm) \(^c\) |
|-----------|-----------------|-------------------------------|---------------------------------|---------------------------------|---------------------------|
| 13X       | 444.8           | 0.1595 \(^d\)                | 0.1053                          | 0.0473                          | 1.4340                    |
| ZSM-5     | 384.2           | 0.1698 \(^e\)                | 0.1438                          | 0.0348                          | 1.3297                    |

\(^a\)Single-point adsorption total pore volume of pores less than 0.9384 nm in diameter at \(P/P_0 = 0.0100\). \(^b\)Single-point adsorption total pore volume of pores less than 372.3423 nm² in diameter at \(P/P_0 = 0.9943\). \(^c\)Total volume of the micropore by the t-plot model. \(^d\)Adsorption cumulative volume of the mesopore calculated using the Barrett–Joyner–Halenda (BJH) model. \(^e\)Adsorption average pore diameter by BET.

**Table 4. Elemental Composition (by XRF, %) of the Zeolites Synthesized from the FAAE Residue**

|          | CO₂   | Na₂O  | K₂O  | CaO  | MgO  | Al₂O₃ | SiO₂   | P₂O₅ | SO₃  | Cl    | TiO₂ | Fe₂O₃ | NiO  | MoO₃ | ZrO₂ |
|----------|-------|-------|------|------|------|-------|--------|------|------|-------|------|-------|------|------|------|
| 13X      | 3.50  | 18.77 | 0.017| 0.011| 0.02 | 32.31 | 44.87  | 0.003| 0.020| 0.453 | 0.036| 0.027 | 0.009| 0.003| 0.006|
| ZSM-5    | 8.31  | 3.87  | 0.049| 0.038| 0.02 | 5.82  | 81.61  | bdl  | bdl  | 0.134 | bdl  | 0.134 | bdl  | bdl  | bdl  |

\(^a\)Below the minimum detection limit.

Production of 13X consumed a large amount of aluminum, which increased the silica/alumina ratio of the liquor to SAR = 32, suitable for the synthesis of ZSM-5. Combined production of high-Si and low-Si zeolites maximized the utilization of both silicon and aluminum in the FAAE residue, reduced requirements for additional materials, and minimized the production of secondary waste. The strategies and practices developed in this study could be used to develop techniques for commercial disposal of fly ash in the future.
Notes
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