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New Synthesis Process of Crystalline Phase - Pure Faujasite Zeolite Material from Sialic Type Coal Fly Ash

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ABSTRACT

Zeolite materials obtained from coal fly ash usually contain undesired mineral impurities and so an effort to achieve the single phase zeolite is important. In this paper, the synthesis of pure faujasite (FAU) type zeolite X from pretreated sialic type coal fly ash was carried out. Special attention was paid to convert coal fly ash into FAU type zeolite X using many pretreatment processes involving water, acid (HCl), base (NaOH) and calcination followed by fusion with NaOH at 550 °C, ageing and crystallization. All the materials were characterized by XRF, XRD, FTIR, SEM-EDX and BET methods. The results indicate that single phase (pure) zeolite NaX crystal (Faujasite type) could be obtained in the process: fusing of acid treated fly ash with NaOH (ratio 1:1.3) at 550 °C, aging 6 h and crystallization temperature 90 °C.

1. Introduction

Coal is composed primarily of carbon and hydrogen, but all coal also contains some mineral matter such as clays, shales, quartz and calcite. The percentage of minerals varies by coal type and source. About 65% of the coal produced in India is used for power generation in some 75 thermal power plants located in various parts of the country [1, 2]. Coal fly ash (FA) is the mineral matter comprising aluminium silicate that is collected after the coal is combusted at high temperature, along with some unburnt carbon. In 2015 world bank has reported that about 160 million tons of fly ash is being generated annually in India, and this is likely to reach 220 million tons by early next century, with 65 000 acres of land being occupied by ash ponds [3]. Such huge quantity does pose challenging problems in the form of land usage, health hazards and environmental dangers. In order to mitigate such challenging problems and considering the generation of large volume of FA from thermal power stations, many possibilities of utilization of FA were considered. Some of the currently adopted fields of utilization of FA are in the building and highway construction, manufacture of cement, floor and wall tiles, insulator, sintered pozzolanic aggregate, mineral wools, ceramics etc. In addition, the utilization of FA has been well-accepted for zeolite synthesis [1-6].

Many technical articles have reported and proposed synthesis of different types of zeolites from FA [7-12]. Most of these methodologies are based on the alkali activation (mainly by means of NaOH or KOH solution) [11-13] followed by crystallization. The alkali fusion procedure has been improved to obtain high porous size and high cation exchange capacity zeolites [14, 15]. In this line there is a wide number of studies focusing on the synthesis of zeolite from coal fly ash without any purification or pretreatment. Since the zeolite materials obtained by such methods usually contain unwanted materials like residual fly ash in significant amount, a little amount of carbon and other minerals, mullite, quartz etc., Such zeolites are chemically and morphously impure [16-19]. Generally, unpurified fly ash consists of quartz, mullite and other minerals together with metal oxides such as Na₂O, K₂O, TiO₂, Fe₂O₃, CaO, MgO, BaO etc., [4, 5, 18]. These metal oxides are known to restrict the formation of pure zeolite form fly ash. Purification of fly ash involves restricting the metal oxide and undesired phases of fly ash during the synthesis of zeolite. However, there

are only few studies focusing on the synthesis of zeolite from chemically modified fly ash [16-19]

Zeolites are crystalline, micro-porous, hydrated aluminosilicates that are built from an infinitely extending three-dimensional network of [SiO₄]⁴⁻ and [AlO₄]⁵⁻ tetrahedra linked to each other by the sharing of oxygen atom. Hence they are considered as inorganic repetition polymers with channels or cavities. They provide large surface area and most of degradable molecules easily diffuse into the channels and cages of the order of 4–14 Å resulting in their enhanced degradation [17]. Especially, zeolites X with low Si/Al ratio in the framework possess high ion exchange capacity, sorption and relatively large pore size. These features impart the highest catalytic activity and make zeolite X ideal adsorbent among the various zeolites [17, 19, 20].

Synthesis of zeolite from FA is superior to other techniques (chemical synthesis of zeolite) due to its low cost including material and operation, simplicity of design, ease of synthesis and non-toxic nature. However, to the best of our knowledge, no systematic investigation has been performed on zeolite synthesis from different types of purified or pretreated sialic type coal fly ash. With this view we have focused a detailed study on the synthesis of crystalline phase-pure zeolite (without any fly ash mineral phase) from purified fly ash by reducing the content of metal oxide. This paper reports the results of suitable experimental conditions such as the effect of alkali, fusion temperature, aging time and crystallization temperature toward the synthesis of highly crystalline, single phase and high surface area zeolite.

2. Experimental Methods

2.1 Materials

The main raw material, coal fly ash sample was collected from electrostatic precipitators of nearby thermal power plant, Tuticorin, Tamilnadu. The commercially available chemicals used in the present work were hydrochloric acid (Merck, Mumbai) and sodium hydroxide (LOBA chemie, Mumbai). All the procured chemicals were used without further purification as they were of analytical grade. Throughout the experiment DD water was used.

2.2 Pretreatment of Fly Ash

The raw fly ash sample or untreated fly ash (UFA) was first screened through BSS Tyler sieve of 75 μ mesh size to eliminate larger particles. The screened fly ash sample was dispersed in different treating reagents such

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as water, 0.1 M HCl and 0.1 M NaOH. A mixture of 10 g screened UFA and 100 mL treating reagent was mechanically stirred at room temperature for 30 min. The solid product was repeatedly washed with treating solution, filtered and dried at 100 °C overnight. These treated fly ashes (FAs) are denoted as WFA (water treated fly ash), AFA (acid treated fly ash) and BFA (base treated fly ash). Another FA sample was obtained by calcination of UFA (unpurified form) at 800 °C for 2 h, referred to as calcined fly ash (CFA). This pretreatment is to estimate the leaching behavior of metal oxide, to assess the Si/Al molar ratio and to facilitate synthesis of zeolite in pure form without any mineral phase of fly ash.

2.3 Synthesis of Zeolite

Zeolite NaX was synthesized from pretreated fly ash by fusion method, which involved alkaline fusion followed by hydrothermal treatment [13, 15]. A typical procedure is given below: Different types of pretreated FAs (WFA, CFA, BFA, AFA) were ground with NaOH pellets at 1:1.3 ratio (wt/wt) and fused at 550 °C for 1 h. The fused product was ground again and was followed by the addition of DD water at solid to water weight ratio of 1 g pretreated FA/10 mL water. The slurry was agitated for 20 h and then it was subjected to crystallization at 100 °C for 6 h without any disturbance. The solid product was recovered by filtration and washed thoroughly with DD water until the pH of the samples was about 9. The product was then dried at 100 °C in a vacuum oven. The products are designated as WFAZ, CFAZ, BFAZ and AFAZ respectively.

2.4 Material Characterization

The chemical compositions of UFA, treated FAs and zeolites were determined by wavelength dispersive X-ray fluorescence spectroscopy (XRF) S8 TIGER from Bruker, Germany. Mineralogical analysis of the synthesized materials was conducted with XRD by the use of a Philips X'Pert Pro- MPD X-ray diffractometer with Cu-K α radiation ($\lambda = 1.5406 \text{ \AA}$) at 40 kV and 30 mA by scanning the samples from 10 to 80°. Mineralogical analysis of the acid treated fly ash converted zeolite (AFAZ) was conducted by XRD of a Philips X'Pert Pro- MPD X-ray diffractometer with Cu-K α radiation ($\lambda = 1.540 \text{ \AA}$) at 40 kV and 40 mA by scanning the sample from 5.0 to 100°. FTIR spectra of the samples were recorded (in spectral grade KBr pellet) with FTIR spectrometer (JASCO FTIR-410) in the wavenumber range 400 – 4000 cm $^{-1}$ at a resolution of 2 cm $^{-1}$. The specific surface area of the powder samples was determined by BET and Langmuir nitrogen adsorption/desorption method in Micromeritics ASAP 2420 surface area analyzer. Morphological analysis of the solid samples was done with Scanning Electron Microscope (VEGA 3 TESCAN). The integrated energy dispersive X-ray (EDX) spectroscopy was applied to identify the elements in zeolite material using BRUKER instrument.

3. Results and Discussion

3.1 Material Characterizations

3.1.1 XRF Studies

The XRF derived chemical compositions of raw fly ash (untreated fly ash, UFA), water treated fly ash (WFA), calcined fly ash (CFA), base (NaOH) treated fly ash (BFA), acid treated fly ash (AFA) and the synthesized respective zeolite materials, WFAZ, CFAZ, BFAZ and AFAZ are summarized in Table 1. According to the American Society for Testing Materials (ASTM) [21] the FA material used in this study is classified as sialic type because it contains less than 5% CaO and >90% of the three main components (SiO $_2$, Al $_2$ O $_3$ and Fe $_2$ O $_3$) combined. The loss of ignition (LOI), 0.06% and SO $_3$ content do not exceed 6% and 5% respectively [22]. It is commonly produced from the burning of higher-rank bituminous coals and anthracites and, therefore, this type of FA is pozzolanic in nature (hardening when reacted with Ca(OH) $_2$ and water) [22]. The properties and chemical composition of FA vary according to coal source and power plant operation, which makes fly ash zeolite even more versatile.

XRF analysis shows significant variation in compositions between the untreated FA and treated FAs. Treatment with water (WFA) does not avoid impurities such as calcium oxide and other metal oxides and it does not improve the purity of silica and alumina. This result suggests that water treatment of UFA and the resultant unremoved impurities may interfere in the synthesis of zeolite in pure phase. As shown in Table 1, base treated fly ash and calcined fly ash have almost similar compositions. The chemical composition of AFA has high levels of silica, alumina and a low level of sodium, potassium and other metal oxides; also the portion of calcium oxide (CaO) in AFA is decreased from 1.24% to 0.81%. This may be due to the dissolution of metal oxides into HCl during treatment and their removal during washing. XRF analysis reveals that AFA is a possible material as a silica source for the synthesis of pure form of zeolite. This

observation is in good agreement with the study by Pengthamkeerati et. al [16], who also reported a decrease in CaO content in acid treated fly ash.

XRF data of different zeolites, WFAZ, CFAZ, BFAZ and AFAZ (Table 1) reveal that alkali fusion and hydrothermal treatment of all treated FAs have considerably increased the percentage of Na $_2$ O in the zeolite materials compared to pretreated FAs (from mere 0.16 to 18.26%). This is due to the capture of sodium ions needed to neutralize the negative charges on the aluminate in zeolite. Other metal oxides have also undergone considerable decrease in their content. Thus the expected chemical change in each and every step of zeolites formation is confirmed and validated by XRF analysis. The amounts of SiO $_2$ and Al $_2$ O $_3$ in zeolites are lower than the corresponding oxides in the treated FAs caused by the dissolution of the amorphous aluminosilicate into the alkaline solution during treatment. As a result, SiO $_2$ /Al $_2$ O $_3$ ratios of WFA, CFA, AFA and BFA are 1.93, 1.98, 1.97, and 2.03 which are reduced to 1.50, 1.50, 1.50 and 1.48 respectively in zeolites and this result confirms the formation of zeolite materials. Other remarkable feature of zeolite formation from treated FAs is the maximum reduction/vanishing of P, V, Cr and S oxides.

Table 1 Chemical compositions (wt%) of unpurified fly ash (UFA), pre-treated FAs (WFA, CFA, BFA and AFA), water treated fly ash zeolite (WFAZ), calcined fly ash zeolite (CFAZ), base treated fly ash zeolite (BFAZ) and acid treated fly ash zeolite (AFAZ) derived from XRF studies

Chemical Compound	% Composition								
	Raw fly ash	Purified fly ash				Synthesized zeolite material			
	UFA	WFA	CFA	BFA	AFA	WFAZ	CFAZ	BFAZ	AFAZ
SiO $_2$	58.0	58.2	59.1	59.1	59.7	42.8	43.1	43.2	42.8
Al $_2$ O $_3$	29.9	29.9	29.9	30.0	29.5	28.7	28.7	28.7	28.7
Na $_2$ O	0.16	0.15	0.16	0.15	0.13	18.1	18.1	18.3	18.3
Fe $_2$ O $_3$	5.58	5.43	5.23	5.23	5.23	5.59	5.66	5.48	5.42
TiO $_2$	1.63	1.65	1.66	1.64	1.64	1.73	1.73	1.74	1.75
CaO	1.24	1.20	0.87	0.87	0.81	1.30	0.96	0.93	0.87
MgO	1.01	1.01	0.88	0.88	0.87	0.93	0.89	0.82	0.75
K $_2$ O	1.41	1.39	1.41	1.41	1.38	0.57	0.55	0.54	0.55
BaO	0.08	0.08	0.06	0.07	0.07	0.08	0.09	0.06	0.06
P $_2$ O $_5$	0.51	0.50	0.32	0.32	0.31	0.02	0.01	0.01	0.00
V $_2$ O $_5$	0.04	0.04	0.04	0.03	0.03	0.02	0.00	0.00	0.00
MnO	0.05	0.05	0.04	0.04	0.04	0.06	0.05	0.05	0.04
Cr $_2$ O $_3$	0.02	0.02	0.02	0.02	0.02	0.00	0.00	0.00	0.00
SO $_3$	0.17	0.08	0.03	0.04	0.02	0.02	0.02	0.02	0.00
SiO $_2$ / Al $_2$ O $_3$	1.94	1.93	1.98	1.97	2.03	1.50	1.50	1.50	1.48
Crystallite size (nm)	33.4	40.9	48.9	49.4	64.7	41.2	50.1	52.7	51.8

3.1.2 XRD Studies

The X-ray diffraction (XRD) patterns of untreated fly ash, UFA and treated FAs, WFA, CFA, BFA and AFA are shown in Fig. 1a. The XRD pattern of UFA indicates that the major mineral phases in fly ash are quartz, mullite and hematite together with an amorphous component. The presence of amorphous phase is identified as a broad diffraction "hump" in the region between $2\theta = 20$ to 30° [15]. As shown in Fig. 1a, there is no significant difference in XRD patterns of UFA and WFA, which is consistent with the data of XRF analysis. This result suggests that water treatment cannot induce bulk phase changes. CFA, BFA and AFA have almost similar XRD patterns; the major crystalline phases in the treated fly ashes (CFA, BFA and AFA) are found to be quartz (SiO $_2$) and mullite (3Al $_2$ O $_3$ ·SiO $_2$) with small quantities of magnetite and hematite. The diffraction patterns of all treated fly ashes are in good agreement with JCPDS file No. 001-0649 (quartz) and 002-0430 (mullite).

The XRD patterns of all synthesized zeolites, WFAZ, CFAZ, BFAZ and AFAZ are shown in Fig. 1b. Pretreatment process plays a significant role in determining the structural property of the final zeolite product. In the XRD pattern of WFAZ, the quartz peak is disappeared, while few mullite peaks are still appearing. The major crystalline phase of WFAZ is identified as zeolite X, with small amounts of mullite. This result implies incomplete dissolution of quartz and mullite phases, because of their mineralogical differences, that had been noted from XRF. The failure to obtain pure phase zeolite NaX using WFA means that the water treatment does not improve the purity of silica and alumina. SiO $_2$ to Al $_2$ O $_3$ ratio of CFA, BFA and AFA is nearly 2 and the values are 1.98, 1.97 and 2.03. This high value is responsible for the formation of zeolites of different types, quantities and purities as discussed below. As shown in Fig. 1b, zeolite NaX is mainly formed in all the samples but the XRD patterns of CFAZ and BFAZ also contain some quartz (Q), mullite (M) and/or sodalite (s) peaks. CFA and

BFA contain the high content of elements such as calcium and iron (Table 1) which inhibit the formation of zeolites in high crystalline and pure form.

On the other hand, the XRD pattern of AFAZ has high intense sharp signals which are all assignable only to zeolite NaX. The characteristic lines located at 2θ values of 10.1, 11.8, 15.5, 18.4, 20.09, 23.3, 26.65, 29.21, 30.3, 31.1, 32.6, 37.4, 41.2° etc., observed from XRD pattern AFAZ can be indexed to zeolite NaX crystalline planes ($\text{Na}_{88}\text{Al}_{88}\text{Si}_{104}\text{O}_{384}\cdot 220\text{H}_2\text{O}$) by matching with the zeolite patterns in the library of XRD pattern [JCPDS No. 39-0218] and also with reports in literature [13, 14, 19]. The most distinct changes in the XRD pattern of AFAZ when compared to AFA fly ash are the disappearance of the quartz and mullite peaks (showing complete conversion of FA into zeolite) and the appearance of new crystalline phase corresponding only to zeolite X. Complemented by Silica/Alumina ratio of 1.48 (i.e. ratio < 1.5) [23] from XRF data in Table 1, this XRD pattern definitely indicates that AFAZ contains only one mineral phase, zeolite X (or faujasite) and does not have any other mineral phase even to the small extent. Hence acid treatment of FA, alkali fusion and crystallization at the previously mentioned condition all lead to phase pure, crystalline faujasite or zeolite X. The average crystallite sizes (D) (Scherrer method) of the materials are UFA = 33.44 nm, AFA = 64.73 nm, AFAZ = 51.84 nm (Table 1).

$$\% \text{ Crystallinity} = \frac{\text{Sum total of relative intensities of fly ash zeolite}}{\text{Sum total of relative intensities of standard Zeolite X}} \times 100 \text{ ----(1)}$$

The crystallinity of acid treated fly ash zeolite is 100%. Further evidence for phase pure zeolite X was obtained from the small angle XRD depicted in Fig. 2. The XRD pattern of AFAZ in Fig. 2 is exactly similar to the pattern in Fig. 1b except that the highest intense XRD signal is obtained at $2\theta = 6.5$ which is an additional peak and is a definite characteristic of faujasite mineral [15].

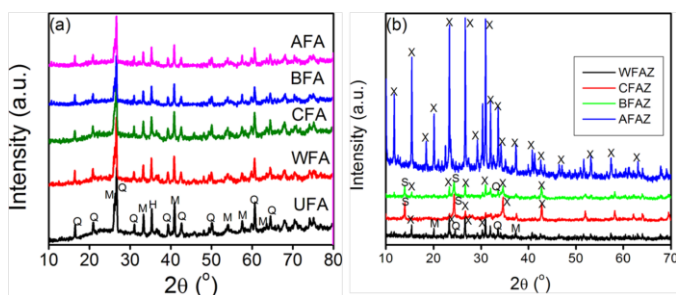


Fig. 1 XRD patterns of (a) unpurified fly ash (UFA), pre-treated FAs (WFA, CFA, BFA and AFA) and (b) water treated fly ash zeolite (WFAZ), calcined fly ash zeolite (CFAZ), base treated fly ash zeolite (BFAZ) and acid treated fly ash zeolite (AFAZ)

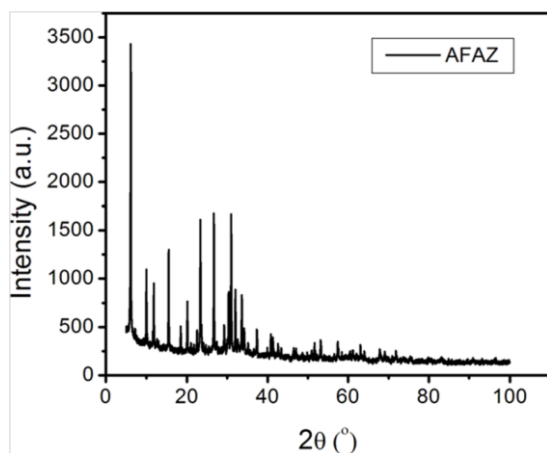


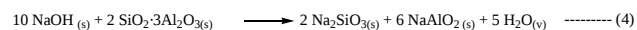
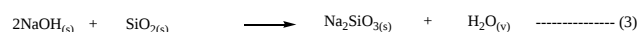
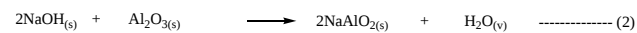
Fig. 2 XRD pattern of acid treated fly ash AFAZ for the angle 5 to 100°

3.1.2.1 Reaction Mechanism in Zeolite Synthesis

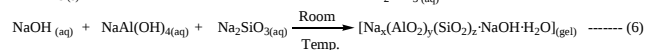
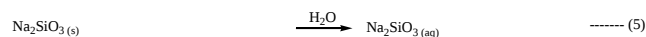
Synthesis of zeolite from treated FA involves three steps [15]. 1. In the fusion step with NaOH at 550 °C the fly ash is transformed into amorphous sodium silicate and aluminosilicate. Crystalline compounds quartz and mullite in fly ash are decomposed completely in NaOH and they enhance the dissolution of fly ash. 2. The aging process is intended to form homogeneous slurry to facilitate crystallization. In the aging process, the fusion product in DD water results in the formation of solutions of Na, Si, and Al salts. The zeolitic phase does not form in the aging step; however,

this step plays an important role and facilitates zeolite formation. Direct hydrothermal treatment without aging step leads to slow nucleation and increases the crystal size and hence such process is not followed. 3. In the hydrothermal treatment the aluminosilicate gel is converted into pure crystalline zeolitic material in a short period and most of the impurities remain dissolved in alkaline solution [15]. The general reaction scheme describing the zeolite synthesis process is as follows:

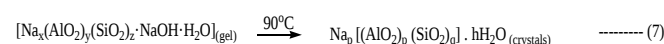
Step 1: Fusion at 550 °C



Step 2: Ageing and gelation in DD water



Step 3: Hydrothermal treatment and Crystallization



3.1.3 FTIR Studies

The FTIR spectra of UFA and treated fly ashes (WFA, CFA, BFA, and AFA) are shown in Fig. 3a. FTIR spectra indicate that treated fly ashes have significant changes in relative intensities and width of vibration bands. FTIR spectra of WFA, CFA and BFA exhibit similar broad band at 1098 cm^{-1} which is attributed to the asymmetrical stretching of TO_4 (= SiO_4 or AlO_4). IR spectrum of AFA shows relatively sharp vibration bands with high intensity. Increase in its sharpness confirms acid treatment with avoidance of maximum impurities. FTIR spectral result of AFA coincides with XRF and XRD results of AFA.

Fig. 3b shows the FTIR spectra of WFAZ, CFAZ, BFAZ and AFAZ. These spectra are mainly located in the mid infrared region. FTIR spectrum of zeolite X exhibits two classes of vibrations: internal tetrahedral vibrations of the framework of TO_4 (where T= Si or Al) and vibrations related to external linkage between tetrahedra at 467, 563, 668, 755, 983 and 1645 cm^{-1} . Strong band at 983 cm^{-1} and moderate band at 755 cm^{-1} are attributed to Si-Al-O asymmetric stretching vibration and single four member ring (S4R) TO_4 external linkage symmetric stretching vibration respectively [15]. Typical band of zeolite X at 668 cm^{-1} corresponds to internal tetrahedral symmetric stretching vibration of (Si, Al) O_4 . Transmission peak at 563 cm^{-1} is attributed to the double six member rings (D6R) T-O-T symmetric stretching and high intense peak at 467 cm^{-1} is assigned to TO_4 S4R symmetric bending vibration. The strong band at 1645 cm^{-1} indicates to H-O-H bending mode of water molecule. These observed sharp bands demonstrate that amorphous fly ash materials are converted into crystalline zeolite materials. The most distinct changes in the FTIR spectra of treated fly ashes and zeolitic products are (i) band at 1098 cm^{-1} in treated fly ash is shifted to lower wavenumber 983 cm^{-1} indicative of the formation of zeolites and (ii) appearance of sharp peaks, particularly in 1000-400 cm^{-1} region [15, 24, 25]. IR spectrum of acid treated fly ash zeolite (AFAZ) shows sharp and high intense bands of FAU-type zeolite X compared to other zeolites WFAZ, CFAZ and BFAZ. IR spectral study, thus, indicates the distinct nature of AFA and the corresponding AFAZ zeolite X. Acid treatment and the removal of impurity mineral phases from FA and the formation of pure crystalline faujasite are all confirmed.

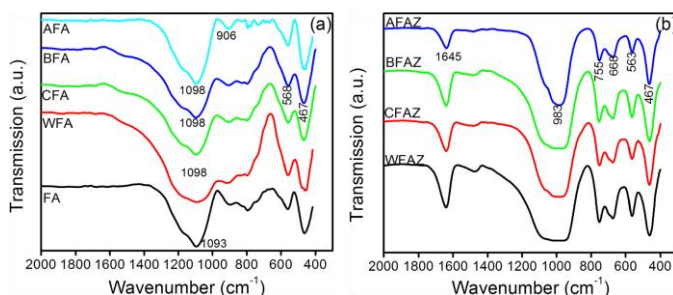


Fig. 3 FTIR spectra of (a) unpurified fly ash (UFA), pre-treated FAs (WFA, CFA, BFA and AFA) and (b) water treated fly ash zeolite (WFAZ), calcined fly ash zeolite (CFAZ), base treated fly ash zeolite (BFAZ) and acid treated fly ash zeolite (AFAZ)

3.1.4 SEM-EDX Studies

Fig. 4 shows the SEM images of fly ash and zeolites WFAZ, CFAZ, BFAZ and AFAZ along with reference material zeolite X. The raw coal fly ash consists of smooth-surfaced spherical particles with a broad particle size distribution in the range of submicron to 2 μm (Fig. 4a) [25]. Contrarily, zeolite has marked changes in the surface morphology due to alkali activation. The smooth spherical particles of UFA are reformed into agglomerations of roughly spherical shape. 1. WFAZ has smaller secondary particles of about 1.0 μm while CFAZ has the largest aggregate (Fig. 4b and c). Nevertheless, spherical primary particles of about 1.0 μm size are clearly seen in the image of CFAZ. Probably the pre-calcination has induced particle-particle adhesion and caused extensive agglomeration. BFAZ has morphology (Fig. 4d) very similar to that of WFAZ. In contrast, SEM image of AFAZ (Fig. 4e) shows white shiny crystalline grains with some agglomeration of primary particles observable as white dots/patches [15]. Yaping et al [26], Ojha et al [15], Chenfeng et al [23] and Thuadaij et al [24] have showed that zeolite NaX has octahedral crystals with particle sizes ranging from 0.5 – 1 μm . 2. Present work also reports similar grain size. SEM image of AFAZ (Fig. 4e) is well matched with SEM image of Zeolite X from Chenfeng et al [23]. The absence of spherical particles in zeolites indicates conversion of fly ash into highly crystalline zeolites on hydrothermal treatment. This shape transformation may provide large surface area with tiny pores and the same is discussed next.

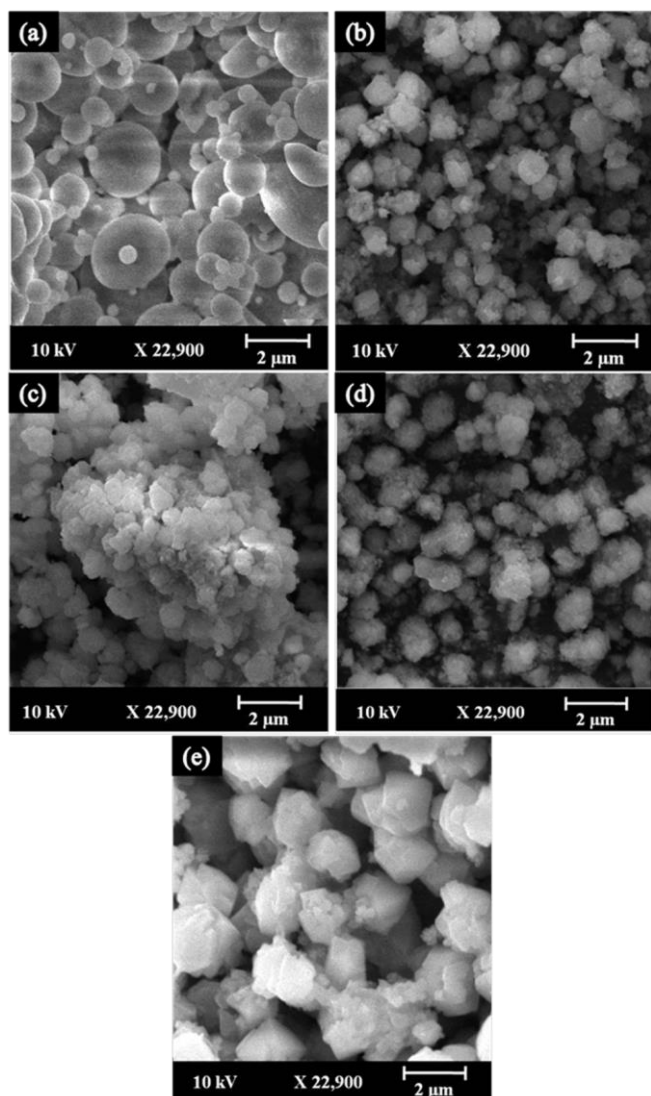


Fig. 4 SEM images of (a) UFA (b) WFAZ (c) CFAZ (d) BFAZ and (e) AFAZ

The EDX spectra of UFA and AFAZ are shown in Fig. 5. The results show sharp signals corresponding to all the expected elements. Thus EDX analysis provides evidence for the elemental composition of untreated fly ash and synthesized zeolite materials (AFAZ).

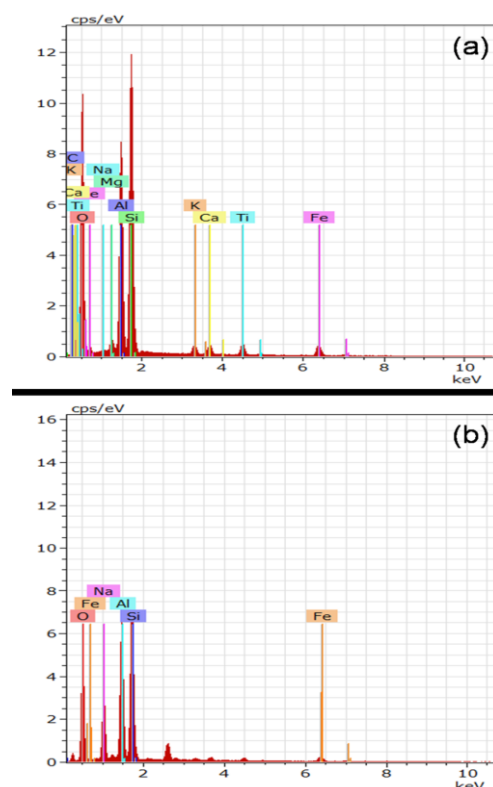


Fig. 5 EDX spectra (a) UFA and (b) AFAZ

3.1.5 BET Studies

The surface area, pore volume and pore size of FAs and zeolites were studied by BET N_2 adsorption/desorption method (Table 2). The specific surface area of zeolites are many folds higher than that of fly ash (UFA). The rough and irregular surfaces of the zeolites results in a significant increase in surface area and pore volume compared to the treated fly ash particles. Especially, AFAZ has the highest surface area of all zeolites (more than 10 times). This may be attributed to the surface modification of FA by acid solution and increase in micropore after dissolution of metal oxides. Tiny pores which probably exist in large volumes inside the particles / grains may increase the surface area. All the zeolite products provide micropore structures with one broad capillary condensation step at P/P_0 of 0.05-0.95, representing the broad pore size distribution which is normally observed with zeolite X [24, 26]. This indicates that AFAZ is zeolite X or faujasite type. Surface area and pore properties of AFAZ are thus distinct from those of other zeolites, an observation in consistent with other characterizations of XRF, XRD, FTIR and SEM.

Table 2 Surface area, Pore volume and Pore radius of UFA, WFAZ, CFAZ, BFAZ and AFAZ

Material	Surface area (m^2/g)	Total pore volume (cm^3/g)	Average pore radius ($\text{\AA}$)	P/P_0
UFA	0.882	0.003	183.33	0.99
WFAZ	34.42	0.078	39.81	0.99
CFAZ	31.19	0.108	58.93	0.98
BFAZ	27.13	0.067	Nil	0.98
AFAZ	413	0.221	29.06	0.98

4. Conclusion

The impact of pretreatment (purification) process using water, calcination, NaOH and HCl of fly ash has been evaluated on the synthesis and properties of zeolite. It is found that acid treatment of fly ash removes maximum impurities. Highly crystalline, pure or single phase faujasite type zeolite NaX (AFAZ) with crystal structure was obtained from acid treated fly ash (AFA) and is confirmed by XRF, XRD, FTIR and SEM micrographs. BET surface area of acid treated fly ash converted zeolite (AFAZ) is very high compared to other zeolites. All characterizations establish that acid treatment of FA followed by alkali fusion and hydrothermal treatment at specified condition facilitates formation of pure, single phase faujasite type zeolite crystals. Thus the present work contributes a new development / synthesis process for crystalline, phase-pure faujasite type zeolite material.

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