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Synthesis and characterization of zeolite-Y from natural clay of Wadi Hagul, Egypt

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Abstract

Zeolites were synthesized from alternative cheap raw material as clay minerals which used as a common source for Al_2O_3 and SiO_2 for synthesis zeolite-Y. Synthetic zeolite-Y has many characteristics, high purity and larger cavities that distinguish it from its counterparts. This study concerned with evaluation of the viability of producing low-silica zeolite through the process of hydrothermal calcination (metakaolinization) of raw material (kaolin) from Wadi Hagul, Egypt at 800ºC for 6 h. Metakaolinite was synthesized under hydrothermal treatment with 1.0 M NaOH solution without additional any catalysts as silica source at 90ºC/2d under atmospheric pressure. Zeolite Na-Y of molar ratio of 2.23 was achieved. The Cation Exchange Capacity (CEC) test showed an impressive value of 401.09 meq/ 100 g clay for prepared zeolite-Y. The raw kaolin, metakaolin and zeolite were characterized by XRF, XRD and SEM. The results confirmed that Zeolite was successfully produced from the raw material extracted from Wadi Hagul with an ideal percentage which approximately equal to 100% of the raw material used. The synthetic zeolite-y can be applied for remediation of wastewater and polluted soil.

Keywords: Clay minerals; Kaolinite; Hydrothermal reactions; Calcination; Zeolization; Zeolites; Remediation

1. Introduction

Zeolites are highly crystalline of microporous aluminosilicate minerals that used in many industrial applications. Their unique selective ability is the direct reflection of their microporous crystal structure that has uniformly sized interconnected pores, and cavities [1-4]. The cation presence is needed in the zeolite framework to balance the negative charge created as a result of isomorphous substitution of Si^{4+} by Al^{3+} in the framework.

This cation's mobility is mainly accountable for zeolite's distinctive ion exchange and catalytic characteristics [5-7]. There are over 150 synthetic zeolites and about 40 naturally occurring zeolites are known. Initially, only natural zeolites were used, but more recently synthetic forms have been made on industrial scale. Faujasite class of zeolites (zeolite- X and zeolite-Y) are known for remarkable stable and solid structure with the large empty space [8]. Zeolite-Y is a crucial active component in catalysts for its uniform cavities, high activity and good stability. It is widely used in the catalytic cracking; hydro racking and isomerization process [9]. Zeolite-Y is a faujasitic structure in which a sodalite cage is linked with double 6-rings resulting in a super cage with 12 rings pore opening. It has 3-dimensional

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channel system and can be imagined as stacking layers of sodalite cages [10].

Zeolite-Y has Si/Al ratio from 2-5 [11, 12]. Commercial synthesis of zeolite-Y was pretended by [13] after the first industrialization of zeolite A and X types [14, 15]. Zeolite Na-Y appears to be topologically identical with the type X aluminosilicate framework. The cubic unit cell of all these alumosilicates contains 192 (Si, Al) O_4 tetrahedrons,Breack [13] suggested that the change of zeolite- X to Y modulation occurs at a silicon to aluminum ratio of 1.5. More studies on synthetic zeolites currently are focused mainly on the use of cheap initial materials [16]. The basic raw material used to obtain zeolites with low content of silicon dioxide containing (like Na-X and Na-Y) kaolin. Kaolin belongs to the large groups of minerals known as clays [17]. The quality and purity of zeolite synthesized is governed by the nature of raw materials used. Clay minerals have been used as a common source for Al_2O_3 and SiO_2 for the synthesis of zeolite. However the production of zeolite from cheap raw material is an economic importance [2, 6]. Kaolinite as clay minerals is profusion worldwide and has a Si/Al ratio similar to that of zeolites and can be modified into zeolites by hydrothermal action [18, 19].

There are two basic steps for obtaining zeolites from kaolinite: the first step is thermal treatment of kaolinite to obtain an amorphous material called metakaolinite which involves to removes a considerable amount of quartz, a major impurity present in kaolin and the second step is the hydrothermal reaction of the metakaolinite with an aqueous alkali medium at a significantly low temperature and for a short time to form the zeolite structure [2,4,5,20]. Kaolinite is a kind of clay mineral with the silica: alumina molar ratio of 1: 1 and it is available as natural form in abundant amount, and it is very unreactive in its natural form and typically transforms during the process of zeolitization into hydrosodalite [21]. Metakaolinization offers a step towards increasing the reactivity of low-cost material to synthesize various zeolites.

Metakaolinization has been discussed by several research studies, the variety of compositions of kaolin over the world mean that it is important to explore the metakaolinization of kaolin before its use in the synthesis of zeolites [22]. However, there is disagreement on the conditions required to produce metakaolin [5]. Several studies concur on the use of 700–950°C as the temperature to achieve metakaolinization in the synthesis of different zeolites [5, 23, 24]. The time of exposure of kaolin at 900°C varies from one study to the others. The effect of calcination temperature on the zeolite synthesis increases the temperature of metakaolinization beyond 700°C can eventually lead to a higher yield of zeolite Na-X- (a high alumina faujasite) [10, 25, 26]. In this study Zeolite-Y was synthesized by using one of the cheapest natural clay minerals (kaolin) which available in Wadi Hagul, Suez, Egypt it has been carried out to prepare zeolite-Y from kaolin which had the SiO_2/Al_2O_3 ratio approximately 2.43. The time of the treatment was two days using a dry oven at 90ºC, this treatment without using any catalysts as silica source [27, 28]. All characterizations were studied by XRD, XRF and SEM.

2. Materials and methods

Experimental for the synthesis of zeolite-Y

Kaolin preparation

Raw representative sample of natural, poorly crystalline clay mineral (kaolin) was collected from Wadi Hagul, Suez, Egypt Latitude 29˚ 44' 13.5" N and longitude 32˚ 14' 08.2" E. The measured molar ratio of the major oxides (SiO_2/Al_2O_3) present in the kaolin is 2.43, which nearly the ideal ratio for obtaining low-silica zeolites as mentioned by [28, 29] Raw samples were air dried to prepare by grinding the rocks to fine powder by using standard sieves to obtain the fine powder less than (40 µm). The location of the study area is shown in (Fig. l).

Calcination (metakaolinization)

Metakaolinite produced by calcination of Egyptian kaolin at 800ºC/6h in muffle furnace decomposition occur leading to the destruction of the structure and removal of the unfavorable volatile matter. Through this process kaolin converted into the more reactive amorphous metakaolin phase [3, 29-31].

Zeolization

Metakaolinite and (1.0M and 3.0M) NaOH solutions were used as starting material without any additional silica to the reaction mixture [32]. NaOH is utilized as homogeneous catalyst which is a non-renewable and becomes consumable chemical in the process [21]. The solid/liquid ratio of metakaolinite to alkaline solution was (10 g/100 mL) in covered Teflon cups.

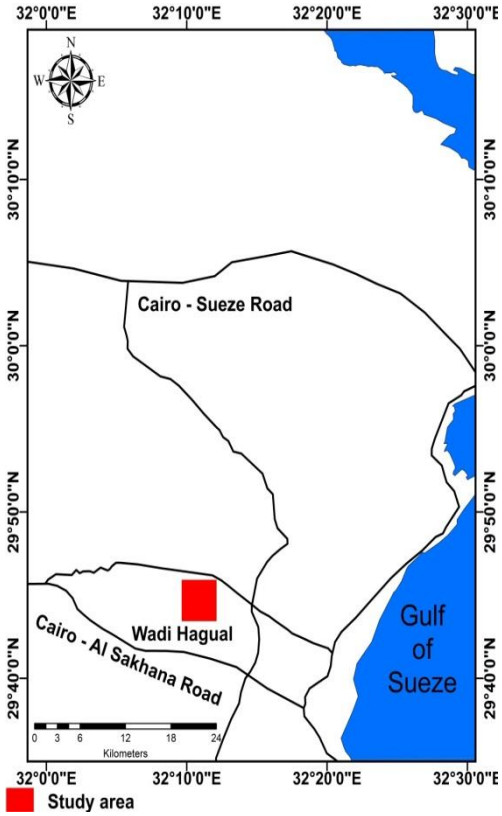

Fig.1. Location of clay mineral (kaolin) in Wadi Hagul, Suez, Egypt

The suspension was stirred at 800 rpm at room temperature for 24 h to obtain a homogenous fusion of solid as presented in [1-3]. The slurry was transformed to the dry oven for treatment at 90°C /2d under ambient pressure to synthesize zeolite-Y [33]. The residue product was washed several times with distilled water to remove excess alkalinity by using centrifuged several times with distilled water to obtain the (pH less than 8). The residue was dried overnight at 100ºC collected and utilized to different characterizations [34, 35]. The steps of synthesis zeolite are shown in (Fig.2).

Characterizations

Chemical and mineralogical composition of raw material (kaolin), metakaolin and final zeolite product were determined by X-ray fluorescence (XRF) instrument model AXIOS, WD-XRF Sequential Spectrometer (for the elemental composition) [36, 37]. (XRD) X-ray diffraction (for the crystaicllographic structural of the entire component in a material) using an Empyrean PANalytical diffractometer with Cu, Kα radiation at 40 kV and the current was 30 mA. The diffractograms were recorded in 0.0263º2θ. The diffraction data were analyzed of both the kaolinite and the resulting zeolite product and the morphology was examined using scanning electron microscopy (SEM). The Scanning Electron Microscope using SEM Model Quanta 250 FEG (Field Emission Gun) attached with EDX Unit (Energy Dispersive X-ray Analyses), with accelerating voltage 30 K.V., magnification14x up to 1000000 and resolution for Gun.1n.

3. Results and discussion

Chemical and mineralogical analysis

The chemical composition (in wt. %) of kaolin before and after calcination and synthesis zeolite contains SiO_2, Al_2O_3, Fe_2O_3, TiO_2, MgO, CaO and K_2O which obtained from XRF analysis was summarized in Table 1. Zeolite–Y is directly prepared from metakaolinite without any additional silica [8, 32, 36]. It is obvious that the clay is mainly composed of Si and Al who contains also iron oxide affects greatly the brightness of the product. The iron oxides, probably present as hematite, were not identified by XRD. The synthesis zeolite from kaolin formed in two steps, the first step is the metakaoliniziation described in Equation1 [37] and the second is zeolization which presented in Equation 2. The volatile matter of the clay developed and di hydroxylation takes place in the course of calcination at 800°C/ 6 h. Calcination process converts kaolin into metakaolin by removal of structural OH ion making it reactive.

Kaolinite Metakaolinite

$$2Al_2Si_2O_5(OH)_4 \xrightarrow{>550°C} 2Al_2Si_2O_7 + 4H_2O \qquad (1)$$

(Fig. 3a) shows the XRD pattern of raw material (kaolin) it is contains kaolinite (K), Na-montmorillonite (Mo) and quartz as major impurities (Qz) with their main peaks at 2θ = 21º, 26.8º and 36.5º, while the kaolinite and montmorillonite peaks are detected at 2θ = 12.5 º, 25 º, 35.2 º and 5 º, 20,25 º.5, respectively. The kaolinite structure breaks down during the heating and the clay becomes an amorphous but highly reactive and metastable phase that a very sufficient source for synthetic zeolite. This amorphous form the clay is known as me-takaolin [5, 27]. The metakaolinization of kaolin by heating for 6 h at 800ºC resembled all the kaolinite peaks expect for the peaks due to the admixed impurities.

Table 1. The chemical composition of kaolin, metakaolinite and synthesis zeolite-Y

Main Constituents	Clay mineral (Kaolin)	Metakaolinite	Synthesis zeolite-Y
	(Wt. %)	(Wt. %)	(Wt. %)
SiO_2	48.58	55.93	39.46
TiO_2	2.77	1.33	1.46
Al_2O_3	19.96	24.47	17.68
Fe_2O_3	16.10	10.92	13.80
MnO	0.04	2.11	0.02
MgO	0.64	N.D.	1.41
CaO	1.33	1.03	0.86
Na_2O	0.08	1.26	7.79
K_2O	0.88	0.67	0.53
P_2O_5	< 0.01	0.09	0.05
Cl	0.51	0.07	0.04
SO_3	0.03	0.01	0.02
L.O.I*	8.84	1.90	16.59
SiO_2/Al_2O_3	2.43	2.29	2.23

(Fig.3b) shows that the characteristic peaks of kaolinite disappear during the calcination step and reduce in peaks intensity of quartz. The small zigzag peaks found in the spectrum refer to the presence of amorphous material. Because of its disordered composition, metakaolin is more reactive than kaolin. Kaolin thermal treatment leads the kaolinite crystal lattice to collapse, forming a structure that is extremely highly disordered and amorphous [38]. The crystallinity of the zeolite-Y increase with increasing the time at 2d of metakaolinite [20, 33]. The only crystalline form in the metakaolin was quartz, as the thermal treatment during calcination does not affect quartz [22]. Synthesis of zeolite-Y depending on the SiO_2/Al_2O_3 molar ratio can be considered as a zeolite-Y if SiO_2/Al_2O_3 up to 2 according to Equation 2 [7, 39]. Two different concentrations of NaOH (3M) at 90°C/2d) and NaOH (1.0 M) at 90°C/2d) used to produce the synthetic zeolite-Y. (Fig. 3c) shows the peaks of zeolite-Y with small peaks of zeolite-P. The zeolite Y in pure phase was crystallized at 90ºC at a time of crystallization of 48 hours, and this result is agreement with Ayoola et al., and Belaabed et al., [20, 40]. Zeolite formation at the concentration NaOH (1.0M) 90°C/2d was observed from (Fig. 3c). The chemical formula of

Zeolite Na-Y is: $Na_{60}\ H_{32}\ (Si_{100}\ Al_{92}\ O_{384})$

Zeolite- P is: $Na_6\ Al_6\ Si_{10}\ O_{32} \cdot 12\ H_2O$ is

The transformation equation from metakaolinite to zeolite-Y is:

$$Al_2O_3 - 2SiO_2 + 2NaOH \rightarrow Na_2O - Al_2O_3 - 2SiO_2 - 4.5H_2O \quad (2)$$

Scanning Electron Microscopy (SEM)

The SEM micrographs and crystallite size of the synthesized material was analyzed using Scanning Electron Microscopy (SEM). (Figure 4) shows the evolution of zeolite Na-Y characteristic cubes from the extensively reacted metakaolinite platelets after 2d, the cubes produced as minute crystals with rounded-edges (cube edge) which then grew with time to bigger euhedral-cubes to reach a larger size at the expense of the original platelets.

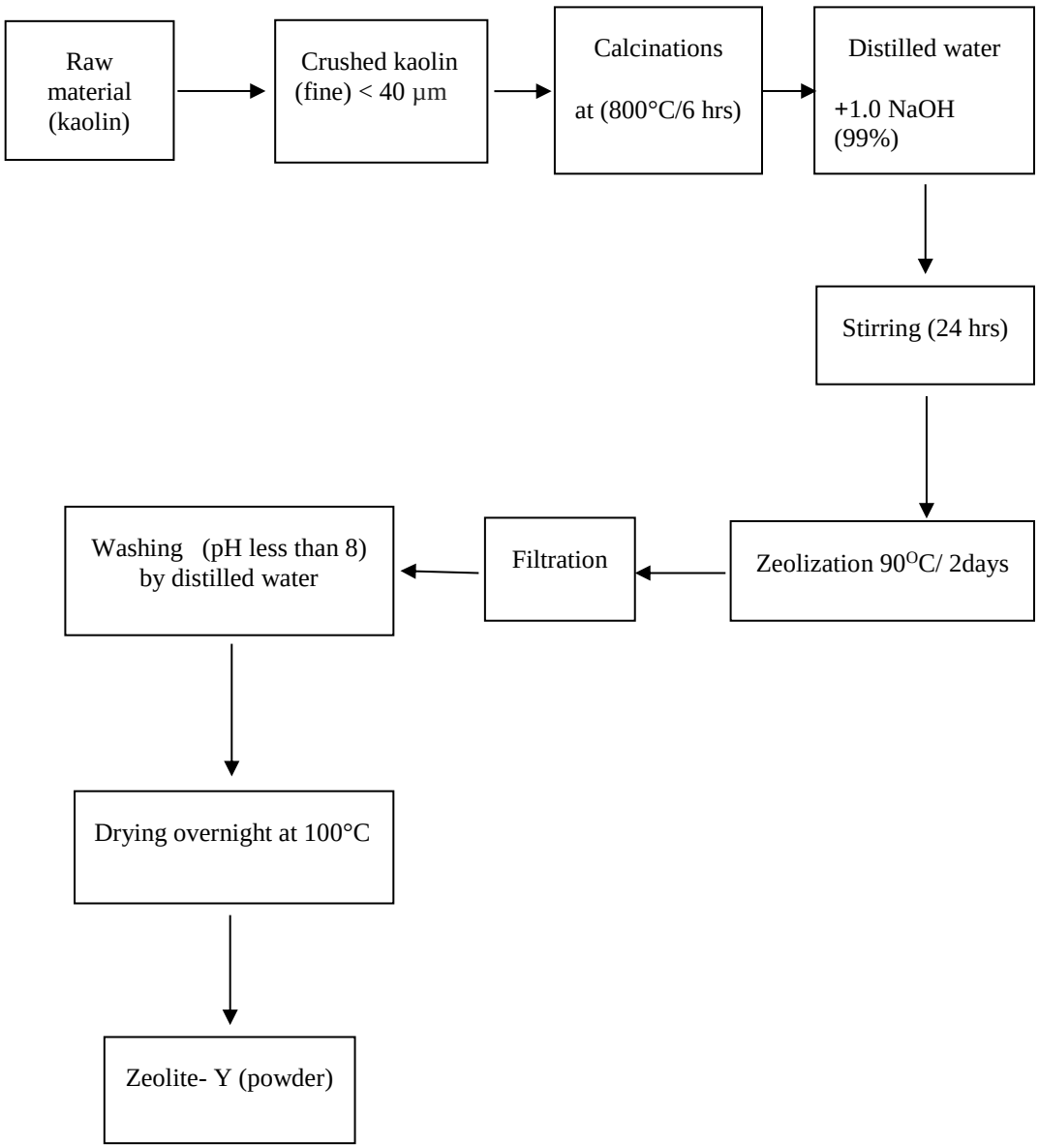

Fig.2. Schematic diagram for the preparation of zeolite -Y from kaolin

At the later stages of crystallization, the well-defined crystals appear of perfect cube edges with abundant quantities from very small amounts of metakaolin debris indicating the final phase of zeolite-Y crystallization [35. 37]. SEM images also show the Na-Y crystallite which appears as an octahedral structure, in uniform aggregate size and some crystallite cluster to form large aggregates. The crystallites of zeolite-Y were obvious from SEM images obtained and compared with similar images of zeolite-Y in previous studies [41, 42]. The presence of remaining kaolin as a result of incomplete dissolution which was indicated by XRD analysis was observed as a thin layer occurs around the Na-Y crystals. It should be noted that the incomplete dissolution of kaolin also affected the Si/Al ratios of the synthesized zeolite-Y and the general picture of the synthetic zeolite shows the cavities between the grains at (200 µm).

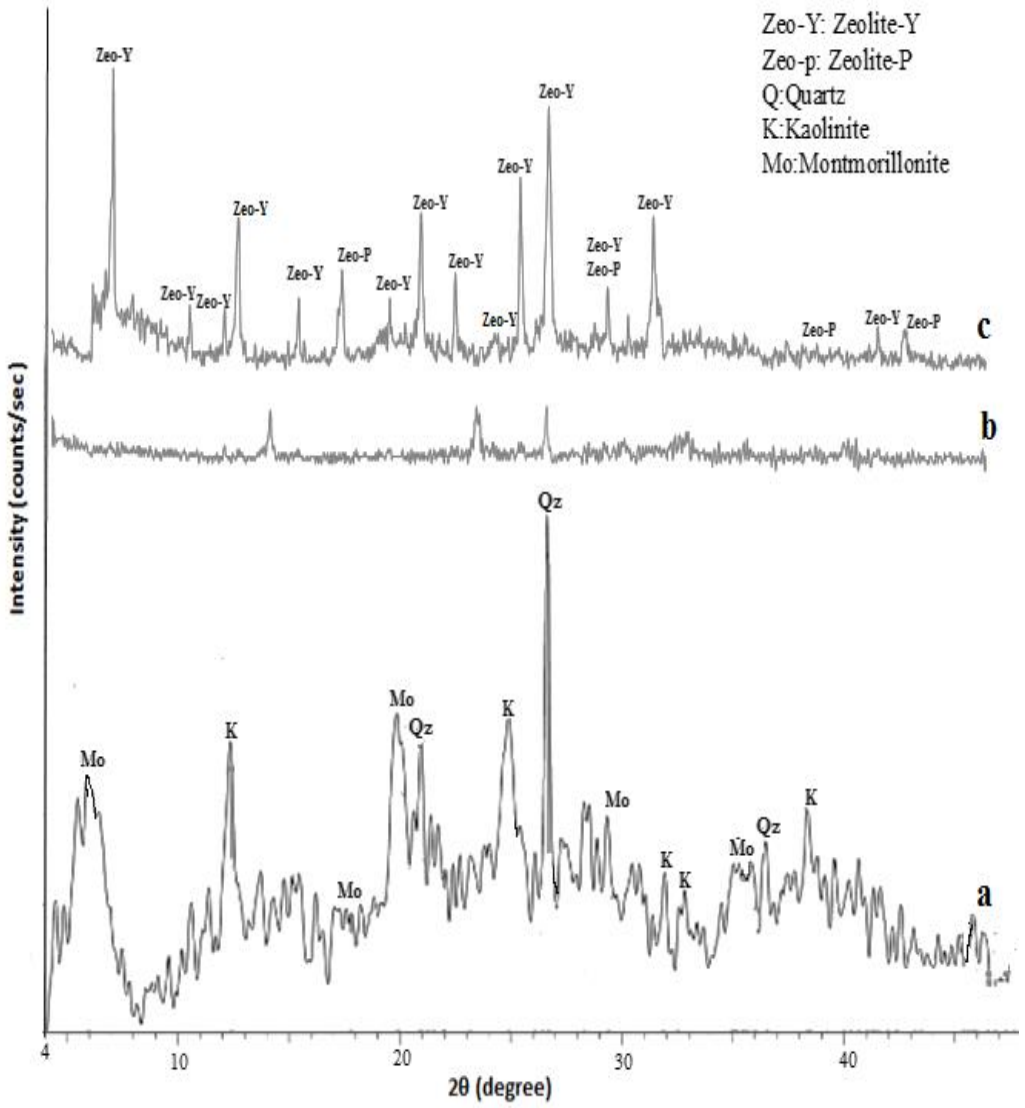

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Fig. 3.

XRD pattern of raw material (kaolin) (a), metakaolinte (b), and synthetic zeolite–Y (Zeo-Y) (c)

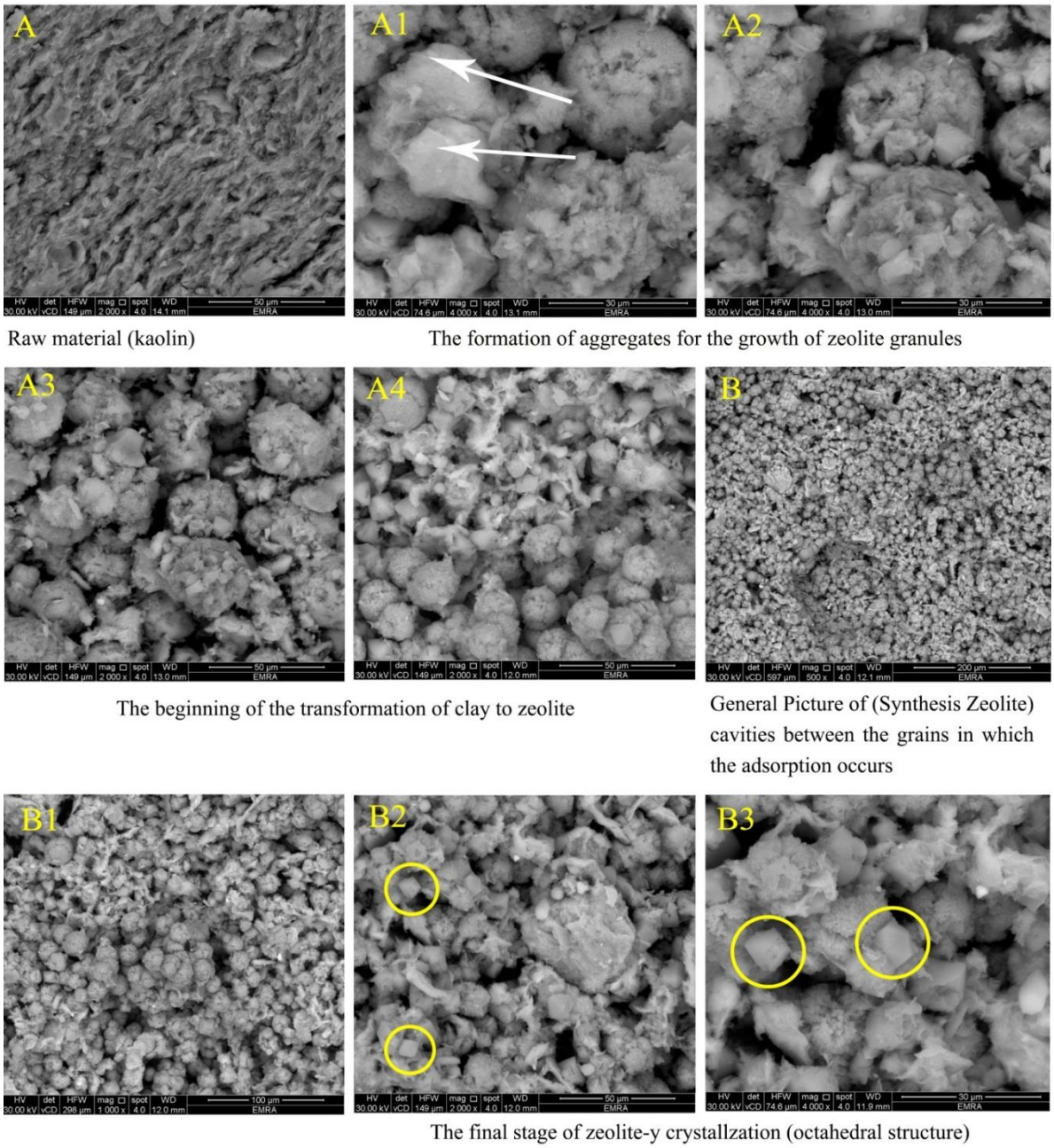

Fig. **4.** SEM micrograph hydrothermal conditions with NaOH (1.0M) / 90ºC /2days; (B, B1) cavities between the grains of zeolite; (B2, B3) crystal of synthetic zeolite Na-Y (octahedral structure).phs of the prepared zeolite A, A1 from kaolin raw material; (A2, A3, A4) metakolinte under

4. Conclusions

This study discussed the production of zeolite-Y from natural Egyptian kaolin in Wadi Hagul , Suez Governorate. Using dry oven instead of microwave and without any catalysts as usual in previous research. The calcination of kaolin was prepared at 800ºC/ 6 h. with 1.0 M NaOH, (pH less than 8). Metakaolin crystalline was formed at 90ºC/ 2d for synthesize of zeolite-Y. It has been successful in producing zeolite-Y with a high efficiency from the raw material and in less time than normal in this treatment, where it reached about two days at most. Zeolite-Y has been characterized by XRD, XRF and SEM analysis techniques as previously mentioned and illustrated in the study. From SEM it was noted that the crystals formed characterized with high quality in form, size and identical to the known form of zeolite-Y. CEC was 401.09 meq/100g clay. Synthetic Zeolite-Y can be used efficiently to remediate the wastewater and waste soil in the future study.

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