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Synthesizing of Zeolite Particle Using Alkaline Plant Extract

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Abstract: The study explored the hydrothermal synthesis of zeolite using kaolin clay precursor in the presence of three different solvents namely, garlic extract, watermelon extract and 2M NaOH solution, with the view of testing their efficacy and potency for green synthesis. Before the zeolitization, the kaolin precursor was activated at a temperature of 850°C to produce metakaolin. The crystallization was achieved in an oven at a temperature of 90°C for 12 hours. The final product is grounded to a fine powder and subsequently undergo testing and characterization via X-ray Diffraction (XRD) and Field Emission Scanning Electron Microscopes (FESEM) methods. The purity and grade of the synthesized products vary accordingly with the suitability of the reaction solvent. The synthesized product using a solvent of watermelon plant extract was able to produce zeolite LTA of a good grade. However, the presence of secondary phases informed the effect of the accompanied impurities that might originate from the kaolin precursor or the solvent. The result also portrays the possibility of obtaining a well crystalline zeolite from the Malaysian kaolin without using any structural directing agent or chemical solvents.

Keywords: Zeolite particles, plant extract, synthesis

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1. Introduction

Zeolites are known to be "fully cross-linked crystalline, microporous, alumino-silicate materials." This class of materials has three-dimensional open-framework structures that form a uniformly sized pore of molecular dimensions. Zeolite minerals are commonly differentiated by their chemical compositions, as well as the differences in size and the arrangement of their crystal structures [1, 2].

The zeolite mineral has a huge industrial, scientific and academic, interest in the areas of ion exchange (treatment of liquid waste, detergent industry, and radioactive waste storage), petroleum refining along with petrochemicals, coal and fine chemical industries [3, 4] and separation (purification, drying, and environmental treatment). In a nutshell, zeolite can be considered as an eco-friendly alternative to many addictive minerals and products.

The ability of zeolite to act as multi-functional materials in many industrial applications is due to their inherent properties such as uniform pore size/shape, catalytic activity, mobile cation and hydrophilicity/hydrophobicity [5]. The pore size of a zeolite plays an important role in adsorbing molecules of different sizes, like a molecular sieve. Therefore, molecules of a certain size are adsorbed by a zeolite and others which are larger than the pore size are not adsorbed [6].

Zeolite is a good adsorbent of heavy metal ion and other toxic substances. Therefore, zeolite is a valuable material for use as an adsorbent or catalyst for reducing the hazard of produced waste from the ecosystems and aquatic environment [7].

Although there are varieties of techniques used to reduce the hazard of the produced waste from the ecosystems and aquatic environments such as vapor compression distillation (VCD) [8], electrodialysis (ED), [9] and membrane separation [10]. The operational cost of these technologies is still considered expensive and sometimes involve complex steps and process. The adsorption separation as reported by [11] is the most popular and invariably the cheaper and simple alternative methods.

On other hand, the conventional methods of producing zeolite are not green, because it heavily rely on the use of chemicals as solvents and templates [12] such as tetraethylorthosilicate (TEOS), sodium silicate and aluminum sulfate that are always accompanying with huge discharge of highly toxic waste to the environment [13] as well as high energy consumption especially during the removal of templates. Recent studies highlighted the potentials of synthesizing zeolite via green routes such as using clay precursor [14], recycled waste [7], and free template method [15].

However, there are no sufficient information in the open literature on using alternative reaction solvents for the zeolite synthesis. To that extent, this study employed the use of two different types of alkaline plant extract (watermelon and garlic extract) as the reaction solvent to produce zeolite particles and compared with the usual NaOH solution with the purpose to investigate the characteristics of the produced zeolite particle and towards achieving a green synthesis process.

2. Methodology

2.1 Materials

At the beginning of the experiment, the kaolin precursor was activated at 850 °C temperature to produce metakaolin following Abdullahi, Harun [16]. The watermelon extract was prepared by cutting the melon peel into small size and then blend to get the extract. Similarly, fresh garlic buds were washed, sliced, and blended to get the liquid extract.

2.2 Synthesis of the Zeolite Particles

Before the crystallization, a part of the activated kaolin was used to prepare the reaction mixture in a 100ml glass vessel containing a 75ml alkaline solvent of garlic extract, watermelon extract and 2M NaOH solution were used, with the view of testing their efficacy. The reaction mixtures undergo ageing for 12h at room temperature.

Immediately at the end of the ageing process, the reaction mixtures are transferred into three different 100ml Teflon-lined autoclaves. The samples were crystallized in an oven at a temperature of 90°C for 12 hours. After the crystallization process, the white precipitates were collected and washed using deionized water for several times until the PH change to neutral and then, centrifuged at 7500 RPM for 10 min. At the end of the washing, the product obtained is dried in the oven at 60°C for 12 hours. The final product is grounded to a fine powder and subsequently undergo testing and characterization via X-ray Diffraction (XRD) and Field Emission Scanning Electron Microscopes (FESEM) methods.

3. Results and Discussions

The FESEM images shown in Fig. 1(a) described the raw kaolin material prior to the thermal activation. The sample consist of small flaky morphology kaolin particles. The EDX spectra in Fig 1 (b) shows the peaks corresponding to the element of the kaolin powder. From the spectra, Al, and Si elements were found at high strong peaks, that confirm the existence of kaolinite in the sample.

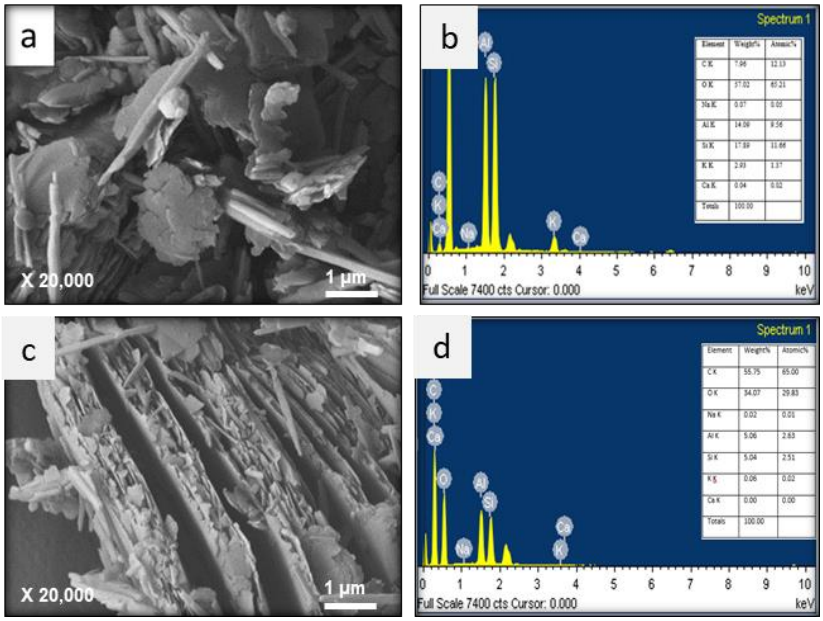

Fig. 1 - FESEM image and EDX spectra for (a-b) kaolin (c-d) metakaolin

On the other hand, Fig 1 (c) revealed the platy structure of the activated kaolin. The thermal activation of the kaolin has therefore produced a highly disordered metakaolin that is characterized by sheet-like morphology and a highly disordered and amorphous structure, which according to Abdullahi, Harun [17], Srilai, Tanwongwal [18] is necessary to attain as a precondition to successful crystallization of zeolite particles.

The metakaolin obtained at the end of the hydrothermal activation operation was used for the crystallization process. The crystallization was carried out at 90 °C for a time of 12 hours. The temperature for the crystallization was taken based on previous reported studies that indicated the minimum temperature for the structural transformation of metakaolin phase to hydroxy-sodalite phase [2, 19]. The purity and grade of the synthesized products vary accordingly with the suitability of the reaction solvent. The synthesized product using 75 ml 2M NaOH solution produced Zeolite LTA with 96.9% phase purity as shown in the X-ray diffractogram in Fig. 2(a). Further analysis revealed that quartz and indium chloride are the accompanied impurities in the produced zeolite Fig 2(b).

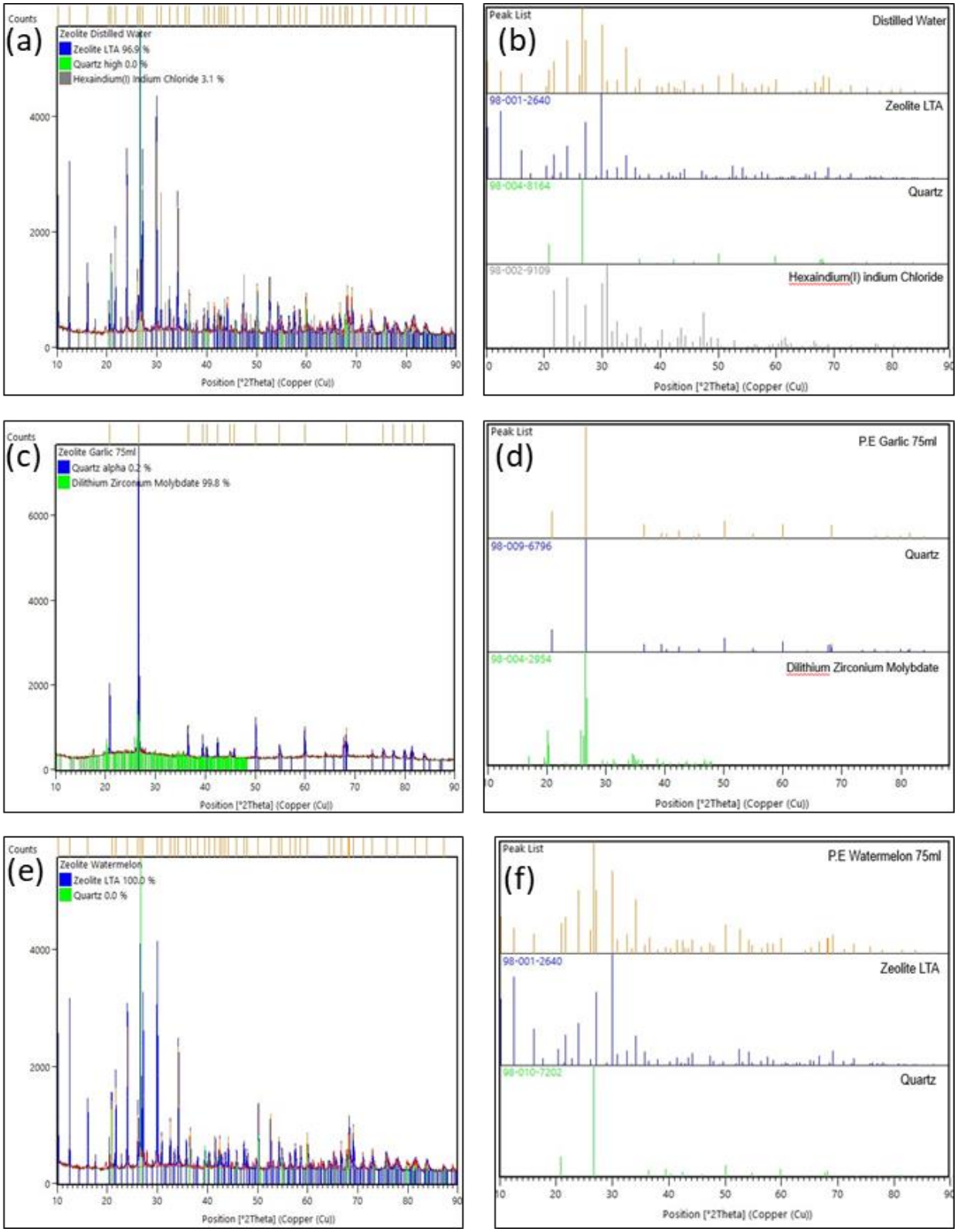

Fig. 2 - XRD Diffractogram of the synthesized product for (a-b) sample using 2M NaOH solution (c-d) sample using garlic plant extract (e-f) sample using watermelon plant extract

Fig 2(c-d) described the synthesized product using 75 ml garlic plant extract. The product yielded a high content of quartz and traces of dilithium zirconium. This implies that the solvent (garlic plant extract) was not able to provide the required alkaline environment necessary for the zeolite crystallization. This finding corroborates with that reported by Wang, Huang [19] where the authors emphasized that crystallization of zeolite particles from metakaolin can only be

successful when the reaction mixture is appropriately kept in an adequate alkaline environment with effective control of the temperature and time.

Similarly, the failure to produce the required zeolite under this experimental parameter can be rightly linked to the interaction between the main elements that build up the aluminosilicate structure of the zeolite. Regarding Fig. 3(d), the elemental weight % value of Al, Si and Na is 16.22%, 18.19% and 1.30% respectively. Compared with the synthesized product using 75 ml 2M NaOH solution which has the elemental weight % value of Al, Si and Na as 7.29%, 15.15% and 6.14% as shown in Fig. 3(b) as well as the product obtained from the sample of 75 ml watermelon plant extract having the elemental weight % value of Al, Si and Na as 6.48%, 18.28% and 5.24% respectively, Fig. 3(f). It is obvious that the sample made with the garlic plant extract was not able to develop the crystallization reaction that may yield the required result.

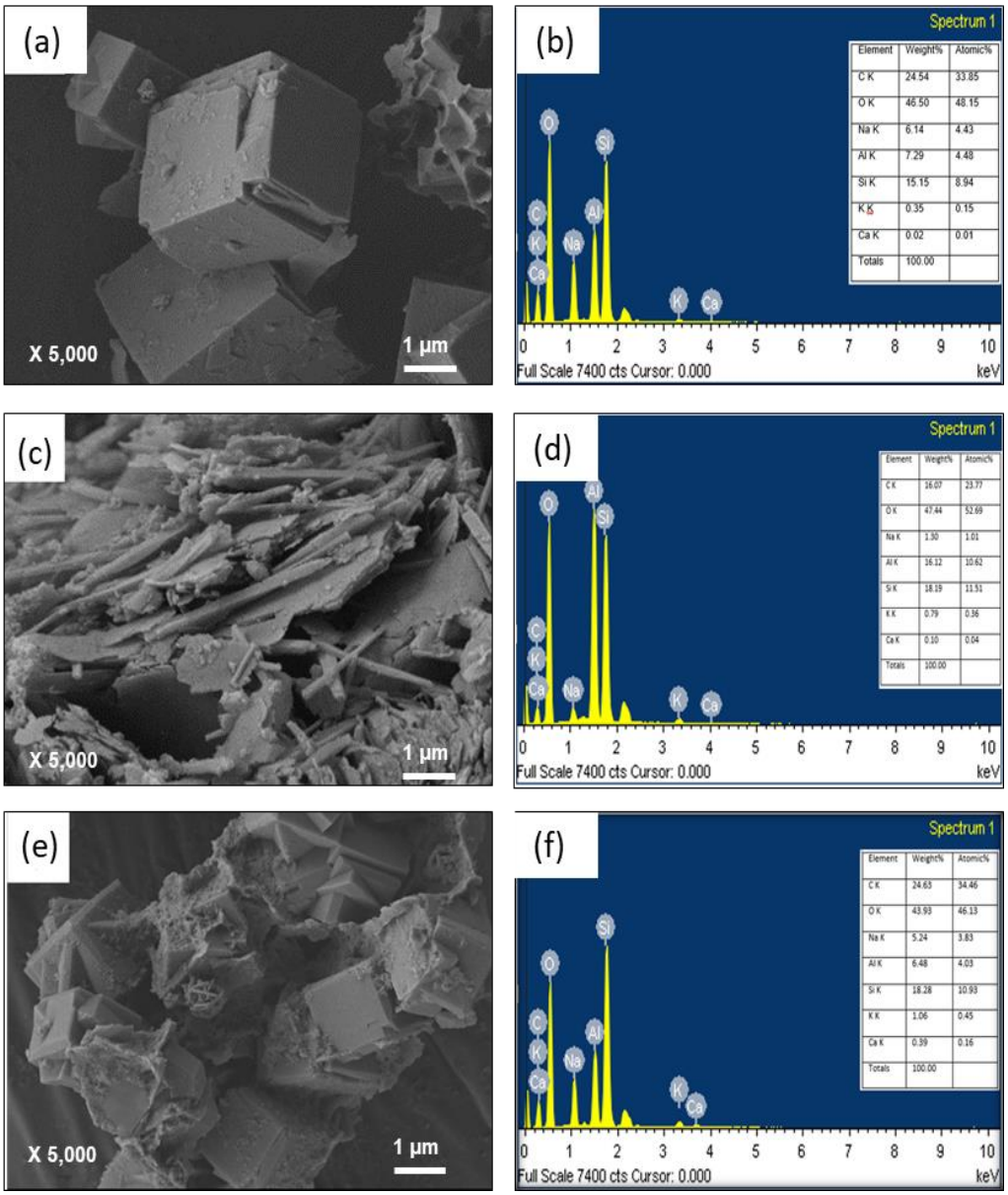

Fig. 3 - FESEM result of the synthesized product for (a-b) sample using 2M NaOH solution (c-d) sample using garlic plant extract (e-f) sample using watermelon plant extract

Considering Fig. 2(e-f) the XRD pattern shows that the zeolite obtained from the sample of watermelon plant extract has yielded a stable zeolite LTA with a little presence of quartz. Nonetheless, Fig. 3(e) revealed that the shape of the produced zeolite under this experimental parameter is not fully cubic as obtained previously by [7]. Thus, indicating that there is a possibility of hydroxy-sodalite secondary phase growth due to the presence of the sodalite cages in the synthesized product.

4. Conclusion

The synthesis process conducted using a solvent of watermelon plant extract was able to produce zeolite LTA of a good grade compared with the garlic plant extract. However, the presence of secondary phases informed the effect of the accompanied impurities that might originate from the kaolin precursor, leading to unsuitable interactions between the main elements that build up the aluminosilicate structure of the zeolite or from the solvents, which failed to provide the required alkaline environment necessary for the zeolite crystallization.

The result also portrays the possibility of obtaining a well crystalline zeolite from a low-grade kaolin using alkaline plant extract, without adding any structural directing agent or chemical solvents. Therefore, it is highly recommended to further study the influence of temperature and time in relation to the concentration of the alkaline plant solvents.

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