

Preparation, Characterization and Application of Zeolite-Y (Na-Y) for Water Filtration

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Abstract: Advancement and innovation in developing water filtering system enhances immensely in the field of water purification. In this study the acid treated rice husk ash was used as SiO₂ source in the preparation of zeolite Y, a medium siliceous zeolite used as an ion exchangers in water treatment process. Purity, pore size distribution, FT-IR and SEM micrographs of treated and non-treated samples are presented. The powdered (semi solid) zeolite Y products were characterized by XRD and FT-IR and pore size distribution was characterized by Micromeritics Instrument. Obtained zeolite Y, BET surface area 621.18 square meters per gram, pore volume is 0.33 cubic centimeters per gram and its average pore diameter is 21.22 Å. The DDW was produced by developed a new type of filtering system using most effective and highly efficient nano-materials. The aforementioned system involved coagulation, chlorination, sand treatment, and activated carbon purification and nylon polisher layers.

Key words: rice husk, zeolite Y, ion exchangers, highly efficient etc.

INTRODUCTION

Innovation in the development of novel technologies to desalinate water is among the most exciting and promising. Recent advances suggest that many of the issues involving water quality could be resolved or greatly ameliorated using nanoparticles, nanofiltration or other products resulting from the development of technology. Utilization of specific nanoparticles either embedded in membrane or on other structural media that can effectively, inexpensively and rapidly render unusable water to potable is being explored at a variety of institutions. In addition to obvious advantages for industrialized nation, the benefits for developing countries could also be enormous. Many factories generate large amount of waste water. Removal of contaminants and recycling of the purified water would provide significant reduction in cost, time and labour to industry and result in improved environment stewardship. Aquifer and groundwater remediation are also critical issues, becoming more important as water supplies steadily decreases and demand continues to increase. Most of the remediation technologies available today, while effective, very often are costly and time consuming, particularly pump-and treat methods. The ability to remove toxic compounds from subsurface and other environments that are very difficult to access in situ, and doing so rapidly, efficiently and within reasonable cost is the ultimate goal; N. Savage *et al.*, (2005) proved it. Nanoparticles have some properties that make them attractive particularly as sorbents i.e. large surface area than bulk particles. N. Savage *et al.*, (2005) can also functionalize with various chemical group to increase their affinity toward target compounds

Zeolite is a crystalline hydrated aluminosilicate of alkaline and earth metals. It is an effective sorbents and ion-exchange media for metal ions which was evaluated as an ion exchange media for the removal of heavy metals from acids mine waste water. Researcher, (Alvarez-Ayuson *et al.*, 2003, and N. Savage *et al.*, 2005) reported the successful use of synthetic NaPI Zeolite to remove Cr(III), Ni(II), Zn(II), Cu(II) and Cd(II) from metals electroplating waste water.

Zeolite can be naturally occurring or synthetic aluminosilicate of varying Si/Al ratio and channel structures. It was estimated by E. Izni *et al.*, (2007) that over 40 different kinds of natural zeolites and over 100 synthetics are found in various part of the world. However, synthetics zeolites hold some key advantage over their natural analogs. According to L. Tagami, *et al.*, (2001) zeolite can be manufactured in a uniform, pure form and require less time to manufacture in comparison to natural analogs which are rarely found in pure form and always contaminated to varying degree by other minerals such as Fe, quartz, amorphous.

Adsorption Property:

Zeolite is used as molecular sieve. Species with kinetic diameter which make them too large to pass through a zeolite pores are effectively sieved. This sieve effect can be utilized to produce sharp separations of molecules by size and shapes. However, a particular affinity a species has for an internal zeolite cavity is electronic considerations. Strong electrostatic field within a zeolite cavity results in a very strong interaction with polar molecules such as water. Furthermore, Non polar molecules are also strongly absorbed due to the polarizing power of this electric field. Thus, excellent separations can be achieved by zeolites even when no steric

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hindrance occurs. Therefore, adsorption based on molecular sieve, electrostatic fields and polarity ability accounts for the considerable economic values of zeolite in adsorptive application (*in the website*, 2009).

Ion Exchange Application:

Because cat ions are free to migrate in and out of zeolite structures, zeolites are often used to exchange their cat ions for those of surrounding fluids. The preference of a given zeolite among the available cat ions can be due to the ion sieving or due to a competition between the zeolite phase and aqueous phase for the cat ions that are present. Sodium zeolite A for example is among the world's most efficient removers of water hardness ions. This is its principle function as a detergent builder (*in the website*, 2009).

Zeolites make extremely active catalysts. Steric phenomena are very important in zeolite catalysis, and a new term "selective catalysis" was coined to describe these effects. Extremely selective reactions can be made to occur over zeolites, when certain products, reactants or transition states are kept from forming within the pores because of the size or shape.

Other Uses:

High heat water adsorption capacity enables zeolite to be used in refrigerators and to store energy during off-peak period and release it during peak period. Zeolites can also be used in refrigeration and air cooling system to reduce water in the air to very low concentrations, allowing very effective evaporative cooling to occur. Natural zeolites are used in the radioactive waste treatment to treat low and intermediate aqueous waste. It is also used in the municipal waste water treatment. However, natural zeolites are uniquely effective in adsorbing ammonia and hydrogen sulfide. This makes them usable in pet litter to prevent irritating odour (Schutte, 2001). In this study, the synthesis of zeolite Y using the rice husk ash as the main raw material was focused. In order to obtain the appropriate $\text{SiO}_2/\text{Al}_2\text{O}_3$ mole ratio, the sodium silicate solution was added to adjust the mole ratio of raw silica source. From the safety and economic point of view, the sodium silicate solution prepared from rice husk ash was used instead of commercial ones because there is an abundant supply of rice husk in Malaysia. By optimizing the anion concentrations of the reaction mixture (S. Mintova, *et al.*, 1999, Q. H. Li, *et al.*, 2002, Zhu, *et al.*, 1998, Mintova *et al.*, 1999 and Schoeman, *et al.*, 1994), they successfully minimize the size of highly crystalline zeolite Y nanocrystals, while simultaneously increasing the yield. Detailed characterization of the zeolite Y nanocrystals is also presented, this includes powder X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FT-IR), nitrogen adsorption, scanning electron microscopy (SEM), and inductively coupled plasma mass spectrometry (ICP MS).

MATERIALS AND METHODOLOGY

Preparation of Zeolite:

Zeolite Na-Y was successfully synthesized by seeding method as described below:

The preparation involved three major steps, namely: Seeding gel, Feedstock gel and Over all gel.

Seeding Gel:

NaOH pellets were weighed (1.6616g) and transferred to the PTFE beaker, distilled water (7.5mL) was added and continuously stirring with magnetic stirrer until a clear solution was obtained after dissolving the pellets. The aluminate solution was prepared by adding NaOH solution (2.0mL) to the sodium aluminate NaAlO_2 (0.7517g) followed by stirring and heating gently until the mixture becomes an apparent solution. The silicate solution was also prepared by mixing (1.536g) of silicate with NaOH solution in the PTFE beaker and subsequently stirred and heated on the water bath at boiling H_2O temperature. The aluminate and silicate were then mixed in the PTFE beaker and stirred for half an hour to achieve homogenization. Then the mixture was transferred to the Teflon bottle and capped for the ageing process to take place by leaving it at room temperature for 24hrs. Afterward, the loose brown gel was appeared and applied as seed gel for seeding of the feedstock gel (Nik Ahmad Nik Malek *et al.*, 2007).

The Feedstock Gel:

NaOH solution was prepared by dissolving NaOH pellet (7.7585g) with distilled water (142.5mL) in the PTFE beaker, stirred with magnetic stirrer until a clear solution emerged. For the preparation of aluminate (13.7711g) was dissolved in the NaOH solution (42.5mL) by stirring and heating gently on the hot plate until a clear solution appeared. In the preparation of silicate solution, NaOH solution (100mL) was added to the (28.1463g) of silicate in the PTFE beaker. The mixture was then stirred and heated in a hot water bath. The aluminate and silicate solutions were mixed together in the PTFE beaker, subsequently stirred for 2hrs with the purpose of making it completely homogenized. This combination of solution was used as the feed stock gel (Nik Ahmad Nik Malek *et al.*, 2007 and M Rahman *et al.*, 2009).

The Overall Gel:

This was prepared by mixing the feed stock and the seed stock. The feed stock was stirred magnetically and at the same time, the seed gel was being added slowly and was continuously stirred for 2hrs at room temperature. The mixture was then transferred into the Teflon bottle and left for ageing for 24hrs at room temperature. After ageing, the mixture in the Teflon bottle was heated in an oven at 100°C for 22hrs. Teflon bottle was taken out; the cap was quickly opened and left to cool to room temperature. Subsequently, the solid product was separated by sucking filtration and washing with hot distilled de-ionized water, then dried overnight in the oven at 100°C^[11]. Finally, the dried zeolite Na-Y was weighed and placed in the plastic bottle. The synthesis was repeated for 10 batches and was labeled according to the zeolite Na-Y. All the synthesized zeolite Na-Y samples were mixed together in the plastic bottles and closed tightly. The mixture was then homogenized for 12 hrs to ensure good homogenization of the samples. The homogenized sample was sieved (250mesh) to obtain desired size of the zeolite samples and ready to be used for characterization and modifications steps and was labeled as zeolite Na-Y gel (Nik Ahmad Nik Malek *et al.*, 2007 and M Rahman *et al.*, 2009).

RESULT AND DISCUSSION**Characterization of Zeolite Na-Y:**

In the identification of Zeolite Na-Y, the following analyses were conducted: Powder XRD (Siemens D-500 diffractometer using Cu K α radiation) was used to determine sample crystallinity. The formation of zeolite Y phase was confirmed by comparing the diffractograms of all the as synthesized samples to the diffractogram of the reference zeolite Y (B. Wang *et al.*, 1998 and Brett *et al.*, 2003). The as-synthesized Y sample gave similar scanning electron microscopy morphology and FTIR spectrum pattern to that of the reference sample zeolite Y (B. Wang *et al.*, 1998 and A. H. Brett *et al.*, 2003). FTIR spectrum, Diffractogram of XRD and morphology are given in the Fig. 1,2 and 3 respectively. Magnifying morphologies are shown in Fig. 3 (a and b) and it shows a very clear and sharp crystallinity. These indicate that the as-synthesized Y sample has a zeolite Y like structure.

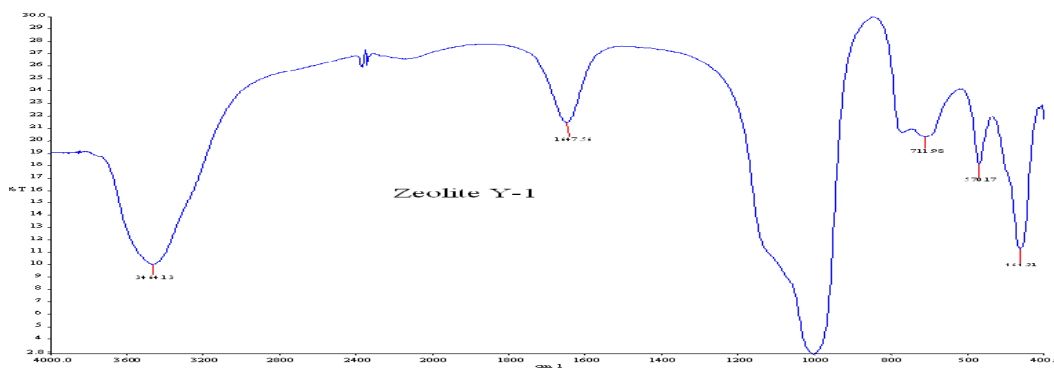


Fig. 1: IR-Spectrum from zeolite Y.

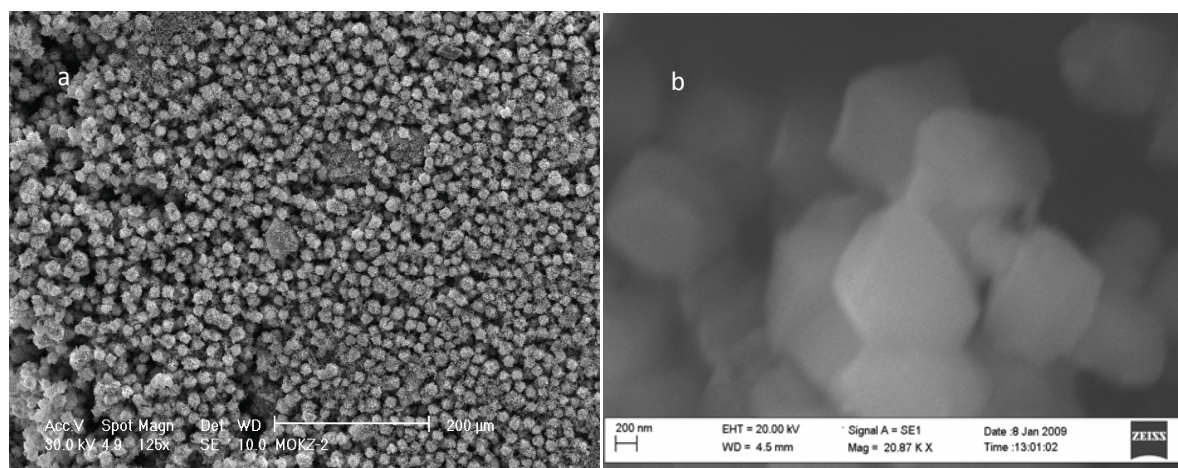


Fig. 2 (a and b) : Scanning electron microscopy morphology from zeolite Y in different magnifications.

Surface Area and Pore Volume of Zeolite Y:

Nitrogen adsorption-desorption measurements were carried out at $-196\text{ }^{\circ}\text{C}$ on a Micromeritics ASAP 2010 instrument to determine the Brunauer Emmett Teller (BET) surface area and to estimate the mesopore size distribution using the Barrett Joyner Halenda (BJH) calculation procedure. Before each measurement, samples were evacuated overnight at $100\text{ }^{\circ}\text{C}$. Higher temperatures were not used so that occluded template in the zeolite structure would not decompose or volatilize. The gas N_2 adsorption-desorption isotherm for synthesized Y is shown in Figure 4. This isotherm Figure indicates that it is an excellent micro porous material. The initial adsorption step at low relative pressure indicates complete filling of the micropores on the sample. The synthesized zeolite Y exhibited a BET surface area as high as $621.18\text{ m}^2\text{ g}^{-1}$ and micropore volume of $0.33\text{ cm}^3\text{ g}^{-1}$ was determined by the t-plot method and the micropore diameter was determined to be 21.22 \AA using BJH method.

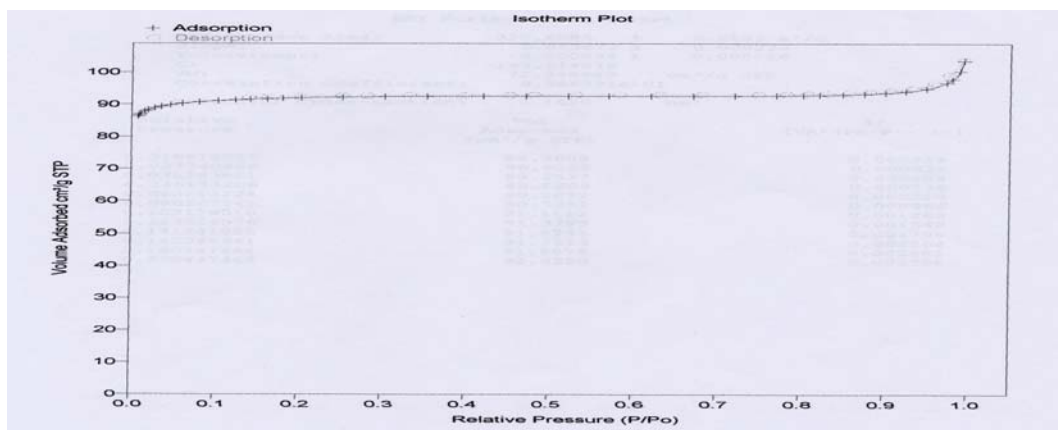


Fig. 4: Adsorption-desorption isotherm from zeolite Y.

Application of Zeolite in Water Purification:

Column adsorption studied was conducted using glass column. The column used in this experiment was filled with Zeolite bed, the bottom of each of the column contained a plug glass wool to support the zeolite bed, distribute back wash and promote uniform collection of the treated water. A glass tube ($0.5 \times 10\text{ cm}$) was packed with 0.125 g of zeolite, the average particle size of zeolite is in range of $90 < \mu\text{s} < 125\text{ m}$. A volume of 25 mL of waste water containing toxic elements in the concentration range of $5\text{--}20\text{ ppm}$ was passed through the column separately, the flow rate was (0.5 mL/min) and the temperature at which experiments were carried out was 27°C to 29°C . The equilibrium concentration of each of the metal ion was established by using flame atomic absorption spectrophotometer C.F Schutte, 2001). The study on adsorption capacity of zeolite Y using artificial polluted water sample was filtered through a column filled by zeolite. Artificial polluted (toxic) 200 ppb samples were prepared mixing As, Cr, Cd, Pb etc. into 1 L deionized water. All of these pollutants were prepared 5 to 10 times higher than WHO standards (Drinking Water Guideline. WHO 1996). This artificial polluted water was filtered by the filtering column maintaining different flow rate (4 and 8 mL min^{-1}). The experiment results of the adsorption are presented in the Table 1. Filtrate water was analyzed by ICP-MS. The metals As, Cd, Co, Cr, Cu and Pb were quantitatively removed from the spiked water through filtering column. So, it is very clearly observed in the Table 1, when filtrations flow rate is (4.00 mL min^{-1}), maximum analytes (heavy metals) removed from water samples were excellently done by zeolite Y through ion exchange.

Table 1: Obtained adsorption capacity of zeolite-Y column in different flow rate ($\text{mL}^{-1}\text{ m}^{-1}$).

Elements	$4.0\text{ mL min}^{-1}(\mu\text{g L}^{-1})$	$8.0\text{ mL min}^{-1}(\mu\text{g L}^{-1})$
As	6.83 ± 0.11	8.35 ± 0.37
Cd	ND	10.83 ± 0.24
Co	ND	8.76 ± 0.20
Cr	1.78 ± 0.03	10.31 ± 0.19
Cu	ND	6.95 ± 0.96
Pb	ND	3.99 ± 0.30

Conclusions:

XRD and SEM shows that the zeolite Y nanocrystals obtained are highly crystalline, and its pore volume is moderate figure and its pore diameter 18.68 \AA ($>20\text{ \AA}$) prove that it is a nanoporous material. The synthesized zeolite Y nanocrystals show high N_2 adsorption; and BET surface area and micropore volume are determined to

be $320.61\text{m}^2\text{ g}^{-1}$ and $0.15\text{ cm}^3\text{ g}^{-1}$ respectively. Finally fabricated zeolite Y filter unit has been found to be significantly improved for its adsorption and ion exchange capability towards arsenic, chromium, cadmium, lead and copper etc. heavy metals. This filter unit provided soft and mineral water and removed heavy metals (toxic) hazardous from the filtrated water.

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