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Preparation of Zeolite NaA for Uranium Removal from Radioactive-Contaminated Water

For the sake of environmental safety, radioactive pollutants in water must be removed, especially in locations where uranium contamination is present. The efficient removal of uranium ions from contaminated water using zeolite NaA is the aim of this study's preparation and application. Zeolite NaA, a microporous aluminosilicate with a large surface area and cation exchange capacity, was created via a hydrothermal technique. Under varied pH levels, solution volumes, and contact times, the uranium ion adsorption effectiveness was investigated. The findings show that uranium ions can be effectively removed by zeolite NaA, with the best removal happening at a pH that is somewhat basic. Zeolite NaA has the potential to be a sustainable and efficient material for radioactive water cleanup, as evidenced by the method' notable reduction of uranium ions. The results demonstrate its applicability for large-scale water treatment in hazardous areas, providing a viable way to lessen the risks that radioactive contaminants cause to the environment.

***Keywords*:** Zeolite NaA; Uranium; Radioactive contaminated water; Sol-gel method
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1. Introduction

Radioactive contaminated water is an aqueous waste that typically contains elevated levels of radionuclides that are released during regular operations at nuclear facilities, such as laundry, decontamination, and decommissioning [1]. Because an unintentional release can seriously contaminate the environment and pose risks to human health, radioactive effluent from nuclear power plants needs to be controlled safely [2]. The amount of radiologically contaminated aqueous solutions can be significantly reduced and a significant decontamination process can be achieved by treating radioactive wastewater using a number of techniques, including flocculation, precipitation, membrane separation, evaporation, ion exchange, and adsorption [3,4]. According to previous works, zeolites are good adsorbents for removing heavy metals from aqueous solutions through ion exchange [5]. The zeolite's network-like structure and uniformly sized pores allow its cations to freely interchange with those in aqueous solutions, which is the process by which radionuclides are removed [4,5]. Zeolites are a class of naturally occurring minerals that are referred to as "natural molecular sieves" because of their porous crystalline structure [6]. Zeolite is crystalline hydrated aluminosilicates, a naturally occurring porous mineral [7].

Alkali or alkaline-earth cations are reversibly trapped in the cavities inside the zeolite framework structure, making it easy for surrounding positive ions to exchange them, giving the structure a high exchange capacity and removal efficiency for certain cations [8]. Toxic and radioactive materials are removed from wastewater through the use of rocks containing zeolite [9].

2. Methods of Preparation

The hydrothermal method used in conventional zeolite synthesis is a laborious process that takes place in an alkaline atmosphere for 24 to 96 hours in a closed system [10,11] between 90 and 150 °C in pressure ranges of 1 to 15 bar [12]. The aluminosilicate hydrogel, organic compounds, and metal cations are transformed into crystalline aluminosilicate through a number of processes [13]. The crystallization process results in the development of crystal nuclei at various stages, with the highest rate of formation occurring during the early phase. When zeolites are synthesized hydrothermally at approximately 100 °C, crystals ranging in size from 0.1 to 10 μm typically form [11].

The preparation of zeolite was done as follows:

1. Raw Material Selection: Sodium hydroxide (NaOH), silica sources (like sodium silicate or fumed silica), and aluminum sources (like aluminum hydroxide or alum) are the main raw materials used in the production of NaA zeolite. For the synthesis of NaA zeolite to be successful, the molar ratio of these ingredients is essential.
2. Gel Preparation: A gel is created by combining the silica and alumina sources with sodium hydroxide and water. To obtain the appropriate Si/Al ratio, the gel's composition is changed, which affects the zeolite's characteristics [14].
3. Hydrothermal Treatment: The gel is heated to high temperatures (usually between 80°C and 200°C) in an autoclave for a number of hours. NaA zeolite crystallization is aided by this method.
4. Washing and Drying: To obtain the finished product, the zeolite is first cleaned of unreacted compounds using distilled water and then dried at a predetermined temperature, as shown in Fig. (1),

which is represented the synthesis method to create NaA zeolite [15].

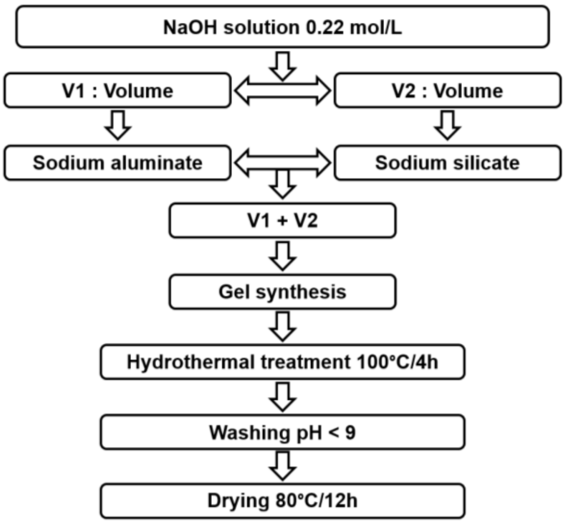


Fig. (1) Schematic for the traditional technique of synthesis

3. Results and discussion

The Zeolite skeleton contains chambers and channels, which offer it several advantageous physicochemical features and a broad range of application possibilities. Zeolites possess oxidation-reduction or acid-base surface-active centers. Their remarkable adsorption and catalytic activity can be attributed to these [16,17]. The existence of micropores with widths between 0.3 and 1.0 nm [18] and volumes between 0.10 and 0.35 $cm^3\ g^{-1}$ [19] is a defining trait of zeolites.

Several methods, such as X-ray diffraction (XRD), Scanning Electron Microscopy (SEM), Fourier Transform Infrared Spectroscopy (FTIR), Energy Dispersive Spectroscopy (EDX) and Scanning Electron Microscopy (SEM) are used to analyze the produced NaA zeolite. These techniques verify the surface area, shape, and crystalline structure—all of which are essential for the material's ability to adsorb substances [20].

Figure (2) shows the Zeolite NaA features, which is type of alkali metal aluminosilicate, features a three-dimensional framework composed of silica (SiO_2) and alumina (Al_2O_3) tetrahedra. It is known for its uniform pore structure, which allows for selective adsorption and ion exchange, making it useful in applications like catalysis, gas separation, and water purification. The high value of intensity refers the high ability to adsorb the heavy radioactivity such as uranium. Changes in the electron density distribution within the zeolite unit cell could be the reason for the abrupt variations in the intensity of each peak in the XRD patterns of Y and X. The degree to which the pores in the electron density distribution. The guest molecules and their characteristics are packed into the zeolite molecules. The guest molecule H_2O fully fills the XRD method's pores. If Y and X's cations are partially or completely swapped out for different cations, the additionally, intensity will shift [21].

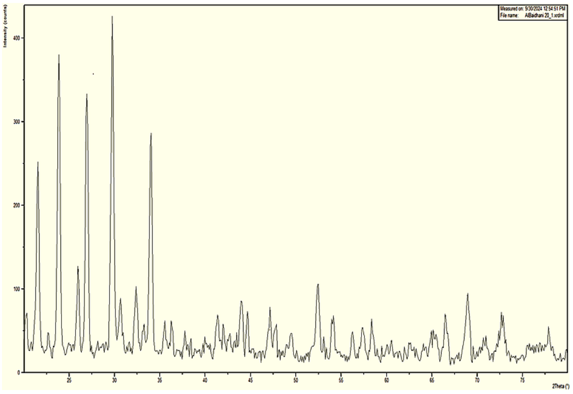


Fig. (2) XRD pattern of zeolite NaA

FTIR spectroscopy is a good device to analyze zeolite NaA, providing many information about its chemical and structural properties. By examining the characteristic absorption bands, researchers can gain insights into the framework, hydration, and potential applications of this versatile material. Due to Al-O and Si-O stretching, the primary absorption bands are associated with the stretching vibrations of the Si-O and Al-O bonds, as shown in Fig. (3). In zeolite NaA, these typically appear in the range of 600 to 800 cm^{-1} (Al-O), and around 1000 to 1200 cm^{-1} (Si-O). Al-O-Si tetrahedral vibrations, they were represented the characteristic bands which can be observed between 420 and 620 cm^{-1}, corresponding to the bending vibrations of the tetrahedral units. Adsorbed water, because zeolite NaA is aqueous, the spectrum show broad absorption bands around 3000 to 3700 cm^{-1} due to O-H stretching vibrations from water molecules. Cation Interactions, also appear this Peaks may in the region of 800 to 1000 cm^{-1}, which can indicate the presence of cations (e.g., sodium ions) within the zeolite framework.

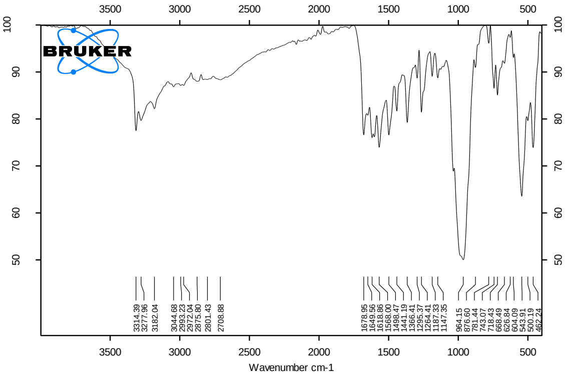


Fig. (3) FTIR spectrum of zeolite NaA

EDX is an analytical device used in conjunction with SEM to identify the elemental composition of materials. In the context of zeolites, EDX can provide detailed information about the elemental constituents of the zeolite framework. The EDX spectrum of zeolite NaA is shown in figures (4) and (5), which is show the presence of cations (like sodium in zeolite NaA) facilitates ion exchange processes. Uranium ions (UO_2^{2+}) can replace sodium ions in the zeolite framework, effectively trapping the uranium. Also EDX allows the elemental composition of zeolite samples determination of, including the ratios of silicon (Si), aluminum (Al), sodium (Na), and other elements present in the framework. EDX can reveal how elements are distributed on the surface or within different regions of the zeolite, providing insights into its adsorption properties.

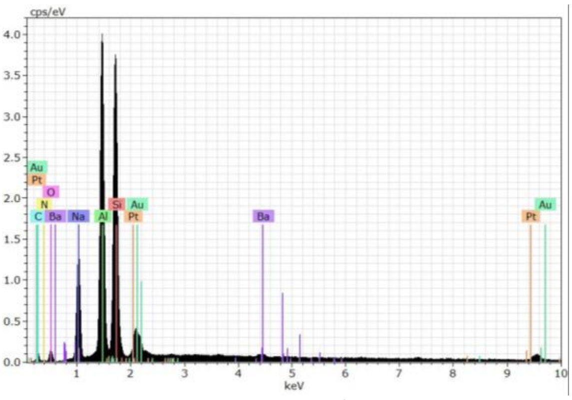
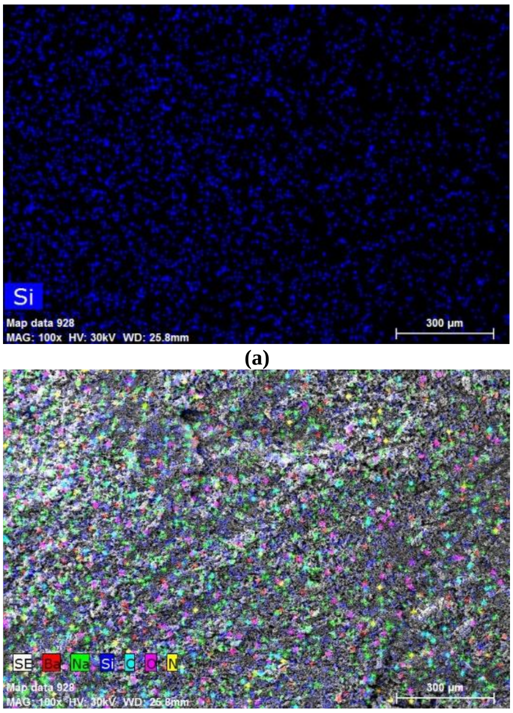
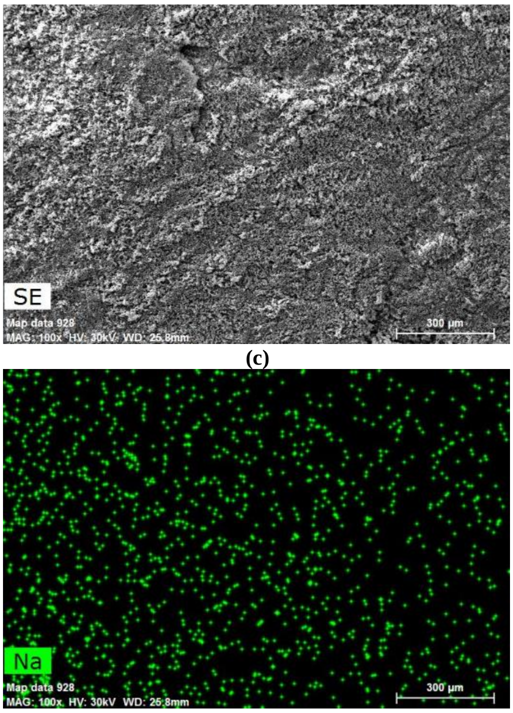


Fig. (4) EDX spectrum of zeolite NaA



(a)

(b)

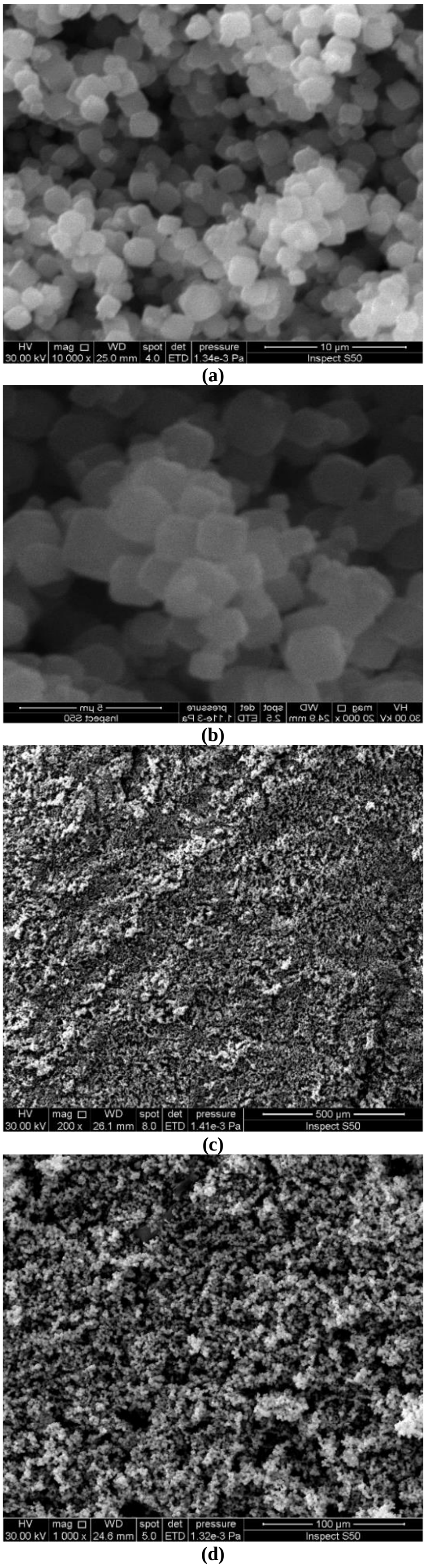


(c)

(d)

Fig. (5) Mapping distribution of elements in zeolite NaA

SEM tool is an effective tool to exam the morphology of zeolite NaA. Figure (6) provides insights into zeolite NaA structural characteristics in various magnifications. At low magnification ×200, zeolite NaA appears as aggregates of crystalline particles, showing a roughly spherical to cubic shape. The larger aggregates may exhibit a rough, porous surface, characteristic of zeolitic materials. In this range of magnification the particles range from a few nanometers to tens of nanometers in diameter, depending on the synthesis method and conditions used. Individual crystals can be discerned as distinct entities within the aggregates. Also the porous nature of the zeolite framework becomes apparent, indicating potential sites for adsorption. At medium magnification (×1000 to ×5000), the individual crystal structures of zeolite NaA are more clearly visible. The crystals often show well-defined edges and faces, characteristic of zeolite structures. The surface may display a combination of smooth and rough areas, with small pits and indentations indicative of the porous structure. These features are essential for the zeolite's ion-exchange and adsorption properties. But at high magnification (×10000 to ×50000), the atomic arrangement within the crystals can be studied. The regularity of the lattice structure becomes apparent, showcasing the orderly arrangement of AlO_4 and SiO_4 tetrahedra. Therefore Fine surface textures, such as small voids and roughness, can be observed. These features play a critical role in enhancing the adsorption capacity by increasing the surface area available for interaction with adsorbates.



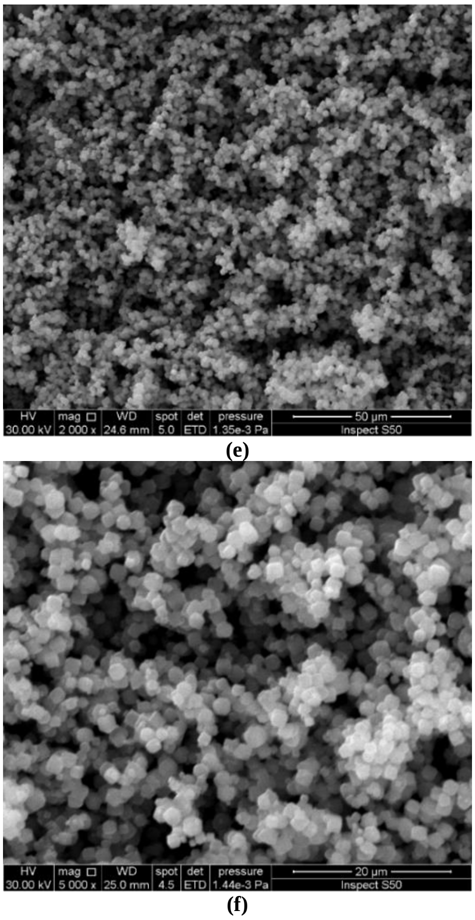


Fig. (6) SEM images of zeolite NaA morphology at different magnifications

NaA zeolite uses ion exchange and adsorption to efficiently remove uranium from contaminated water. Uranium ions can replace sodium ions in solution due to NaA's high cation-exchange capability. According to studies, given the right circumstances, NaA zeolite may remove uranium with an efficiency of over 90%. Table (1) represents the results for efficient removal of uranium using zeolite NaA from contaminated water, in this work the acidity or alkalinity (pH) of the solution, which have varies values from 3, 5, 7, and 9 at different experiments. The time of contact between the uranium contaminated water and zeolite NaA, volume of solution (ml), dose weight (The weight of the dose in milligrams applied during the process) (mg), and initial concentration (ppm) were stay constants (4 hours, 20 ml, 10 mg and 100 ppm) respectively. Also the temperature remain constant throughout all experiments.

From these results it can see the final concentration of a substance in parts per million after the experiment or process were decrease at increases the pH vales.

- The uranium removal efficiency vs. pH as shown in Fig. (7): As the pH increases from 3 to 9, the uranium removal efficiency increases from 17.7% to 84.5. This suggests a significant relationship between the pH value of solution and the effectiveness of the process in reducing the concentration.

- Consistent Dose and Conditions: The dose weight, time, and volume remain constant across all experiments, indicating that the variable being tested for its effect is likely pH.

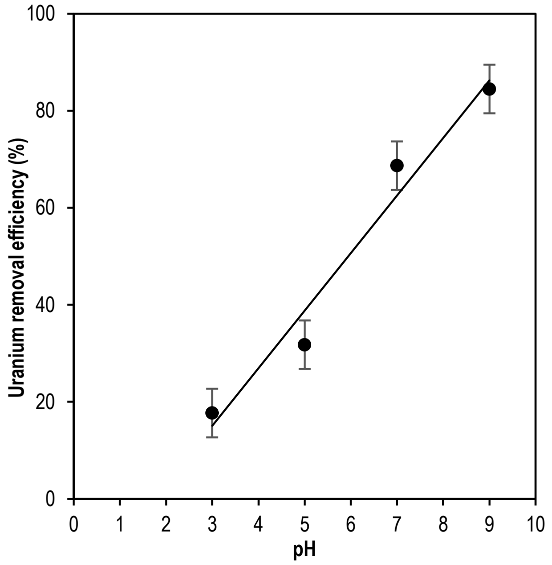


Fig. (7) Relationship between the concentration of final substance after adding Zeolite NaA nanoparticles in ppm after the experiment and pH

4. Conclusions

In order to maximize the adsorption efficiency of zeolite NaA, meticulous synthesis, activation, and possible functionalization are required for the removal of uranium from radioactively polluted water. Zeolite NaA has the potential to be a useful material for reducing uranium contamination in water systems with careful testing and optimization. Therefore it can be used to treating the contaminated water which is stored in Altwitha site [22].

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Table (1) Results of efficient removal of uranium using zeolite NaA from contaminated water, at different values of pH

Initial Concentration (ppm)	Volume of Solution (ml)	Time (hours)	pH	Dose weight (mg)	Final Concentration (ppm)	Uranium Removal Efficiency (%)
100	20	4	3	10	82.3	17.7
100	20	4	5	10	68.2	31.8
100	20	4	7	10	31.3	68.7
100	20	4	9	10	15.5	84.5