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Efficient Uranium Removal from Contaminated Water Using Laboratory-Prepared Zeolite

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Abstract

Due to radioactive uranium pollution of water posing serious risks to human health and the environment, effective and sustainable cleanup techniques are required. Zeolite, a porous alumina-silicate mineral with a large surface area and ion-exchange capacity, has drawn interest as a successful uranium removal adsorbent. This study the removal of uranium from radioactively polluted water using laboratory prepared zeolite. To increase the adsorption efficiency, a number of variables were adjusted, such as pH, contact time, initial uranium content, and temperature. According to experimental studies, zeolite exhibits a great affinity for uranium ions and, in the right circumstances, can remove uranium ions with high efficiency. The results show that zeolite is an environmentally acceptable, inexpensive, and promising material for uranium decontamination in aquatic environments. By developing scalable techniques for treating radioactive wastewater, this research helps to ensure safer water supplies.

Keywords: Zeolite (NaA), Removal Uranium, radioactive contaminated water.

الإزالة الفعّالة لليورانيوم من المياه الملوثة باستخدام الزيوليت المُحضَّر مختبريًا

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الخلاصة

يشكل التلوث الإشعاعي للمياه باليورانيوم خطرا جسيما على صحة الإنسان والبيئة، لذلك هناك حاجة ملحة إلى تطوير تقنيات فعّالة ومستدامة لإزالة هذا التلوث. ويُعد الزيوليت وهو معدن سيليكات الألومنيوم المسامي ذو المساحة السطحية الكبيرة وقدرة التبادل الأيوني العالية، من المواد التي حظيت باهتمام واسع بوصفه مادة مازة فعّالة لإزالة اليورانيوم. هدفت هذه الدراسة إلى إزالة اليورانيوم من المياه الملوثة إشعاعيًا باستخدام الزيوليت المحضَّر مختبريًا. ولتحسين كفاءة الامتزاز، جرى دراسة وضبط عدد من العوامل المؤثرة، بما في ذلك الأس الهيدروجيني(pH) وزمن التلامس والتركيز الابتدائي لليورانيوم ودرجة الحرارة. وأظهرت النتائج التجريبية أن الزيوليت يمتلك ألفة عالية تجاه أيونات اليورانيوم، وأنه قادر على تحقيق كفاءة إزالة مرتفعة في ظل الظروف التشغيلية المثلى. وتشير النتائج إلى أن الزيوليت يُعد مادة واعدة ومنخفضة التكلفة وصديقة

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للبيئة لإزالة نويدات اليورانيوم من الماء الملوث. وتسهم هذه الدراسة في تطوير تقنيات قابلة للتطبيق على نطاق واسع لمعالجة المياه العادمة المشعة، بما يضمن توفير مصادر مياه أكثر أمانًا وحمايةً للصحة العامة والبيئة

Introduction

Radioactive materials have been utilized for many different reasons, which has led to the contamination of various locations worldwide, including buildings, soils, rocks, waterways. The affected locations may vary from tiny urban areas to bigger areas in the environment, encompassing several tens or hundreds of square kilometers [1]. In order to safely release radioactive waste produced by using radionuclides in companies, hospitals, and research institutes, it must be treated to lower the quantity of radioactive pollutants. Conformity with national regulations and international agreements requires managing radionuclides appropriately: short-lived radionuclides must be stored to allow their radioactivity to disintegrate, whereas for longer-lived radionuclides, specific measures must be implemented to separate and concentrate the radioactive materials from the aqueous phase [2]. Most often, liquid radioactive waste treatment involves the use of a variety of techniques, including filtration, sedimentation, adsorption, ion exchange, evaporation, and membrane separation in order to meet the requirements for the release and use of liquids after radioactive waste disposal in various aspects of life [3].

Ion exchange is switching out liquid ions for solid ones. It works incredibly well to reduce the radioactive content of a large liquid to a small amount of solid [4].

Owning to its chemical toxicity and radioactive nature, uranium is regarded as one of the most dangerous pollutants [6]. In addition to the naturally existing quantity, uranium is present in the environment due to nuclear industry emissions, discharge from mill tailings, burning of coal and other fossil fuels, and use of phosphate fertilizers containing uranium [7]. The degree of mobility of uranium is mostly determined by its valence state, which has a significant impact on the hazard it poses on the environment [8].

Because of its great efficiency and ease of handling, adsorption is widely employed in metal separations, hydrometallurgy, water and wastewater treatment, and the removal and recovery of metal ions [9, 10]. The available surface area and pore volume of the adsorbent have a significant impact on the process, which is dependent on the chemical makeup of both fluid and solid [11]. Adsorbents come in various forms, including inorganic, composite, and natural and synthetic organic materials. Some extremely common sorbents include zeolite, apatite, activated carbon, calcined phosphate, oxides, and hydroxides [12].

Numerous stable cations, gases, and radionuclides, such as radium isotopes, can be adsorbed by natural zeolites, which are aluminosilicate minerals with a porous structure. Consequently, these substances are referred to as "molecular sieves." Mordenite, chabazite, clinoptilite, natrolite, and stilbite are the most prevalent natural zeolites. There are currently around 40 distinct types of natural zeolites known to exist. Natural zeolites cannot be used in several applications because they are contaminated with other minerals. Synthetic zeolites are made for these uses [13]. Zeolites are employed as catalysts in the nuclear industry to purify water.

NaA zeolite effectively extracts uranium from tainted water using ion exchange and adsorption. Because of NaA's high cation-exchange capability, uranium ions can take the place of sodium ions in solution. Studies have shown that NaA zeolite can extract uranium with an efficiency of more than 90% under the correct conditions [14].

Experimental Works

Adsorption test

The adsorption tests were conducted in a batch system by combining a predetermined amount of (uranyl nitrate) with a known volume of synthetic solution (water) for a predetermined amount of time in order to reach equilibrium. The impact of four operating parameters—solution pH, zeolite particle size, adsorbent quantity, and retention time—was investigated during the experimental testing. Based on previous research, the pH of the solution was determined to be between 3 and 9. The tests are often not conducted at pH values higher than 9 since a further increase in pH usually results in the precipitation of uranium, thorium, and rare earths [15-19].

Furthermore, zeolite breakdown is observed at pH values below 3, leading to inefficient adsorption and a reduced adsorption capacity [12]. To modify the solution chemistry and attain various pH values, a 2 M sodium hydroxide (NaOH) solution was employed.

For the tests, contact times of one, two, and three hours were established in accordance with earlier research [10,14]. After the tests were finished, filter sheets with pore sizes of 0.45 µm were used to separate the sorbent. Following that, the adsorption rate of uranium was measured using X-ray fluorescence. The following parameters were used in this work:

1. **Concentration (ppm)** – Refers to the final concentration of a substance in parts per million.
2. **Dose weight (mg)** – Refers to the fixed weight of the nanoparticles NaA zeolite used in the experiment, which is 10 mg for all entries.
3. **Temperature (°C)** –the temperature was constant during the experiment (room temperature).
4. **pH** –the pH level was fixed at 9 across all experiments.
5. **Time (hours)** – Refers to the duration of the experiment, which was 4 hours in all cases.
6. **Volume of solution (ml)** – The volume of the solution used, which was consistently 20 ml.
7. **Initial Concentration (ppm)** –is the starting concentration of the substance before the experiment begins.

Results and Discussions

Table 1 shows the results of the first experiment which was done by decreasing the initial concentration of uranium keeping the other parameters constant at: 20ml volume of the solution, 4h contact time, a pH value of (9), 10mg NaA weight, and 25 °C temperature of the contaminated water and dose weight.

Table 1: The effective uranium removal from contaminated water using zeolite NaA, for various values of concentration.

Initial Concentration (ppm)	Final Concentration(ppm)
50	48.2
100	31.4
150	102.4
200	106.7

Figure 1 shows the relationship between the initial concentration and the final concentration of uranium. The highest value of adsorption was seen at an initial concentration of 100ppm, the uranium concentration decreased to 30 ppm.

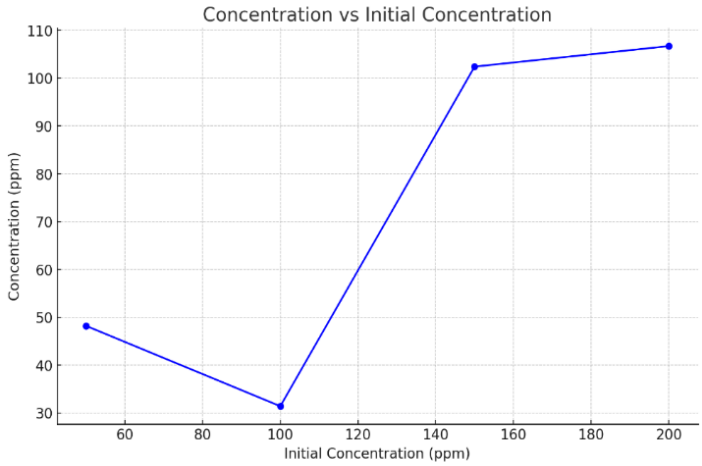

Figure 1: The relationship between the initial concentration and the final concentration of uranium.

Table 2 presents the results of the effect of increasing the absorbent-contaminated water contact time on the final uranium concentration, keeping all the other factors constant at: 20 ml of solution volume, 100 ppm of the uranium initial concentration, a pH of 9, the adsorbent volume = 10ml, the temperature of contaminated water was (25 °C), and a dose weight of 10 mg. The uranium concentration decreased as the time of contact increased due to the increase of the probability of adsorbing uranium from water**.** The process follows a mechanism where adsorption occurs in two stages: rapid adsorption during the initial phase, followed by a slower phase as the active sites become saturated. At first, there are many available sites on the adsorbent, so the adsorption rate is high. Over time, as these sites are occupied, the adsorption rate slows down, but the overall concentration of uranium in the water continues to decrease until equilibrium is reached.
This phenomenon is commonly described using adsorption isotherms such as the Langmuir or Freundlich models, which account for the changes in the amount of adsorbate (uranyl nitrate) as a function of time, and can demonstrate that longer contact times result in greater uranium removal [20,21].

Table2: The same initial concentrations of uranyl nitrate (100 ppm) and at constant volume of adsorbate (nano NaA zeolite, 20ml), fixed pH (9) and constant temperature (25°C) were used in all experiments, relying on different times to measure adsorption.

Time (hours)	Final Concentration(ppm)
2	82.4
4	61.3
6	48.3
8	36.9

As time progresses (from 2 to 8 hours), the concentration decreases, showing a potential reduction in concentration over time from 82.4 ppm to 36.9 ppm as shown in Figure 2**.** This figure displays a line plot of "Concentration vs. Time", showing how a measured concentration in parts per million (ppm) changes over time in hours. The concentration decreases steadily over time.

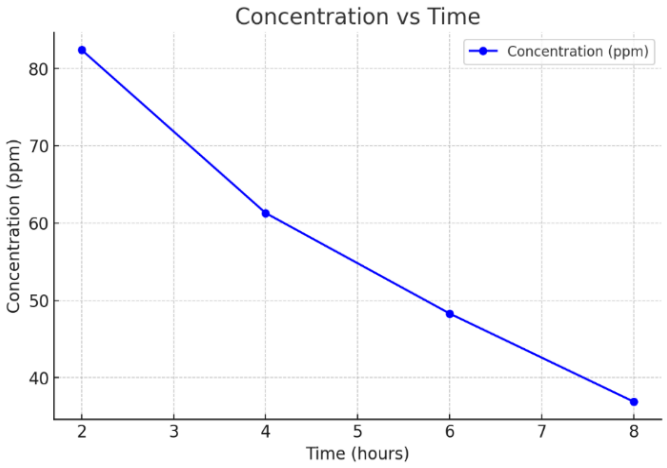

Figure 2: The relationship between the uranium concentration (ppm) vs the contact time (hours).

Table 3 presents the results of the effect of the contaminated water temperature on the adsorption process at a contact time of 4 hours, keeping all the parameters constant at the same values. The concentration of uranium decreased (the uranium adsorbed from water increased) as the temperature increased due to the increase of the uranium nucleus mobility in water.

Table 3: The same initial concentrations of uranyl nitrate (100 ppm) and at constant volume of adsorbate (nano NaA zeolite, 20ml), fixed pH (9) and constant time contact between uranim and nano solution (4 hours)) were used in all experiments, relying on different temperatures of contaminated water to measure adsorption.

Temperature °C	Final Concentration (ppm)
50	64.5
55	59
60	52.1
65	37.4

As the temperature changed from 55°C, to 65°C, the concentration decreased from 64.5 ppm to 37.4 ppm, as shown in Figure 3. The graph shows a negative linear relationship between temperature and final concentration: as the temperature increases, the final concentration decreases, which might indicate a temperature-dependent process. The observed decrease in uranium concentration suggests that the adsorption process is temperature-dependent. This can be explained by the fact that temperature effects the rate of adsorption in various ways. As the temperature increases, the kinetic energy of both the uranium ions and the adsorbent particles increases. This results in enhanced interaction between the adsorbent (zeolite) and the adsorbate (uranium ions), which may accelerate the adsorption process initially. However, at higher temperatures, the adsorption sites on the adsorbent may become more dynamic, potentially causing desorption or competition for available sites, reducing the overall efficiency of uranium removal. This trend is typical of many adsorption processes, which often exhibit a maximum efficiency at an optimal temperature, beyond which the efficiency may decline.

Additionally, temperature changes can affect the solubility of ions in solution and the viscosity of the water, which also influences how effectively the adsorbent interacts with the uranium ions. Therefore, the decrease in uranium concentration with increasing temperature may indicate that the adsorption process is not purely physical but involves some degree of temperature-sensitive chemical interaction, such as ion exchange or surface complexation.

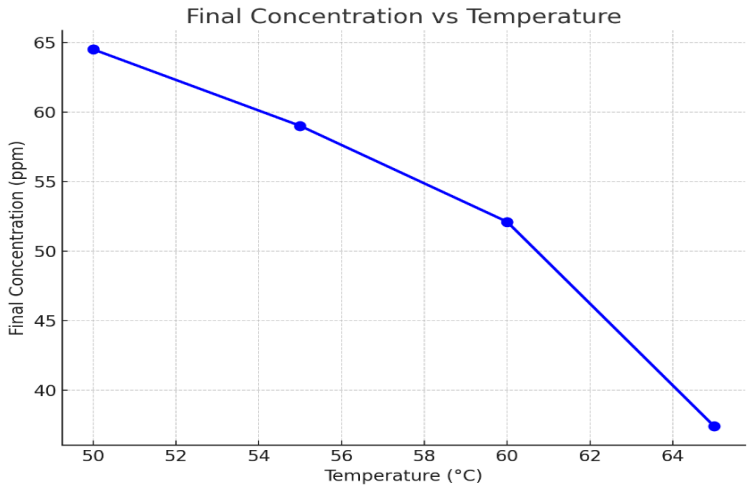

Figure 3: Final uranium concentration vs the temperature of contaminated water

Table 4 presents the results of changing adsorbent volume when the same values of initial concentration, the contact time between absorbent and contaminated water, pH, temperature and dose weight were used. The concentration of uranium decreased when the volume of absorbent increased due to the increase of the probability of uranium adsorbed from water.

Table 4: The same initial concentrations (100ppm), constant time contact (4hours), constant pH (9) and constant temperature (25 °C) were used in all experiments, relying on different volumes of adsorbent.

Volume of solution(ml)	Final Concentration (ppm)
15	84.5
20	69
25	53.3
30	36.2

As the volume of the adsorbent material (nano NaA zeolite) increased (from 15 to 30 ml), the concentration decreased from 84.5 ppm to 36.2 ppm, as shown in Figure 4. This figure represents a graph that shows the relationship between the volume of a solution (measured in milliliters, ml) and the uranium concentration (measured in parts per million, ppm). The inverse relationship suggests that when the volume of the polluted solution increases, the solute's concentration decreases, which aligns with the principle of dilution: increasing the volume of solvent reduces the concentration of solute in a solution.

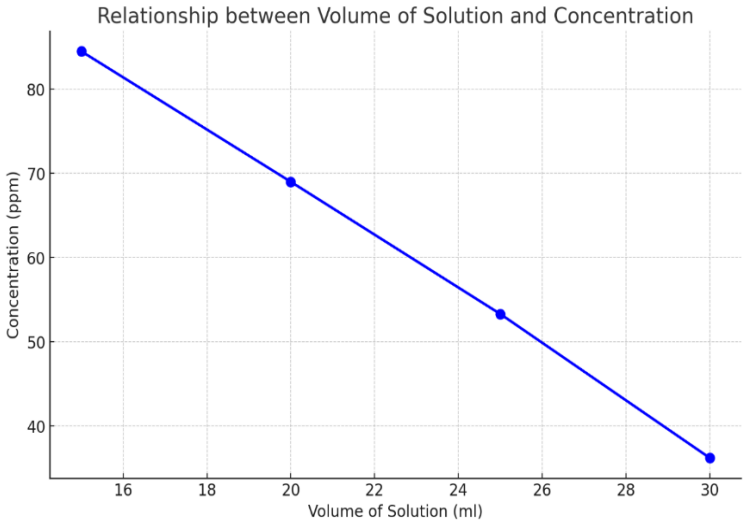

Figure 4: The relationship between the volume of a solution (measured in milliliters, ml) and its concentration (measured in parts per million, ppm).

Conclusions

The present study highlights the potential of laboratory-prepared zeolite (NaA) as an effective and sustainable adsorbent for the removal of uranium from contaminated water. The experimental results demonstrate that zeolite can efficiently extract uranium ions through ion exchange and adsorption, achieving removal efficiencies exceeding 90% under optimized conditions. Key parameters, such as pH, contact time, temperature, and adsorbent volume, significantly influence the adsorption process, with higher contact times, increased adsorbent volumes, and specific temperatures enhancing uranium removal efficiency. This makes zeolite a promising candidate for the decontamination of radioactive water, offering a cost-effective, environmentally friendly solution. Further research and scale-up studies are necessary to optimize the process for real-world applications, ensuring the safe treatment of radioactive wastewater and contributing to cleaner water sources and better public health protection. Furthermore, this method is more efficient than other methods which are done to decrease the radionuclides activity using different materials [22].

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