

Radioactive Wastewater Treatment using Magnetic Nanocomposite Material

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ABSTRACT

Radionuclide contamination has become a big problem after the development of nuclear power plants and the use of nuclear waste in making weapons used in recent wars. Here, using the electrical exploding wire method, the core-shell iron@carbon@magnesium nanocomposites were synthesized. Two vital properties make the nanoparticles very profitable as an absorbent material. On a mass basis, nanoparticles have much larger surface areas than the total molecules. It can also be reinforced with different reactor groups to increase their chemical affinity towards targeted compounds. Adsorption is among the different technologies available, one of the best technologies thanks to its ease of processing, low cost, and high efficiency. The fate of the environment and the toxicity of materials are essential issues in selecting materials and designing a wastewater purification system. Concrete actions included for the first time the treatment of particles on the surface of the negatively charged Fe_3O_4 nanoparticles, then the nucleus through the carbon cation round, then the growth of a thin layer of Mg^+ (~ 30 nm). SEM characterization techniques indicate that the $\text{Fe}_3\text{O}_4 @ \text{C} @ \text{Mg}$ was very stable and magnetic with a specified large surface area (~600 sq/g). It was employed to remove uranium radionuclides from radioactively contaminated water.

Results indicate the decreases in uranium concentration from contaminated water from 50 ppm to 10 ppm using $\text{Fe}_3\text{O}_4 @ \text{C} @ \text{Mg}$ nanostructure. This research has suggested that $\text{Fe}_3\text{O}_4 @ \text{C} @ \text{Mg}$ can achieve stability and favorable functions at one time and can be easily separated by an external magnetic field after being used in radioactive wastewater treatment, which was of great practical benefit and cheap in radioactive waste extraction.

Keywords: $\text{Fe}_3\text{O}_4 @ \text{C} @ \text{Mg}$, Adsorption, Plasma Jet Method, Water treatment

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INTRODUCTION

Radionuclides have demonstrated irreplaceable properties in the fields of basic science, medicine, agriculture and industry. Radionuclides can be present in the surrounding air, soil, and water that they emit from sources, so some radionuclides are discovered in soil, sediments, air, and aquatic environments. Previous air force experiments, some emissions from active nuclear power plants, and radioactive fuel reprocessing activities could transport radionuclides to the ground through rainwater. It can participate in complex reactions with soil organic matter. Their availability on the top layer of soil and sediments is a primary source for their introduction into the food chain. Some naturally occurring anthropogenic radionuclides such as ^{222}Rn , ^{226}Ra , ^{228}Ra , ^{238}U , ^{234}U , and ^{232}Th are found in groundwater worldwide. Exposure to radionuclide contaminated water may have harmful health result. These radionuclides cause diseases.¹ Most countries today face drinking water problems and very severe conditions,

especially in developing countries. The world faces enormous challenges in meeting the growing demand for clean water as the available freshwater supplies are depleted due to (1) the prolonged drought, (2) the population growth, (c) stricter health regulations, and (d) competing demands from a variety of users.² Clean water (i.e. water free from toxic chemicals and pathogens) is essential for human health.

To date, radionuclide disposal methods have included ion exchange, membrane filtration, photocatalysis, electrolysis, chemical precipitation, reverse osmosis, and absorption.³⁻⁶ Among these methods, adsorption technology gained widespread concern due to its excellent economic efficiency, simplicity of operation, and strong practical applicability. In treating environmental pollution, the adsorption method has become one of the most widely used and effective means of removing radionuclides from aqueous systems. Chitosan magnetic nanoparticles pick up heavy metals and dyes.⁷ Synthesized $\text{Fe}_3\text{O}_4 @$ carboxymethyl cellulose uses it to

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eliminate highly efficient Eu (III) ions.⁸ Fabricated magnetic / poly chitosan (acrylic acid) Microsphere to eliminate copper ions (II).⁹

Therefore, it was expected that the magnetic materials' surface would be modified with Mg nanoparticles, enhancing the efficiency of radionuclide removal in treating radioactive contamination. Based on the results mentioned above, herein, we designed an uncomplicated approach to fabricate core-shell $\text{Fe}_3\text{O}_4@\text{C}@\text{Mg}$ structure. The aims of this work were: (1) to construct and characterize core-shell $\text{Fe}_3\text{O}_4@\text{C}@\text{Mg}$ structure nanomaterials; (2) to apply the materials for the removal of uranium radionuclides from contaminated water under various contact time; (3) to discuss the fundamental interaction mechanism of radionuclides with the composites from batch experiments.

Experimental Work

Synthesis of Magnetite-Based Adsorbents

Before experiments, the Fe_3O_4 , Mg and, C wires were cleaned (dia: 0.3 mm; purity: 99.998%) and plates (opaque: 20, 20, 1 mm; purity: 99.998%) cleaned with sandpaper, followed by acetone. The 20 mm wires were detonated on plates of the same type of material in the distilled water using the EEW array shown in Figure 1. These wires were detonated by making the metal wire in sudden contact with the metal plate. The above mentioned wire length stands for several sparks and is replaced after several contacts. Each of the electrodes (wire and plate) experiences a potential difference of 80 V -100 A DC power supply. The resulting nanoparticles (NP) remains suspended in water (colloidal form). Fe_3O_4 particles were suspended in distilled water, where 30 mg of these particles were suspended in 50 cm^3 of water. These particles are decorated with magnesium and carbon particles using EEW method.

Independent experiments were conducted to prepare each type of NP. These NP were used to investigate adsorption properties for uranium radionuclides removal experiments. The EEW process has been energy-intensive ever since relatively low voltages are applied, and also lead to large amounts of NP that are produced from both electrodes consumed.

Determination of Uranium Radionuclides

CR-39 has a chemical composition of $(\text{C}_{12}\text{H}_{18}\text{O}_7)$ and density $(1.32\text{g}\cdot\text{cm}^{-3})$ and is an organic detector used in this work. The use of CR-39 plastics to detect nuclear particles has become

generalized in dosimetry, spectroscopy, and environmental science due to its high sensitivity.¹⁰ The reagent sheets of 500 μm thickness were cut into small pieces 1 cm x 1 cm each. CR-39 detector was placed in the contaminated water for a month. Uranium concentrations were measured in water samples by comparison of track density recorded on unknown sample detectors and those of standard samples according to

$$C_x = C_s \left(\frac{\rho_x}{\rho_s} \right)$$

where

C_x : Uranium concentration in the unknown sample (ppm). C_s : Uranium concentration in the standard sample (ppm). ρ_x : track density of unknown sample ($\text{tracks}/\text{mm}^2$). ρ_s : track density of standard sample ($\text{tracks}/\text{mm}^2$). C_s (standard)/ ρ_s (standard) deduced from the calibration curve shown in Figure 2.

(CR - 39) detector was drilled etched in NaOH ($\text{Na} = 6.25$) solution at a temperature of 60°C for a period of (7 hours) and then washed with distilled water and dried. The intensity of the track was recorded using an optical microscope.

Then using the calibration curve Figure 2¹¹ the uranium concentration was determined in this water. Then we put $\text{Fe}_3\text{O}_4 @ \text{C}@\text{Mg}$ in polluted water for 24 hours, after which by the external magnetic field, the nanomaterial is collected from the water. Then the uranium concentration in the filtered water was measured in the same way.

RESULTS AND DISCUSSION

SEM Images Analyses

Figure 3, illustrates that the Fe_3O_4 nanospheres have a uniform shape and smooth surface with an average diameter of about (400 nm). Thus figure represents how the magnesium nanoparticles are formed in the form of rods and are entirely bound to Fe_3O_4 and C nanoparticles. This type of shape increases the surface area of the particles.

Uranium Concentration

Radionuclides, the chemical elements emitting either α , β , or γ - rays, or neutrons can be classified based on their origin, such as naturally occurring and anthropogenic. Uranium is a natural component of groundwater in some parts of the environment that emits alpha particles. However, there is little information about the whereabouts of uranium. Uranium

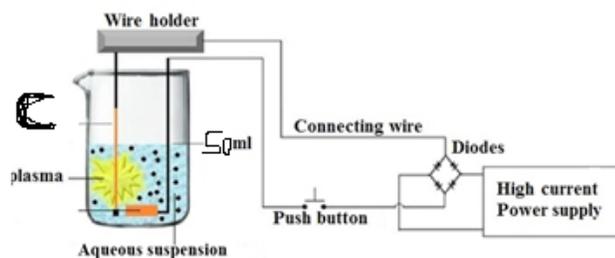


Figure 1: Experimental set-up used to fabricate the core shell $\text{Fe}_3\text{O}_4 @ \text{C} @ \text{Mg}$ nanostructure.

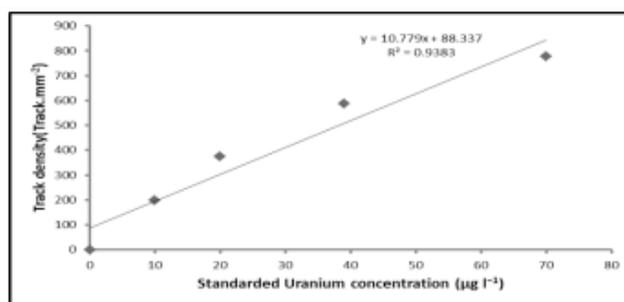


Figure 2: The relation between track density and uranium concentration ($\mu\text{g}\cdot\text{l}^{-1}$) for standard geological groundwater samples using (CR-39) track detector¹⁰

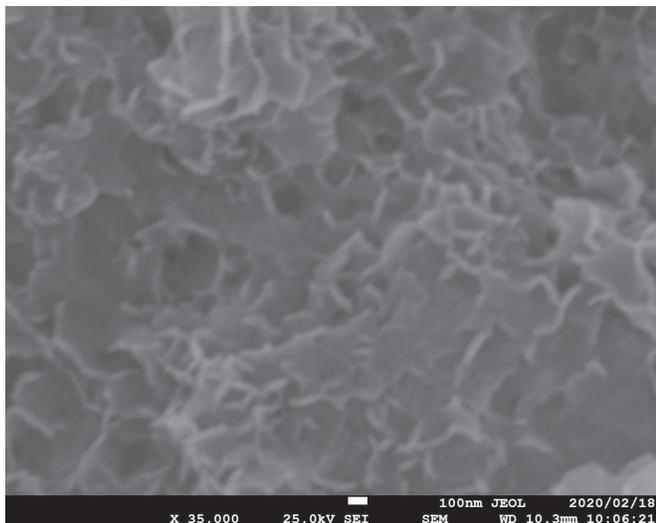


Figure 3: SEM images of core shell Fe₃O₄ @ C @ Mg

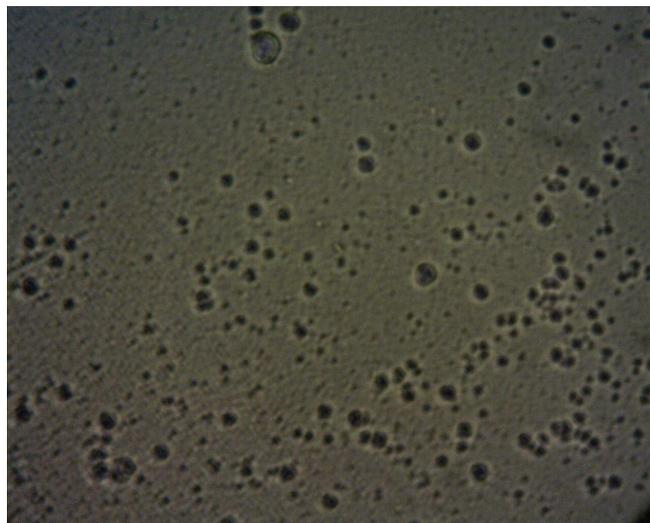


Figure 5: The concentration of tracks in treatment water for 1h

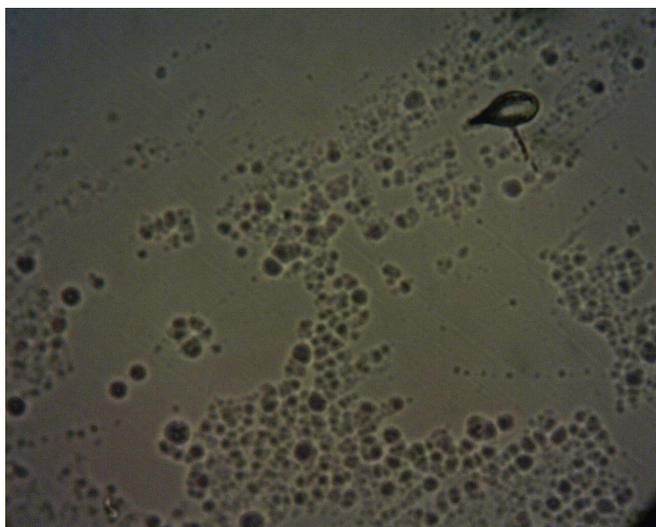


Figure 4: The concentration of tracks in un-treatment water

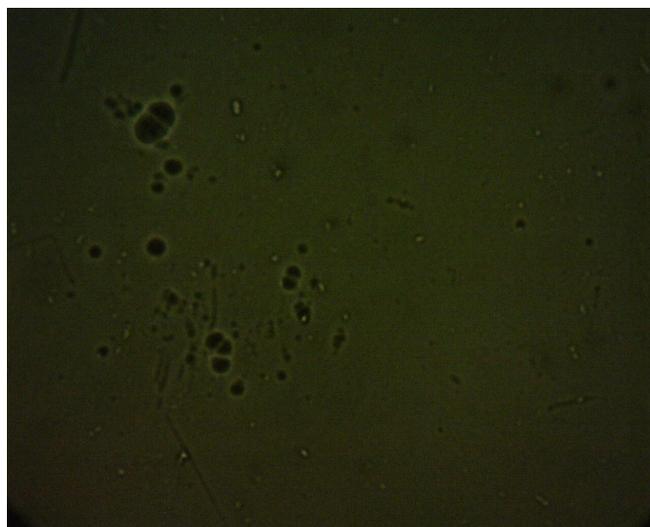


Figure 6: The concentration of tracks in treatment water for 4h

enters drinking water when groundwater dissolves minerals containing uranium. The amount of uranium in the water varies according to its concentration in the first place. However, even within regions containing uranium base types, there is a large variability within relatively small regions.

Owing to the dual toxic nature, the U.S. Environmental Protection Agency (EPA) recommended maximum contaminant levels for uranium equal to 30 mg L⁻¹ (0.74 Bq L⁻¹) have been established as drinking water standards.¹² Figure 4 represents the un-treatment water, the concentration of uranium is 50 ppm (1.17Bq/L), then Figure 5 shows the treatment water for 1-hour by Fe₃O₄ @ C @ Mg nanostructure, the concentration of water is 40 ppm (0.93Bq/L). Figure 6 shows that uranium concentration in treated water for 4 hours is equal to 10ppm (0.23 Bq/L). The concentration of uranium nuclides is decreased with increasing the time of treatment. At 4 hours, uranium concentration less than the EPA recommended maximum contaminant levels for uranium equal to 0.74 Bq L⁻¹. Kinetic adsorption is important for understanding the reaction

process. As shown in Figure 7 and 8, the adsorption ratios increased quickly with enhanced time at the initial contact time from 1 hour active sites with abundant absorption on Fe₃O₄ @ C @ Mg surfaces it was favorable for binding radionuclides. Balance was achieved in approximately 5 hours for U. Also, the greater the connection time and the adsorption rate became slow. And time-dependent the kinetic data were synthesized by a second-order pseudo model Pseudo-class model of first order,¹³ respectively, which is expressed as follow:

$$\text{Pseudo-first-order model : } \ln(Q_e - Q_t) = \ln Q_e - k_1 t$$

$$\text{Pseudo-second-order model : } \frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{t}{Q_e}$$

where k_1 (min⁻¹) and k_2 (g / (mg min)) indicate kinetic rate constants of the two models. Q_e (mg/g) and Q_t (mg/g) correspond to absorption radionuclides loaded to Fe₃O₄ @ C @ Mg in equilibrium and contact state Time t .¹⁴

Fe₃O₄ @ C @ Mg nanomaterials contains a magnetic core which is very susceptible to agglomeration, which causes

significant changes in magnetic properties. Moreover, $\text{Fe}_3\text{O}_4 @ \text{C} @ \text{Mg}$ are prone to undergo autoxidation and potential Fe^{2+} leaching and not well target selective under ambient conditions. An appreciated method involves modifying the magnetic core with the deposition of desirable chemical compounds acting as protecting shell to compensate for this problem. Its practical application to various contaminated sites is the chemical compounds for the magnetic nanoparticles shell carbon.¹² The resulting magnetic nanoparticles exhibit higher stability and adsorption capacity by hampering possible oxidation and aggregation under complete modification. Also, the protecting chemical compounds can be further conjugated with various chelating agents, eventually making magnetic nanoparticles a more versatile precursor for a broad range of environmental applications.^{15,16} Besides the surface modification or functionalization of magnetic nanoparticles, an alternative approach can be achieved by situating magnetic nanoparticles with carbon materials to synthesize magnetically retrievable composites. The resulting magnetic composites represent astounding output for radionuclides removal, challenging to attain by individual substrate components. Convincingly, nanoparticles can become ideal candidates for water treatment because of their high potential and definitive characters and may resolve upcoming problems in respective technologies.¹⁷

CONCLUSIONS

Here, the $\text{Fe}_3\text{O}_4 @ \text{C} @ \text{Mg}$ nanomaterials were synthesized and applied as adsorbents for U radionuclides from wastewater. The characterization results show that $\text{Fe}_3\text{O}_4 @ \text{C} @ \text{Mg}$'s saturation magnetization was 39.32 emu/g, which can be efficiently separated from the water with purification methods. The large BET specific surface area (106.41 m^2/g) of $\text{Fe}_3\text{O}_4 @ \text{C} @ \text{Mg}$ provides abundant sorption sites. Adsorption of U on $\text{Fe}_3\text{O}_4 @ \text{C} @ \text{Mg}$ was primarily attributed to electrostatic attraction and inner-sphere surface complexation. The versatile $\text{Fe}_3\text{O}_4 @ \text{C} @ \text{Mg}$ was a potent and promising candidate for the effective elimination of radionuclides out of wastewater.

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