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Preparation of new nano magnetic material Fe_3O_4@g-C_3N_4 and good adsorption performance on uranium ion

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Abstract. A new nano magnetic material Fe_3O_4@g-C_3N_4 was prepared by deposition reduction method, which performed good adsorption performance to uranium ion. Characterization results showed that the g-C_3N_4 particles were wrapped around the nano magnetic Fe_3O_4 particles, and the textural properties of this material was improved, so the adsorption performance to uranium ion was good. Adsorption experiments of this material demonstrated that the optimum pH value was 10, the optimum mass of adsorbent was 6.5 mg and the optimum adsorption time was 150 min in the initial concentration of 140 mg/L uranium ion solution system, and the maximum adsorption capacity was up to 352.1 mg/g and the maximum adsorption rate was more than 90%.

1. Introduction

With the development of nuclear technology and nuclear power, Uranium is one of the most common non renewable clean energy nuclear fuels, which is the one of most important strategic resources [1]. According to the statistics, the total uranium which has been known is only about 5 million ton, but the amount of which can be used for natural uranium nuclear reaction is less than 5%, and most of the nuclear industry utilization rate of nuclear fuel uranium is generally lower than 10%. Also, It is inevitably that the large amounts of radioactive waste or wastewater contains uranium ion was produced in the utilization of uranium resource, this is not only pollute the surrounding vegetation, soil and water irreversible, but also threat to human health [2]. Therefore, both the recovery of trace uranium from wastewater in order to improve the utilization of uranium resource and reduce the harm of uranium in the environment is very important, the new researching or exploring the function adsorption material of uranium ion are significant.

Many researchers have used natural mineral, microorganism and biomass material to form adsorption material, such as "YuccaMountain project", which means using natural mineral soil directly adsorb to U, Sr or Cs ions. The adsorption percentage of radionuclides by different materials was calculated, and the solution composition, concentration of radionuclides, adsorption temperature and solid particle sizes were discussed [3]. Combined with IPN theory of surface complexation model, EricSimoni have done many adsorption tests for radioactive elements as uranium or thorium, the surface chemical behavior in wastewater solution and the important regularity of adsorption was summarized [4]. However, we have not obtained an ideal material for the industrialize adsorption material to uranium, the key factor is the adsorption efficiency is not ideal or the cost of adsorption

material is too high. Therefore, exploring new efficient functional material to adsorb uranium is difficult and challenge [5].

A novel magnetic nano material with good adsorption performance which contain the cheap precursor, and it is prepared through special preparation technology to form unique nano functional structure. Generally, it contains the Fe_3O_4 particles in order to exert magnetic properties, this excellent adsorption properties have been recognized [6]. Recently, a new unique material g-C_3N_4 has been caused many scholars' attentions, because it can perform better electron hole recombination rate of good light, and it can be derived directly from decomposition of melamine [7]. Herein, we have prepared the new Fe_3O_4@g-C_3N_4 nano magnetic materials which can be used for purifying uranium ion, and good results have been achieved. So this material can perform better scientific significance in the development of nano magnetic materials.

2. Experimental

2.1 Reagents and instruments

Uranyl nitrate, Arsenazo III, melamine, cyanuric acid, urea, sodium twelve, ferric chloride, ammonia, ethanol, acetic acid, sodium acetate, buffer reserve of different concentrations of acetate were prepared.

2.2 Prepared method of Fe_3O_4@g-C_3N_4

Melamine, Cyanuric acid and Urea were mixed carefully by the mass ratio of 30:10:1, the powder was removed into the tube furnace, and calcination at 873 K for 6 h under the continuous flow of nitrogen gas. After cooling to room temperature, the light yellow block solid was polished to powder named as g-C_3N_4.

Some distilled water was added into the clean beaker, some ferric chloride solid was added (the mass was 10% of g-C_3N_4), stirring continuous at room temperature to form uniform ferric yellow liquid. 0.2 g twelve sodium and the amount of g-C_3N_4 powder were added at the same time. Then, the sensor pH meter was connected to the liquid solution, some ammonia was added carefully until the pH value was 3.5. After stirring for 6 h at room temperature, it was washed 1 h in the microwave oscillation cleaning at room temperature. Then, drying over night at 383 K, cooling and polished into powder carefully, the solid powder was washed with distilled water several times, drying over night at 383 K again. The dry powder was reduction 2 h under the under the continuous flow of hydrogen gas, and it was named as Fe_3O_4@g-C_3N_4.

2.3 Adsorption experiment

First of all, 1 mL uranium standard solution (100 mg/L) was putted in 1000 mL flask. Much distilled water was added to configure 100 μg/L uranium ion liquid solution, which was placed in Teflon bottle. 10 mL uranium standard solution was putted into 50 mL flask, some buffer solution were added into the mixture until the certain assignment pH value was displayed. A certain amount of adsorption materials were added into this system after deposited 24 h at room temperature. After ultrasonic dispersion for 30 min, this system which placed in a constant temperature water bath shaker was vibrated continuous at 160 r/min until the reaction reached equilibrium.

The adsorption liquid was centrifuged, and then it was putted into the 10 mL flask which contained 1 mL Arsenazo III 1 mL. Determining the value of pH, some buffer solution were added to adjust to 10 mL system for each testing. Determination about the mass concentration of uranium ion by absorption spectrophotometry, the best standard wavelength was chosen as 652 nm, and the linear relationship were built up by the adsorption results of different concentrations of uranium standard solution. So, we can measure directly the mass concentration by determination value in the adsorption equilibrium. The adsorption amount as Q(mg/g) and the adsorption rate as E(%) could been calculated easily as following formula: $Q=(C_0-C)\times V/m$, $E=(C_0-C)\times 100\%/C_0$. C_0 and C was the initial concentration uranium ion and concentration of uranium ion after adsorption respectively. The V was volume (L) and m was the mass of adsorption material (g).

3. Results and discussion

3.1 Characterization result

The infrared spectra of g-C_3N_4 and Fe_3O_4@g-C_3N_4 are shown in Figure.1.The two figure shape is basically same, there is a large absorption peat at 3130 cm^{-1}, and it decreased by the addition of Fe_3O_4 nano magnetic particles, which is attributed to the fusion of two kinds of particles. It is not different of peak shape at the 1340 cm^{-1} which is attributed to surface g-C_3N_4 has not been changed clearly. So, the combination of g-C_3N_4 and Fe_3O_4 particles maybe very good.

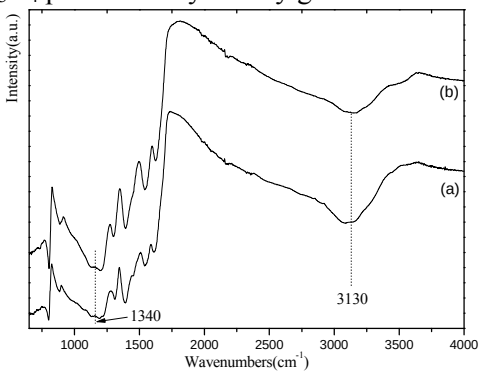

Figure 1 The infrared absorption spectrum of two samples.

The textural properties of adsorption material can be shown directly in Table 1.The surface area of g-C_3N_4 is small, but the aperture is so larger that accept and accommodate Fe_3O_4 magnetic articles. The aperture become smaller indicate that Fe_3O_4 particles stay in the channel of g-C_3N_4, which can lead the increasing of specific surface area, so the pore volume is increased significantly because the development of the micro structure,which is helpful to playing the better adsorption performance.

Table 1 The BET characterization results

Sample	Specific surface area /(m^2 • g^{-1})	Average pore size /nm	Average pore volume /(cm^3 • g^{-1})
Fe_3O_4@g-C_3N_4	23.13	16.70	0.10
g-C_3N_4	8.03	20.95	0.04

The pore size distribution is shown in Figure 2.The pore size distribution of g-C_3N_4 material is not centralized, but the pore size distribution mainly concentrate at the 4~6 nm after it is combined with the nano magnetic Fe_3O_4 particles, which can demonstrate that Fe_3O_4@g-C_3N_4 can be formed successfully.

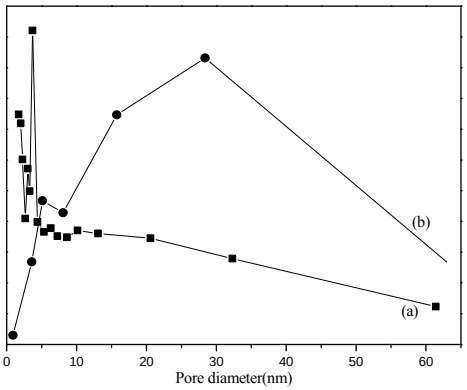

Figure 2 The pore size distribution image: (a) Fe_3O_4@g-C_3N_4 (b) g-C_3N_4

The XRD spectrum of the samples are displayed in Figure 3 exhibit characteristic diffraction peaks of Fe_3O_4 at 2θ=28.2°, 43.4°, 63.1°, 82.7° , which is corresponding to the literature [8]. It is indicate that there is no Fe^0, Fe^{2+}, Fe^{3+} single diffraction peaks existed, so the magnetic adsorption nano Fe_3O_4@g-C_3N_4 material is holistic and it has been prepared successfully.

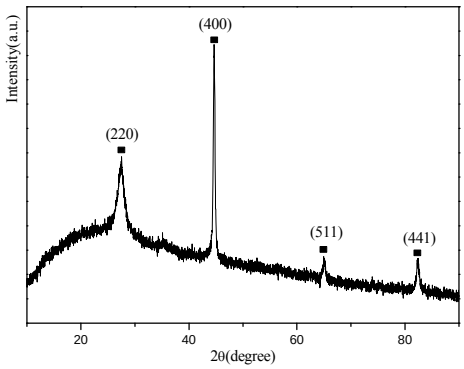

Figure 3. The XRD spectrum of Fe_3O_4@g-C_3N_4

The scanning electron microscope SEM of this kind of material is shown in Figure 4. When the surface of this material is magnified 5000 times, we get the bright and translucent material surface shadow. It is clearly that g-C_3N_4 have been coated nano Fe_3O_4 particles completely.

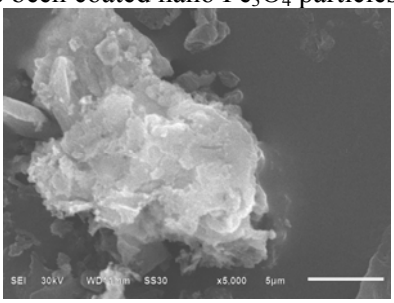

Figure 4. The SEM image of Fe_3O_4@g-C_3N_4

3.2 Effects of pH value

6 mg adsorption material was added in 20 mL liquid solution which contained uranium ion (the initial concentration is 140 mg/L). After adjusted the different pH values of the solution, the different adsorption properties were obtained. The experimental data were shown in Figure 5.

It is clearly that the maximum adsorption was performed when the pH value was 10, the adsorption capacity decreased distinctly with the continuous increament of the pH value. The adsorption activity was not ideal both in strong acidic or alkaline environment, but Fe_3O_4@g-C_3N_4 could be performed better adsorption performance in the neutral weak alkaline solution, so the optimum pH value was 10.

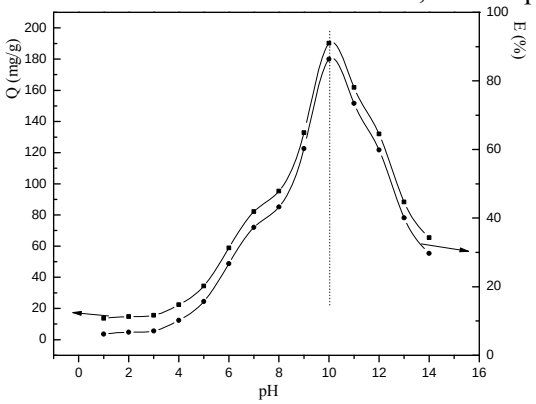

Figure 5. The effects of pH value

3.3 Effects of adsorbent dosage

On the above basis, many different mass of Fe_3O_4@g-C_3N_4 were added into 20 mL liquid solution which contained uranium ion (the initial concentration is 140 mg/L). The pH value was fixed as 10 by the addition of buffer solution, and the varied adsorption performance were shown in Figure 6.

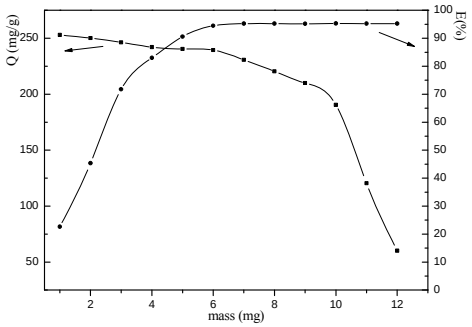

Figure 6. The effects of adsorbent dosage

With the increament of adsorbent, the adsorption rate increased carefully, and the adsorption rate reached the maximum value when the adsorbent mass was 6~7 mg. Therefore, the optimum adsorbent mass was 6.5 mg.

3.4 Effects of adsorption time

Similarly, 6.5 mg Fe_3O_4@g-C_3N_4 was added into 20 mL liquid solution which contained uranium ion (the initial concentration is 140 mg/L) and the pH value was fixed as 10 by the addition of buffer solution. The effects of adsorption time was shown in Figure 7. With the increament of adsorption time, the adsorption capacity and adsorption rate increased. The maximum adsorption rate reached the stable value when the adsorption time was 140 min and the adsorption capacity was 332.1 mg/g. Although the adsorption speed was not fast, but the maximum adsorption rate was more than 90%. So the optimum adsorption time was 150 min.

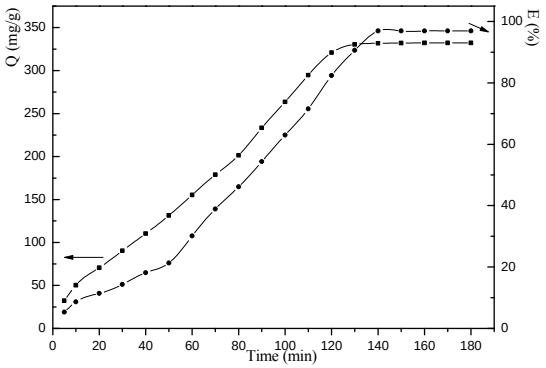

Figure 7. The effects of adsorption time

3.5 Effects of initial uranium concentration

The initial concentration of uranium has great influences in this system, the required adsorbent mass was large and the adsorption time was long when the initial concentration of uranium was high. The effects of initial uranium concentration was shown in Figure 8.

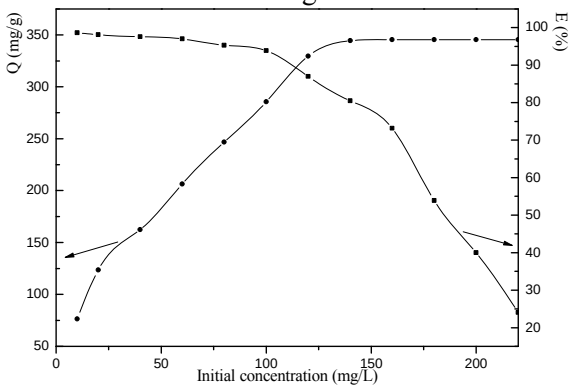

Figure 8. The effects of the initial uranium concentration

With the increament of the initial uranium concentration, the adsorption rate decreased which was attributed to the mass of Fe_3O_4@g-C_3N_4 was fixed, the concentration of uranium ion was also low and the active sites of adsorption material was sufficient. If the adsorption speed was large, the adsorption rate was high. After the adsorption reached to the saturation, the adsorption amount did not increase with the continuous increament of the initial concentration of uranium ion, so the adsorption rate decreased distinctly. The results showed that the optimal initial concentration of uranium ion was 140 mg/L and the maximum adsorption capacity had been reached 352.1 mg/g.

Generally speaking, it was good efficient adsorption material when the adsorption capacity was more than 200 mg/g. We also had chosen the activated carbon, molecular sieve, γ–Al_2O_3, silica as the carrier, the effects were not better than Fe_3O_4@g-C_3N_4 as same as encapsulated nano magnetic Fe_3O_4 particles. The reasons may be mainly relate to the correlate function, which is needed to be further explored and verified in the future.

4. Conclusion

A new nano magnetic material Fe_3O_4@g-C_3N_4 was prepared successfully, the special textural properties was certified by characterizations. Many experimental results demonstrated that it was a good adsorption material which could adsorb uranium ion effectively. It was clearly that g-C_3N_4 particles could be used to encapsulate nano magnetic Fe_3O_4 particles. The surface area of Fe_3O_4@g-C_3N_4 was ideal, and the pore structure distribution was uniform, so the adsorption performance was good in liquid solution contained uranium ion. The optimum pH value was 10, the optimum initial concentration of uranium ion was 140 mg/L, the optimum mass of adsorbent was 6.5 mg and the optimum adsorption time was 150 min. The maximum adsorption capacity has been reached 352.1 mg/g and the maximum adsorption rate was more than 90%.

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