

Мохаммед Нур Алхедер

Аспирант факультета наук

Химический факультет Университета Аль-Баас,

Хомс, Сирия.

Юсеф Джаммоал

Комиссия по атомной энергии,

Гидрометаллургическое отделение,

Дамаск Сирия.

Хер Алдин Алхатеб

Химический факультет,

Факультет наук, Университет Аль-Баас

Хомс, Сирия

Mohammad Nour Alkheder

PHD student, Faculty of Sciences

Department of Chemistry, Al-Baath University,

Homs, Syria.

Yousef Jammool

Atomic Energy Commission,

Hydrometallurgy Office,

Damascus, Syria.

Kher Aldeen Alkhateb

Department of Chemistry,

Faculty of Sciences, University of Al- Baath

Homs, Syria

REMOVAL OF URANIUM (VI) FROM AQUEOUS SOLUTION

Abstract: *A new amine-functionalized graphene oxide (GO-NH₂) nanosheet was prepared via covalently grafting reaction. The removal of U(VI) from aqueous solution with GO and GO-NH₂ was compared using batch method. The adsorption of uranium*

ions could be well-described by the Langmuir isotherm and pseudo-second kinetic model. The adsorption capacities of GO and GO-NH₂ were found to be 41.3 and 112.2 mg/g⁻¹ at RT, respectively. Results showed that adsorption capacity of GO was significantly improved by amine functionalization.

Keywords: Uranium (VI), Graphene oxide, Adsorption.

СИНТЕЗ КОМПЛЕКСА КОБАЛЬТА (II) С НОВЫМ ЛИГАНДОМ (1-АЗОФЕНИЛ, 4-АЗОМЕТИН ВАНИЛИН) ФЕНИЛ

Аннотация: Новый керосиновый нанолит из оксида графена (GO-NH₂) был получен с помощью реакции ковалентной прививки. Удаление U (VI) из водного раствора с помощью GO и GO-NH₂ сравнивали с использованием периодического метода. Адсорбция ионов урана может быть хорошо описана с помощью изотермы Ленгмюра и псевдосекундной кинетической модели. Было установлено, что адсорбционные способности GO и GO-NH₂ составляют 41,3 и 112,2 мг г⁻¹ при RT соответственно. Результаты показали, что адсорбционная способность ГО была значительно улучшена за счет функционализации амина

Ключевые слова: Уран (VI), оксид графена, адсорбция.

1. Introduction

Uranium plays an important role in the development and utilization of nuclear power [1]. However, large amounts of waste water containing uranium are produced by the nuclear industry, ore mining and industries [2, 3], which poses a threat in the environment because of uranium has chemical toxicity and radioactivity. Uranium can cause harmful diseases once it enters the food chain of human beings, such as lung, pancreatic and liver cancer [4], even at trace levels. Various technologies explored for the removal of uranium from aqueous solution include precipitation [5], electrolytic reduction [6], solvent extraction [7], membrane dialysis [8]. The GO and GONH₂ were characterized and sorption of uranium ions from aqueous solution was explored at different conditions of pH, contact time, temperature, as well as ionic strength. The

batch kinetics and equilibrium data were obtained. The effectiveness of these two sorbents for U(VI) sorption was compared.

2. Experimental:

2.1. Materials:

Nature large-flake graphite (325 mesh, 99.8 %) obtained from Qingdao Jinrilai Graphite Co., Ltd., China. PPD, EDC. And NHS were procured from J&K Scientific Co., Ltd., China. A stock solution of U (VI) were prepared by dissolving $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (Sinopharm Chemical Reagent Beijing Co., Ltd., China) into deionized water at the required initial concentrations. pH adjustments were performed with 0.1 mol L^{-1} Nitric acid and 0.1 mol L^{-1} sodium hydroxide solutions. All chemicals and reagents used in this study were of analytical grade and Without any further purification.

2.2. Preparation of GO nanosheets:

The GO nanosheets were prepared using improved Hummers method, typically, natural large-flake graphite (6 g) and NaNO_3 (3 g) were mixed with concentrated H_2SO_4 (70 mL) in a 500 mL three neck flask kept in an ice-water bath. KMnO_4 (18 g) was slowly added and the mixture was heated to $40 \text{ }^\circ\text{C}$ under stirring for 20 min. Deionized water (100 mL) was gradually added into the mixture and the temperature was kept at $95 \text{ }^\circ\text{C}$ for 20 min. The mixture was further treated by adding 300 mL deionized water and 7 mL H_2O_2 (30 %), then filtered and washed with 5 % HCl followed by deionized water. The product was isolated by centrifugation at 10,000 rpm for 60 min and washed repeatedly using deionized water. The GO nanosheets were obtained after drying under vacuum at $80 \text{ }^\circ\text{C}$ for 24 h.

2.3. Characterization of GO and GO-NH₂:

The prepared materials were characterized by Fourier transform infrared spectroscopy (FTIR) and Raman spectrometry. FTIR spectra were performed with Fourier transform infrared spectrophotometer, KBr pellets were used.

3. Results and Discussion:

3.1. Characterization of GO and GO-NH₂:

The main characteristic bands of the GO were found at 3414, 1739, 1624, 1231 and 1076 cm⁻¹. The band at 3414 cm⁻¹ is assigned to the stretching vibrations of –OH bonds, and the band at 1739 cm⁻¹ is due to the stretching vibration of C=O in carboxylic acid. The peaks at 1624, 1231 and 1076 cm⁻¹ correspond to the skeletal vibrations of aromatic C=C bonds, C–O–C and C–O in epoxy or alkoxy, respectively. Compared to pristine GO, the FTIR spectrum of the GO-NH₂ shows a distinct peak at around 1508 cm⁻¹ corresponds to N–H bending. The peak at 3348 and 3222 cm⁻¹ represent N–H asymmetric and symmetric stretching for amine-modified GO. The peaks at 1062 cm⁻¹ is ascribed to the C–N stretching vibration. The appearance of a band near 839 cm⁻¹ indicates the N–H bond out-of-plane bending vibration.

3.2. Effect of time and kinetics of adsorption:

Effect of contact time on the adsorption of U(VI) on GO and GO-NH₂ is illustrated in Fig. 1. The sorption rate increased rapidly at the beginning of contact time, which is attributed to the large number of vacant adsorption sites available on the sorbents at this stage. As time proceeds, the vacant adsorption sites are gradually filled up by uranyl ions, and adsorption becomes slow. For GO-NH₂, the gradual increase in adsorption capacity was observed with time at 1-3 h. This is because the kinetics will be more dependent on the rate at which intraparticle diffusion occurs which probably need a longer time to reach equilibrium. U(VI) sorption on GO and GO-NH₂ reaches equilibrium after 1h and 3h min, respectively. At equilibrium, the change of adsorption capacities for the both sorbents show no remarkable effects. Usually, adsorption kinetics includes two phases: the first initial sorption phase is rapid and contributed significantly to equilibrium uptake. It is interpreted to be an instantaneous adsorption stage or external surface adsorption due to the availability of a large number of adsorption sites. The second phase is slower and considered to be a gradual adsorption stage where intraparticle diffusion controls the uptake rate until the adsorption reaches equilibrium.

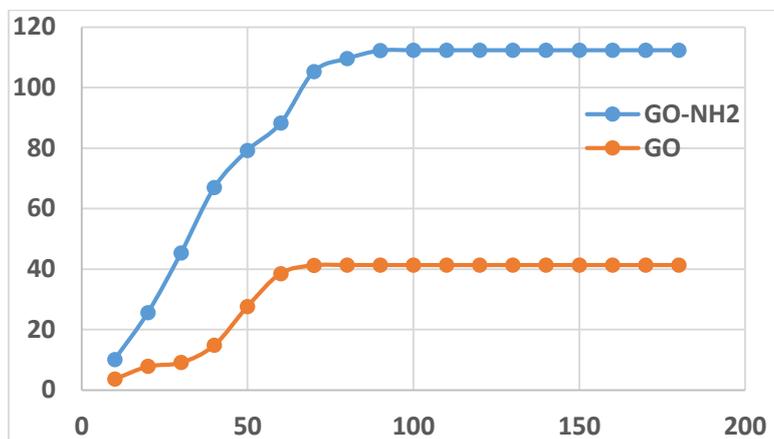


Figure 1. The effect of time on U(VI) adsorption onto GO and GO-NH₂
(C[U(VI)] = 60 mg L⁻¹, pH = 5.5, T = 25 °C)

4. Conclusion:

The GO was successfully amine-functionalized. The adsorption properties of U(VI) by GO and GO-NH₂ were investigated. In comparison with the GO nanosheets, the GO-NH₂ showed a better adsorption capacity onto U(VI) ions as the monolayer adsorption capacity was as high as 114.2 mg/g⁻¹, which was larger than that of GO nanosheets. Thermodynamic parameters demonstrate the adsorption of U(VI) ions by the GO nanosheets and the GO-NH₂ composites were spontaneous and endothermic nature of the process. The results indicate that the GO-NH₂ is a potentially useful adsorbent for removal uranium ions from aqueous solution with higher efficiency.

References:

- [1]- Manos MJ, Kanatzidis MG. Layered metal sulfides capture uranium from seawater. *Journal of the American Chemical Society*. 2012 Sep 25;134(39):16441-6.
- [2]- Shao DD, Hou GS, Li JX, Ren XM, Wang XK. PANI/GO as a super adsorbent for the selective adsorption of uranium(VI). 2014, *Chem Eng J* 255:604–612.
- [3]- Fiedor JN, Bostick WD, Jarabek RJ, Farrell J. Understanding the mechanism of uranium removal from groundwater by zero-valent iron using X-ray photoelectron spectroscopy. 1998, *Environ Sci Technol* 32:1466–1473.

- [4]- Anirudhan TS, Radhakrishnan PG. Improved performance of a biomaterial-based cation exchanger for the adsorption of uranium(VI) from water and nuclear industry wastewater. 2009, *J Environ Radioact* 100:250–257.
- [5] Zhang C, Dodge CJ, Malhotra SV, Francis AJ. Bioreduction and precipitation of uranium in ionic liquid aqueous solution by *Clostridium* sp. 2013, *Bioresour Technol* 136:752–756.
- [6] Ohashi Y, Harada M, Asanuma N, Ikeda Y. Feasibility studies on electrochemical recovery of uranium from solid wastes contaminated with uranium using 1-butyl-3-methylimidazolium chloride as an electrolyte. 2015, *J Nucl Mater* 64:119–127.
- [7] Elsayed HM, Fouad EA, El-Hazek NMT, Khoniem AK. Uranium extraction enhancement from phosphoric acid by emulsion liquid membrane. 2013, *J Radioanal Nucl Chem* 298:1763–1775.
- [8] Kedari CS, Pandit SS, Gandhi PM. Separation by competitive transport of uranium(VI) and thorium(IV) nitrates across supported renewable liquid membrane containing trioctylphosphine oxide as metal carrier. 2013, *J Membr Sci* 430:188–195