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Uranium(VI) adsorption on graphene oxide nanosheets from aqueous solutions

Zijie Li [1], Fei Chen [1], Liyong Yuan, Yalan Liu, Yuliang Zhao, Zhifang Chai, Weiqun Shi *

Key Laboratory of Nuclear Analytical Techniques and Key Laboratory for Biomedical Effects of Nanomaterials and Nanosafety, Institute of High Energy Physics, Chinese Academy of Sciences, Beijing 100049, China

HIGHLIGHTS

- Single-layered graphene oxide (GO): an effective sorbent material for UO_2^{2+} removal.
- The sorption is pH-dependent and ionic strength independent.
- Oxygen-containing groups of GO play crucial roles in the sorption of UO_2^{2+}.
- Formations of inner-sphere complexes of UO_2^{2+} on GO with oxygen-containing groups.

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ABSTRACT

Single-layered graphene oxide (GO) prepared by the Hummers method was used to adsorb U(VI) ions from aqueous solutions. The U(VI) sorption was studied as a function of solution pH, ionic strength, and initial concentration of U(VI) using a batch method in air. It is found that the sorption is rapid, strongly dependent on pH, attaining a plateau at pH 4.0–7.5, and independent of the ionic strength. This suggests the formation of inner-sphere surface complexes of U(VI) on GO. The maximum sorption capacity of GO for U(VI) was evaluated to be 299 mg/g at pH 4.0, ranking it among the most effective sorbents reported for U(VI) so far. The abundant oxygen-containing functional groups of GO were demonstrated to play crucial roles in the sorption. Using the distribution coefficients obtained at different temperatures, thermodynamic parameters were also calculated, showing that the sorption is endothermic and spontaneous. GO could be a promising sorbent material applied in the environmental cleanup of uranium pollution and the enrichment of uranium from large volumes of solution.

1. Introduction

Currently, global climate change and fossil fuel depletion make more and more countries develop nuclear fission energy to ensure their energy security. In 2008, the global uranium production for power generation was recorded as 40,000 tons, supplying approximately 15% of the world's electrical energy [1]. At the same time, large amounts of uranium are inevitably released into the environment due to nuclear fuel cycle activities, imposing long-term threat to human beings and bio-organisms [2,3]. The recent Fukushima nuclear leakage shocked the world heavily, causing enormous setback to the credibility of nuclear power. On the other hand, due to the expected shortage of uranium in near future, the recovery of uranium from unconventional resources such as sea water and waste solutions becomes necessary for the sustainable development of nuclear energy [4]. Hence, highly efficient enrichment of uranium as well as its removal from aqueous solutions is of extreme importance from the point of view of full utilization of uranium resources and environmental protection.

Solid-phase extraction techniques based on carbon materials, such as activated carbon and carbon nanotubes (CNTs) have been used in the removal and recovery of uranium from aqueous solutions [5]. Carbon materials are chosen for this purpose because of their higher thermal and radiation resistance than organic exchange resins and better chemical stability than familiar inorganic sorbents in strongly acidic solutions in the most cases of nuclear wastewaters. It was reported that oxidized multi-walled carbon nanotubes (MWCNTs) are suitable materials in the pre-concentration and solidification of U(VI) from large volumes of aqueous solutions [6]. It is fast to achieve sorption equilibrium and the sorption is mainly dominated by the surface complexation and cation-exchange of UO_2^{2+} with oxygen-containing functional groups, i.e. hydroxyl, carbonyl, and carboxyl groups on MWCNTs.

Graphene is a carbon layer only one atom thick, composed of sp^2-bonded carbon. Since the first report of electrical properties of graphene in 2004 [7], it has attracted intense interest because

* Corresponding author. Tel.: +86 10 88233968; fax: +86 10 88235294.
E-mail addresses: lizijie@ihep.ac.cn (Z. Li), chenfei@ihep.ac.cn (F. Chen), yuanly@ihep.ac.cn (L. Yuan), liuyalan@ihep.ac.cn (Y. Liu), zhaoyuliang@ihep.ac.cn (Y. Zhao), chaizf@ihep.ac.cn (Z. Chai), shiwq@ihep.ac.cn (W. Shi).
[1] These authors contributed equally to this work.

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