

Contents lists available at ScienceDirect

Carbon

journal homepage: www.elsevier.com/locate/carbon

Oxidatively modified carbon as efficient material for removing radionuclides from water

Artur Khannanov^a, Vadim V. Nekljudov^a, Bulat Gareev^a, Airat Kiiamov^a,
James M. Tour^{b, **}, Ayrat M. Dimiev^{a, *}^a Laboratory of Advanced Carbon Nanomaterials, Kazan Federal University, Kremlyovskaya Street 18, Kazan 420008, Russian Federation^b Department of Chemistry, Department of Material Science and Nano Engineering, and NanoCarbon Center, Rice University, MS-222, 6100 Main Street, Houston, TX 77005, USA

ARTICLE INFO

Article history:

Received 18 October 2016

Received in revised form

29 December 2016

Accepted 9 January 2017

Available online 10 January 2017

ABSTRACT

There is a constant need to develop advantageous materials for removing radioactive waste from aqueous systems. Here we propose a new carbon-based material prepared by oxidative treatment of various natural carbon sources. The as-prepared oxidatively modified carbon (OMC) has an oxygen-rich surface, and retains its particulate granular texture. It has relatively low cost and can be used in traditional filtration columns. The sorption ability of OMC toward several metal cations is demonstrated. It is especially efficient toward Cs⁺ cations, the species that are among the most difficult to remove from the waters at the Fukushima nuclear plant.

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1. Introduction

Radioactive waste imposes a serious problem to modern society, threatening life and health of many worldwide. The accidental releases of radioactive waste from military or civil nuclear activities can contaminate large areas, as it happened in 1957 at Kyshtym, or in 1986 in Chernobyl, both in the former USSR. The recent accident at the Fukushima nuclear power plant in Japan underscores the magnitude of the problem [1]. Currently, hundreds of thousands tons of water contaminated by radionuclides are stored in massive tanks on the plant site awaiting efficient methods of purification. Among the radionuclides most difficult to remove from water is ¹³⁷Cs⁺ [1]. The ease with which radionuclides can be removed from water decreases in the order of trivalent, divalent to monovalent metal cations. While trivalent elements can be almost quantitatively removed by numerous known absorbents, the sorption of monovalent ¹³⁷Cs⁺ remains an enormous challenge [1–4]. Sorption with porous minerals [5–12], and ion-exchange media [13] are among the most effective methods for the Cs⁺ removal.

One type of materials effectively sorbing Cs⁺ are synthetic ferrocyanides [11,12]. However, these materials suffer from their

nanometer or micrometer scale size that makes them difficult to be used in large-size continuous systems [12]. This is why they need to be immobilized in suitable organic or inorganic matrices [12]; this additionally increases their cost. Among the known Cs⁺ sorbents that found practical implementation are synthetic zeolites [5–10]. However, the relatively high cost of synthetic zeolites lowers their performance-to-cost ratio. In addition, after sorption, Cs⁺ cannot be removed from ferrocyanides and zeolites as easy as from the ion-exchange resins. Thus, the spent materials need to be compartmentalized and stored along with sorbed radionuclides until their near complete decomposition by the natural radioactive decay.

From this perspectives, carbon-based materials have advantages over the minerals. In practice, after sorption, carbon can be burned in a nuclear furnace leaving only radioactive ash, which results in an ultra-small volume of radioactive waste. Alternatively, carbon-based materials can be regenerated and be used in a new purification cycle. But traditional carbon-based absorbents such as activated carbon and charcoal are not efficient scavengers of metal cations. While being effective in gaseous phases and for removing some organic contaminants from water, they do not efficiently bind metal cations from aqueous solutions. For this purpose, the carbon surface needs to be decorated with oxygen functional groups.

Recently, the method of removing radionuclides by graphene oxide (GO) was reported [14–18]. GO was demonstrated to be very efficient toward several trivalent and divalent metal cations due to

* Corresponding author.

** Corresponding author.

E-mail addresses: tour@rice.edu (J.M. Tour), AMDimiev@kpfu.ru (A.M. Dimiev).