



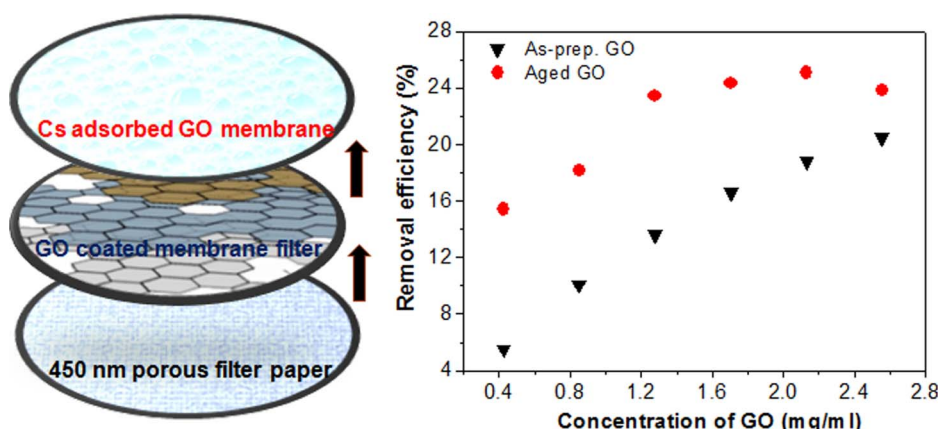
Graphene oxide supported filtration of cesium from aqueous systems

Pavan K. Narayanam*, Jishnu A., K. Sankaran

Materials Chemistry Division, Materials Chemistry & Metal Fuel Cycle Group, Indira Gandhi Centre for Atomic Research, Kalpakkam 603102, India



GRAPHICAL ABSTRACT



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ABSTRACT

Graphene Oxide-Cesium (GO-Cs) composites, in the form of dispersions as well as solid state thin films, were prepared by introducing different concentrations of Cs into aqueous GO dispersions or subphase. Interaction mechanism of Cs with various functional groups of GO and the extent of Cs uptake on GO sheets under different ambient conditions were investigated using FT-IR, Raman, PL and XPS techniques. Cs was noticed to exhibit preferential interaction with hydroxyl groups of GO and furthermore, incorporation of Cs causes significant change in relative ratios of functional groups, inferring a major structural reorganization in the composite sheets. FT-IR and PL studies showed that aged GO samples can accommodate relatively larger quantities of Cs compared to fresh GO sheets, owing to largely available $-\text{COOH}$ and free/associated hydroxyl groups in aged GO sheets. Intercalation studies on GO multilayers indicated that extent of Cs uptake on GO layers increases with duration of intercalation as well as quantity of GO available for interaction. Membrane filters of GO were prepared by controlled coating of GO sheets on filter papers and their adsorption and filtration abilities were tested by filtration of aqueous Cs solutions. Aged GO coated membrane filter displayed superior adsorption abilities compared to those coated with fresh GO sheets, which corroborate well with the inferences drawn from the spectroscopic investigations. In the present work, the prepared GO membrane filters showed a maximum Cs adsorption capacity of ~ 148 mg/g.

* Corresponding author.

E-mail address: pavankumarn@igcar.gov.in (P.K. Narayanam).

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1. Introduction

Leakage of radionuclides into environment is a serious public concern, as it can contaminate the air-water-soil, enter into the food chain and ultimately become a part of living beings, which can be a potential threat for biodiversity and human health. During the reactor accidental conditions, venting of radioactive gases and burning of spent fuel rods may take place, which cause leakage of several volatile radioactive nuclides such as, ^{137}Cs , ^{134}Cs , ^{131}I , ^{235}U , ^{90}Sr etc. Among the several radioactive nuclides and fission products generated, radionuclides of cesium (Cs) are considered as potential threat due to their long half lives and other destructive effects on environment. The radio isotopes of cesium, ^{134}Cs and ^{137}Cs are highly volatile in nature, display radiation stability and possess high ion-exchange capacity. The radioactive liquid waste generated from the reprocessing of spent nuclear fuel contains significant amount of radioactive cesium, which emits beta and gamma rays. Radioactive cesium is chemically analogous to sodium and potassium, and hence can be easily bio-accumulated in the human body, gets deposited in the soft tissues and creates internal hazard such as thyroid cancer. Further, absorption of high dosage of ^{137}Cs in human body can cause medullar dystrophy, disorder of reproductive function and bad effect on liver and renal functions [1]. Apart from its negative impacts, ^{137}Cs is an excellent source for gamma irradiation, which has potential medical applications such as sterilization of medical ware, equipment etc. Selective removal of radioactive Cs from low level liquid waste and aqueous systems not only reduces the radioactivity from the targeted systems, but also recovers a strong source for gamma irradiation [2].

Various chemical separation methods and waste treatment technologies such as, ion-exchange [3], solvent extraction [4], electro-dialysis [5], chemical precipitation [6], adsorption [1], sand filtration [7], membrane processes [8] etc. have been adopted for the separation/removal of Cs from the radioactive waste, contaminated surface/ground water. Among these, adsorption method was reported to have definite advantages in terms of cost, flexibility, simplicity of design and operation, adaptability to different types of liquid wastes, insensitivity to toxic pollutants [1] etc. Further, adsorption processes do not leave out large volumes of harmful disposals at the end of the process and the adsorbents can be regenerated, wherever reversible adsorption is possible.

Removal of cesium from contaminated systems was investigated using variety of inorganic adsorbents including clays [9], zeolites [10], biopolymers [11], activated carbon [12], silica [13] etc. Adsorption of Cs on a variety of Turkish clays, including bentonite, sepiolite, kaolinite and natural mordenite were investigated and the observed superior adsorption was attributed to the involved ion exchange mechanisms [14]. Adsorption of Cs on adsorbents made up of mesoporous silica, layered manganese oxide were investigated earlier and it was reported that these conjugate adsorbents are highly pH sensitive and able to selectively remove the Cs in neutral pH region [13]. Bio-sorbent materials such as, *Funaria hygrometrica* and its composites with polysilicates were tested for adsorption of ^{137}Cs from low strength radioactive material and it was reported that the radionuclide adsorption is supported by –carboxylate groups in the composite [11]. Prussian blue (PB, ferric hexacyanoferrate, $\text{Fe}_7(\text{CN})_{18}$) is another important material used as an adsorbent in decontamination of radioactive cesium, owing to its high selectivity towards cesium. PB nanoparticles with higher density of hydrophilic defect sites were reported to possess the superior Cs adsorption capability, which is attributed to adsorption of Cs^+ ions onto the defect sites of nanoparticles by proton-elimination reaction from the coordinated waters [15].

Considering the exceptional and superior characteristics such as, large surface area, low specific mass, unique amphiphilic nature and chemical stability, graphene oxide (GO) and its composites have been investigated as potential materials for attenuation of radionuclides. Diverse and abundant oxygen functional groups of GO can provide

sorption sites for selective removal and exhibit optimal sorption towards variety of radionuclides. There are several reports on removal of radionuclides using GO composites, especially on sorption of uranium (U), thorium (Th), europium (Eu), cesium (Cs) and strontium (Sr) from radioactive wastes as well as contaminated aqueous systems [16–18]. These reports primarily describe the preparation of GO based materials for sorption of radionuclides, their sorption kinetics and capacities, sorption isotherms for various radionuclides etc. Romanchuk et al. carried comparative studies on interaction of GO with set of actinides including Am(III), Th(IV), Pu(IV), Np(V), U(VI) and typical fission products such as Sr(II), Eu(III), Tc(VII) and their sorption kinetics. It has been reported that coagulation of cation/GO results in formation of nanoparticle aggregates of GO sheets, which facilitates the removal of radionuclides [16]. Sun et al. investigated sorption of U(VI), Th(IV), Eu(III), Sr(II) and Cs(I) radionuclides on to various functionalized/composite forms of GO as a function of pH, ionic strength, initial concentration and reported that chemical affinity of radionuclides for nitrogen-containing functional groups is stronger than that for oxygen-containing functional groups [18–21]. Pan et al. investigated the adsorption of Th(IV) on reduced graphene oxide (RGO) as a function of pH, contact time, adsorbent dosage, initial concentration, temperature and reported that the adsorption of Th(IV) on RGO is a spontaneous process and endothermic in nature [22]. In a recent work on competitive sorption mechanisms of U(VI) and Sr(II) on GO, Cheng et al. reported that GO exhibit superior chemical affinity towards U(VI) compared to Sr(II) and increase of U(IV) concentration supply new sorption sites for Sr(II) due to the shift of inner-sphere to outer-sphere surface complex formations [23].

There are few reports on removal of radionuclides of Cs from radioactive wastes as well as contaminated aqueous systems using GO composites. Tan et al. investigated the adsorption of Cs^+ on GO as a function of reaction time, pH, ionic strength and temperature. In these studies, adsorption of Cs^+ on GO was found to be dependent on the ionic strength, while the process of formation of corresponding outer- and inner-sphere surface complexes as sensitive to pH [24]. Deming Li et al. synthesized the magnetic graphene oxide using chemical co-precipitation method and reported that the adsorption of Cs^+ on magnetic graphene oxide increase with increasing pH in the range of 2–6. At low pH conditions, adsorption of Cs^+ was reported to take place through ion-exchange mechanism and inner-sphere surface complexation was observed to take place at pH 5 [25]. Romanchuk et al. investigated the sorption mechanism of Cs^+ on GO under various pH and competing cationic conditions. These studies revealed that ion exchange is the predominate mechanism for Cs sorption [26]. Jang et al. prepared prussian blue (PB)/reduced graphene oxide foam (RGOF) composite using one-step hydrothermal reaction and the prepared composite showed enhanced removal efficiency of ^{137}Cs from the contaminated water, compared to that of precursor RGOF. The enhanced adsorption efficiency was attributed to large surface area of the 3D porous RGO foam and superior ion-exchange properties of the PB nanoparticles [27].

Yang et al. synthesized graphene oxide and prussian blue (PB) coated Fe_3O_4 magnetic nanoparticles for removal of cesium from contaminated water systems. The higher adsorption efficiencies exhibited by composite adsorbents were attributed to selective Cs removal by PB and the cooperative action between PB and graphene oxide [28]. Kadam et al. synthesized pectin-stabilized magnetic graphene oxide Prussian blue (PSMGPB) nanocomposites for cesium removal from waste water. The optimum temperature and pH conditions for Cs adsorption by these composites were reported at 300 °C and pH 7, respectively. Increased adsorption capacity of PSMGPB nanocomposites was attributed to the pectin-stabilized separation of graphene oxide sheets and enhanced distribution of magnetites on graphene oxide surface [29]. Jang et al. prepared PVA-alginate encapsulated Prussian blue-graphene oxide (PB-GO) hydrogel beads as adsorbent for the removal of cesium from aqueous solutions. Cs adsorption capacity of PB-

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