Synthesis of Graphene Oxide Based Material for Removing Radioactive Iodine

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1. Introduction

Radioactive iodines (125/129/131I) are released into environments during nuclear fuel processing, nuclear power plant operation and severe accidents such as Chernobyl and Fukushima. Environmental concern is mostly for ^{129}I due to its high toxicity and long half-life, $t^{1/2} = 1.6 \times 10^7$ years [1]. Usually, to remove the radioactive iodine, activated carbon based organic materials and silver exchanged zeolite were used as sorbents in aqueous phase. However, these materials have the limits in removal efficiency, cost, and high selectivity [2]. Therefore, this study investigated the bismuth functionalized graphene oxide (Bi-GO) to test high removal efficiency and selectivity of iodide and iodate species under the various Cl^- background solution conditions.

2. Methods

2.1 Synthesis of graphene based sorbent

To functionalize the sorbent, the graphene oxide (GO) has been synthesized from natural graphite powder (99.95%) by the modified Hummers method [3], and then graphene oxide was functionalized with $Bi_2O_3 \cdot 2H_2O$ in deionized water.

2.2 Characterizations

After synthesis, sorbent was characterized using X-ray diffraction (XRD) to identify the crystalline minerals, Scanning Electron Microscope (SEM) / Energy Dispersive Spectrometer (EDS) for microstructure and elemental chemistry of samples, and Fourier transformed-infrared (FT-IR) for the functionalized group identification.

2.3 Iodine removal experiments

Iodine batch adsorption experiments were conducted using a 10 mL glass amber vial with non-radioactive iodine solutions such as iodide (I^-) and iodate (IO_3^-). The Bi-GO (0.05 g) was mixed with 10 mL of Cl^- (1, 10, and 100 ppm) background solutions with different iodide or iodate concentrations (0.1, 1, and 4 ppm). Using Bi-GO, iodine adsorption experiments were carried out for different reaction times (15 min, 30 min, 1 h, 1 day, 3 days, and 7days) to check the adsorption kinetics. After each reaction, the supernatants were separated by syringe filter (0.45 μm). The final concentration of iodine in batch solution was analyzed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The removal efficiency of iodine and the results were compared with 35% silver exchanged zeolite from Sigma Aldrich.

3. Results & Discussion

3.1 Characterizations

Graphene oxide is a layered material consisting of hydrophilic oxygen functional groups on their basal planes and edges. The bismuth-functionalized graphene oxide used in this study was a major sorbent material from the synthesis of bismuth and graphene oxide; photographic images of the graphene oxide and bismuth-functionalized graphene oxide samples are shown in Fig. 1.

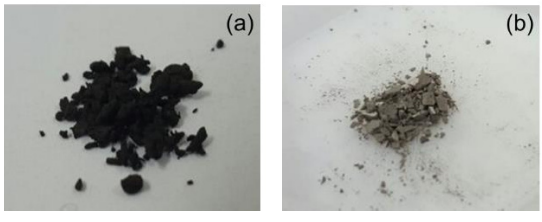

Fig. 1. (a)Graphene oxide and (b) Bismuth-functionalized graphene oxide.

After functionalization, graphene oxide color has been changed from deep black to light grey.

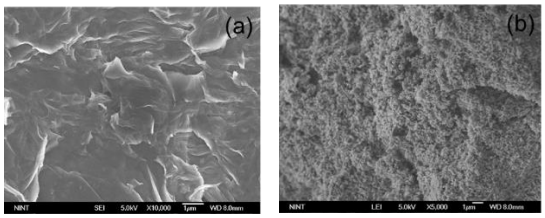

Fig. 2. SEM images of (a) graphene oxide and (b) bismuth-functionalized graphene oxide.

In Fig. 2, SEM images showed the morphology of the graphene oxide and the presence of functionalized bismuth on graphene oxide surfaces. The bismuth in these images appears to adhere to the surface of the graphene oxide, forming discrete Bi particles.

3.2 Iodine removal efficiency

The results of iodine removal experiments under the Cl^- background solution condition and iodine initial concentration (4 ppm) are presented in Fig. 3. These experiments were performed to compare the removal efficiency between the developed material and commercial Ag-zeolite sorbents. The grapheme oxide based material with Bi presented high removal efficiencies (≥95%) for iodide and iodate regardless of background solution concentrations. In contrast, the Ag-zeolite showed similar tendencies for iodide without effect of background solutions. However, removal efficiencies for iodate was decreased to less than 25% with increasing Cl^- concentrations in Ag-zeolite.

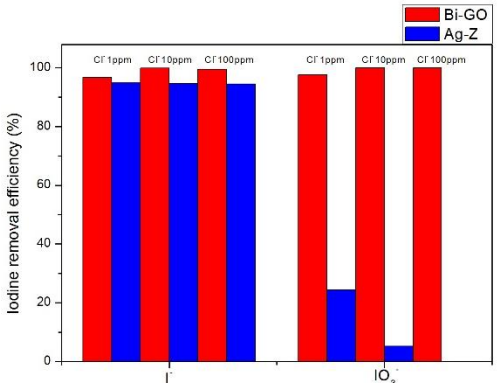

Fig. 3. Selective adsorption for iodide and iodate under the Cl^- background solution condition.

These results suggest that the bismuth-functionalized graphene oxide demonstrate higher iodine removal performance (especially for iodate) than the Ag-zeolite and the possibility of selective iodine removal from wastewater containing various radionuclides generated from severe accident or operation of nuclear power plant.

4. Conclusion

In summary, we report bismuth-functionalized graphene oxide synthesis as novel absorbent for iodine removal and compare the results with the commercial Ag-zeolite under different ionic strength conditions. The bismuth based materials show higher iodine removal efficiencies (≥95%) than Ag-zeolite for iodide and iodate. In contrast, the Ag-zeolite has a limitation for removal capacity for iodate. Our study provides potential role of the grapheme based material on selective removal of both radioactive iodide and iodate in wastewaters.

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