Fabrication of silver-graphene fiber composite for radioactive iodine adsorption

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1. Introduction

Although a plenty of advanced developments in nuclear technologies have been achieved since nuclear reactors were initiated for electricity generation, it is still not able to earn trust from the public due to the potential environmental problems. Various man-made radionuclides produced from uranium fissions can significantly affect the environment and human health when they are released. Radioisotopes of iodine, which are elements directly related to the biological activities, can be easily transferred through ground water, accumulated in human thyroids, and cause cancers. Isotopes of iodine from ^{127}I to ^{144}I are yielded from fission of ^{235}U. Only ^{129}I has a considerably long half-life (1.6×10^7 years), while ^{127}I is stable and the others just a few days of half-life (^{131}I, 8.04 days) at the longest. Eventually, 76.5% of ^{129}I and 23.5% of ^{127}I remain in a spent fuel within 2 years of cooling [1]. However, during the reprocessing process, at least 94% of iodine is evaporated and released to the gas streams in the form of mostly I_2 (g) [2]. To prevent the release of radioactive iodine gas from a reprocessing facility, various techniques have been developed for removing radioactive iodine in the off-gas stream, scrubbing or using solid sorbents. Nowadays, the use of solid sorbents is regarded as more attractive method than scrubbing due to no requirement of large capacity for toxic and corrosive chemicals.

Table 1. A variety of solid sorbents and their iodine capture abilities.

Sorbent	*Maximum mass increase (% mass)*	
$AgNO_3$-Al_2O_3 (Ag-10 wt%)	20	[3]
Ag-Z (mordenite)	17	[4]
Ag-silica aerogel	32	[5]
Graphene powder	45	[6]
Graphene aerogel	51	[6]

Silver (Ag) and ceramic (alumina or zeolite) composites materials [3-5] have been extensively studied for iodine capture due to the chemical properties of silver, which can react with iodine compounds and capture iodine atoms by forming AgI (s), and the excellent chemical and thermal stability of the ceramics. Carbon-based sorbents can be also good candidates for iodine capture materials due to the physisorption properties of the graphite basal plane. Recently, large surface carbon-based nanomaterials, graphene powder and aerogel, are also researched and showed much better performance for iodine removal than other silver composite materials [6]. Since the physisorption is a non-selective and reversible reaction, however, the iodine capture ability of the carbon materials would be degraded due to other volatile gases such as NOx or volatile organic compounds and non-immobilization of iodine. Also, the fine powders or coarse granules (or aerogel) are not suitable form of sorbents for gas streams [7]. In the case of a powder-type sorbent, its pores become narrow due to the pressure of gas flow, and then the removal efficiency of iodine from the gas flow is eventually dropped. Even though a granular-type sorbent is not compacted by the gas flow, it is also inefficient for iodine capture because the diffusion of adsorbate from the surface to the inner pores of the sorbent is slow**.**

In this study, the iodine sorbents was prepared using graphene fiber, which is a new materials invented recently and has been received attention due to its high mechanical, electrical, and thermal properties. For achieving high iodine removal efficiency from the gas stream, fiber-type sorbents were prepared, which can overcome the compaction problem of powder sorbents and the slow kinetics problem of granule sorbents. Also, the fibers were hybridized with silver nanoparticles to increase selectivity and immobilization ability of the sorbents for iodine.

2. Experiment preparation

2.1 Preparation of graphene oxide (GO) solution

Graphene oxide solution was first prepared with large graphite flake (-10 mesh) by using the combination of two oxidation methods, Kovtyukhova's and Hummers' methods [9]. After the reaction, the prepared GO solution was washed several times with water and HCl and concentrated by 8mg/L with a centrifuge, then GO liquid crystal can be obtained.

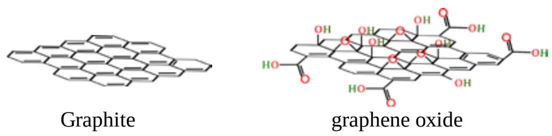

Figure 1. The schematic structure of graphite and graphene oxide.

2.2 Preparation of GO fibers

The GO solution was doped on a syringe pump and injected (~600 μL/min) through a thin (below 18 gauge) spinneret into a coagulation bath contained a solution of 5 wt% $CaCl_2$ dissolved in the mixture of H_2O and EtOH at a ratio of 3:1 (volume) [10]. After 30min of coagulation, GO fibers were stretched and winded up in a specific frame as shown in Figure 2. And then the GO fiber was dried in a vacuum oven at room temperature for 12 hours.

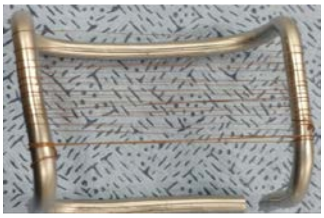

Figure2. Graphene oxide fibers

2.3 Preparation of silver/graphene fiber composites

The silver/graphene fiber composites were prepared by reducing Ag^+ and GO fiber with $NaBH_4$, simultaneously. If alkaline solution is mixed before adding the reducing agents, it can motivate the hybridization of Ag(s) and graphene because Ag_2O (s) is formed at high pH and precipitated on the functional groups of GO.

2.4 Material characterization

The morphology and the atomic distribution of the Ag-graphene fiber composite were observed using SEM and EDS. To verify the existence of Ag(s), the crystalline structure of the composite were characterized with XRD. The weight loading of Ag can determined from the XPS spectrum. The specific surface and pore size distribution were estimated by nitrogen isotherm.

3. Conclusions

Decontamination of radioactive nuclides has been investigated to secure the environmental reliability of nuclear power. Radioactive iodine, especially ^{129}I, must be removed due to its carcinogenic effects on human health. Various types of adsorbents which contains silver have been reported for iodine removal. Graphene-based large-surface-area materials also suggested for iodine capture, but the low selectivity and revolatilization could be a significant problems for current applications. Furthermore, the compaction or the slow kinetic of particular-form of adsorbents is not suitable for efficient iodine capture. In order to overcome the problems, graphene fiber-based silver hybridized adsorbents were prepared. The new adsorbents materials are expected tohave the better performance for iodine capture than other researched materials.

ACKNOWLEDGEMENT

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