

# Graphene Fiber and its Silver Functionalization for Radioactive Iodine Adsorption

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## 1. Introduction

Among different challenges in the radioactive waste management at each stage of the nuclear fuel cycle, capture of radioactive iodine is a growing priority for research and developments. The major source of radioiodine is the aqueous reprocessing of used nuclear fuel (UNF) as it is dissolved in nitric acid (PUREX process) [1]. Iodine radioisotope  $^{129}\text{I}$  is a greatest concern due to its long half-life of  $1.6 \times 10^7$  y and high mobility in environment. The development of the possible methods for radioiodine capture is necessary.

There are two types of iodine capture technologies such as solid sorbents and wet scrubbing. Solid sorbents, based adsorption techniques, are effective and advantageous than the wet processing [2]. Activated carbon (AC) is a potential solid sorbent due to its pore size and high specific surface area. But the presence of nitrogen oxide in off-gas decreased the capacity of AC and leads to explosive reactions [3]. Other solid sorbents are Ag-loaded alumina (AgA), Ag-loaded silica (AgS), and Ag-loaded zeolite (AgZ).

Graphene, a two-dimensional material, has been investigated for different kinds of energy and environmental application. The theoretical specific surface area of a single layer surface area is very large ( $2600 \text{ m}^2 \text{ g}^{-1}$ ) [4]. Graphene-based nano-materials also show high adsorption for iodine [5].

This work presents graphene fiber for the adsorption of gaseous radioactive iodine from off-gas stream of nuclear fuel cycle-related facilities. This work investigates the iodine adsorption efficiency of graphene oxide (GO) fiber, reduced graphene oxide (rGO) fiber, and silver functionalized graphene (Ag/G) fiber. The synthesis and characterization of the fibers are also described.

## 2. Methods and Results

In this section, the graphene fiber fabrication process and silver functionalization process are described. Adsorption experimental procedure, characterization techniques and the adsorption results are also described.

### 2.1 Graphene oxide (GO) fiber fabrication

Graphene oxide (GO) solution was purchased from Grapheneall Co. Ltd and fibers were fabricated through the wet-spinning process. Graphene oxide is injected at a rate of  $300 \mu\text{l}/\text{min}$  into a rotating coagulation bath

where 5 wt% of  $\text{CaCl}_2$  is mixed with the solution of ethanol/water (1/3 v/v). After 10 min coagulation, these fibers are washed with the solution of ethanol/water (1/3 v/v) and dried at room temperature. Final graphene fibers are obtained through drying in a vacuum oven at  $60^\circ\text{C}$  for 12 h.

### 2.2 Chemical reduction of GO fiber

Graphene oxide fibers are reduced by L-ascorbic acid (Vit. C). Fibers are immersed in 0.1 M L-ascorbic acid solution and heated at  $90^\circ\text{C}$  for 12 h. The reduced graphene oxide fibers are washed with distilled water and ethanol. Finally, the reduced graphene fibers are collected by vacuum drying at room temperature.

### 2.3 Silver (Ag) functionalized graphene fiber fabrication

Silver nitrate ( $\text{AgNO}_3$ ) (1 wt%) is added with GO solution and through the wet spinning process described in section 2.1, the silver functionalized graphene oxide (Ag/GO) fibers are fabricated. This Ag/GO fibers are then reduced by the by L-ascorbic acid (Vit. C), which is described in section 2.2.

### 2.4 Iodine adsorption experiment

Iodine adsorption experiment had been carried out in saturated iodine environment inside a vacuum desiccator. The fibers were allowed to adsorb non-radioactive iodine at  $150^\circ\text{C}$  for different time. The iodine adsorption efficiency is calculated using the following equation  $w\% = (M_{\text{final}} - M_{\text{initial}})/M_{\text{initial}} \times 100\%$ , where  $M_{\text{initial}}$  and  $M_{\text{final}}$  are the mass of graphene fibers before and after iodine adsorption, respectively.

### 2.5 Characterization techniques

Scanning electron microscope (SEM) was used to observe the surface morphology of the fibers. X-ray photoelectron spectroscopy (XPS) show the elemental composition and chemical state of the different fibers. Thermogravimetric analysis (TGA) will be carried out to determine the thermal stability of the fibers.

### 2.6 Results and discussion

Fig. 1 shows the SEM images of the rGO fiber and Ag/G fiber. The compact form of graphene sheets is

observed. Presence of silver oxide ( $\text{Ag}_2\text{O}$ ) on the Ag/G fiber before adsorption is observed in Fig. 1(d).

Fig. 1(c) and Fig. 1(f) are the EDS elemental maps of the cross-sectional image Fig. 1(b) and Fig. 1(e) respectively. These EDS shows presence of the iodine in the fiber after the adsorption experiment.

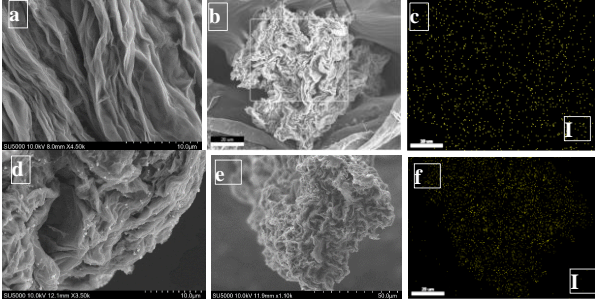


Fig. 1. SEM images and EDS of (a) (b) (c) rGO fiber; (d) (e) (f) Ag/G fiber

Fig. 2 shows the XPS results of the fibers after iodine adsorption. In the rGO fiber, Presence of iodine ( $\text{I}_2$ ) is observed in the Fig. 2 (a)(b), which means the physisorption of iodine. Fig. 2 (c)(d) shows the presence of iodine in the form of silver iodide ( $\text{AgI}$ ) in the Ag/G fiber after iodine adsorption. Both physisorption and chemisorption have happened in the Ag/G fiber which may show low desorption at higher temperature.

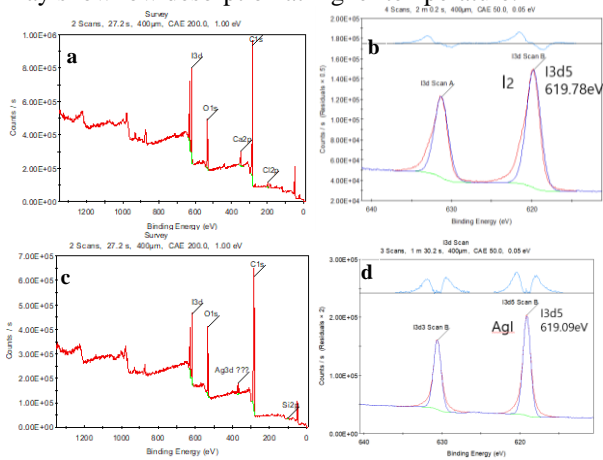


Fig. 2. XPS of (a) (b) rGO fiber (c) (d) Ag/G fiber after iodine adsorption

All the graphene-based fibers (GO fiber, rGO fiber and Ag/G fiber) in this work shows good iodine adsorption capability. Table I shows some of the results of iodine adsorption experiment.

Table I: Iodine adsorption of different fibers

Types of fiber	Weight gain (%) at 150 °C	
	6h	8h
GO	82.05	90.64
rGO	89.31	99.56
Ag-G		99.59

### 3. Conclusions

In this work, GO fiber, rGO fiber and Ag/G fiber have been reported as potential sorbent of radioactive iodine. Compared to GO fiber, rGO fiber and Ag/G fiber showed better iodine adsorption. Ag/G fiber may show better desorption characteristics than the other fibers at higher temperature.

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### REFERENCES

- [1] T. Fukasawa, K. Funabashi, Y. Kondo, Separation Technology for Radioactive Iodine from Off-Gas Streams of Nuclear Facilities, *J. Nucl. Sci. Technol.* 31 (1994) 1073–1083. doi:10.1080/18811248.1994.9735261.
- [2] S.U. Nandanwar, K. Coldsnow, V. Utgikar, P. Sabharwall, D. Eric Aston, Capture of harmful radioactive contaminants from off-gas stream using porous solid sorbents for clean environment – A review, *Chem. Eng. J.* 306 (2016) 369–381. doi:10.1016/j.cej.2016.07.073.
- [3] D. Reports, Htiiaim Y NOTICt A major purpose of the Techni - cal Information Center is to provide, (n.d.).
- [4] C. Zheng, X. Zhou, H. Cao, G. Wang, Z. Liu, Synthesis of porous graphene/activated carbon composite with high packing density and large specific surface area for supercapacitor electrode material, *J. Power Sources.* 258 (2014) 290–296. doi:10.1016/j.jpowsour.2014.01.056.
- [5] S.M. Scott, T. Hu, T. Yao, G. Xin, J. Lian, Graphene-based sorbents for iodine-129 capture and sequestration, *Carbon N. Y.* 90 (2015) 1–8. doi:10.1016/j.carbon.2015.03.070.