Removal of Radioactive Iodine with Graphene fiber-based adsorbents

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1. Introduction

Due to the high volatility and biological affinity, radioactive iodine, especially ^{129}I which has the longest half-life (1.57 x 10^7 yr), should be decontaminated from off-gas streams of reprocessing facilities before releasing environments. Most iodine gas can be captured via wet-chemical methods such as caustic scrubbing, mercurex, or iodox, and a trace amount of non-captured iodine is removed with solid sorbents [1].

Since silver can selectively capture iodine through chemical sorption, many Ag impregnated sorbents such as Ag-zeolite, Ag-alumina, and Ag-silica have been extensively investigated [2]. Carbon nanomaterials such as activated carbon or graphene [3] can also physically adsorb iodine gas, and show excellent performance for iodine removal.

In this paper, graphene-based materials were fabricated in a fibrous shape and hybridized with silver for developing iodine filter materials. By taking synergetic effects of physisorption from graphene and chemisorption from silver, better iodine removal performance would be expected. Also, fibrous adsorbents have advantages of low pressure drop and high adsorption kinetics which are suitable for continuous treatment of waste gas streams.

2. Methods and Results

2.1 Fabrication of graphene fiber (GF) and its silver functionalization (Ag-GF)

Graphene fibers were fabricated with typical wet-spinning methods [4]. 13 g/L of graphene oxide (GO) solution was doped on 10 mL syringe and spun in 5 wt.% of $CaCl_2$ solutions. After collecting and washing as-spun GO fibers, they were immersed in $AgNO_3$ and reduced with L-ascorbic acid (Vitamin C). As shown in Fig. 1, wrinkled and porous structures of GFs were observed, and nano-sized silver particles were evenly distributed on the surface of Ag-GFs.

2.2 Adsorption test

Solid iodine crystal (I_2), Graphene fibers, and desiccant were placed in a vacuum desiccator and heated up to 150 °C to make a saturated iodine atmosphere. Iodine uptake was estimated by measuring the mass increase of the adsorbents (**Table 1**).

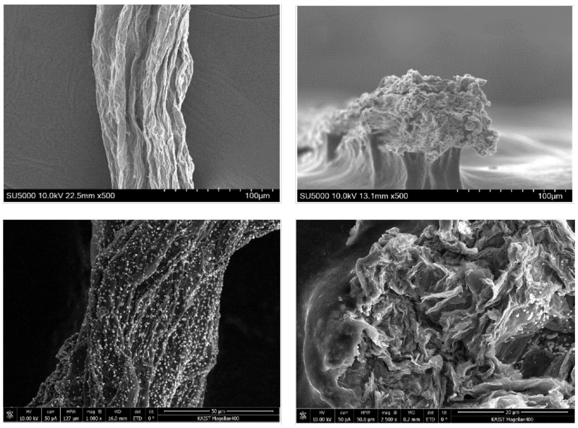

Fig. 1. Axis and cross-section SEM images of graphene fibers(up) and Ag-graphene fibers(down)

Table1: Contents of iodine after adsorption test (%)

Adsorbents	Time (hour)		
	6	8	10
GF	47.9±1.5	49.9±2.1	52.5±2.9
Ag-GF	48.1±2.2	49.9±2.8	52.8±3.2

2.3 Iodine adsorption properties of Ag-GFs

From the EDS mapping results (**Fig. 2**), it was confirmed that most iodine was adsorbed on the surface where silver was mainly distributed, and some iodine was diffused into the fiber and adsorbed. Also, XPS and XRD analysis data in **Fig. 3** and **Fig. 4** indicated that iodine was adsorbed in both physical form (I_2) and chemical form (AgI) on the adsorbent.

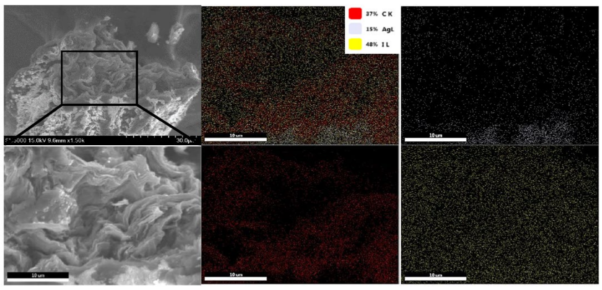

Fig 2. SEM image of iodine adsorbed Ag-GFs and EDS mapping

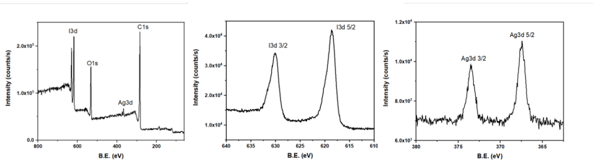

Fig 3. XPS of iodine adsorbed Ag-GFs

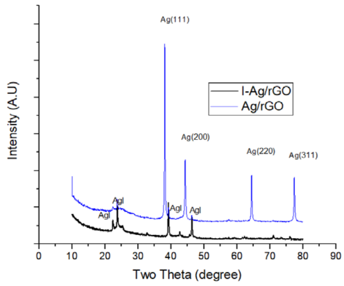

Fig 4. X-ray diffraction of Ag-GFs before and after iodine adsorption test

2.4 Thermal stability of iodine adsorbed Ag-GFs

There were two transient temperatures from the TGA-MS curve (Fig 5). First transient near 200 °C was due to physical desorption of I_2 and second transient near 1000 °C was due to thermal decomposition of AgI.

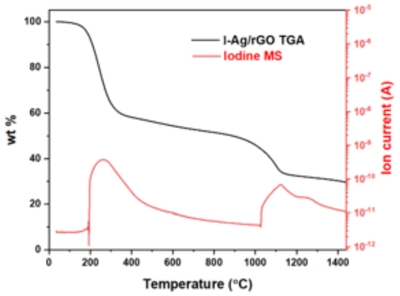

Fig 5. TGA-MS of iodine adsorbed Ag-GF

3. Conclusion

For the capture of radioactive iodine gas in reprocessing facilities, there have been lots of studies about solid sorbents such as silver-impregnated chemisorption sorbents and graphene-based physio-sorption sorbents. We prepared GF and Ag-GF for adopting advantages of Ag-graphene synergies and geometric merits of fiber. Both physical and chemical iodine adsorption properties of Ag-GFs were observed, and their thermal stabilities were also analyzed. In future work, pressure drop and adsorption kinetics of Ag-GFs will be investigated for verifying geometric effects.

Acknowledgement

This study was supported by the Ministry of Science, ICT and Future Planning. (NRF-2016M3A7B4905630)

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