Green immobilization of Cs-adsorbed MXenes to form durable waste matrix

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1. Introduction

Nuclear power is a carbon-free, clean energy solution for the world's ever-growing energy demand. Like all industries, whatever fuel is used, the generation of electricity produces waste. The waste management strategies must be adopted to safeguard human health and minimize the impact on the environment. Radioactive waste is not unique to the nuclear fuel cycle. Radioactive materials are also used extensively in medicine, agriculture, research, manufacturing, non-destructive testing, and minerals exploration. The safety perceptions of public are related to the operation, radioactive waste handling and disposal of radioactive waste.

Conditioning of separated radionuclides is an important step to ensure the safe long term disposal. Radioactive cesium (Cs^+) is one of the most important fission by-product elements present in nuclear waste. It has very high mobility in the aqueous environment and biologic affinity. Among the different reported separation techniques, adsorption is considered the most effective one due to its removal efficiency, cost and simplicity. The fixation of as-spent adsorbent is usually carried out by the vitrification process. This is a high temperature process and cause the volatilization loss of the adsorbed Cs^+. High temperature, long processing times and volatilization issues make vitrification a less viable immobilization option for radioactive Cs.

In this study, for the first time, we are reporting the green immobilization of Cs^+ adsorbed MXenes ($Ti_3C_2T_x$) in a calcium hydroxyapatite (HAp) matrix. Dried calcium hydroxyapatite nanopowder loaded with 40 wt.% of $Ti_3C_2T_x$ as-spent adsorbent (Cs-MX) is sintered using a low temperature cold-sintering technique without causing any volatilization of adsorbed radionuclide.

2. Materials and Method

The adsorbent $Ti_3C_2T_x$ was synthesized by the HF etching process of Ti_3AlC_2 powder (MAX phase). The synthesis details were reported elsewhere[1]. For exfoliation, the synthesized $Ti_3C_2T_x$ was dispersed in deionized water and sonicated for 20 minutes. Overnight drying of the exfoliated $Ti_3C_2T_x$ sheets was carried out in a vacuum oven at 80 °C.

HAp nanopowder was synthesized following reference[2]. Cesium chloride (CsCl) was used as surrogate source of radioactive Cs^+ ions. 0.2 g of dried $Ti_3C_2T_x$ powder was added in aqueous solutions of CsCl having 10 ppb and 10 ppm concentration of Cs^+, respectively. The as-spent adsorbent (Cs-MX) was then separated using centrifugation at 4000 rpm for 10 min and dried at 90 °C in a vacuum oven for 12 h.

HAp and Cs-MX were mixed with a 60 to 40 wt.% ratio to make the composite waste form. Homogenized mixing of the composite was carried out using pestle and mortar.

1 g of composite was used to prepare the cold sintered samples and cold-sintering was carried out as per reference[3,4].

Microhardness of the cold sintered samples was measured using the Vickers hardness method. The 7-day product consistency test (7-day PCT) as per standard ASTM C1285 is being carried out to investigate the chemical durability of the cold sintered composite.

3. Results and discussions

$Ti_3C_2T_x$ is a novel 2D material where T_x corresponds to terminal atoms/molecules (O, F or OH). Very high Cs^+ adsorption capacity (25.4 mg/g) was reported for this material[1]. The thermogravimetric-mass spectrometry data (TG-MS) analysis of the $Ti_3C_2T_x$ shown weight gain at temperatures higher than 300 °C which corresponds to the oxidation of the material at these elevated temperatures. Therefore, despite the higher adsorption capacity, high temperature fixation of Cs-MX can be a problem causing degradation of the adsorbent due to oxidation. Thus a low temperature consolidation process was proposed. The study is not only the first low temperature immobilization as well as is the first conditioning study of cesium adsorbed MXenes.

The scanning electron microscopy (SEM) images of the fractured surface of the cold sintered samples showed good consolidation with high densification (Fig. 1). The cold sintered samples presented microhardness value > 200 GPa.

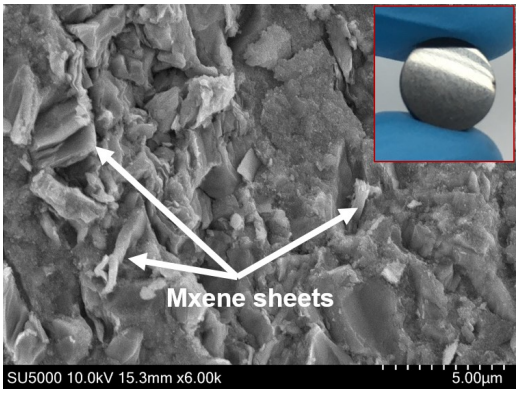

Fig. 1: Cold sintered pellet (inset) and SEM image of the fractured surface of cold sintered sample.

4. Conclusion

For the very first time immobilization of simulated radioactive cesium adsorbed MXenes was reported. Moreover, the immobilization was carried out using a green, low temperature sintering that is novel cold-sintering. There was no volatilization loss of the loaded simulated waste as well as the sintering temperature was less than the measured oxidation temperature of the MXene. Cold sintered samples showed good microhardness with high density. Leaching tests and further characterization are being carried out to ensure that the developed matrix can be a good candidate for radioactive waste immobilization.

Acknowledgments

This study is supported by the KUSTAR-KAIST Institute and NRF Korea (NRF-2018M2B2A90657456).

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