Development of Graphene Reinforced Calcium Hydroxyaptite Matrices for the Immobilization of Radioiodine

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1. Introduction

The presence of volatile radioisotopes like radioiodine-129 (I-129) makes the immobilization of ILW somehow difficult through vitrification and other high temperature techniques because of its volatilization above 500^{O}C. This is being one of the challenges suffered by the used nuclear fuel (UNF) reprocessing industry and requires the development of such matrices which can effectively immobilize the I-129 below its volatilization temperature as well as exhibit good durability in the geological repository environment. This is not only necessary to fully establish the worth of used nuclear fuel processing facilities in nuclear industry, also to counter the radiological effects of this long lived radioisotope. In this regard the development and adoption of effective methodologies/techniques to capture and immobilize the volatile radionuclides being evolved in the off-gas system may be one of the major licensing requirements for UNF processing facility [1].

Calcium hydroxyapatite $\{Ca_{10}(PO_4)_{10}OH_2\}$-HAP is calcium based phosphorus mineral having good corrosion resistance properties in ground environment as well as resistance against radiation damages of the loaded radioactive material [2,3]. HAP has the properties to accommodate iodine ions having an ionic radius of 196 pm at channels within its crystal structure [4].

In this study we have successfully sintered the synthesized graphene reinforced HAP at an extraordinary low temperature of <200^{o}C by using the technique of cold sintering [5] as the first attempt. In this effort we have got 90% relative dense sintered pellet at a temperature of 180^{o}C under a uniaxial pressure of 300MPa and has started the work for the consolidation of radioiodine loaded graphene within this ultra-low temperature sintered ceramic matrices.

2. Methods and Results

2.1 Apatite Synthesis

Initially the crystalline apatite has been synthesized by using the wet precipitation method as described elsewhere [6]. Two solutions, A & B, were prepared by dissolving each di-ammonium hydrogen phosphate $\{(NH_4)_2HPO_4\}$ and calcium nitrate $\{Ca(NO_3)_2{\cdot}_4H_2O\}$ in 100ml of double deionized water respectively. The pH of the solutions was adjusted at 10.5 by using concentrated ammonia $\{NH_4(OH)_2\}$. All used materials were of reagent grade. Then solution "A" was mixed with solution "B" dropwise during one hour under continuous stirring at the rate of 150RPM and temperature was maintained at 35 ^{o}C. The pH of the solution was continuously monitored by using digital pH meter and was maintained at 10.5 throughout the synthesis period by the dropwise addition of concentrated ammonia. At the end of synthesis, precipitates were filtered and thoroughly washed with double deionized water. Finally the filtrate was room temperature dried for 12hrs in a vacuum oven.

2.2 Powder Characterization

XRD pattern of the apatite powder was obtained by using SmartLab, RIGAKU, high resolution powder X-ray diffractometer and the 2Θ range was between 10 and 70◦.

2.3 Cold Sintering of Apatite

The sintering of apatite powder by using cold sintering technique has been investigated first time in. 3 g of apatite powder was mixed with 10wt% of deionized water by using pestle and mortar. 0.5wt% of graphene nano powder reinforced was used to study the effect of this enforcement on the textural strength and on the density of the sintered matrix. The powder was then poured in a steel mold and uniaxial pressed under the pressure of 300 MPa for 3 hrs. In order to provide controlled heating during the pressing, heating tape was wrapped around the steel mold. The pressing time was divided into three steps.

Step-I
Pressing at room temperature for 10 minutes

Step-II
Pressing with heating (25 °C to 180 °C) for 120 minutes

Step-II
Pressing during cooling for 50 minutes

After 3hrs, pressure was released and a sintered/dense pellet of the reinforced HAP was obtained.

3. Conclusion

In this work, ultralow temperature sintering of graphene reinforced apatite by using cold sintering technique was demonstrated. This leads to our final goal of development of an ultralow temperature sintered graphene reinforced matrices for the immobilization of I-129 for long term geological disposal.

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