Ultra-Low Temperature, Non-Conventional Sintering of Iodate Substituted Hydroxyapatite (IO-HAP) for the Immobilization of Radioiodine

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1. Introduction

Immobilization of radioiodine (I-129) cannot be carried out by using conventional vitrification route by using borosilicate glass because of its low volatilization temperature. A number of different low temperature methods are being investigated in order to resolve the loading as well as volatilization issues during the loading process. Among all those on going and suggested waste matrices the use of hydroxyapatite has an advantage of not only effectively hold the loaded radioiodine as well as its decay product i.e. Xe-129. It also has good corrosion resistance properties in ground environment and resistance against radiation damages of the loaded radioactive material [1].

Iodate substituted hydroxyapatite (IO-HAP) $\{Ca_{10}(PO_4)_6(OH)_{2-x}(IO)_x\}$ has been synthesized by using the wet precipitation process with 10 wt% of iodine loading. Since the synthesized material is a porous material with high surface area which may accelerate the leaching of loaded iodine due to large surface area in contact with the ground water. In order to overcome this issue sintering of the IO-HAP has been carried out by using spark plasma sintering at temperatures higher than 200°C [2].

In our work we have demonstrated and shown that IO-HAP dried powder can be sintered at temperatures ≤ 200°C by using a steel mold, a uniaxial press and heating band. We have also studied the sintering behavior under the presence of added as well as adsorbed water in the dried IO-HAP. We have optimized the sintering conditions to achieve the sintered relative density of ≥ 90% without loss of loaded iodine.

2. Methods and Results

2.1 Iodate substituted Apatite Synthesis

Initially the crystalline apatite has been synthesized by using the wet precipitation method as described elsewhere [3]. Two solutions, anionic and cationic, were prepared by dissolving each di-ammonium hydrogen phosphate $\{(NH_4)_2HPO_4\}$+ ammonium iodate (NH_4IO_3) 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 anionic solution was mixed in cationic solution dropwise during one hour under continuous stirring at the rate of 200RPM and temperature was maintained at 35°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 left inside the solution for 12hrs for ageing. Later on precipitate was filtered and thoroughly washed with double deionized water. Finally the filtrate was dried for 12hrs in a vacuum oven at 50°C.

2.2 Powder Characterization

XRD pattern of the IO-HAP powder was obtained by using SmartLab, RIGAKU, high resolution powder X-ray diffractometer and the 2Θ range was between 10 and 70° (Fig. 1).

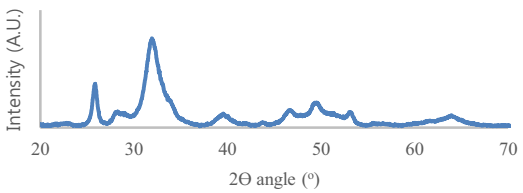

Fig. 1. XRD patterns of IO-HAP synthesized at 35℃.

FTIR spectra of the sintered IO-HAP sample was also obtained to verify the formation of HAP and presence of main apatite groups (Fig. 2).

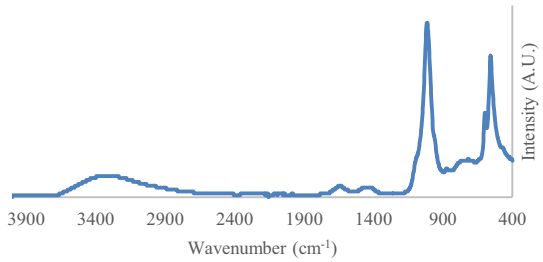

Fig. 2. FTIR spectra of IO-HAP synthesized at 35℃.

Raman spectra of the sintered IO-HAP sample was also obtained to verify the stability and presence of iodine in the form of iodate in the sintered samples (Fig. 3).

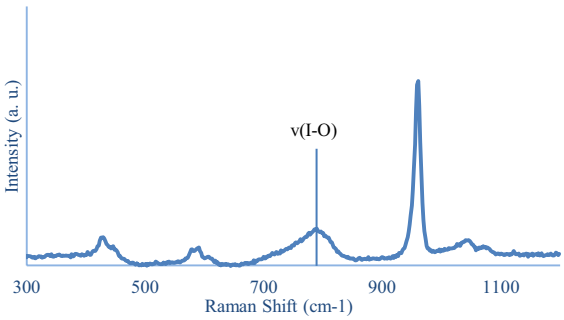

Fig. 3. Raman Spectra of the IO-HAP sintered at 200℃.

2.3 Sintering of IO-HAP

The ultra-low temperature sintering of IO-HAP dried powder has been investigated first time. Fo sintering purpose 0.5g of IO-HAP dried powder was poured in a steel mold with internal diameter of 17mm. In order to provide controlled heating during the pressing, heating jacket of 0.5 kW connected with a temperature controller was tighten around the steel mold. An optimized uniaxial pressure of 500 MPa was applied for an optimized holding time of 10 minutes only.

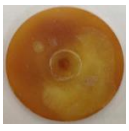

Fig. 4. IO-HAP pellet sintered at 200°C under 500 MPa uniaxial pressure at holding time of 10 minutes.

After 10 minutes of holding at 200°C, pressure was released and a sintered/dense pellet of the IO-HAP was obtained (Fig. 4).

3. Conclusion

In this work, ultra-low temperature, non-conventional sintering of iodate substituted hydroxyapatite was demonstrated for the first time. We have studied the role of adsorbed water present in the dried powder even after drying at 50°C under vacuum conditions and found that adsorbed water has vital role in the realization of this ultra-low temperature sintering. We have optimized the sintering temperature as 170°C in order to get sintered relative density upto 88% whereas for relative densities ≥ 90% the sintering temperature has been optimized as 200°C. The uniaxial pressure and holding time has been optimized as 500 MPa and 10 minutes. This leads to our final goal of development of an ultralow temperature sintered waste matrices for the immobilization of I-129 for long term geological disposal.

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