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ScienceDirect

Procedia Environmental Sciences 31 (2016) 330 – 334

Procedia Environmental Sciences

The Tenth International Conference on Waste Management and Technology (ICWMT)

Electron irradiation effects of $SrZrO_3$ ceramic for radioactive strontium immobilization

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Abstract

The perovskite structure is widely considered ideal host phases for Sr immobilization. To investigate the radiation tolerance on ceramic waste forms, the as-prepared $SrZrO_3$ ceramics were irradiated by external electron beam. The results indicated that the electron irradiation had different influence on the surface and inside of the $SrZrO_3$ ceramic. The no-crystallizing can be produced on the surface of $SrZrO_3$ ceramic, and electron beam heating promoted grain growth in the inside of $SrZrO_3$.

Peer-review under responsibility of Tsinghua University/ Basel Convention Regional Centre for Asia and the Pacific

Keywords: Ceramics;mmobilization; Electron irradiation;Microstructure

1. Introduction

The stabilization and immobilization of radioactive waste continues to attract much attention due to radioactive hazards. In order to reduce threat to the environment, radioactive wastes must be treated or immobilized prior to final disposition, especially to the high level waste (HLW) such as long half-time minor actinides and fission products[1]. In the immobilization process, a critical scientific contribution to this problem is the development of robust materials science solutions that encapsulate radioisotopes for long periods of time. In the past few decades, various glasses had been extensively employed as candidates for immobilizing HLW[2-4]. However, glass is unsuitable for radioactive waste disposal due to the poor radiation tolerance and leaching resistance. In recent years, most results indicated that ceramic forms exhibit excellent chemical durability, long-term stability and radiation tolerance to glass forms, and have been in a hot research topic to disposing high-level radioactive waste[5,6]. As

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Peer-review under responsibility of Tsinghua University/ Basel Convention Regional Centre for Asia and the Pacific
doi:10.1016/j.proenv.2016.02.044

crystalline waste-forms, ceramics are stable compounds with definite thermodynamics and kinetics properties, and expected to be more robust than for glasses[7, 8]. Therefore, it is considered an excellent host for immobilizing complex actinide and fission wastes.

It is well known that ^{90}Sr have relatively long half-life of 28.1 years, it is typical high level radioactive wastes generated from spent nuclear fuels. In ^{90}Sr host phases, ^{90}Sr decay to daughter ^{90}Y, the ^{90}Y decay to stable ^{90}Zr by emitting beta particles.The stability of crystalline waste forms comprised of ^{90}Sr is likely to be affected by daughter product formation and beta particles. As an oxide ceramic, $SrZrO_3$ contains parent ^{90}Sr and stable daughter ^{90}Zr, so it is useful to investigate beta irradiation effects of $SrZrO_3$ ceramic. In this work, the prepared $SrZrO_3$ ceramic was irradiated by external electron beam, used to simulate beta irradiation of strontium. Based on characteristics of the microstructure, the radiation resistance of the $SrZrO_3$ ceramic was investigated before and after electron irradiation.

2. Materials and methods

$SrZrO_3$ ceramics were synthesized from powders prepared by Sol-thermal spraying method (STSM). Firstly, 0.1 M solutions of Sr^{2+} and Zr^{4+} were prepared by dissolving $Sr(NO_3)_2$ and $Zr(NO_3)_4{\cdot}3H_2O$ in deionized water with heating and stirring on a hot stirrer set at 60 ℃ for 2 h. Subsequently, the two solutions were mixed corresponding to nominal compositions of $SrZrO_3$ and stirred at 60±5 ℃ for 1h. To obtain a stable precursor solution, citric acid and polyethylene glycol were added to the solution containing Sr and Zr ions according to 8g $C_6H_8O_7H_2O$ and 5 g in 100 ml Solution. The precursor solution prepared was added into spray pyrolysis equipment and pyrolysised at 823 K under air environment by a heater. A black precipitate was obtained which was further heated in a conventional furnace at 900 ℃ for 2 h. Green The circular plates with 15 mm diameter and 2 mm thickness produced by hydraulic pressing (HP) at a pressure of 15 MPa.The plates were then sintered at 1250 ℃ for 12 h in air.

To evaluate irradiation effects of $SrZrO_3$ ceramic, the circular plates sintered were subjected to an electron irradiation with energy of 1.8MeV and flux of 1.5×10^{13} electrons/cm^2/s. The effects of electron irradiation on the microstructure of $SrZrO_3$ ceramics were identified by X-ray diffraction analysis (XRD), Fourier transform Raman (FT-Raman) and field scanning electron microscope (FSEM).

3. Results and discussions

Fig. 1 shows the XRD patterns of the $SrZrO_3$ ceramics before and after electron irradiation. It was clear found that no secondary phase was observed by XRD on the $SrZrO_3$ before electron irradiation, the $SrZrO_3$ sintered is a single phase with orthorhombic perovskite structure, and consistent with previous reports[9-11]. The sharp and well-defined diffraction peaks indicated that the obtained ceramic has a high degree of crystallinity. The lattice parameters of $SrZrO_3$ determined from XRD are a = 0.5795 nm, b = 0.5817nm, c = 0.820 nm.

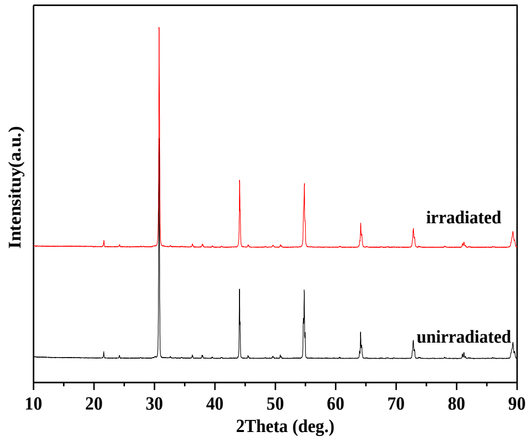

Fig. 1. XRD patterns of $SrZrO_3$ ceramics before and after electron irradiation