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Microstructural study of Ytterbium Zirconate Pyrochlore synthesized by the mixed method: sintering/chemical infiltration

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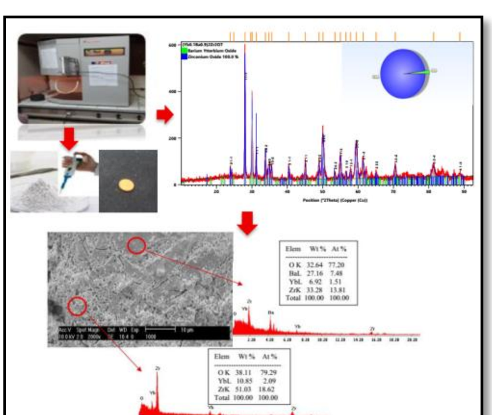
ABSTRACT

In this study, a zirconate pyrochlore with the chemical formula $(Yb_{0.1}Ba_{0.9})_2Zr_2O_7$ was synthesized and stabilized by ytterbium nitrate. The used synthesis method combined between calcination and infiltration procedure where the considered infiltration parameters were concentration of ytterbium solution set at C=390 g/L, infiltration temperature and T= 105°C. However the thermal cycle of sintering in two stages, calcination at 550°C for 4 h, then a sintering of 1200°C for 24h. The characterization of the synthesized material was carried out by several analysis techniques. Archimedes' density using a hydrostatic balance gave a value of 4.001 g/ cm3 for the raw ceramic and 4.665g/cm3 for the sintered one. X-ray diffraction (XRD) analysis was used to track the progress of the synthesis and confirm its success through the formation of the ZrO_2 structure, the main framework of the pyrochlore material. Observation using a scanning electron microscope (SEM) allowed us to observe the distribution of grains and pores and estimate the quality of sintering, combined with analysis using X-ray energy dispersive spectrometry (EDX), which allowed us to check the global chemical composition of the sintered matrix. Fourier transform infrared (FTIR) analysis of pyrochlore $(Yb_{0.1}Ba_{0.9})_2Zr_2O_7$ shows essential absorption bands between 400-4000 cm^{-1}.Two absorption bands can be seen at 470 cm^{-1}corresponding to Zr-O vibrations, and an absorption band at 682 cm^{-1} corresponding to Ba-O vibrations.

Keywords: pyrochlore; ytterbium; zirconate; infiltration; XRD; SEM; FTIR.

Graphical abstract



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1. Introduction

The current nuclear fuel cycle produces a variety of radioactive wastes [1]. The most dangerous of these are intermediate- and high-level wastes (ILWs and HLWs), which are primarily produced when uranium fuels fission in nuclear reactors as separated wastes from the reprocessing of spent nuclear fuels that contain fission products and transuranic elements with long half-lives [2]. The most prevalent medium for handling reprocessed HLWs has been intensively investigated: glass [3]. For the immobilization of various actinide-rich radioactive wastes, a variety of ceramic formulations based on zirconolite ($CaZrTi_2O_7$) and pyrochlore ($Ln_2Ti_2O_7$: Ln = Y or lanthanides or $CaATi_2O_7$: A = actinides) have been developed [4]. $A_2B_2X_6Y$ is the general formula for pyrochlore, a derivative of the fluorite structure, where X and Y are 4-coordinated anion sites and A and B are 8- and 6-coordinated cation sites [5].Transition metal ions like Ti, Zr, Nb, Hf, Ta, Sn, and W can be incorporated on the B site, while alkaline earth, lanthanide, and actinide cations can be incorporated on the A site [6]. Pyrochlore is therefore a very adaptable and appealing host phase that can incorporate a variety of cations for nuclear applications, particularly for the immobilization of radioactive wastes that are rich in actinides [7]. Furthermore, the dissolution rates of fully amorphous titanate pyrochlores are very similar to those of their crystalline equivalents, according to radiation damage studies. This suggests that the chemical durability of pyrochlore-structured materials is maintained even in their fully amorphous state [8]. The aim of this work is to prepare $(Yb_{0.1}Ba_{0.9})_2Zr_2O_7$ zirconate pyrochlore using mixed method: sintering/chemical infiltration. And consolidate the final product via Microstructural study, with the aim of verifying its suitability as a radioactive waste storage material.

2. Materials and Methods

2.1. Samples preparation

To synthesize 20 g of a zirconium-stabilized DR confinement matrix loaded with Yb with the chemical formula $(Yb_{0.1}Ba_{0.9})_2Zr_2O_7$, the starting reagents: ZrO_2 (ALDRICH, 99% purity), Yb_2O_3 (ALDRICH, 99.9% purity), BaO (prepared in the laboratory from $BaNO_3$ FLUKA, 99.6% purity), are ground manually in an agate mortar. The quantities are weighed. The powders are formed into pellets by mixing the fine powders of the starting reagents with 4% organic binder (zinc stearate, Aldrich) in the target stoichiometric quantities. The powder obtained is mixed in an Automatic Sieve Shaker D403 homogenizer for 4 hours and 30 minutes to ensure proper dispersion of the mixture. It is then dry-compacted into pellets in a Sodemi RDDE uniaxial hydraulic press. After filling the mold and closing the upper punch, pressure of 310 MPa is applied for 20 seconds. The resulting pellets are sintered in air in a Carbolite Furnace RHF 1800 at 1200°C for 24 hours, with a thermal cycle in which the heating and cooling rates are 5 and 10°/min, respectively. The obtained pellets are ground into a fine powder using an automatic agate mortar micro-grinder, model Controlab N6 87-60K. They are then infiltrated using an aqueous solution of Yb nitrate at 390 g/L. Infiltration is carried out at a temperature of 105°C for 24 hours to achieve the final composition of the product. The powder obtained is calcined at 550°C for 4 hours, with a thermal cycle such that the heating and cooling rates are 5 and 10°/min respectively. 4% zinc stearate is added to the product, which is then pelletized under a pressure of 310 MPa. The pellets are then sintered using the following thermal cycle: Finally, from 50 to 1200°C, the heating rate is 2°C/min, with a heating plateau of 24 hours.

2.2. Characterization

Both green (dc) and sintered (df) geometrical densities were determined utilizing the Archimedes method, which involved immersing the samples in distilled water as the immersion liquid. The materials phases' identification is performed by X-ray diffraction analysis (XRD) with a Philips X-Pert Pro spectrometer. The X'Pert High Score Plus software, 4.1 version, is used for phase' identification [11]. Observation using a scanning electron microscope (SEM) highlighted the distribution of grains and pores and estimated the quality of sintering, combined with energy dispersive spectroscopy analysis (EDS), the global chemical composition of the sintered matrix was checked. The FTIR spectra are recorded from 4000 to 400 cm^{-1} (wave numbers) by a standard KBr pellet method using a NICOLET 380 spectrometer. The pellets are prepared by mixing pyrochlore powder of about (1–2 mg) with anhydrous KBr (100–200 mg). The resolution of the device is 4 cm^{-1}. The spectra data processing is performed by the OMNIC software [12].

3. Results and Discussion

3.1. *Material density*

The both densities, the green and the sintered one, yielded a value of 4.001 g/cm³ and 4.665 g/cm³ respectively. The sintered density is greater than the green density. This is due to the densification of the grains through the sintering process.

3.2. *XRD phases identification*

XRD analysis of the Yb-doped zirconium oxide sample produced the diffraction pattern shown in Figure 1. Phase identification of the Ba-doped zirconium oxide infiltrated with Yb revealed that the main crystalline phase represents approximately 98% of the total composition. This phase corresponds to monoclinic ZrO_2, in agreement with JCPDS card No. 98-007-2694 [11], and crystallizes in the P21/c space group. This phase exhibits a preferred orientation along the (11-1) plane, indicating a partial texturing effect likely induced during synthesis or thermal treatment. A minor secondary phase, identified as barium ytterbium oxide ($Ba_3Yb_4O_9$), was detected at about 2%, corresponding to JCPDS card No. 01-075-2476 [11]. The average crystallite size, estimated using the Scherrer equation [13], was found to be approximately 32 nm.

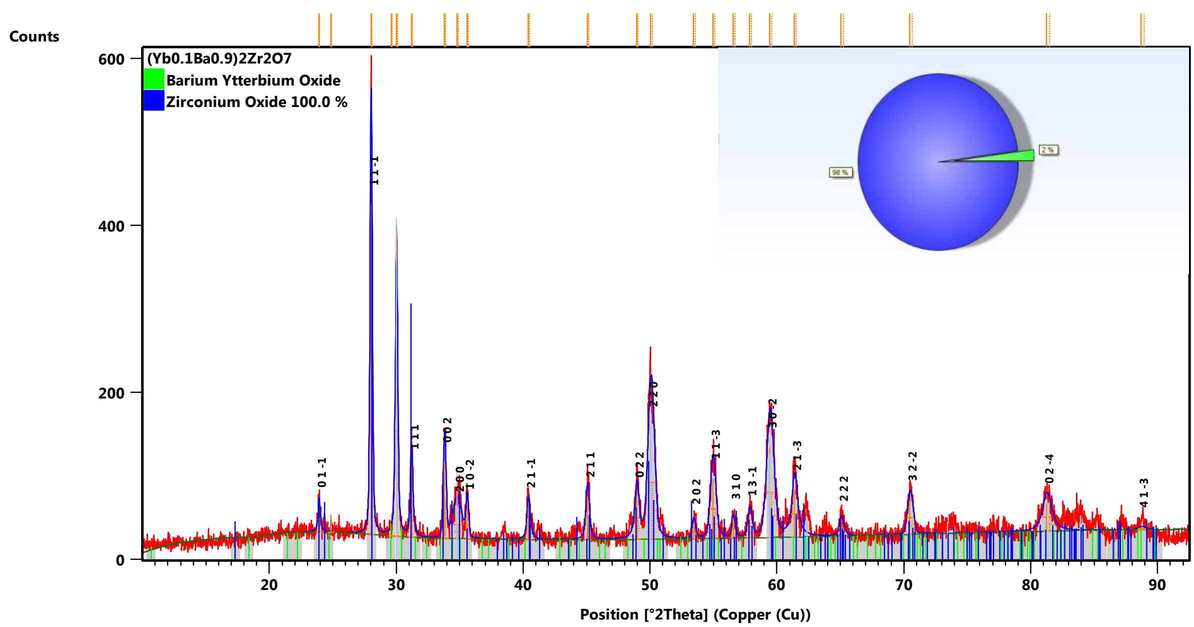

Fig 1. Diffractogramm of the final Yb-doped stabilized zirconium oxide product.

3.3. *Scanning electron microscopy and EDS*

The micrograph is shown in Fig.2. It allowed us observing both types of phases: a first showing a light contrast and the second a dark contrast. The chemical EDS analysis confirmed the presence of the ZrO_2 phase, corresponding to the main matrix, along with a minor secondary phase identified as barium ytterbium oxide ($Ba_3Yb_4O_9$). This result is consistent with the XRD findings, further supporting the coexistence of these two phases in the synthesized material. The grain distribution of these two phases is uniform. Their average sizes are 1 and 2µm, respectively.

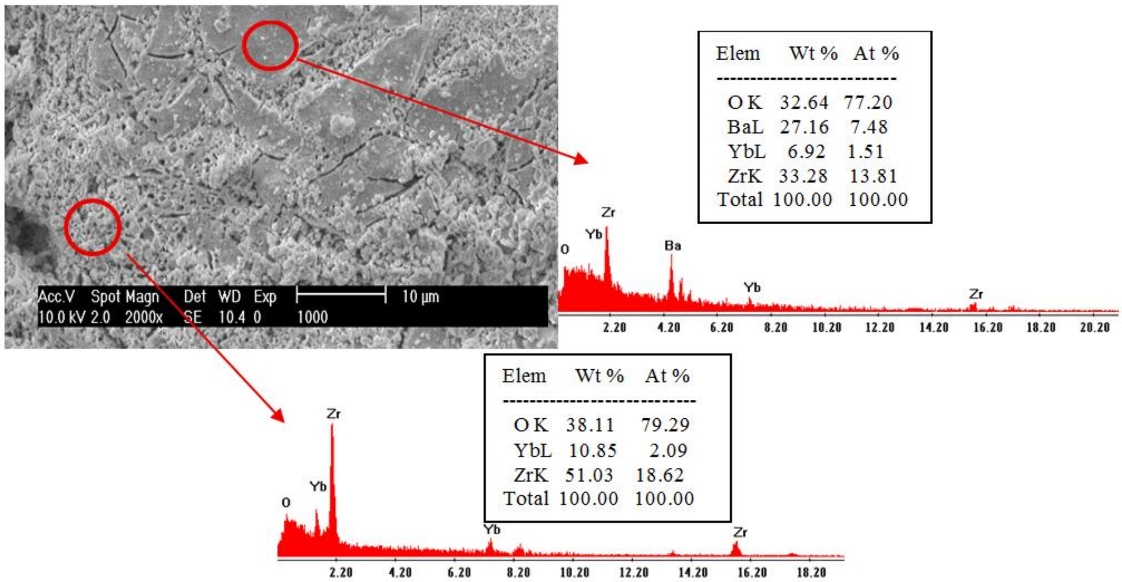

Fig 2. SEM micrograph of the final Yb-doped stabilized zirconium oxide product and EDS analysis.

3.4. Ftir analysis

Figure 3 presents the FTIR analysis of the Yb-doped barium zirconium oxide sample revealed several characteristic absorption bands, confirming the formation of metal–oxygen bonds. The broad band observed around 2400 cm^{-1} is attributed to the O–H stretching vibration of adsorbed water molecules on the surface. The weak band near 1630 cm^{-1} corresponds to the H–O–H bending vibration. In the low-frequency region, a strong absorption band detected around 500–700 cm^{-1} is assigned to the Zr–O stretching vibration [14]. characteristic of zirconium oxide. Additionally, an absorption band corresponding to the **Ba–O vibration** was observed, indicating the incorporation of barium into the oxide lattice. A minor contribution in this region may also arise from **Yb–O vibrations [**15], confirming the successful doping of ytterbium into the structure. These results are consistent with the XRD and EDS analyses, further supporting the coexistence of ZrO_2 and $Ba_3Yb_4O_9$ phases in the synthesized material.

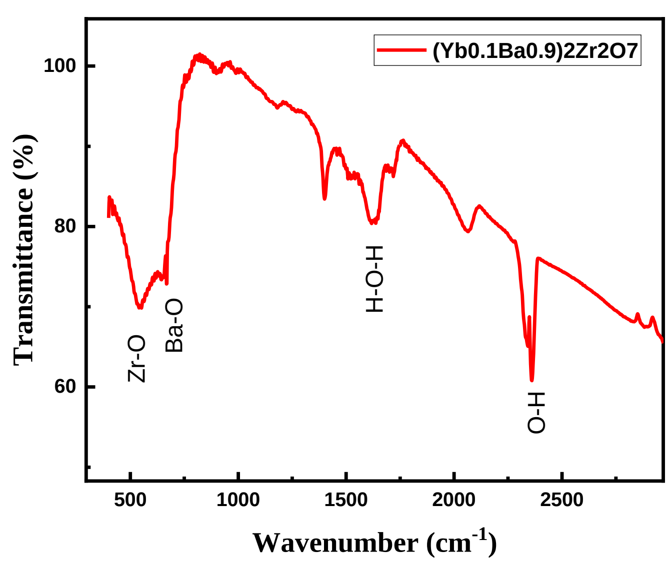

Fig 3. FTIR of the final Yb-doped stabilized zirconium oxide product.

4. Conclusion

The simple approach employed to develop the confinement matrix for radioactive wastes (DRs) with the chemical formula $(Yb_{0.1}Ba_{0.9})_2Zr_2O_7$, combining sintering and infiltration, has produced significant and reproducible results. X-ray diffraction (XRD) analysis confirmed the successful synthesis and revealed the presence of two crystalline phases: a predominant ZrO_2 phase and a minor $BaYbO_4$ phase, providing clear evidence of ytterbium incorporation in both structures. Scanning electron microscopy (SEM) coupled with EDS analysis further confirmed the coexistence of two distinct phases, corresponding to a Zr-rich phase and a Ba-rich phase, in agreement with the XRD findings. In conclusion, the use of a cost-effective and straightforward method has demonstrated the suitability of this natural analogue for nuclear waste confinement, opening promising prospects for future applications in the field of radioactive waste management.

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Ethical Statement
This study does not contain any studies with human or animal subjects performed by any of the authors.

Conflict of Interest
The authors declare that they have no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available within the article

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