



Impact of distinct sintering temperatures in pellets to strontium immobilization

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1. Introduction

The study of methods to immobilize radioactive waste has been carried out for several decades. Although it is an old topic, it is still relevant due to the increase for energy demand in the world, which has consequences in all types of energy generation, in the case of the use of nuclear energy the consequences start from the extraction to the disposal of nuclear waste [1].

The need for a safe final disposal is a challenge and has not yet been well defined. One of the fission products of ²³⁵U is the ⁹⁰Sr, it is founded in the spent nuclear fuel reprocessing and in the nature caused by nuclear accidents like Chernobyl and Fukushima, and nuclear weapons. This isotope has a long half-life of 29 years and his similarity to calcium can causing many of problems, being present on nature the population may ingest contaminate foods and water leading to an accumulate in the bones of the human body [2, 3, 4].

This work proposes a way to immobilize high-level radioactive waste (HLRW) containing radioactive strontium generated in wastewater decontamination process. To achieve this, a zeolite type A was synthesized and contaminated with a simulate solution containing strontium ions and then, mixed with a glass, which was studied before by Costa-Silva for nuclear immobilization application. The pellets obtained were analysed to meet the requirements of density and water absorption [4, 5].

2. Methodology

Firstly, borosilicate glasses containing niobium from 2 to 8 mol% were prepared by traditional melt-quenching technique. Samples from the system SiO₂-Na₂O-CaO-B₂O₃-Al₂O₃-Nb₂O₅ were produced and their compositions obtained by X-ray fluorescence (XFR) showed the substitution of silica for niobium in quantities of 2, 4, 6 and 8 mol% resulting in the samples V2, V4, V6 and V8. The raw materials were manually homogenized, to obtain the batches, which were submitted to melting in an alumina crucible at 1,300°C for 2 hours. The glasses were cooled down to room temperature and reduced to powder manually in an agate mortar [6].

In parallel, type A zeolites were produced [7] to perform as an adsorbing material for the liquid effluent, in this case strontium ions as a simulated of HLRW. The zeolites were contaminated in a solution of distilled water and strontium chloride, so the final sorption was approximately 19 mol% on the composition.

The glasses and contaminated zeolite (ZAC_Sr) were mixed in proportion of 60% of glass and 40% of zeolite forming pellets called VxZAC_Sr in the configuration where “x” is the number of 2, 4, 6 or 8 according to the proportion. The pellets were submitted to heating treatment in three different temperatures 800, 900 and 1,000 °C for 2 hours and each composition samples were subjected to the ASTM C373 standard to measure density, porosity, and water absorption [8].

3. Results and Discussion

In Fig. 1 the green pellets obtained are shown, with 12mm in diameter and 2,2 high in average. By Differential Thermal Analysis (DTA) as shown in Fig. 2 the temperatures of crystallization and fusion were determined to do the heat treatment.



Figure 1: Pellets of glass and zeolite before sintering: V2ZAC_Sr; V4ZAC_Sr; V6ZAC_Sr; V8ZAC_Sr, from right to left.

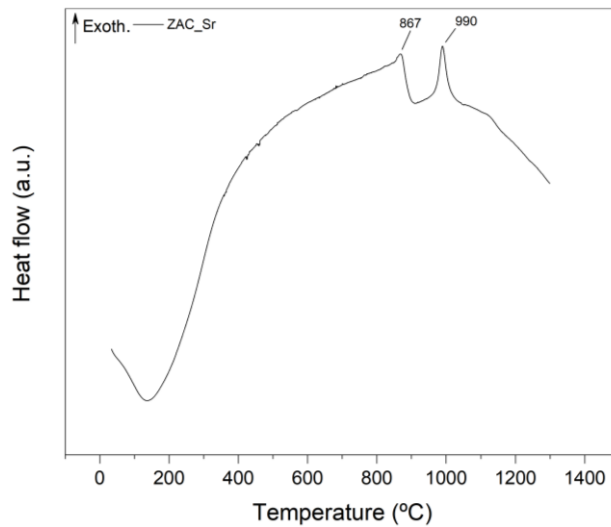


Figure 2: DTA of saturated zeolite.

After the heat treatment at the three temperatures, the samples obtained at 1000 °C were excluded because they melted, and this result is not compatible with the objective of this work. The other two were submitted to ASTM C373-88 standard, the results are show on Table I. The pellets sintered in 900 °C clearly had the smallest absorption of water and a high density.

Table I: results of the ASTM C737-88 standard.

	Composition	V2ZAC_Sr	V4ZAC_Sr	V6ZAC_Sr	V8ZAC_Sr
Apparent density (g.cm ⁻³)	800 °C	1.94	1.93	1.97	1.99
	900 °C	2.16	2.19	2.02	1.92
Water absorption (%)	800 °C	14.41	15.24	13.76	13.31
	900 °C	2.80	1.21	1.58	3.18

Apparent porosity (%)	800 °C	22.97	29.45	27.10	26.57
	900 °C	6.45	2.66	3.20	6.14

The illustrate the difference of topography according to the temperature, the same composition (V8ZAC_Sr), but in different temperatures can be observed in Scanning Electron Microscopy (SEM) in Fig. 3. The densification was noted by the lack of porosity and the smother surface.

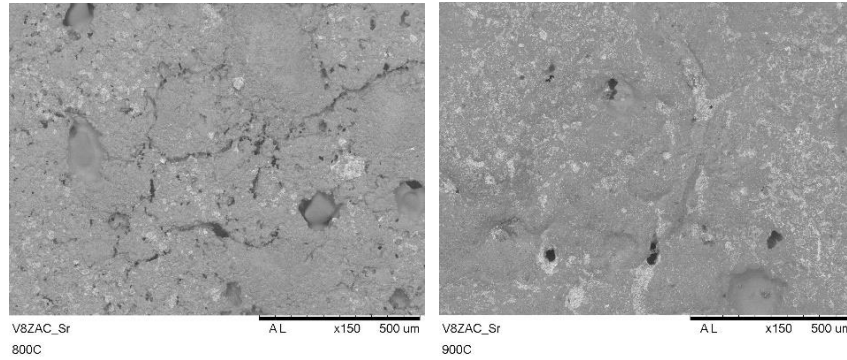


Figure 3: SEM of V8ZAC_Sr in 800°C (left) and 900°C (right).

Following the SEM results, differences were also observed from the X-ray diffraction (XRD) as shown Fig. 4, the same composition at the two temperatures showed patterns of crystallization process. However, at 800°C the structure is analogous to a feldspar containing strontium, $Al_{1.69}Na_{0.03}O_8Si_{2.29}Sr_{0.84}$ (6255-ICSD) and at 900 °C a $Fe_{0.5}Nb_{0.5}O_3Sr$ (8409-ICSD) phase were detect.

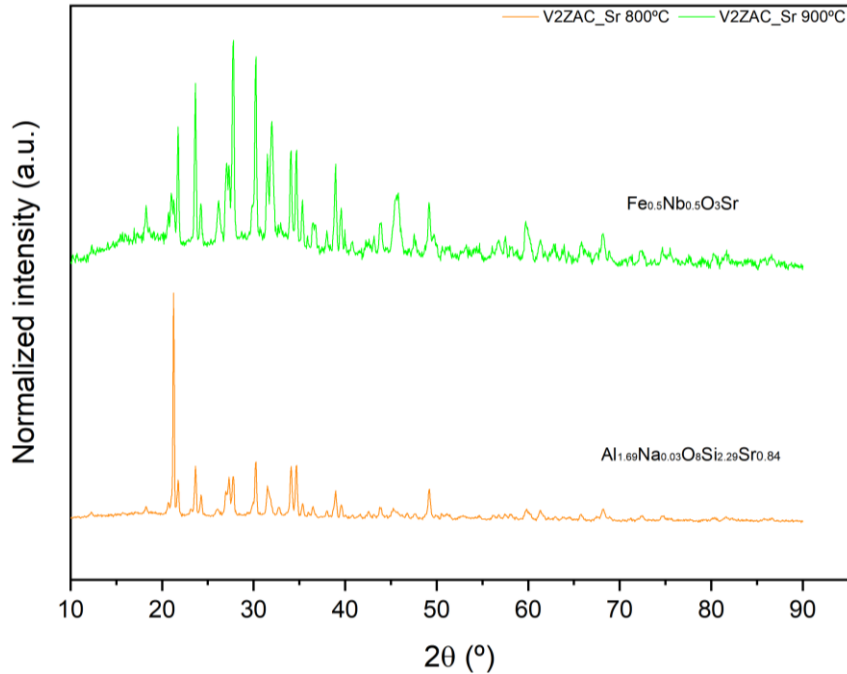


Figure 4 – XRD of V2ZAC_Sr at 800 °C and 900 °C after heat treatment.

4. Conclusions

This study concludes that regardless of the composition, all four configurations absorb less water at 900 °C than at 800 °C, with the XRD results the temperature contributes to formation of phases that interact with strontium. For the application the best composition and temperature is the V4ZAC_Sr at 900 °C for the best performance in densification, it has a low water absorption and the lowest apparent porosity. For future work, the zeolites can be contaminated with more ions to verify which phases are form with the glass and check if the temperatures of heat treatment will change.

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