doi.org/10.15392/2319-0612.2024.2652
2024, 12(4A) | 01-10 | e2652
Submitted: 2024-08-28
Accepted: 2024-12-11

Impact of distinct sintering temperatures in pellets to strontium immobilization

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Abstract: The immobilization of radioactive strontium (Sr) from liquid effluents, a byproduct of nuclear fission and accidents, presents significant environmental and public health challenges. This study evaluates the immobilization of Sr in a glass matrix by comparing the effects of different sintering temperatures. The analyses started from three temperatures 800ºC, 900ºC and 1000ºC for each of the four compositions, which vary the addition of niobium (Nb) content in the vitreous matrix by 2%, 4%, 6% and 8% in mol. The highest temperature was excluded because its deformation did not match the final objective, at 900ºC the compositions had better results in density and water absorption, so that at the same temperature the composition with 4% niobium responded better to the results. The study underscores the significance of treating temperature and Nb content in the glass matrix for effective Sr immobilization, offering valuable insights for the development of safe and sustainable nuclear waste management strategies.

Keywords: liquid effluent, strontium immobilization, niobium, zeolite.

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Impacto da temperatura em pastilhas imobilizadoras de estrôncio

Resumo: A imobilização de estrôncio radioativo (Sr) proveniente de efluentes líquidos, um subproduto da fissão nuclear e de acidentes, representa desafios significativos para o meio ambiente e a saúde pública. Este estudo avalia a imobilização de Sr em uma matriz vítrea, comparando os efeitos de diferentes temperaturas de sinterização. As análises partiram de três temperaturas: 800ºC, 900ºC e 1000ºC, para cada uma das quatro composições, nas quais varia a adição de nióbio (Nb) na matriz vítrea em 2%, 4%, 6% e 8% em mol. A temperatura mais alta foi descartada porque sua deformação não correspondeu ao objetivo final; a 900ºC, as composições apresentaram melhores resultados em densidade e absorção de água, de modo que, à mesma temperatura, a composição com 4% de nióbio obteve os melhores resultados. O estudo destaca a importância de tratar a temperatura e o teor de Nb na matriz vítrea para a imobilização eficaz de Sr, oferecendo insights valiosos para o desenvolvimento de estratégias seguras e sustentáveis para a gestão de resíduos nucleares.

Palavras-chave: efluente líquido, imobilização de estrôncio, nióbio, zeólita.

1. INTRODUCTION

The study of methods to immobilize radioactive waste has been ongoing for several decades. While this topic is not new, it remains highly relevant due to the increasing global demand for energy, which affects all forms of energy generation. In the case of nuclear energy, the challenges begin with uranium extraction and extend 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 ^{235}U is the isotope ^{90}Sr, which can be found in the spent nuclear fuel, as well as in the environment, produced during nuclear accidents such as Chernobyl and Fukushima, or through nuclear weapons. This isotope has a long half-life of 29 years, and it is similarity to calcium can cause many problems associated with environment and public health, due to the possibility of ingestion by contaminated foods and water, leading to its accumulation in the bones of the human body [2, 3, 4].

This study proposes a method to immobilize contaminated zeolite containing strontium, classified as high-level radioactive waste (HLRW), generated during wastewater decontamination process. To achieve this, a zeolite type A was synthesized and contaminated with a simulated solution containing strontium ions. The contaminated zeolite was the mixed with a specific type of glass previously investigated by Costa-Silva for its potential in nuclear waste immobilization. The resulting mixture was formed into pellets, which were then evaluated to ensure they met the necessary standards for density and water absorption, critical factor in long-term radioactive waste storage [4, 5].

2. MATERIALS AND METHODS

Firstly, borosilicate glasses from the system SiO_2-Na_2O-CaO-B_2O_3-Al_2O_3-Nb_2O_5 containing Nb_2O_5 from 2 to 8 mol% were prepared by traditional melt-quenching technique according to Costa-Silva [5]. 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. The compositions of the obtained glasses were confirmed by X-ray fluorescence (XRF) ensuring the niobium in quantities of 2, 4, 6 and 8 mol% resulting in the samples V2, V4, V6 and V8, respectively.

Simultaneously, type A zeolites were synthesized by conventional hydrothermal method according to previous works [6] to perform as an adsorbing material for effluent treatment, in this case to selective removal of strontium ions from HLRW. Simulated nuclear effluent containing strontium ions were obtained from a mixture of distilled water and strontium chloride, then, 5 g of zeolite was added to the solution. The solution was stirred for 24 h and followed by filtering and frying overnight at 105 °C. The obtained sample was labeled as ZAC_Sr. To obtain a semi-quantitative assessment of the amount of strontium adsorbed, chemical analysis was conducted using an energy dispersive X-ray fluorescence (ED-XRF) spectrometer, specifically the SHIMADZU EDX 720 model. The fundamental parameters (FP) method was employed for the analysis of the powdered samples.

The evaluation of the thermal behavior of the zeolites during their heating was carried out by differential thermal analysis (DTA), using synthetic air, heating rate of 10 °C/min up to a temperature of 1,300 °C.

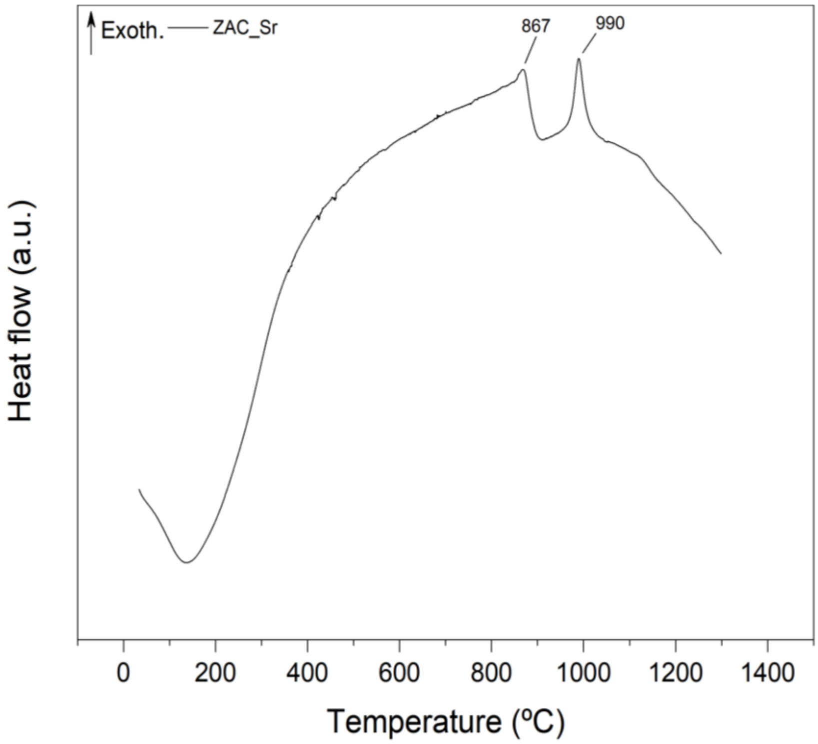
The glasses (V2, V4, V6 and V8) and contaminated zeolite (ZAC_Sr) were mixed in proportion of 60% of glass and 40% of zeolite forming pellets called VxZAC_Sr where “x” is the quantity of Nb_2O_5 in the glass [7].

The pellets were submitted to heat treatment at three different temperatures 800, 900 and 1,000 ºC for 2 hours and all samples were subjected to the ASTM C373 standard to measure density, porosity, and water absorption [8].

3. RESULTS AND DISCUSSIONS

After the adsorption experiment, the retention of SrO was up to 19 mol% obtained by X-ray fluorescence spectroscopy (XRF). The temperatures of crystallization and fusion were determined by Differential Thermal Analysis (DTA) shown in Fig. 1 and selected for the heat treatments.

Figure 1: DTA of contaminated zeolite.



The green pellets obtained are shown in Fig. 2, with an average diameter of 12 mm and height of 2.2 mm.

Figure 2: Pellets of glass and zeolite before sintering V2ZAC_Sr, V4ZAC_Sr, V6ZAC_Sr and V8ZAC_Sr from left to right.

After the heat treatment at the temperatures of 800, 900 and 1,000 °C, the samples obtained at 1,000 ºC were excluded due to melting, which was incompatible with the objective of this work. The remaining samples, treated at 800 and 900 ºC, were submitted to ASTM C373-88 standard experiment, and the results are shown on Table 1.

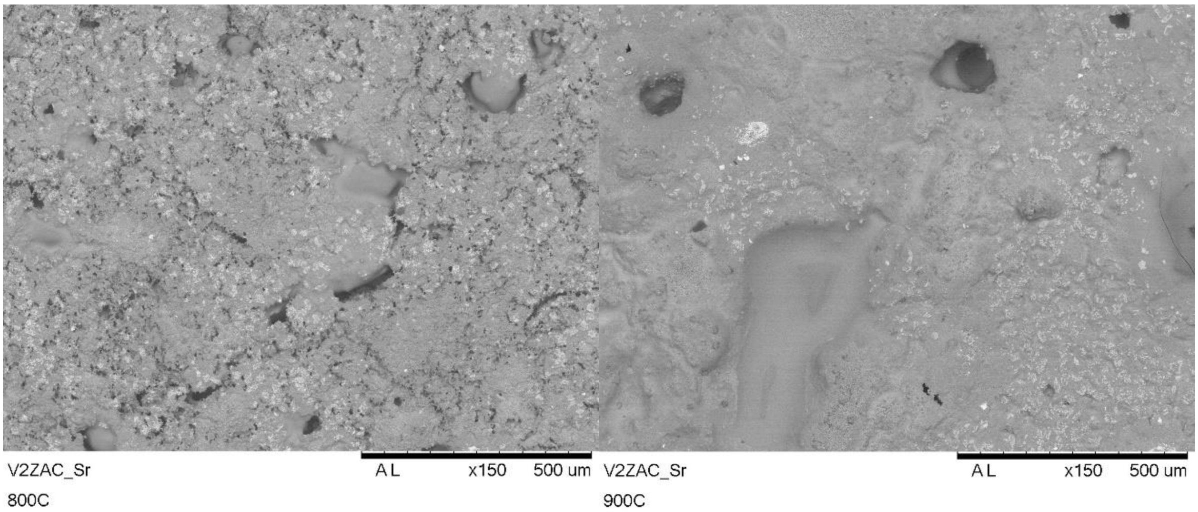
Table 1: Results of the ASTM C737-88 standard experiment.

	Composition	V2ZAC_Sr	V4ZAC_Sr	V6ZAC_Sr	V8ZAC_Sr
Apparent density (g.cm^{-3})	800ºC	1.94	1.93	1.97	1.99
	900ºC	2.16	2.19	2.02	1.92
Water absorption (%)	800ºC	14.41	15.24	13.76	13.31
	900ºC	2.80	1.21	1.58	3.18
Apparent porosity (%)	800ºC	22.97	29.45	27.10	26.57
	900ºC	6.45	2.66	3.20	6.14

The pellets treated at 800 ºC had a higher water absorption compared to the 900ºC. The density generally increased with rising temperature. However, the V8ZAC_Sr composition showed a decrease in density at 900 °C compared to 800 °C. Table 1 indicates a maximum densification point for the V4ZAC_Sr composition, suggesting the existence of a curve with maximum and minimum densification as a function of different niobium contents and temperature variations. The apparent porosity was similar in the compositions with lower and higher amount of niobium, which may have new phases formation resulting in intern caves. The reduction in porosity not only improves the material's physical integrity but also contributes to its effectiveness in immobilizing radioactive strontium by minimizing potential pathways for leaching. In all results of the technique applied the composition V4ZAC_Sr demonstrated the betters results in all aspects.

To illustrate the changes of surface at the different temperatures, the composition V2ZAC_Sr was observed by Scanning Electron Microscopy (SEM) and the micrographs are shown in Fig. 3.

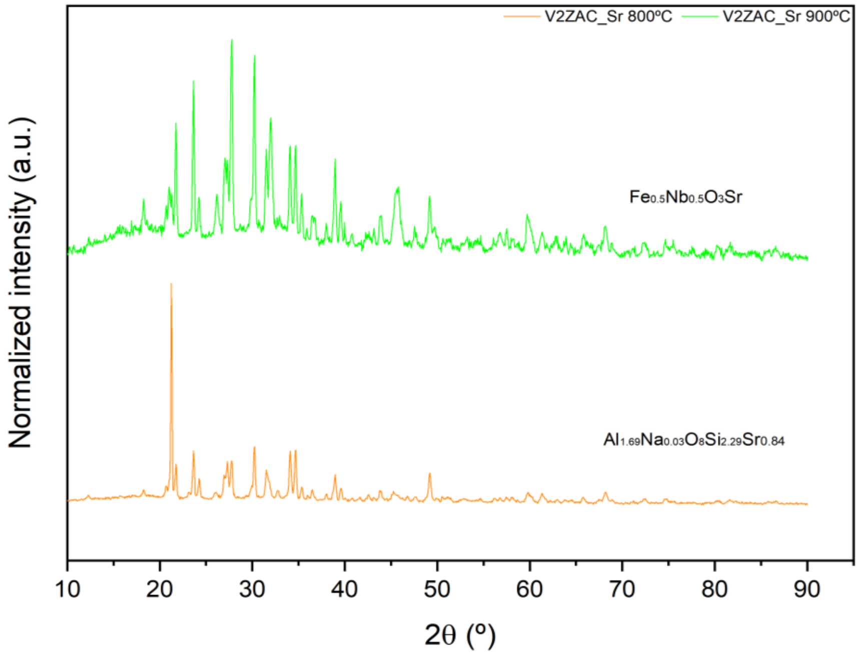
Figure 3: SEM of V2ZAC_Sr at 800ºC (left) and 900ºC (right).



The SEM micrographs of the V2ZAC_Sr composition provide visual evidence of the microstructural changes induced by the heat treatments. The sample treated at 800°C exhibits a surface marked by pores, indicative of its higher porosity. In contrast, the sample treated at 900°C shows a significantly smoother and denser surface, with a marked reduction in visible pores. This observation directly correlates with the measured decrease in water absorption and apparent porosity at the higher temperature, confirming the enhanced densification achieved through sintering.

Following the SEM results, differences were also observed from the X-ray diffraction (XRD) patterns, as illustrated in Fig. 4.

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



The results for V2ZAC_Sr composition at the two temperatures showed patterns of crystallization process. However, at 800ºC 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 ºC a $Fe_{0.5}Nb_{0.5}O_3Sr$ (8409-ICSD) phase were detected, indicating the different interaction of strontium with the glass matrix at the distinct temperatures.

4. CONCLUSIONS

The study successfully demonstrates that elevating the sintering temperature from 800 °C to 900 °C significantly reduces water absorption in all the tested compositions, irrespective of their niobium content. The X-ray diffraction analysis further reveals that this temperature increase facilitates the formation of crystalline phases that effectively interact with and immobilize strontium. While all compositions exhibited potential for immobilization applications, the V4ZAC_Sr sample, sintered at 900°C, stands out due to its optimal combination of low water absorption and minimal apparent porosity. These properties are crucial for ensuring the long-term integrity and containment of the immobilized radioactive waste. However, to gain a more comprehensive understanding of the immobilization process and the long-term behavior of these materials, further characterization techniques, such as leaching studies and microstructural analysis, are recommended.

ACKNOWLEDGMENT

This research was partially supported by the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior - Brasil (CAPES) Finance Code 001 and the Instituto de Pesquisa Energéticas e Nucleares (IPEN) for all structure and materials.

FUNDING

This research financed by the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior - Brasil (CAPES) Finance Code 001.

CONFLICT OF INTEREST

All authors declare that they have no conflicts of interest.

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