Vitrocerus: An alternative for processing MTR spent fuel from research reactors

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ABSTRACT

Research reactors spent fuels disposal is a problematic area that conduces to the quest of feasible solutions for garantee the safe destiny of the spent fuels. In this work, a new method for processing spent fuel from MTR reactors is presented. The main objective of this process is the immobilization of principal radioactive elements present in the spent fuel in order to achieve a suitable material which could be temporally stored safely. The Vitrocerus method involves simple physical procedures performed in a hot cell. It differs from conventional vitrification because there is a reduction in volume of glass material added.

The process consists of the ceramization of a mixture of milled and calcined spent fuel plates with natural uranium oxide (U_3O_8) to obtain the desired isotopic dilution (to low the U^{235} enrichment). At the same time, a small fraction of vg98/12 glass [1] is added to enhance low temperature sintering. The treatment and conditioning tasks proposed in Vitrocerus were tested on MTR fuel miniplates that simulate a real U_3Si_2 dispersed in Al fuel, which were successfully transformed into ceramic sintered pellets [2,3] with low enrichment, structural integrity, good mechanical properties and water corrosion resistance. Eventually these pellets could be stored safely in an interim dry storage facility.

INTRODUCTION

Argentina counts with three research reactors whose work since a few decades with MTR fuels. Disposal of spent fuel has been an investigation issue, searching isolate them of biosphere Nowadays , spent fuels are in cooling pools at the reactor, and showing corrosion hints, with a possible radionuclide release. One objective of the argentinean Programa Nacional de Gestión de Residuos Radiactivos (Radioactive Wastes Management National Program) is to provide a solution for safety location of these elements. In the last years, processing methods for spent fuels type MTR from research reactors has developed in Nuclear Materials Division at CAB (Bariloche Atomic Center), these processes involved the creation of ceramic matrix conteining fuel and natural uranium in an isotopic solution, this project has named CERUS (Spanish acronym of Ceramización de Elementos Radioactivos en Uranio Sinterizado). This process works with reduced volumes than a conventional vitrification, however because of some issues on the material resistance; we propose the addition of small amounts of glass to enhance the lixiviation and resistance properties of the final material.

The objective of this condittioning method is to obtain ceramic pellets of low enrichment, with a significant resistance to possible external factors in the interim storage. In that view it must be evaluate the quality of materials: leaching and strenght resistance, hardness, sintering. Vitrocerus material stills being an uranium sintered ceramic.

EXPERIMENTAL PROCEDURE

Specimen preparation

In order to imite the MTR fuel, it was used a non-irradiated miniplate which simulates the size and structure of the actual plates used. The morphology and composition of the miniplate are proportional to the standard plate in the reactor operating. Figure 1 shows the scheme of procedure for obtaining samples.

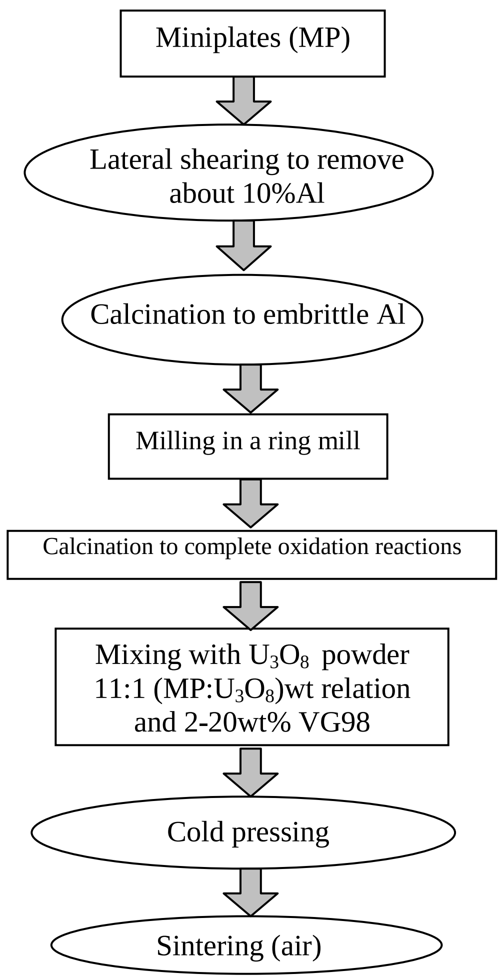

Figure 1. Diagram procedure for miniplates processing

The plates are two layers of pure aluminum (AL 6061) containing a fuel natural uranium silicide (U_3Si_2) powder – but is supposed enriched to 19.75% - and about 40% aluminum powder, these are shown in figure 2. Burned on average, 50% of ^{235}U, then it continues with an enrichment of 10%. MTR fuel element of reference (like the OPAL rector in Australia), contains 18 fuel plates, 2270g of uranium and the whole occupies a volume of 6.5 L.

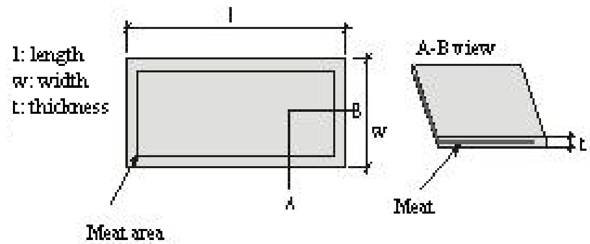

Figure 2. Scheme of miniplate, which is proportional to the standard MTR plate

With a view to achieve an enrichment of about 1,5% to avoid criticality, is necessary to dilute with natural (or depleted) uranium. For a dilution of approximately 1.5% and 50% burned, the mass of natural uranium per unit mass of calcined uranium silicide plate is 11:1 (figure 3).

The Vitrocerus resulting material (48 μm) is a powder mixture of the calcined miniplates (65μm) and natural uranium (45μm) at a rate 11 times greater, for powder particle size distribution it was determined by analysis at 2050 rpm in the Mastersizer micro V2.00. Powders were rotated mechanically, and adding glass VG98 in different proportions. The glass base VG98 borosilicate glass was specially formulated for the vitrification (immobilization) of waste, which was originally developed for the vitrification of wastes from power reactors in Germany [1] and is the basis for numerous formulations matrix (glass particle size: 15μm), the addition of sodium gives it a low viscosity: 1.7 Log $dPas^{-1}$ at the temperature of 1200°C.

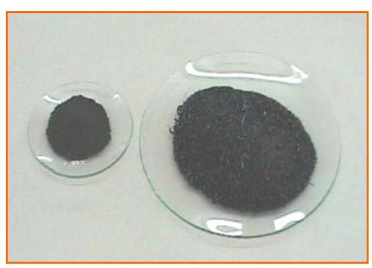

Figure 3. Powders for isotopic dilution

The powder mixture was compacted in cylindrical pellets using a metalic floating matrix, which were sintered in air [4], the final samples are shown in figure 4.

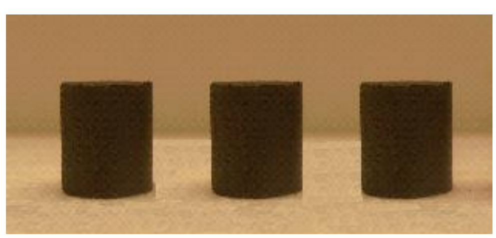

Figure 4. Photograph of sintered pellets

CHARACTERIZATION

Morphology and sintering

Precursor powders, ie the aluminum calcined miniplates (before and after each thermal tratement), U_3O_8 natural uranium, were analyzed by Scanning Electron Microscopy (Philips 515 SEM), EDS (Edax Genesis 2000) and X Ray Difraction XRD (in a Philips Difra PW1700 difractometer with a Cu Kα radiation. Studies were carried out in equipment available in the Materials Characterization Group of the CAB, in order to characterize the starting materials.

Figure 5 shows a heating microscopy test conducted in the high temperature microscope Laboratory of Nuclear Materials to visualize the variation of the area with the temperature of a pellet. In this experiment it generates an image of the sample while is sintered and notes that there is at first an increase in the area due to oxidation of uranium and densification occurs later the same up to 94% of the area.

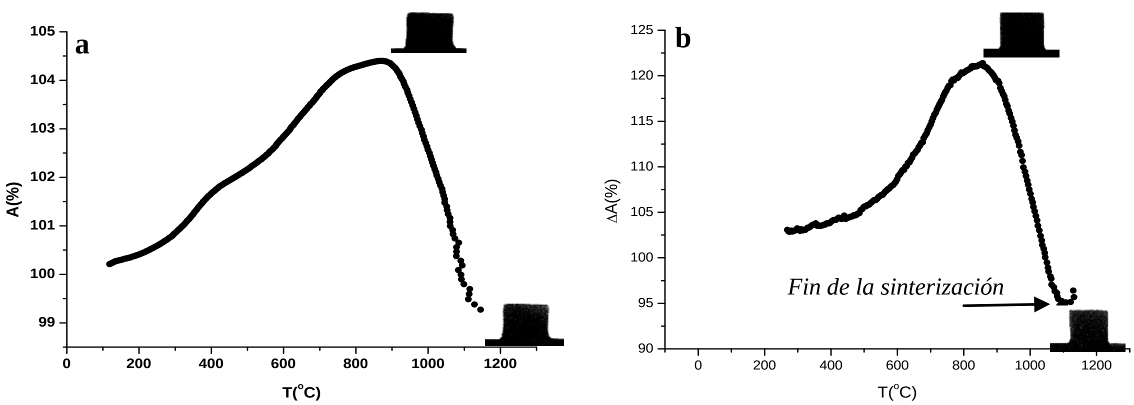

Figure 5. Graph of variation of the area with the temperature of a pellet a) without addition of glass and b) with 5% VG98

In the micrograph obtained by scanning electron microscope SEM (figure 6) depicts the partial sintering of the system for a sample containing 5%compared to a glass without VG98.

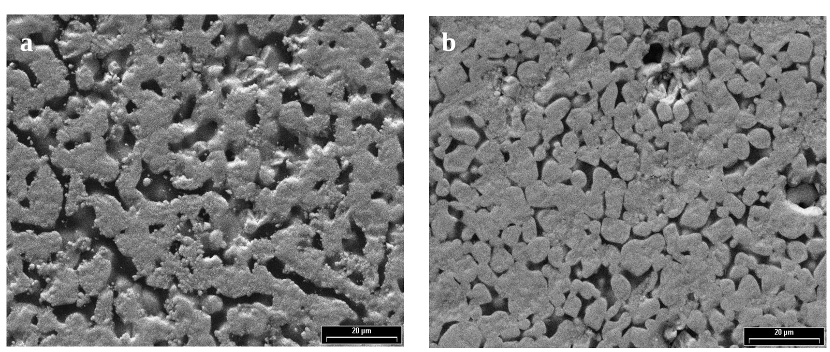

Figure 6. Micrographs of sample a) without VG98/12 glass addition, b)with 5wt%VG98/12

In micrographs at figure 7 it evidences a mechanism of liquid phase sintering. There is some solubility between oxidated miniplates-glass, which allows that glass assists sintering of the compound by the action of a liquid phase.

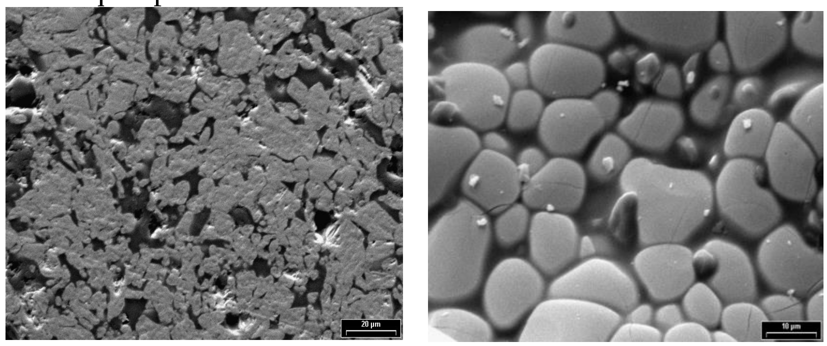

Figure 7. Micrographs of sample with 8wt%VG98/12

<u>Density and porosity</u>

Density values of the samples goes from 6,2 to 4,5 g cm^{-3} for pellets without addition of glass and with 10wt%VG98 respectively. The average of open porosity was 19% for the whole range of samples.

<u>Mechanical Properties</u>

Leaching tests were carried out the protocol MCC1P [5] using monolithic samples, the characterization of mechanical behavior was made using the Brazilian test or diametral compression tests [6-7], respect to hardness test were made indentations in a Mitutoyo microhardness tester, the diamond indenter load was 50g.

DISCUSSION

Successful results were obtained for leaching resistance, in terms of gravimetric dissolution rate (GDR) for10wt%VG98 batches, this GDR is similar to the glass value. This improvement in properties, enhance possibility to be implemented as part of processing and conditioning of spent fuel and versatility to be placed in a dry interim storage [8].

In terms of mechanical strength properties, the materials prepared with 2-10wt% VG98 glass, shows superior quality compared to those not contained. The pressure at which cracks occur in the material with a 2wt%VG98 are 14 times greater than without glass. On the other hand, with Vitrocerus process the volume of a fuel element increases about 25%, but this is far below of the volume in a conventional vitrification [9].

CONCLUSIONS

The process is a viable alternative that could be applied in the processing within the framework of the management of spent fuel from research reactors.

The material obtained is enough resistent to be manipulate and ready to dispose in a dry temporary facility.

The shape and properties of the resulting material are suitable to be conditioned in the same store designed for power reactors spent fuels.

Three inherent problems related with this kind of spent fuel were solved: corrosion of the plates, high enrichment and volumen reduction, using a simple mechanical process, that can be performed in a hot cell.

Small amounts, of glasses would enhanced the resistance, which is comparable to glass, similar to that used in the processes of safe disposal of HLW or spent fuel.

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