RESEARCH ON THE MANAGEMENT OF MTR SPENT FUEL IN THE DMN-CAB-CNEA

A.A. CHAVEZ
Fuel and Waste Division, Nuclear Materials Department, Applied Research Management, Bariloche Atomic Centre, National Atomic Energy Commission (CNEA).
San Carlos de Bariloche, Rio Negro, Argentina.
Email: arielchavez@cnea.gob.ar

A. MAGNONE
Fuel and Waste Division, National Atomic Energy Commission (CNEA).
San Carlos de Bariloche, Rio Negro, Argentina.

F.L. BECKER
Fuel and Waste Division, National Atomic Energy Commission (CNEA).
San Carlos de Bariloche, Rio Negro, Argentina.

C.N. AYALA
Fuel and Waste Division, National Atomic Energy Commission (CNEA).
San Carlos de Bariloche, Rio Negro, Argentina.

R.M. CASTAGNA
Fuel and Waste Division, National Atomic Energy Commission (CNEA).
San Carlos de Bariloche, Rio Negro, Argentina.

Abstract

In 1967, the RA-3 research reactor was inaugurated to cover the Argentinean demand for medical radioisotopes [1]. The spent fuels from this reactor have been stored in wet conditions in CAE facilities, in the past at the DCMFEI deposit and currently in the FACIRI deposit [2].

There are three proposals for the management research reactor spent fuels (RRSF) [2]:

- Uranium isotopic dilution for use in light water power reactors.
- Return to the country that supplied the enriched uranium when this possibility exists.
- Isotopic dilution conditioning for final disposal in the deep geological repository.

That will be defined when the country defines its nuclear fuel cycle.

In our Uranium Laboratory of the Nuclear Materials Department (LU-DMN) located in the Bariloche atomic centre, we worked on proposals for the conditioning and immobilization of these types of spent fuels by isotopic dilution and ceramization [3, 4], the addition of glass to the ceramization process [5], and glasses matrices for immobilization [6, 7]. The proposal of this presentation is to show some of our research lines in the context of the nuclear fuel cycle of the Argentina nuclear energy sector on proposals of the management of RRSF, and in particular, results on the Cerus Process (Ceramization of radioactive elements in sintered uranium).

1. INTRODUCTION

Argentina is home to a number of research reactors situated across the country. The CNEA atomic centres are home to the most important, including RA-1 in Constituyentes, RA-6 in Bariloche, and RA-3 and RA-10 in Ezeiza, the latter of which is still under construction [8]. The spent fuels from these reactors are stored in pools located within the reactor buildings. RA-3, the radioisotope-producing reactor, requires more frequent fuel element replacements than the others. Initially, the spent fuels were stored in the DCMFEI deposit, but as of 2016, they are now stored in FACIRI [2]. Additionally, Argentina has several critical facilities, including RA-0 at the University of Córdoba, RA-4 at the University of Rosario, and RA-8 at the Pilcaniyeu Technological Complex [8]. In the RERTR program, all of the HEU's spent fuels have been returned to their country of origin [2].

Research Reactor Spent Fuel (RRSF) management is challenging due to the enrichment at discharge higher than 11% of ^{235}U [9]. Due to criticality considerations and susceptibility to materials degradation, direct disposal is not considered a viable option for RRSF management [8]. The main idea of the process consists of the use of natural (or depleted) uranium oxide to achieve the ^{235}U isotope dilution down to 1 wt.% [3].

2. SPENT FUEL CONDITIONING

The National Radioactive Waste Management Program (PNGRR) established the Strategic Plan for Radioactive Waste Management, which outlined various proposals for managing spent fuels [2]. The DMN was responsible for conducting thermo-mechanical treatment of plates, isotopic dilution via the dry method, and then immobilizing it in glass matrices through sintering and melting, or in a ceramic matrix through sintering.

2.1. Glass matrices

We evaluated borosilicates, Iron borosilicates, and Iron phosphate glasses as potential matrices for immobilizing calcined RRSF. We conducted sintering tests using various loads of calcined and isotopically diluted materials, pressing pellets at 1.5 kg/cm^2 and sintering them at temperatures ranging from 510 to 755ºC. We determined density through immersion and geometry, and assessed leaching behavior. The samples were then characterized using electron microscopy and EDS [6, 7].

2.2. Ceramic matrices

The U3O8 was selected for two objectives to achieve the ^{235}U isotope dilution and seeking for a monolithic ceramic wasteform with triuranium octoxide as immobilization matrix [3]. The U_3O_8 precursor powder was obtained by two experimental Routes: R1_ by calcination of UO_2-ex Ammonium uranyl carbonate (AUC) powder; R2_ by nitric dissolution of this UO_2 powder, subsequent precipitation of ammonium diuranate (ADU) and final thermal conversion to U_3O_8. Finally, there were pressed at 752 MPa, in a stainless-steel matrix of 9.2mm of diameter. The “green” pellets were sintered in air at four different temperatures 1100, 1200, 1300 and 1400°C [10]. Then samples were characterized by immersion, ceramography, optical and electronical microscopy, and microhardness [4, 10].

3. RESULTS

3.3. Glass matrices

The results of leaching iron phosphate glass with a treated RRSF loading of 7%, 10%, and 15% are presented in Fig. 1, Fig. 2 displays electronic microscopies of the glass with the load, showing areas with uranium in the glassy matrix. Fig. 3 shows the sample after a leaching test of 28 days [6, 7].

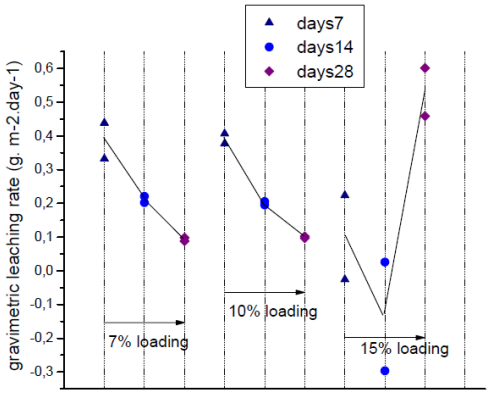

FIG. 1. Gravimetric leaching rate for 7, 10 and 15% loading.

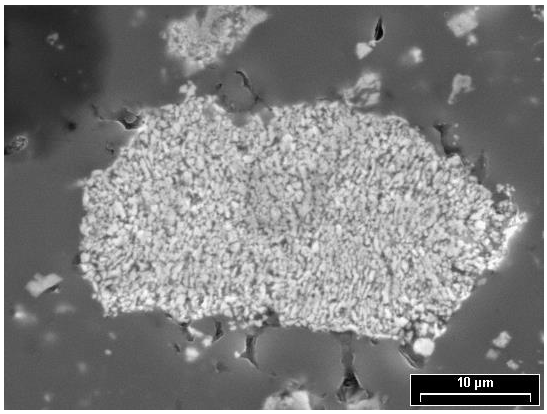

FIG. 2. Iron phosphate glass with uranium, polished sample.

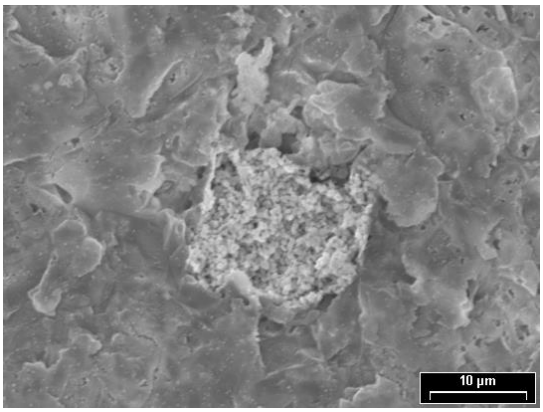

FIG. 3. Iron phosphate glass with uranium, after 28 days leaching.

3.4. Ceramic matrices

The behaviour of precursor powders varies depending on their particle morphology. Fig. 4 compares green density to sintered density, while Fig. 5 displays the ceramographies of pellets made from the two synthesis routes [4, 10].

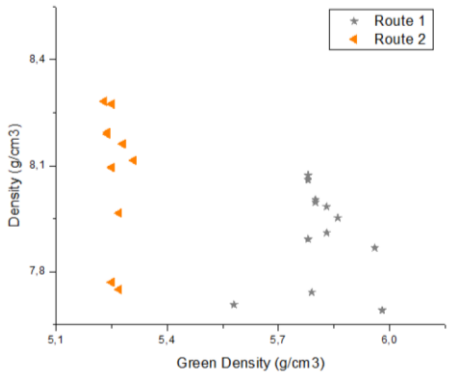

FIG.4. Sintered density vs green density of pellets.

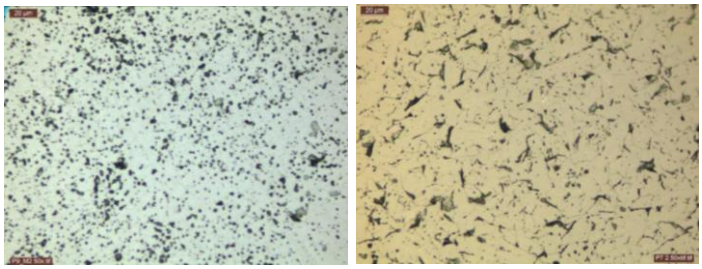

FIG.5. (Izq) pellet Route 1, 1300ºC, 50X. (Der) pellet Route 2, 1300ºC, 50X.

In order to establish the correlation between Vickers and Knoop hardness, we performed microhardness examinations on sintered samples from these two precursor powders. Our findings can be found in Table 1, which showcases the average HV and HK values at varying temperatures, as well as the HV/HK ratio and average HV/HK ratio for each powder. The results of the Knoop and Vickers indentations on the samples are illustrated in Fig. 6 [10].

TABLE 1. AVERAGE VALUES FOR HV, HK AND RATIO HV/HK OF SAMPLES

	T(°C)	HV av (Gpa)	HK av (Gpa)	HV/HK	HV/HK av (mat)	σ(HV/HK av)
Route 2	1100	4,1	3,7	1,10	1,15	0,04
	1200	4,4	3,8	1,15		
	1300	3,7	3,3	1,13		
	1400	3,6	3,1	1,19		
Route 1	1100	3,1	2,5	1,22	1,15	0,06
	1200	3,3	2,9	1,15		
	1300	3,0	2,6	1,13		
	1400	2,6	2,4	1,08		

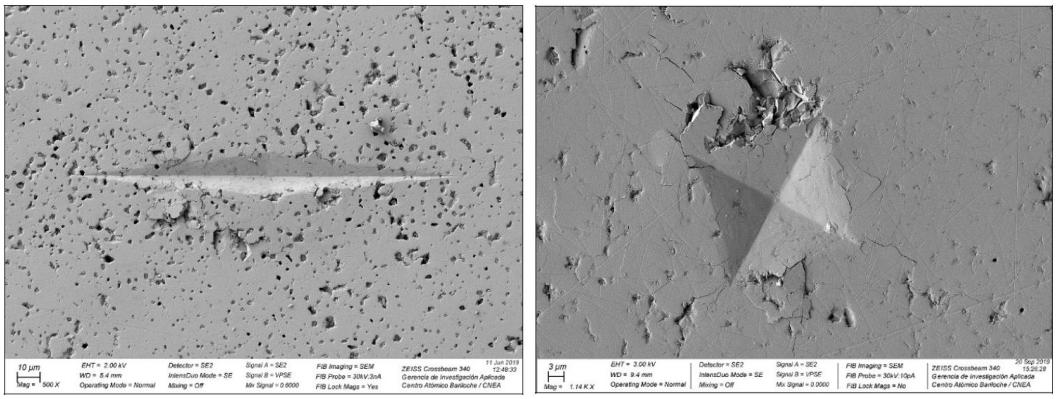

FIG.5. (Izq) Knoop indentation, pellet 1300ºC. (Der) Vickers indentation, R2, pellet 1300ºC.

4. CONCLUSIONS AND OUTLOOK

In Argentina, research reactors are essential for training highly skilled personnel, developing new materials for industry and energy, and providing radioisotopes for radiodiagnosis and disease treatment. These activities align with the UN Sustainable Development Goals, including promoting health and well-being, quality education, affordable and clean energy, work and economic growth, and climate action.

It is important to consider and develop treatments for managing spent fuel from these reactors to ensure proper management and avoid negative impacts on people and the environment. Work related to the back end of spent fuel has been ongoing since the 2000s, focusing on non-irradiated materials. However, future stages should evaluate these processes using irradiated materials in hot cell facilities.

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