Nuclear Fuel Cycle Optimization

with Shielded Electron Probe MicroAnalysis

Investigating new ways to reduce the long-term radiotoxicity of nuclear wastes

The long-lived minor actinides (MA) that result from irradiation of oxide fuels in light water reactors increase the radiotoxicity of nuclear waste. In order to facilitate disposal, scientists investigate various processes to recycle the MA. One consists of separating the MA from the spent fuel and incorporating the MA in novel U-Pu-Zr metallic fuels prior to irradiation under a fast-neutron spectrum.

The recent FUTURIX-FTA[1] experiment aims to assess the safety of MA transmutation. In this study, irradiation was performed in a sodium fast reactor at an average linear heat generation rate of 318 W/cm and peak cladding temperature of 550°C. Shielded EPMA was used to investigate the interactions occurring between fuel and cladding throughout the process.

U-Pu-Zr fuel behavior

The irradiated fuel was composed of 34.1 U — 28.3 Pu — 3.8 Am — 2.1 Np — 31.7 Zr (weight %). The ~5 µm thick Zr rich rind encircling the fuel perimeter at the time of fabrication remains intact after irradiation (Figure 1). A large crack is also visible formed from swelling and expansion of the fuel towards the cladding.

Diffusion from the cladding

Prior to irradiation the fuel was cladded in a variant of 316 stainless steel. EPMA mappings of major elements from the cladding (Fe, Ni, and Cr) reveal penetration of at least 30 µm into the fuel after irradiation. A decrease in concentration is observed at the very beginning of the cladding-fuel interface due to the influx of fission products into the cladding (Figure 2).

Specific interactions at the fuel-cladding interface

The fuel-cladding chemical interaction is of paramount importance to understand diffusion and migration mechanisms. The outer perimeter of the fuel displayed a series of six complex distinct layers (not detailed here). Am and Sm penetrate ~8 µm into the cladding along presumed grain boundaries (Figure 3). Ce and some lanthanide elements constitute linear enrichment zones along the outer fuel periphery.

Instrumentation

CAMECA Shielded EPMA proves a tool of choice for characterizing the distribution and chemical composition of highly radioactive fission products under safe conditions. These data illustrate some its unique analytical advantages: superior energy resolution (10 eV) which is essential for differentiating complex overlapping spectral peaks, high sensitivity spectrometers ensuring excellent detection limits, and quantitative elemental mapping from sub-micron areas providing precise and accurate compositional information.

[1] *https://www.osti.gov/biblio/1356073*

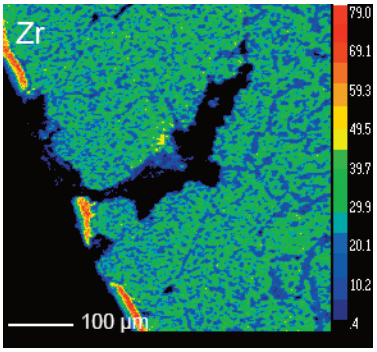
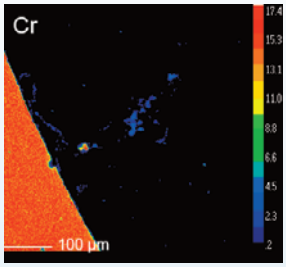
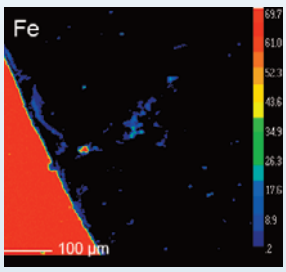


Figure 1

EPMA quantitative X-ray maps showing elemental distribution at the fuel-cladding interface after irradiation. Vertical axis on the right of the image in weight percent units.

Figure 2





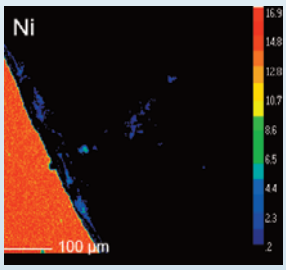
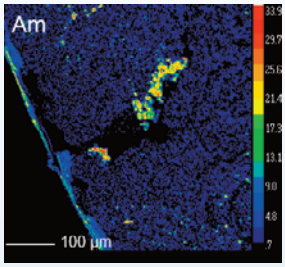
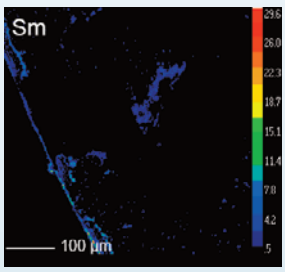
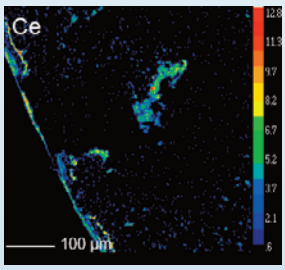


Figure 3



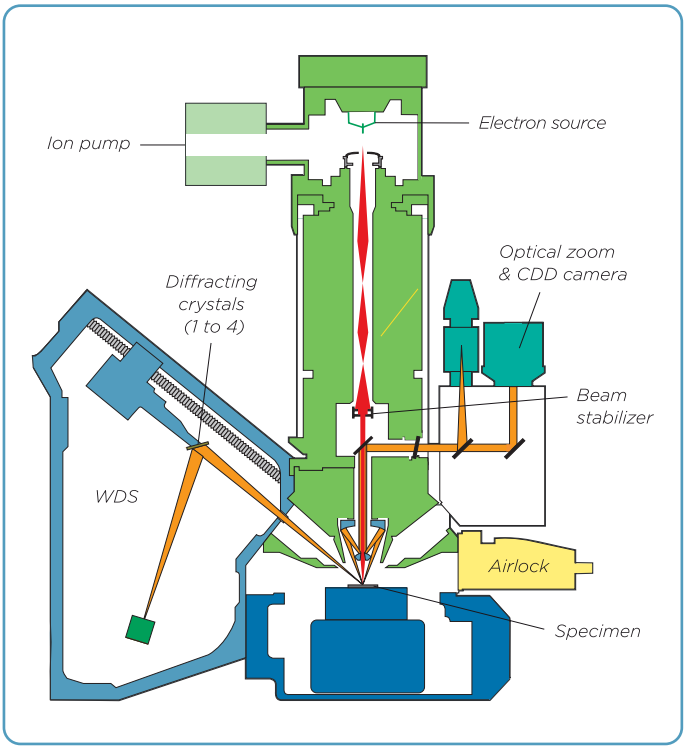




Adapted from K. Wright et al., Journal of Nuclear Materials (2019). DOI: 10.1016/j.jnucmat.2019.1517451. Data collected on SX100R at Idaho National Laboratory, USA.

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The technology behind



Electron Probe Microanalysis (EPMA)

EPMA works by bombarding a micro-volume of a sample with a focused electron beam (typical energy = 5-30 keV) and collecting the X-ray photons thereby emitted by the various elemental species. The wavelengths of these X-rays being characteristic of the emitting species, the sample composition can be easily identified and quantified by recording WDS spectra (Wavelength Dispersive Spectroscopy). Owing to the internal properties of WDS, the general sensitivity, analysis of light elements and interpretation of qualitative spectra are all superior with EPMA than with standard SEM/EDS systems.

EPMA remains one of the most precise and accurate microanalytical technique. It is a non-destructive, qualitative and quantitative method of elemental analysis of micron-sized volumes at the surface of materials, with sensitivity at the level of ppm. Routine quantification to 1% reproducibility is obtained over several days.



SKAPHIA
SHIELDED ELECTRON PROBE ANALYZER

SKAPHIA is CAMECA's latest generation Shielded Electron Probe MicroAnalyzer, providing a safe environment for manipulating and analyzing radioactive materials. It is designed to accept samples emitting beta and gamma radiations with a maximum acceptable g radiation level of 111 GBq at an energy of 0.75 MeV.

- The tool is installed in a hot cell (lead or concrete shielded room).
- Remote-manipulators and/or ball manipulators are used to insert and mount the radioactive samples.
- The instrument is fully remote-controlled (stage, column, diaphragms...), with electronics and computer deported outside the "hot" cell environment.
- All WDS analyzers and detectors are shielded to prevent the background caused by the gamma radiations.
- The stage is made of Denal material.
- The secondary electron detector has a special orientation to avoid gamma ray perturbation.
- The backscattered electron detector is protected by a retractable shield.

A dedicated EPMA software program provides all necessary features for quantitative analysis, X-ray mapping, line profile acquisition and data processing.

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