

Electron probe microanalysis of a high burnup (Th,Pu)O₂ fuel section

Philipp Pöml¹, Stéphane Brémier¹, Jérôme Himbert¹

¹ European Commission, Joint Research Centre, Institute for Transuranium Elements, P.O. Box 2340, 76125 Karlsruhe, Germany

Electron probe micro-analysis (EPMA) is an important technique for a broad range of applications in nuclear sciences. One main target is to improve the safety of the nuclear fuel cycle, by studying the chemical and physical properties of spent nuclear fuel and its fission products, either solids, volatiles, or gases, after the irradiation [1]. Of particular interest for the nuclear scientist is the distribution and quantity of actinides in the fuel, before and after irradiation. The fuel types most commonly studied by EPMA are uranium oxide and Mixed Oxide Fuel (MOX) containing a mixture of uranium and plutonium oxide. Minor actinides (Np, Am, Cm) are produced during irradiation by neutron capture and/or alpha-decay of uranium, and can be added during fuel fabrication (usually containing the fissile material) in the case of transmutation schemes.

As an alternative to widely used UO₂ and MOX fuels, thorium oxide-based fuel can be a very interesting option. A thorium fuel cycle could have several advantages compared to "traditional" fuel cycles:

- Thorium is abundant in the Earth's crust, hence its deployment in a Th-²³³U cycle can be considered as a viable long term energy option.
- In contrast to uranium based fuel, thorium fuel does not breed minor (transuranic) actinides such as plutonium, americium, or curium. Hence, spent fuel containing these latter elements can be avoided.
- ThO₂ is an ideal candidate as fuel matrix to burn surplus plutonium that is currently produced worldwide in light water reactors. During irradiation, the plutonium would be consumed, hence (Th,Pu)O₂ fuel is ideal for diminishing the world's plutonium inventory without having to worry about safe plutonium disposal.

In this contribution we report the results of an EPMA study on an irradiated (Th,Pu)O₂ fuel pellet section (burnup ~40 GWdays/tonne). Figure 1 shows a secondary electron image of the fuel specimen. The sample was investigated along a typical radius (white line in Figure1) to obtain the radial distribution of the elements present in the fuel. The analysed elements were the fission products Ru, Mo, Xe, Cs, Nd, and the actinides Th, U, Pu. Some uranium was present as decay product in the Pu oxide fraction, and additional U was bred by the thorium during irradiation.

The analysis of irradiated thorium fuel differs in some significant points from the analysis of uranium fuel, particularly in terms of line overlaps. For uranium fuel analysis commonly the U M α and Xe L α can be used. In the case of thorium fuel, however, the U M α line is heavily overlapped by the Th M β line, hence the U M β has to be used. The U M β line had to be corrected for interference with the Th M₃-N₄ line. In case of Xe, the Xe L β line has to be used, because the Xe L α line is overlapped by the Th M₂-N₄ line. Close to the Xe L β line, however, is U M₂-N₄ line. Analysis of the peak positions showed that those lines are 150 spectrometer units (~ 0.01 Å) apart. Considering also the very low concentration of U in the sample, it was concluded that the consequence of this overlap is negligible. Additionally, a spectrum was acquired (Figure 2) which shows no significant U M₂-N₄ intensity/overlap issues.

This contribution will show detailed results of the element distribution in the fuel disc. In addition, we will compare our results with theoretically calculated irradiation and fission yield results.

References:

[1] C Walker, *Journal of Analytical Atomic Spectrometry* **14** (1999) p.447.

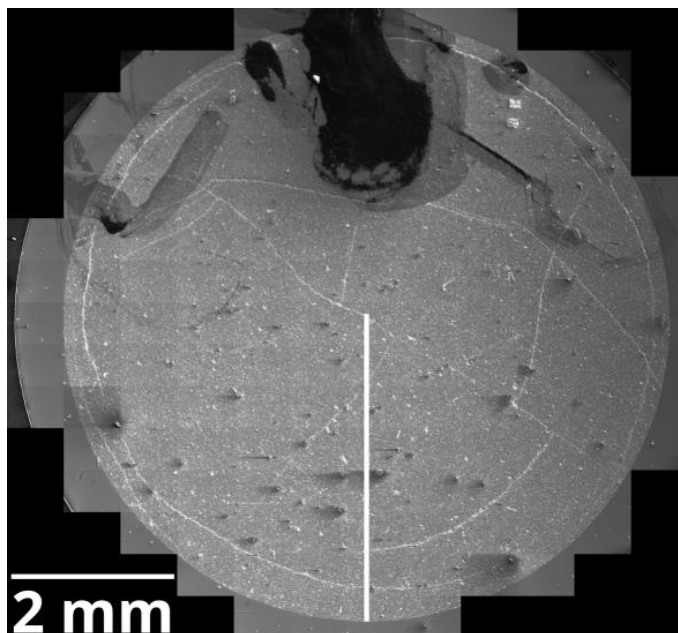


Figure 1. Secondary electron image of the analysed (Th,Pu)O₂ fuel sample. The white line marks the analysed radius.

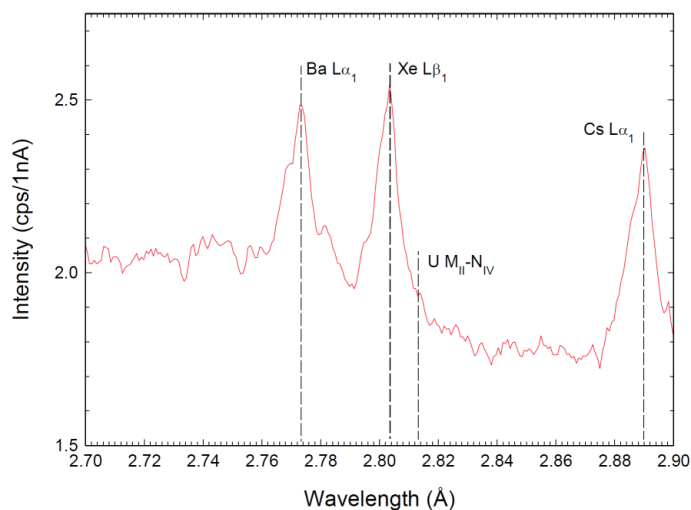


Figure 2. Spectrum of the quartz crystal between 2.7 and 2.9 Å taken on the edge of the fuel sample. The spectrum shows that the overlap of the U M₂-N₄ line on the Xe Lβ line has a negligible effect. The peak of Ba Lα shows that a significant amount of Ba is present in the sample. The Cs Lα line is present at the low energy end of the spectrum.