doi:10.1017/S1431927614010733
Microsc. Microanal. 20 (Suppl 3), 2014

Microbeam analysis techniques for the characterisation of irradiated nuclear fuel

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Microbeam analysis is widely used in the nuclear power industry. It is used for routine postirradiation examination and for research into the mechanisms affecting safe operation of the nuclear fuel. The techniques most commonly used are wave-length dispersive electron probe microanalysis (WDS-EPMA), scanning electron microscopy (SEM) and secondary ion mass spectrometry (SIMS). Other microbeam analysis techniques that have been successfully applied to irradiated nuclear fuel are transmission and replica electron microscopy (TEM and REM), micro X-ray fluorescence (micro-XRF) and micro X-ray diffraction (micro-XRD). SEM, TEM and REM have been mainly used to study the evolution of fission gas bubbles, which cause the fuel to swell during irradiation [1–3], and micro-XRD has been used to investigate the change in lattice parameter caused by irradiation damage and the build-up of fission products during irradiation and to assess their influence on the transformation of the fuel microstructure after prolonged irradiation [4].

WDS-EPMA is the microbeam analysis technique most widely available in nuclear research centres around the world. From the perspective of investigating irradiated nuclear fuel, this technique has a number of limitations. The main drawback is that it does not measure isotopes. A further shortcoming is the detection limit, which for a radioactive sample is at best 200 ppm [5]. In addition, the fission gas krypton cannot be measured because the second order $M\alpha_1$ U X-ray line coincides with the Kr $L\alpha_1$ line and the Kr K X-ray lines have a high critical excitation energy of 14.3 keV. Finally, owing to the shallow electron penetration in nuclear fuel, the fission gas trapped in pores and bubbles larger than about 0.1 μm cannot be detected [6,7]. These deficiencies have been overcome by combining EPMA with micro-XRF [8] and SIMS [9].

This presentation will describe the context of irradiated fuel examinations and in particular the main characteristics of irradiated fuel rods, followed by a description of the features that set apart the microbeam analysis of irradiated nuclear fuel from standard practice on "cold" materials. Finally, specific examples illustrating the past and present use of microbeam analysis in nuclear research are discussed, with emphasis on most valuable and unique sets of results.

Figure 1a shows the typical radial distribution profile of Xe in a conventional pressurised water reactor fuel. The Xe concentration falls sharply close to the surface ($r/r_0 = 1$) as a result of a recrystallisation process during which the fission gas are swept out of the original fuel grains. EPMA can thus be used to estimate the radial extent of the transformation of the microstructure by measuring the distance from the pellet edge over which Xe depletion in the fuel matrix occurs. In Fig.1b the local Xe concentration measured in the outer regions of UO_2 fuel sections is plotted as a function of the local burn-up. The steep fall of the Xe concentration at burn-up around 60 MWd/kgHM marks the onset of the recrystallisation process. At burn-ups over 120 MWd/kgHM the constant low Xe concentration indicates that the fuel microstructure transformation is complete. [10] In figure 2, large area X-ray mapping of Pu, U and Zr of a metal alloy fuel illustrate the heterogeneous distribution of the fuel components after irradiation. The as-fabricated distribution of the base fuel elements U, Pu and Zr in the ingot was

homogeneous at the macroscopic scale. At this scale, EPMA provides clear evidence that significant mass transport occurred during irradiation. [11]

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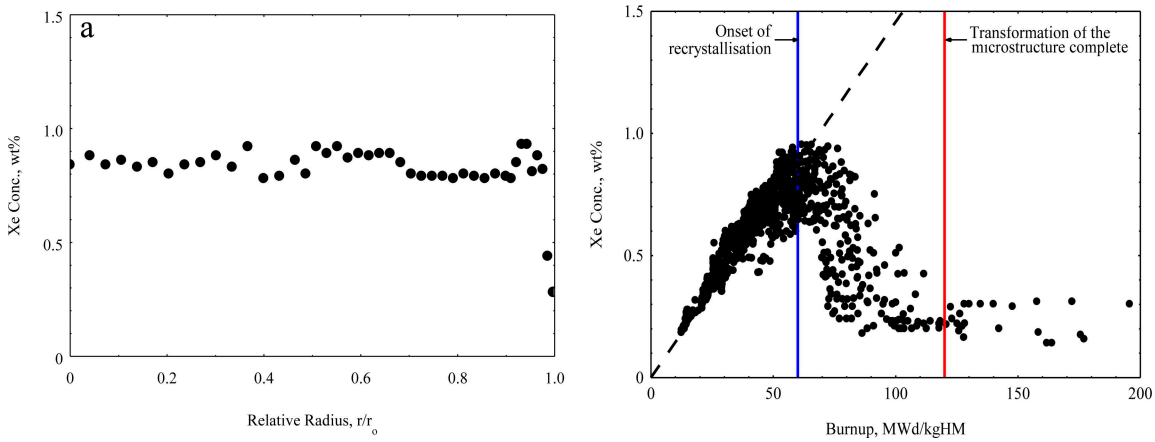

Figure 1: a) Typical radial distribution profile of Xe in a conventional pressurised water reactor fuel. b) Xe concentration measured in the outer regions of UO_2 fuel as a function of the local burn-up.

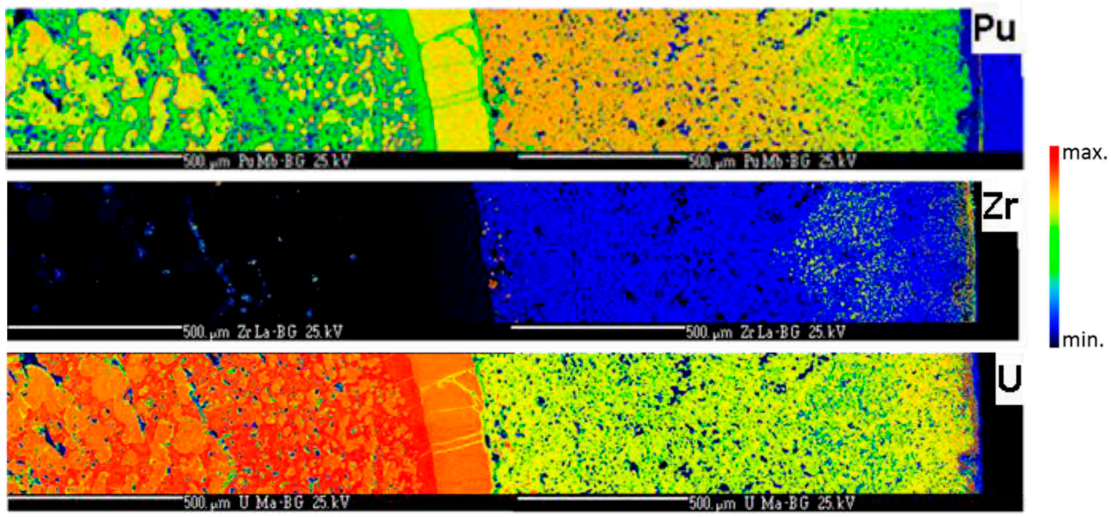

Figure 2: False colour large area X-ray mapping for Pu, U and Zr illustrating the distribution of fuel constituents in a metal alloy fuel sample.

https://doi.org/10.1017/S1431927614010733 Published online by Cambridge University Press