SIMS Analysis of Zircaloy Cladding and Ion Implanted UO₂: First Results

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A CAMECA IMS 6F, magnetic sector, secondary ion mass spectrometer (SIMS) has been installed in the Microbeam Analysis Laboratory of the Institute for Transuranium Elements for the analysis of irradiated nuclear materials (fuel and cladding). The device is specially equipped with heavy metal shielding that will enable the safe examination of irradiated samples with activities up to 75 GBq. A lead cell and a steel glove box protect personnel and electronic systems from radiation and contamination [1].

Lithium and boron present in the primary coolant of water reactors have important roles in cladding corrosion phenomena and are incorporated in the oxide layer on the outer surface of Zircaloy during irradiation. The incorporation of lithium and boron in the cladding oxide layer has been simulated by treating Zircaloy samples in an autoclave under appropriate conditions. The samples were then analysed by SIMS. Ion maps and line scans were produced (Cs^+ primary ion beam) to determine the lithium and boron distributions and to measure the oxide layer thickness. As shown in Fig.1, lithium and boron are apparently uniformly distributed in the oxide layer. The oxide layer thickness measured by SIMS is in excellent agreement with the optical microscopy results.

The second important field of interest is quantification of fission product concentrations in irradiated nuclear fuel. Neodymium analysis is important for fuel local burn-up assessment while xenon characterization in bubbles, which is not possible by EPMA, will allow a better understanding of fuel swelling and gas release mechanisms during irradiation. Thus, Nd and Xe implanted UO₂ single crystals have been studied by depth profiling. Figure 2 shows the profile of ¹⁴³Nd implanted UO₂ and compares it with the theoretical Gaussian implantation profile (energy 400 keV, dose 5.10^6 at.cm⁻²). A small discrepancy is observed at the tail of the curve, attributed to a slight decrease in the UO₂ density due to defect creation during implantation. The result is an increase of the length of implanted ion tracks. As is the case for all rare gases, ionisation of Xe occurs above the sample surface [2] and thus an oxygen leak is used to enhance the secondary ion signal [3]. Figure 3a shows ¹²⁹Xe depth profiles (O₂⁺ primary ion beam) acquired at different current densities, J. In Fig. 3b, the variation in the Xe peak count rate and the dependence of the time to reach the maximum count rate is plotted as a function of J. The ¹²⁹Xe count rate increases in accordance with aJ+bJ², in agreement with theory, and the time required to reach the maximum count rate varies inversely with J. These results are in excellent agreement with the literature [3].

References

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- [2] M.A. Ray et al., J. Vac. Sci. Technol., A 6 (1988) 44.
- [3] L. Desgranges and B. Pasquet, Nucl. Instr. and Meth. in Phys. Res. B 215 (2004) 545.



Fig.1. SIMS results for B and Li trace concentrations in the oxide layer on Zircaloy cladding material; a) and b) Li ion map and corresponding optical micrograph; c) line scans for Zr, Li, B and O. The oxide layer thickness measurements are in excellent agreement with the optical microscopy results.



Fig.2. Depth profile at $J = 0.8 \text{ A/m}^2$ for ¹⁴³Nd implanted in UO₂. The SIMS measurement is in excellent agreement with the theoretical Gaussian implantation profile.



Fig.3. SIMS results for UO₂ implanted with ¹²⁹Xe (energy 400 keV, dose 5.10^{16} at.cm⁻²); a) influence of current density on the measured depth profiles (ion intensity versus time); b) the Xe peak count rate and the time required to reach the peak count rate as a function of current density.