

Radiation effects in oxides foreseen for the immobilization and transmutation of radioactive wastes: case study of zirconia

Sandra Moll, Lionel Thomé

CSNSM Orsay (CNRS-IN2P3), Université Paris-Sud, Bâtiment 108, 91405 Orsay, France

The safe and long-term control of the radioactive residues arising from power plants and the dismantling of nuclear weapons is a crucial problem that developed countries have to solve in the next decades. The long history of research on this topic has shown that ceramic oxides are among the most promising materials for use as matrices for the immobilization and/or transmutation of nuclear wastes. Because of its stability in a radiative environment and its ability to confine the radiotoxic elements, yttria-stabilized cubic zirconia (YSZ) was identified as a potential candidate for the incorporation of radioactive species. Radiation produced in nuclear reactors or in storage forms may be simulated by external irradiations with various types of ions in a broad energy range. The two main slowing-down regimes (S_n and S_e) were distinctly investigated: low-energy heavy ions account for the recoil nuclei arising from the α -decay of actinides (S_n); swift ions aim to reproduce the impact of fission fragments (S_e). In this study, the depth distribution, the degree and the nature of the damage, the microstructural modifications produced in ion-irradiated YSZ single crystals were measured as a function of the ion fluence by combining advanced techniques which probe the sample at various spatial scales (RBS/C, XRD, TEM, AFM).

The damage buildups determined for both low energy (MeV heavy ions) and high energy (GeV heavy ions) irradiations were satisfactorily represented in the framework of a model (MSDA) [1] which was developed to account for the damage production in irradiated solids (Figures 1 and 2). The shape of the buildup and the fluence range where the damage becomes significant ($\sim 10^{13} \text{ cm}^{-2}$ for S_e and $>10^{15} \text{ cm}^{-2}$ for S_n) strongly depend on the ion slowing-down regime.

The damage cascades created by nuclear collisions at low energy (effects of S_n) lead to several steps of disorder accumulation by the creation and relaxation of radiation-induced stresses (Figure 1). A sharp increase of the damage is exhibited in the second step, due to the formation of dislocation loops which overlap to create a dense network of tangled dislocations revealed by TEM. This second step occurs when a radiation dose of a few dpa is achieved independently of irradiating ions [2]. At higher fluences, a third step is exhibited where a decrease of the amount of radiation damage is observed, which may be attributed to the reorganization of dislocations.

Tracks created by electronic excitation at high energy (effects of S_e) lead to a direct transformation of the melt volume into a new structure via a single-step process (Figure 2). The diameter of ion tracks indirectly determined from RBS/C data agrees with that directly measured by TEM ($\sim 4 \text{ nm}$). Larger hillocks of matter ejected from the ion tracks are observed by AFM. Dislocations are created along the ion trajectories and overlap with increasing fluence to form a dense network of dislocations. TEM and RBS/C data have also evidenced the existence of a S_e threshold for track formation (in the range 20–30 keV/nm) [3].

References

- [1] J. Jagielski and L. Thomé, *Vacuum* 81 (2007) 1352.
- [2] L. Thomé et al., *Eur. Phys. J. Appl. Phys.* 24 (2003) 37; S. Moll et al., to be published (2009).
- [3] S. Moll et al., *J Appl. Phys.* 105 (2009) 023512.

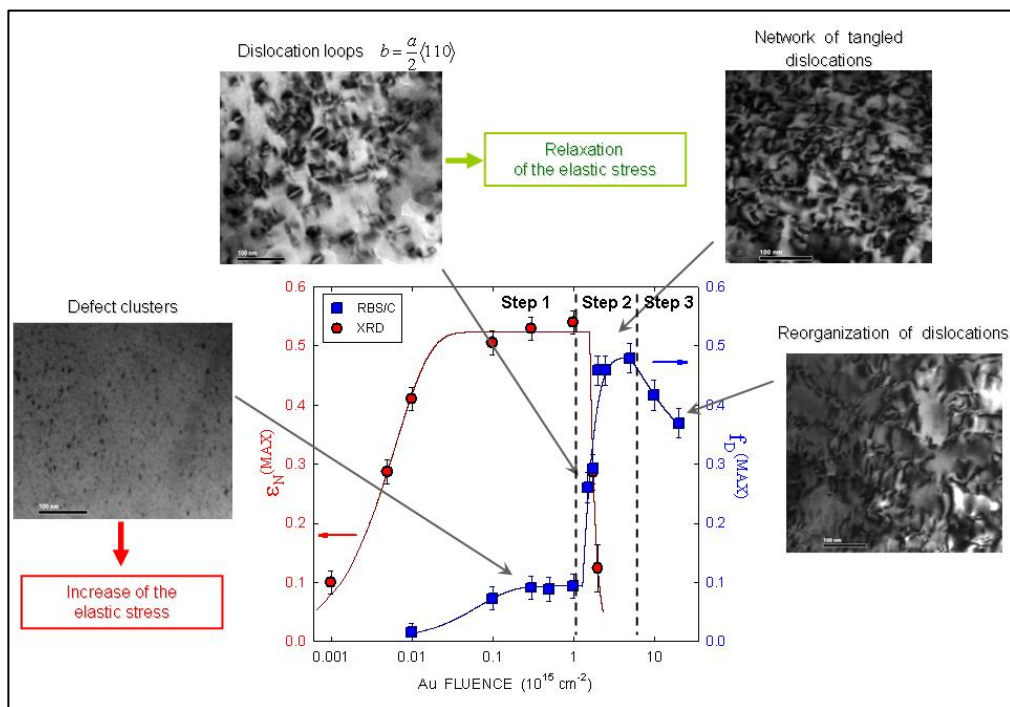


Fig. 1. Buildup of the damage created in YSZ irradiated with 4 MeV Au ions (effect of S_n). RBS/C (squares), XRD (circles) and TEM results. Note the logarithmic scale for the fluence axis. Lines are fits to RBS/C and XRD data according to the MSDA model.

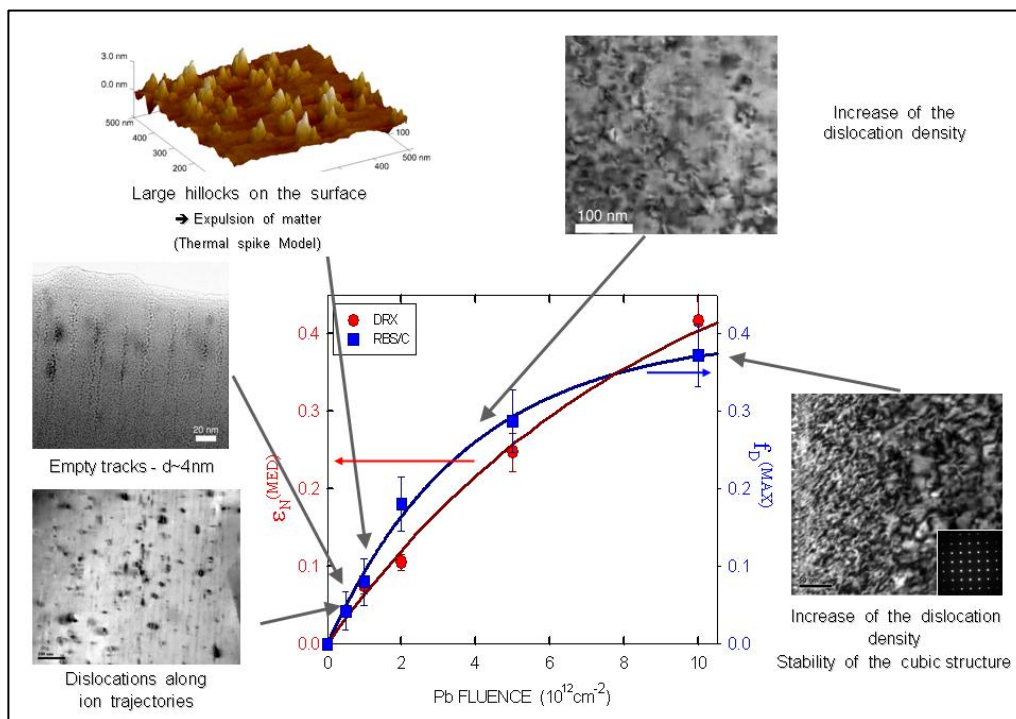


Fig. 2. Buildup of the damage created in YSZ irradiated with 940 MeV Pb ions (effect of S_e). RBS/C (squares), XRD (circles), AFM and TEM results. Lines are fits to RBS/C and XRD data according to the MSDA model