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Complementary use of ERDA and RBS/C for the determination of implanted atom and damage distributions in spinel

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Abstract

Crystalline oxide ceramics (particularly, zirconia and spinel) are promising matrices for nuclear waste immobilization and/or transmutation. The behavior of implanted ions and of radiation damage is a very important issue in the qualification of nuclear matrices. Ion beams provide very efficient tools for such an evaluation. This paper presents the results obtained for spinel single crystals implanted with He and Ar ions at fluences of 10^{17} and $2 \times 10^{16}\ \mathrm{cm}^{-2}$, respectively. He and Ar depth profiles were measured by ERDA using high-energy Cu ions. The Ar depth profile was also determined by RBS. The evaluation of the amount of radiation damage induced in Al, Mg and O sublattices of spinel single crystals by ion implantation was done using both a classical analytical procedure and Monte-Carlo simulations of channeling data. The disorder depth profiles using the two methods are compared and correlated with He and Ar depth profiles.

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1. Introduction

One of today's important challenges in nuclear technology is the reduction of the amount of plutonium and other actinides produced in nuclear

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power plants. A possible way to solve this problem is the incineration of radioactive elements in nuclear reactors. To that purpose their incorporation in inert matrices has been proposed. Nuclear fuel matrices are submitted to a severe radioactive environment due to neutron exposure, gamma and beta irradiations, fission-fragment bombardment and He self irradiation arising from the alpha decay of actinides. Crystalline oxide ceramics, such as zirconia and spinel, were identified as promising matrices for actinide transmutation due to their high melting point, reasonable thermal conductivity and strong resistance to irradiation [1–5].

The final qualification of materials with respect to safety criteria requires an extensive study of their physical and chemical properties. A critical issue is the behavior of He and fission products produced by the disintegration of actinides. It is essential to investigate the influence of various parameters (concentration and location of foreign atoms in the host material, substrate temperature, radiation damage behavior) on the mechanisms of impurity diffusion. The introduction of controlled quantities of atoms in a material can be performed by ion implantation. Whereas the determination of depth profiles of heavy atoms can be easily done by using the RBS technique, problems arise for low and medium mass atoms, i.e. when the RBS peak due to the backscattering of analyzing particles from impurity atoms overlaps with plateaus due to matrix elements. The depth distribution of implanted He atoms is generally determined via NRA. However, it was shown that the ERDA technique using high-energy heavy ions is well adapted to such a task [6,7].

In a recent paper [8] we reported results obtained in the case of spinel single crystals implanted with He ions at several fluences. By using ERDA and RBS/C we were able to correlate He profiles with the damage profiles. In the present work we report new results obtained for spinel single crystals implanted with He ions and with a heavier element, Ar. The objective of this experiment was to extend the range of elements for the study of the correlation between implanted ion and damage depth distributions. The implantation profiles were measured using high-energy ERDA for He and both ERDA and RBS for Ar, whereas the damage induced by implantation was analyzed by RBS/C. To evaluate the radiation damage induced in Al, Mg and O sublattices from channeling data, and thus to obtain damage profiles, we used both a classical procedure based on the two-beam approximation and Monte-Carlo simulations.

2. Experimental

The samples are $\langle 100 \rangle$-oriented magnesium aluminate spinel ($MgAl_2O_4$) single crystals. He and Ar ions were introduced into the spinel samples by 30 and 260 keV ion implantations, respectively, using the IRMA implanter of the CSNSM in Orsay. The ion fluences were of 10^{17} and $2 \times 10^{16}\ cm^{-2}$, respectively. The surface of the samples was covered with a thin ($\sim$15 nm) carbon layer prior to ion beam analysis in order to avoid charging effects.

The amount of damage induced by He and Ar implantation and the Ar depth profiles were measured by Rutherford backscattering and channeling (RBS/C) experiments with the ARAMIS accelerator of the CSNSM in Orsay [9]. A 1.6 MeV ^4He beam was used. The energy resolution of the RBS setup ($\sim$15 keV) corresponds to a depth resolution of $\sim$12 nm.

The ERDA measurements were carried out at the 8.5 MV tandem accelerator of the NIPNE in Bucharest. A 80 MeV ^{63}Cu beam was used. The detection of recoil atoms was made by using a compact ΔE–E telescope, consisting of a ΔE ionization chamber and a residual energy silicon detector [10]. The gains of the amplifiers were adjusted to yield convenient pulse heights for light (H, He) and medium (C, O, Mg, Al) mass elements. The telescope was positioned at 30° with respect to the beam direction. The surface of implanted spinel samples was oriented at 15° with respect to the beam direction. Two energy loss signals and the residual energy signal were fed to three ADCs that are part of a multiparameter acquisition system.

3. Results and discussion

The ΔE–E spectra recorded by ERDA on a spinel crystal implanted with Ar ions at a fluence of