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Ion beam analysis of Cs-implanted zirconia and spinel

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Abstract

Fission products (Cs) were introduced into yttria-stabilized zirconia (YSZ) and magnesium aluminate spinel (MAS) single crystals by room temperature ion implantation. The effect of high-temperature annealing on the depth distribution of implanted species and the surface homogeneity of crystals were investigated by the combination of AFM and RBS using a macro- and a micro-ion beam. The diffusion and release of Cs involve mechanisms which depend on the material and Cs concentration. In YSZ Cs desorbs out of the crystal at lower temperature (~550 °C) than in MAS (~850 °C). In YSZ the surface of the sample remains unaltered when Cs desorption occurs, whereas in MAS Cs desorption is accompanied by the exfoliation of the sample surface.

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1. Introduction

Magnesium aluminate spinel (MAS) and yttria-stabilized zirconia (YSZ) are widely investigated materials for potential use as actinide transmutation matrices [1–3]. Safety considerations require the evaluation of the retention properties of volatile fission products in the selected matrices. Such a study may be performed by introducing the species to be confined via ion implantation and by following the diffusion behavior upon subsequent thermal treatments [4,5]. Rutherford backscattering spectrometry (RBS) and atomic force microscopy (AFM) may then be used to both monitor the depth distribution of implanted elements and characterize the surface homogeneity of annealed samples.

Previous work shows that the determination of the depth profiles of implanted species by conventional RBS experiments using a standard millimeter-sized ion beam (macro-RBS) may lead to mistaken results [6]. This statement is particularly true when the diffusion of these species induces the formation of aggregates. However, this question may be unambiguously addressed by combining surface investigation techniques (such as AFM) and RBS analyses using a micrometer-sized ion beam (micro-RBS). In our case, the depth and lateral distributions of Cs atoms implanted into YSZ and MAS single crystals were accurately measured and the results provide enlightenment about the mechanisms of Cs desorption upon high-temperature treatments.

2. Experimental

The samples are cubic {1 0 0}-oriented YSZ and MAS single crystals. They were implanted at room temperature

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with 300 keV Cs ions at the IRMA facility of the CSNSM in Orsay [7]. Ion fluences ranging from 5×10^{15} to $5\times10^{16}\,cm^{-2}$ lead to average concentrations in the range 1–8 at.%. After implantation the samples were annealed in vacuum for 1 h up to 1000 °C.

The surface of the samples was examined by AFM at the SPCSI in Saclay [8]. The AFM images were acquired in contact mode with a Picoscan+ from Molecular Imaging. The Cs depth profiles were measured by macro-RBS with 1.6 MeV (for MAS) or 3.065 MeV (for YSZ) ^4He beams (size $\sim1\times1\,mm^2$) delivered by the ARAMIS accelerator of the CSNSM in Orsay [7], and by micro-RBS with a 2 MeV ^4He beam (size $\sim3\times3\,\mu m^2$) provided by the Pierre-Süe accelerator in Saclay [9]. In these latter experiments an annular detector was used to collect reasonable charges of backscattered He owing to the small current used ($\sim$1 nA).

3. Results and discussion

Fig. 1(a) and (b) present macro-RBS spectra recorded on YSZ and MAS single crystals implanted with 300 keV Cs ions at $5\times10^{16}\,cm^{-2}$. Note the presence of the resonance peak at 3.04 MeV on ^{16}O (around channel 150) for YSZ analysis. Fig. 1(c) and (d) show the Cs depth distributions obtained (before and after annealing) from the analysis of macro-RBS spectra. The diffusion and desorption of Cs is observed upon annealing at 600 °C in the case of YSZ and at 850 °C in the case of MAS. Previous results (not presented in the figure) show that the Cs profiles are almost unaffected upon annealing up to 1000 °C in both materials when the initial Cs concentration is lower than 2–3 at.% [5,6,10]. The summary of thermal treatments presented in Fig. 2 indicates that annealing results in a sharp drop of

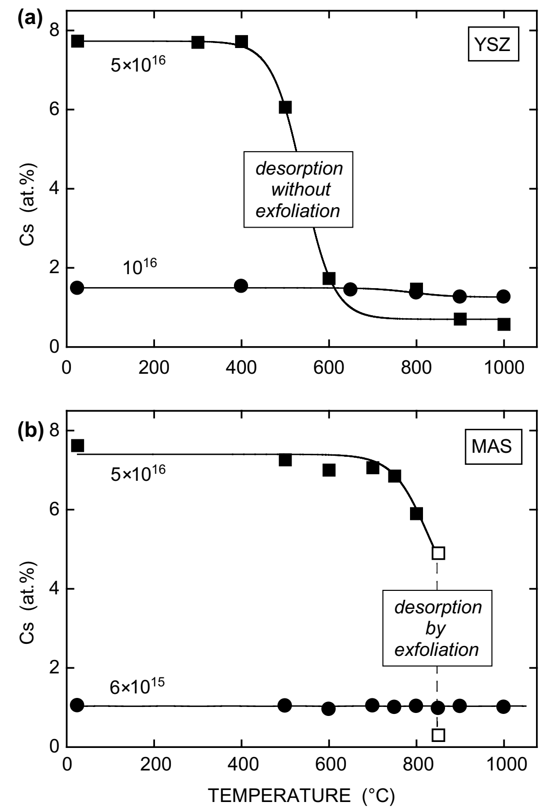

Fig. 2. Cs concentration versus annealing temperature for YSZ (a) and MAS (b) single crystals implanted with Cs ions at the indicated fluences. Filled and open symbols hold for macro- and micro-RBS data, respectively.

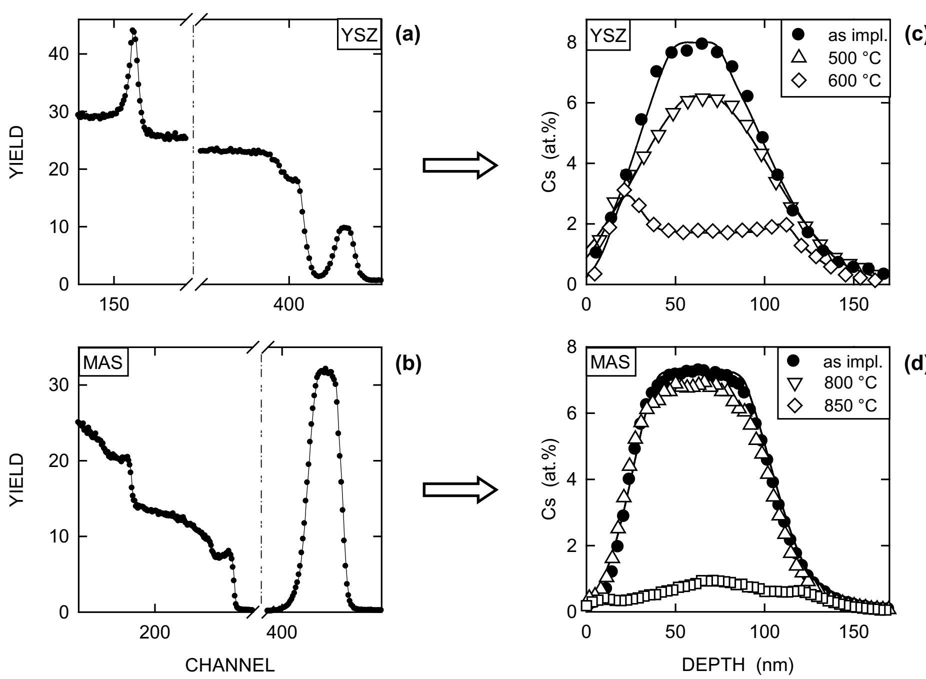

Fig. 1. RBS spectra recorded on YSZ (a) and MAS (b) single crystals implanted with Cs ions at a fluence of $5\times10^{16}\,cm^{-2}$, and depth distributions of Cs ions before and after annealing at the indicated temperatures (c and d).