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Vacancy defects induced in sintered polished UO_2 disks by helium implantation

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Abstract

Vacancy defects have been investigated in sintered polished and annealed uranium oxide disks. Slow positron beam coupled with Doppler broadening spectrometer was used to probe the track region of 1 MeV ^{3}He ions implanted in uranium dioxide (UO_2) disks. The low and high momentum annihilation fractions, *S* and *W*, respectively, were measured in the first micrometer near surface region of the disks as a function of positron energy. The *S* and *W* values indicate that the 1 MeV He ions induce vacancy defects in the track region of their range. The vacancy defect depth distribution is heterogeneous. The positron trapping at these vacancy defects increases with the depth and with the implantation fluence indicating an increase of the vacancy defect concentration. The nature of the induced vacancy defects does not change with the fluence.

Keywords: Slow positron beam; Uranium dioxide; Vacancy defects; Helium implantation; Doppler broadening

1. Introduction

A few experiments have been performed on the behavior of helium gas produced by alpha decay of actinides in spent fuel. Yet, the amount of helium produced after irradiation is large, in particular, in the case of MOX (Pu, U mixed oxides) fuels: in MOX (burnup 47.5 GWatt day/tonnes of Uranium), after a storage of 10,000 years the He amount is evaluated at 4% He at/at$_{iHM}$ (atoms per initial heavy metal atom). In CERI, we are investigating the He migration process, bubble formation and He-vacancy defect interaction using nuclear reaction analysis method to determine the helium depth profile in as-implanted uranium dioxide (UO_2) and positron annihilation spectroscopy to study vacancy defect properties.

The radiation damage in UO_2 has already been studied by using different techniques: Weber has used XRD [1] to measure the lattice parameter change as a function of the alpha decay dose. Matzke and Turos

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have studied the damage induced in UO_2 by heavy ion implantation by using channeling Rutherford backscattering [2]. Soullard has shown with TEM, the formation of interstitial clusters by irradiation with electrons ($E > 1.6$ MeV) at 300 K [3]. While very few results have been published concerning the study of vacancy defects in UO_2 by using positron annihilation spectroscopy [4,5]. The annihilation characteristics (diffusion coefficient, lattice and defect lifetimes) are not known in this UO_2 matrix. This work is a part of the study we have undertaken in CERI to determine these data and the defect properties.

In this work, we have used slow positron beam coupled with Doppler broadening spectroscopy to study the vacancy defects induced in the track region of 1 MeV ^{3}He ions implanted in polished and 1700 °C/24 h/ArH_2 annealed sintered UO_2 disks.

2. Experimental

Up to 15 disks cut from the same set of sintered uranium (0.2 at% ^{235}U) dioxide pellets have been used for this study. After polishing of one side, the disks were annealed at 1700 °C in a wet ArH_2 atmosphere to keep the samples at the stoichiometric composition. The mean grain size is about 18 μm and the mean O/U ratio is 2.0051 ± 0.0001 as determined by polarography. The density of the material is 10.76 ± 0.03 g cm^{-3}. The disks are 300 μm thick and 8.2 mm in diameter.

Ten UO_2 disks have been implanted with 1 MeV ^{3}He ions using the 3.5 MeV Van der Graaff accelerator at CERI Orléans. The implantation is performed by focusing the beam (1 mm × 1 mm) and by sweeping it over the disk surface to ensure a homogeneous fluence (see ref. [6] for details). Seven different fluences have been performed in the range from 2×10^{14} to 1×10^{17} cm^{-2}. For each fluence, two UO_2 disks have been implanted during the same run. The flux were fixed from 1.6×10^{11} ^{3}He cm^{-2} s^{-1} for the implantations at the lowest fluences to 1.7×10^{12} ^{3}He cm^{-2} s^{-1} for the ones at the highest fluences. The temperature was below 80 °C during implantations.

The 1 MeV ^{3}He ion range in UO_2 calculated with SRIM program [7] has a value $R_p = 1.9$ μm. The number of O and U vacancies induced per incident ion and per depth unit is calculated with SRIM as a function of depth, by using as displacement energies in the two sublattices: E_d(U) = 40 eV and E_d(O) = 20 eV [8]. Let us notice that SRIM does not take into account the recombinations that could occur during implantation. In the track region of ^{3}He ions located between surface and approximately 1.5 μm depth, corresponding to the thickness of the ^{3}He implanted UO_2 disk which is probed with the slow positron beam, the total vacancy concentration [V_{tot}] slightly varies as a function of depth (Fig. 1) from 1×10^{21} to 8×10^{21} cm^{-3} for the 1×10^{16} cm^{-2} ^{3}He fluence.

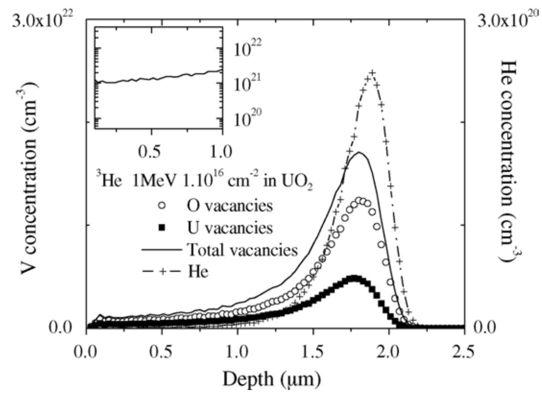

Fig. 1. SRIM calculations of the vacancy and helium concentrations as a function of the depth in 1 MeV ^{3}He implanted UO_2 at the 1×10^{16} cm^{-2} fluence.

The positron–electron pair momentum distribution has been measured at 300 K by recording the Doppler broadening of the 511 keV annihilation line characterized by the low (S) and the high (W) momentum annihilation fraction in the momentum range $(0 - |2.80|) \times 10^{-3}$ and $(|10.61| - |26.35|) \times 10^{-3}$ m_0c, respectively. The energy resolution of the Ge detector is 1.76 keV at 1.28 MeV. To investigate the depth dependence of S and W, the curves $S(E)$ and $W(E)$ were recorded as a function of the positron energy E changed in 0.5 keV steps in the 0.5–25 keV range using the slow positron beam [9] at the CERI laboratory. The positron mean implantation depth in UO_2 varies from approximately 1–670 nm in this energy range [10]. The disk B23 polished and annealed at 1700 °C during 24 h in humid ArH_2 atmosphere has been measured regularly as a reference sample. In this disk, S and W values remain constant between 5 and 25 keV ($S_{B23} = 0.3727(5)$; $W_{B23} = 0.0783(2)$).