Available online at www.sciencedirect.com
ScienceDirect
Journal of Nuclear Materials 357 (2006) 198–205
journal of nuclear materials
www.elsevier.com/locate/jnucmat

A NRA study of temperature and heavy ion irradiation effects on helium migration in sintered uranium dioxide

G. Martin [a], P. Garcia [b,*], H. Labrim [a], T. Sauvage [a], G. Carlot [b], P. Desgardin [a], M.F. Barthe [a], J.P. Piron [b]

[a] Centre d'Etudes et de Recherches par Irradiation, 3A rue de la Férollerie, 45071 Orléans cedex 2, France
[b] CEA, CE de Cadarache, DEN/DEC/SESC/LLCC, Bât. 151, 13108 Saint-Paul-lez-Durance cedex, France

Received 21 December 2005; accepted 23 June 2006

Abstract

Helium implanted uranium dioxide sintered samples were studied using nuclear reaction analysis prior to and following heavy ion irradiations and temperature anneals at 800 °C and 1100 °C. The results show that the heavy ion irradiations do not produce measurable long range movement of helium atoms. However, the ion irradiations do affect the behaviour of helium during subsequent temperature anneals. As regards the 800 °C anneal, the reduced mobility of helium in the ion-irradiated samples is interpreted as resulting from enhanced helium atom segregation produced by the ion-irradiation. Conversely at 1100 °C, the initial heavy ion irradiation appears to produce a greater than expected movement of helium within the bulk of the sample which could be an indication of defect assisted helium diffusion. Thermal diffusion coefficients are also reported at 800 °C and 1100 °C based on an analysis using a one-dimensional diffusion model.

PACS: 61.82.Ms; 66.30.Jt; 66.30.Xj

1. Introduction

The behaviour of fission gases Xe and Kr produced in nuclear oxide fuels has been extensively studied due to their potential effects on the structural and mechanical properties of fuel elements (see for instance [1] or [2]). Helium is another rare gas produced in-pile. Moreover, a substantial quantity is expected to form under long term storage conditions as a result of the alpha decay of certain actinides. To help predict the evolution of irradiated oxide fuel, the behaviour of helium implanted in sintered uranium dioxide (UO_2) disks was looked at in previous studies [3–5]. A NRA (nuclear reaction analysis) technique using the $^3He(d,\alpha)^1H$ reaction enabled the study of ^3He depth profile changes in UO_2 sintered samples at various annealing stages. Results showed that helium is mobile in implanted polycrystalline samples at temperatures as low as 600 °C. He bubble precipitation was then found to

* Corresponding author. Tel.: +33 4 42 25 41 88; fax: +33 4 42 25 13 37.
E-mail address: garciap@drncad.cea.fr (P. Garcia).

0022-3115/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2006.06.021

predominate up to 1000 °C. At 1100 °C however, He release resumed unhindered [5]. Large helium release fractions were also observed in neutron irradiated oxide fuels at or around this threshold temperature and above [6].

However, fission fragments produced in-pile or alpha particles produced as a result of the decay of actinides during the storage of used fuels are likely to impact the behaviour of helium. These effects are studied in the work presented here. In particular, we wish to ascertain whether electronic energy loss mechanisms alone are capable of inducing athermal, long range movement of He atoms. To this end, ^{3}He was implanted at two different depths, 0.3 μm and 2 μm. Samples were irradiated with high energy heavy ions in order to produce defects essentially through electronic energy loss mechanisms at depths of a few micrometers from the samples' surface. The ^{3}He(d,α)^{1}H nuclear reaction analysis method was used to determine helium depth profiles before and after annealing at 800 °C and 1100 °C. The He depth profile changes measured as a function of the annealing temperature and heavy ion irradiation conditions are compared and discussed.

2. Experimental

2.1. Sample preparation and helium implantations

Four sintered uranium dioxide disks were polished on one side. The thickness of the disks was approximately 300 μm and their diameter 8.2 mm. After polishing, the disks were annealed at 1700 °C in a H_2 atmosphere (1.7 vol.% H_2O) for 24 h to remove the damage caused by polishing and insure that the samples were of stoichiometric composition. The average grain size following the 1700 °C anneal was estimated at approximately 24 μm. Two of the four UO_2 disks (G1, G2) were implanted simultaneously at room temperature with 0.1 MeV $^3He^+$ ions to a fluence of 0.56×10^{16} at. cm^{-2} using the 400 kV implanter at CSNSM Orsay. Both other disks (G3, G4) were implanted to a fluence of 1.7×10^{16} at. cm^{-2} with 1 MeV $^3He^+$ ions using the 3.5 MV Van de Graaff accelerator at CERI Orléans. Fluences were chosen to obtain similar helium concentrations (0.3 at.%) at the depth profile peak. Details concerning polycrystalline sample E4 allude to in Section 3.4 can be found in Ref. [5].

2.2. Heavy ion irradiations

Only two samples were irradiated at room temperature with heavy ions, G2 and G4. G2 was irradiated to a fluence of 0.13×10^{16} at. cm^{-2} with 220 MeV $^{79}Br^{12+}$ ions and G4 was irradiated to a fluence of 0.14×10^{16} at. cm^{-2} with 250 MeV $^{127}I^{14+}$ ions. Both ion irradiations were performed at the IRES Vivitron accelerator in Strasbourg. Of course, these energies are greater than fission fragment energies, typically around 120 MeV (respectively 70 MeV) for the lighter (respectively heavier) of the two fission fragments. However, the energy loss corresponding to the bromine irradiation in particular is very similar to that of fission fragments (typically 20 keV/nm) as can be seen from Table 1. The electronic energy loss associated with the iodine irradiation is somewhat greater than that of typical fission fragments, but these irradiation conditions were chosen so as to exacerbate effects associated with electronic excitation.

2.3. Annealing conditions

All the samples were annealed twice under reducing atmospheres (Ar, 10 vol.% H_2) at 800 °C

Table 1
Average electronic and nuclear energy losses in MeV μm^{-1} relative to initial helium implantations and heavy ion irradiations

		Depth x		
		$x < 1$ μm	1 μm $< x <$ 2.2 μm	$x >$ 2.2 μm
Helium 0.1 MeV	dE/dx el.	0.2	–	
	dE/dx nucl.	0.007	–	
Bromine 220 MeV	dE/dx el.	22	21	9
	dE/dx nucl.	0.04	0.04	0.5
Helium 1 MeV	dE/dx el.	0.6	0.3	–
	dE/dx nucl.	0.001	0.005	–
Iodine 250 MeV	dE/dx el.	32	30	15
	dE/dx nucl.	0.11	0.13	0.52