Nuclear Inst, and Methods in Physics Research B 414 (2018) 141–145

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Studies of O^{18} impurity trapping at interstitial dislocation loops in ion implanted Fe (1 1 0) by ion channeling and *ab initio* calculations

Vairavel Mathayan[a,b], Saravanan Kothalamuthu[a], Jaiganesh Gnanasekaran[a], Sundaravel Balakrishnan[a,b,*], Binaykumar Panigrahi[a,b]

[a] Materials Science Group, Indira Gandhi Centre for Atomic Research, Kalpakkam 603102, India
[b] HBNI, Kalpakkam 603102, India

ARTICLE INFO

Keywords:
Ion channeling
Radiation damage
Density functional theory
Lattice location
Dislocation loop

ABSTRACT

The O^{18} and self ions are implanted at same depth in Fe (1 1 0) crystal and annealed to study the oxygen trapping under excess self interstitial defects. Rutherford backscattering spectrometry, nuclear reaction analysis and channeling measurements have been performed to determine the lattice site position of O^{18}. The presence of dislocation loops is confirmed by energy-dependent dechanneling parameter measurements. From the tilt angular scans of Fe and O^{18} signals along ⟨1 0 0⟩, ⟨1 1 0⟩ axes, O^{18} is found to be displaced 0.2 Å from tetrahedral towards octahedral interstitial site in O^{18}. Similar lattice site location of oxygen with the displacement of 0.37 Å is predicted by density functional theory calculations for the interaction of oxygen with ⟨1 0 0⟩ interstitial dislocation loop structure. Our results provide strong evidence on oxygen trapping at interstitial dislocation loops in the presence of excess interstitial defects in iron.

1. Introduction

Body centered cubic Iron and Iron-chromium alloys are the model systems for ferritic/martensitic steels which are proposed to be one of the structural materials for future nuclear reactors. In nuclear reactors, the high-energy neutrons damage the structural materials. Vacancies and interstitials are primary defects which are formed during operation, which changes the microstructure of structural materials [1]. In addition to vacancy and self-interstitial defects, impurities present in the materials change the radiation-induced microstructure due to the interactions with vacancy/interstitial defects. For example, the accumulation of He atoms acts as nucleation sites for void formation in ferritic steels [2].

The presence of impurities also significantly influences the mechanical properties of structural materials under radiation damage. The increase of radiation hardening with increasing N impurity content in mild steels has been reported [3]. The interactions of C and N impurities with radiation-induced defects have been reported to improve the mechanical properties of mild steels at elevated temperatures [4]. The interstitial impurities (C, N) present in low-carbon ferritic steels are found to trap at dislocation defects which strongly influence the mechanical properties [5,6]. Being an interstitial impurity atom, oxygen plays a vital role in the radiation-induced microstructural evolution and corrosion of ferritic steels [7]. However, the direct experimental evidence on oxygen lattice location in the presence of defects in iron is scarce. In this study, the lattice site of oxygen upon self-ion implantation is examined by ion channeling experiments to understand the oxygen interaction better under the radiation damage.

2. Methodology

In this study, bcc Fe (1 1 0) crystal with a chemical purity of 99.98% is implanted with 300 keV O^{18} ions at a fluence of 5×10^{15} ions/cm^2 at room temperature followed by in-situ annealing in the vacuum of 2×10^{-7} mbar at 400 °C for 30 min. The projected range and straggling of 300 keV O^{18} in Fe is 275 nm and 78 nm respectively, which are obtained from Monte-Carlo simulation program SRIM [8]. The SRIM simulated ion and vacancy profiles for 300 keV O in Fe shown in Fig. 1. With the aim of introducing excess interstitials to overlap with oxygen depth profile, 750 keV Fe^+ ions with a projected range of 266 nm are introduced by subsequent implantation in Fe (1 1 0) at a fluence of 1×10^{16} ions/cm^2 and annealed in-situ at 400 °C for 30 min.

Lattice site location of oxygen atoms in O^{18} and 750 keV Fe^+ ion implanted and annealed Fe(1 1 0) crystal is studied with RBS and NRA/channeling measurements. The Fe(1 1 0) matrix signal is obtained by RBS whereas O^{18} signal is measured with O^{18} (p,α) N^{15} nuclear reaction by using 850 keV proton beam. The experimental tilt angular scans of O^{18} and Fe are simulated by FLUX7 program [9]. More experimental

* Corresponding author at: Materials Science Group, Indira Gandhi Centre for Atomic Research, Kalpakkam 603102, India.
E-mail address: bsundar@igcar.gov.in (S. Balakrishnan).

https://doi.org/10.1016/j.nimb.2017.11.021
Received 28 September 2017; Received in revised form 25 October 2017; Accepted 18 November 2017
Available online 23 November 2017
0168-583X/

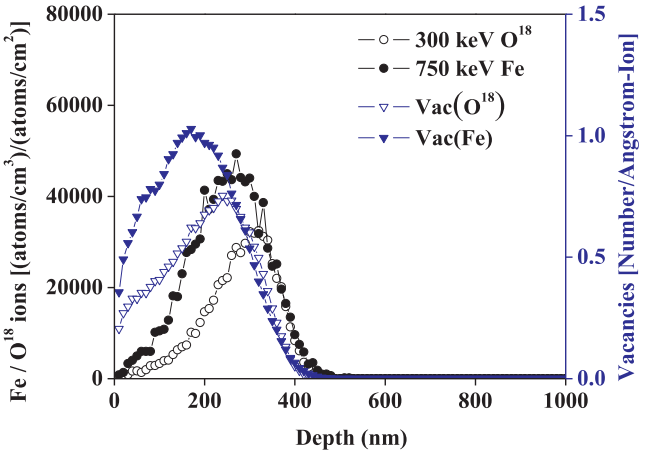

Fig. 1. SRIM calculated range of ions and vacancies in Fe crystal produced by 750 keV Fe ions and 300 keV O^{18} ions.

details could be found in reference [10]. During the fitting of simulated scans with experiment, the calculated O^{18} yield is corrected for additional random fraction of incident beam [11], which is given by,

$$\chi_O^{corr}(\psi) = (1-f_R)\chi_O^{calc}(\psi) + f_R \tag{1}$$

$$f_R = \frac{\chi_{Fe}^{exp}(0)-\chi_{Fe}^{calc}(0)}{1-\chi_{Fe}^{calc}(0)} \tag{2}$$

where $\chi_O^{corr}(\psi)$ is random fraction corrected simulated O^{18} yield used for fitting experimental O^{18} yield, is the calculated O^{18} yield, ψ is the angle of incidence, f_R is the fraction of channeled beam that has become dechanneled due to surface Fe defects, $\chi_{Fe}^{exp}(0)$, $\chi_{Fe}^{calc}(0)$ are the experimental and theoretical yield of Fe in the aligned direction.

The energy dependence of dechanneling parameter is measured to investigate the type of defects formed at the O^{18} ion implanted depth. For this, RBS spectra are taken with He^+ ion with the energy ranges from 1100 keV to 3500 keV along the normal axis (⟨1 1 0⟩). Random and aligned RBS spectra are simulated using FLUX7 program for all incident energies and converted to corresponding spectra to yield versus depth, from which variation of χ_{min} with depth for pristine (χ_V) and ion implanted and annealed (χ_D) samples are obtained. The type of defects at a particular depth can be determined by energy dependence of the dechanneling probability (*DP*) [12] which is given by,

$$DP = \left[-\ln\frac{1-\chi_D}{1-\chi_V}\right] \tag{3}$$

The point defects, stacking faults and dislocations show energy dependence of $E^{-0.5}$, E^0, $E^{0.5}$ respectively.

In order to understand our experimental observations we employ the *ab initio* calculations for finding the oxygen lattice location in the presence of self-interstitial defect structures in bcc Fe using the Vienna Ab-initio simulation package [13–15]. We have used projector augmented wave (PAW) pseudopotentials approach. Generalized gradient approximation based Perdew–Burke–Ernzerhof (PBE) exchange-correlation functional is used [16]. The cutoff energy in the plane-wave basis set is 500 eV which arrived after performing the required convergence tests. Following the Monkhorst-pack scheme [17], Brillouin zone integration is carried at 3 × 3 × 3 *k* point grids. A tolerance of 10^{-6} eV in the total energy is used for the self-consistency convergence criteria. Vosko et al. [18] spin-interpolation scheme is used for the spin-polarized calculations in this system. For all defect studies, we performed constant volume (*CV*) calculations with (128 + defects) atom cell. The binding energy is defined as [19],

$$E_b^{a,b} = E_{defect\text{-}a} + E_{defect\text{-}b} - E_{defect\text{-}(a+b)} - E_{Fe} \tag{4}$$

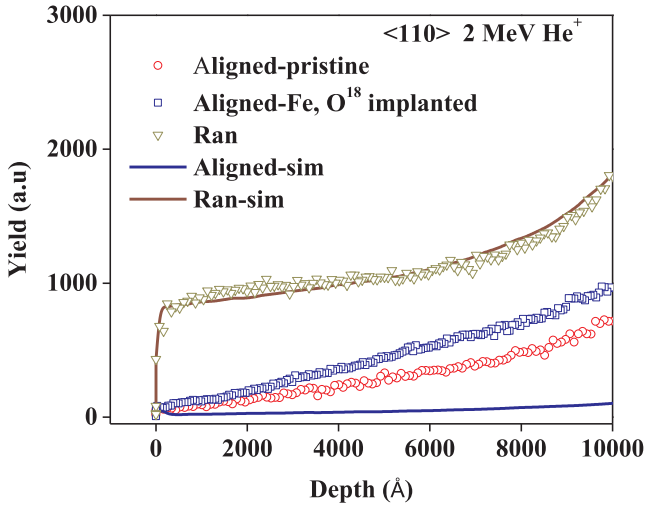

Fig. 2. RBS spectra taken with 2 MeV He^+ ions in pristine Fe(1 1 0) and O^{18} and Fe^+ ion implanted Fe (1 1 0) along random and ⟨1 1 0⟩ orientations, together with FLUX7-simulated random spectrum and ⟨1 1 0⟩ channeling spectrum for ideal Fe (1 1 0).

where $E_{defect\text{-}a}$, $E_{defect\text{-}b}$ and E_{Fe} are the energy of cell containing defect-a, defect-b and defect-free lattice. In all the discussions positive binding energy indicates energy lowering attraction of defects.

3. Results and discussions

To check the crystallinity of O^{18} and Fe ion implanted and annealed Fe(1 1 0), RBS and channeling measurements are carried out with 2 MeV He^+ ions. Fig. 2 shows the RBS spectra after depth conversion using the FLUX7 program from pristine Fe(1 1 0) and ion implanted and annealed Fe(1 1 0) along with random and aligned directions. The random and channeling spectra for ideal Fe(1 1 0) crystal, simulated using FLUX7, are also shown in Fig. 2. The value $\chi_{min} = Y_{aligned}/Y_{random}$ taken in a near-surface region of RBS scan is the measure of crystalline quality [12]. The measured χ_{min} at a depth interval of 100–110 nm is 10% for the virgin sample; it increased to 15% after O^{18} and Fe ion implantation followed by annealing, which indicates that defects are not annealed out completely. The slope of the channeling spectra increases with depth in the implanted and annealed sample due to dechanneling by defects.

The experimental and simulated tilt angular scans of Fe and O^{18} yield along ⟨1 1 0⟩ normal and ⟨1 0 0⟩ off-normal axes, performed in ion-implanted and annealed Fe (1 1 0) are presented in Fig. 3(a) and (b) respectively. Experimental χ_{min} of Fe tilt angular scan is 0.13, 0.3 along ⟨1 1 0⟩, ⟨1 0 0⟩ respectively. The channeling half width ($\psi_{1/2}$) of the experimental scan along ⟨1 1 0⟩ axis is 0.6° which is comparable to the simulated value of 0.67°. In the tilt angular scan of ⟨1 0 0⟩ off-normal axis, axial dip of Fe signal is shallower and narrower. There is also some asymmetry, which is due to the appearance of a nearby planar channeling dip, causing lower yield in one side. This can cause a small reduction in the measured tilt angular width. Dechanneling by defects in the off-normal axis can also cause the reduction in tilt angular width. Limitations in the alignment of the crystal along off-normal axis can also cause a shallow dip and a similar behaviour is observed in our earlier work without self ion implantation [10]. If experiment is carried out in Fe crystal with ⟨1 0 0⟩ axis along normal, a better shape can be obtained. Small humps in either side of the central peak are observed in experimental tilt angular scan of O^{18} signal along ⟨1 0 0⟩. The experimental tilt angular scans from the O^{18} signal is simulated using the FLUX7 software. While fitting the experimental data with the simulated data, the random fraction (f_R) due to the dechanneling of ions from Fe matrix is also taken into account as in Eq. (1). The calculated f_R using Eq. (2) is 0.1, 0.28 for ⟨1 1 0⟩, ⟨1 0 0⟩ axis respectively. The narrowing