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Vacancy type defect formation in irradiated α -iron investigated by positron beam Doppler broadening technique

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Abstract. Vacancy type defects formations have been investigated in virgin and irradiated α -iron samples using slow positron beam Doppler broadening technique. Mono-vacancies and vacancy clusters are observed in 1.5 MeV ^4He ions irradiated Fe samples at varying fluences from 1×10^{13} to $1 \times 10^{17} \text{ cm}^{-2}$. In the 1.2 MeV Yttrium ions implantation at low fluence $1 \times 10^{14} \text{ cm}^{-2}$, vacancy clusters with higher concentration and larger size are formed. In this sample, vacancy defects are detected deeper than predicted by SRIM calculation due to channelling.

1. Introduction

Iron based alloys, and for example, Oxide Dispersion Strengthened (ODS) ferritic steels, constitute good candidates for structural components in fission reactors or future fusion reactors which are submitted to extreme conditions of irradiation and temperatures. Changes in microstructure and macroscopic properties of these nuclear materials are governed by the kinetics of defects produced by irradiation, such as vacancy-type clusters or vacancy-impurity complex clusters. In ODS steels, the Yttrium, Titanium oxide nanoparticles are traps for He and pin dislocations allowing to limit the swelling and creep. These materials are fabricated by mechanical alloying and their interesting properties are very dependent of the nanoparticles distribution in size and in space. Fundamental studies are needed to improve the control of this distribution. One of the key-point in the understanding of the formation of these particles is the role of vacancy defects induced during milling. As a key component of these materials, the understanding of defect formation in irradiated bcc iron (α -Fe) is essential. Many efforts, both experimental and computational works, have been put into this study. Some are carried out by using positron annihilation spectroscopy as its sensitivity to the size and density of vacancy clusters in the range from mono-vacancies up to cavities containing 50-100 vacancies [1]. Recently T. Iwai et al. [2, 3] have studied the damage induced in iron by various ion species (He, C, O, Fe) by using positron beam Doppler broadening. Mono-vacancies and vacancy clusters have been observed but characteristics S/W of positron annihilation in these defects have not been defined which are important to discriminate between the different vacancy types.

In this study, the vacancy-type defects induced by 1.5 MeV ^4He ions at varying fluences in α -Fe are investigated with slow positron beam Doppler broadening (SPBDB) technique at the CEMHTI laboratory. It allowed defining the annihilation characteristics S and W for mono-vacancy and vacancy clusters. Then the defects produced by 1.2 MeV Yttrium implantation in α -Fe have also been studied. This will be the basis for our study on the interaction of vacancy clusters with Y.



2. Experimental

Samples are 99.99% pure iron 7 mm×7 mm×0.5 mm sheets. After polishing, the samples were annealed at 800°C for one hour in vacuum. The grain size is about 20-100 μm . 1.5 MeV ^4He irradiations were carried out with the 3.5 MeV Van der Graaff accelerator at CEMHTI Orleans. Eight different fluences have been performed in the range from 1×10^{13} to 1×10^{17} cm^{-2} . 1.2 MeV Yttrium implantations were done with the 3 MeV Tandetron accelerator at laboratory HZDR Dresden at the fluence of 1×10^{14} cm^{-2} producing a similar order of magnitude of damage dose compared to 1.5 MeV ^4He irradiation at fluence 1×10^{17} cm^{-2} . Both these two types of irradiations were carried out at room temperature.

The momentum distribution of electron- positron pairs 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 $(-2.49, 2.49)\times 10^{-3}$ and $(-24.88, -9.64)$, $(9.64, 24.88)\times 10^{-3}m_0c$, respectively. To investigate the depth dependence of S and W, the S(E) and W(E) were measured as a function of the positron energy E changed in 500 eV steps in the 0.5 - 25 keV range using the slow positron beam. For this energy range, the maximum positron penetration depth in pure Fe is at most ~ 2 μm .

Figure 1 shows the depth profiles of Y and He concentrations and the corresponding damage calculated by using SRIM 2008 [4] for pure Fe irradiated by 1.5 MeV ^4He and 1.2 MeV Y. In these calculations, the Fe displacement threshold energy has been fixed at a value of 40eV [5]. In the same figure, the slow positron implantation profile is also illustrated for different energies [6]. In the track region of ^4He ions located between surface and 2 μm which is probed with the slow positron beam, the damage dose is 0.1 dpa (displacement per atom) and the helium concentration is 4 atomic ppm for the highest fluence 10^{17} cm^{-2} . This concentration is quite low, thus the effect of helium on vacancy defects could be ignored in the following. 1.2 MeV Y ions are implanted between surface and 500 nm in Fe. The projected range (R_p) is 265 nm. Thus, the positrons could detect the entire implanted region. At fluence 1×10^{14} cm^{-2} , the damage dose is 0.14 dpa at the surface and increases up to reach 0.24 dpa at the depth of 170 nm and the Y concentration reaches the maximum value of 58.9 appm at the depth of 270 nm. This concentration is lower than yttrium solubility in iron (290 at. ppm in iron at 800°C [7]). The profile of Y concentration has also been calculated using Marlowe code considering that sample is a polycrystal. It suggests that Y ions stop deeper than R_p up to ~ 1500 nm.

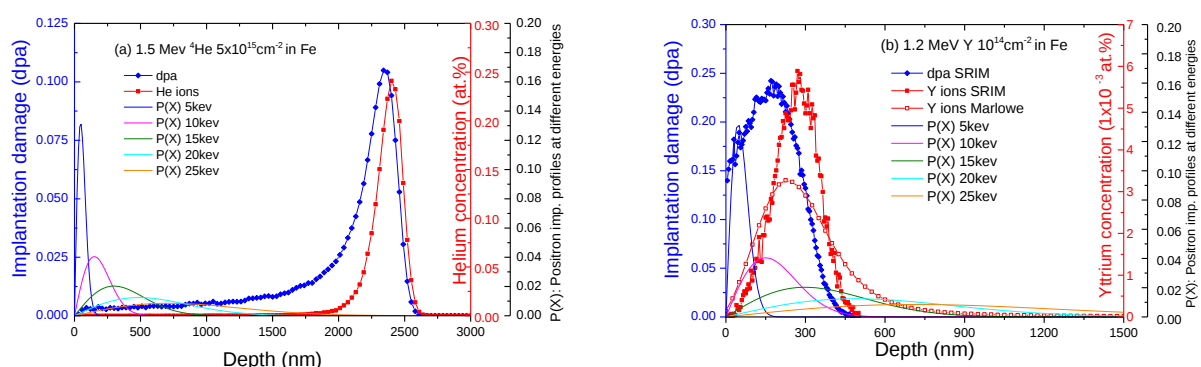


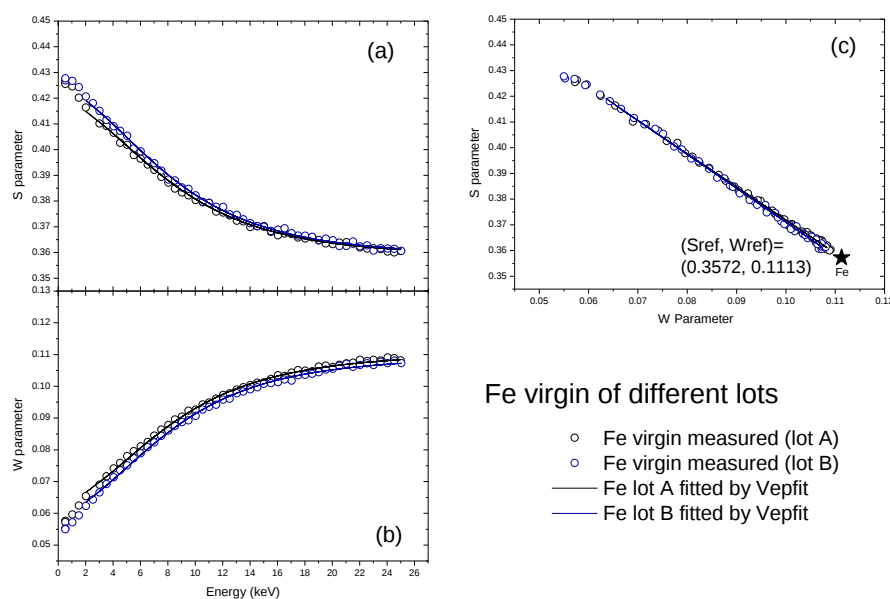
Figure 1. Depth profile of defect production and atomic concentration (a) 1.5 MeV ^4He in Fe (b) 1.2 MeV Y in Fe.

3. Results and discussion

3.1. Virgin iron samples

All as-received polished and 800°C/1 h/vacuum annealed Fe samples have given SPBDB reproducible results and a typical representation is illustrated for two samples in Fig 2. S (respectively, W)

decreases slowly (increases) as a function of positron energy. This slow evolution indicates that a large fraction of positrons diffuses back to the sample surface at intermediate positron energy and annihilation occurs both at surface and in the bulk of sample. The adjustment of experimental results with VEPFIT [8] program using a simple assumption of one homogeneous layer reveals an effective positron diffusion length $L^+ = 160 \pm 2$ nm in the bulk. The positron diffusion lengths reported for defect-free metals are ~ 100 nm [9]. Higher L^+ value of 184 nm, has been obtained by Iwai and *al.*, for pure Ni (99,997 wt.%) annealed at 800°C in vacuum for one hour [10]. All these results reveal that the quantity of open-volume defects in the bulk of the virgin samples is low. The annihilation characteristics of the Fe lattice considered as the reference values in this work are: $S_{\text{ref}} = 0.357(2)$ and $W_{\text{ref}} = 0.111(3)$.



Fe virgin of different lots

- Fe virgin measured (lot A)
- Fe virgin measured (lot B)
- Fe lot A fitted by Vepfit
- Fe lot B fitted by Vepfit

Figure 2. Low (S) (a) and high (W) (b) momentum fraction as a function of positron energy and S as a function of W (c) for the virgin pure Fe samples. VEPFIT is used to fit the (S , W) curves with a model of one homogeneous layer.

3.2. 1.5 MeV ^4He irradiated iron samples

Figure 3(a) shows low momentum fraction S as a function of positron energy for pure Fe samples irradiated with 1.5 MeV ^4He at fluences from 1×10^{13} to $1 \times 10^{17} \text{ cm}^{-2}$ (10^{-5} to 10^{-1} dpa) and for Fe virgin sample. The S and W positron annihilation characteristics in the ^4He irradiated Fe samples differ from the ones measured in the virgin Fe samples. Whatever the fluence, all the $S(E)$ values are higher in the irradiated samples than in the virgin ones when all the $W(E)$ values are, respectively, lower. This indicates that ^4He ions have induced vacancy-related defects in their track region. The general look of $W(E)$ curves is reverse to that of $S(E)$. $S(E)$ curves depend on the fluence. For low fluences from 1×10^{13} to $5 \times 10^{14} \text{ cm}^{-2}$, the curves suggest the diffusion back of positrons to the surface and a large fraction of annihilation in the surface states. For higher fluences, diffusion back to the surface becomes negligible. From approximately 22.5 keV, the $S(E)$ increases as a function of E for low fluences and decreases for the highest ones. In Figure 1.a, we can see that 25 keV positrons probe a region where the damage dose and the He concentration begin to increase because of the accumulation of cascades. $S(E)$ increases with the damage dose. But when the fluence becomes equal or higher than $1 \times 10^{16} \text{ cm}^{-2}$, it decreases. At this fluence the He concentration becomes non negligible in the region probed by high energy positrons. He can be trapped in vacancies and it leads to a change of electron

density and momentum at the trap [11]. This effect of helium is negligible at low fluences when the He concentration is very low. In the following we choose to consider the S and W values measured in the energy range between 20 and 22 keV as the annihilation characteristics in the track region of the ions. In this range S(E) changes very slightly, the diffusion back of positron to surface and the annihilation fraction in cascade region are negligible. In this energy range S increases with the fluence (see arrow in Fig 3.a). It indicates that damage increases when the irradiation fluence increases in the pure Fe samples.

The mean values of S and W estimated in this energy range will be called S and W respectively to lighten the text. The evolution of S as a function of W is plotted on Figure 3(b). At low fluence from 1×10^{13} to $2 \times 10^{14} \text{ cm}^{-2}$ (from 1×10^{-5} to $2 \times 10^{-4} \text{ dpa}$), the S,W points follow the same straight line D1 which goes through the (S_{ref} , W_{ref}) point. It indicates that the same nature of vacancy defects has been created for these fluences. In these conditions of irradiation, SRIM predicts that the energy of the Primary Knocked-on Atoms (PKA) is low (91.8 eV) so the recoiled atoms collisions should lead to the formation of small cascades with essentially single vacancies as molecular dynamics modelling predicts [12]. Thus, we will propose that this D1 line is more likely characteristic of annihilation in single vacancy in Fe.

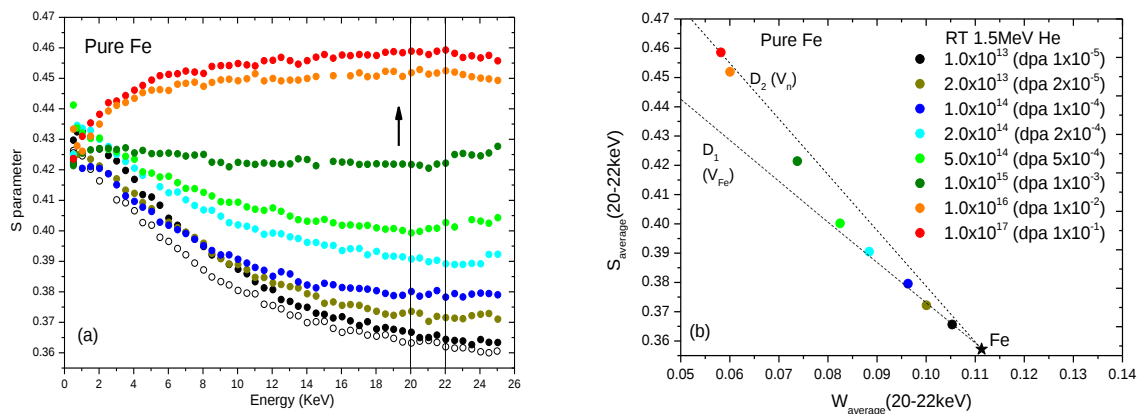


Figure 3. Low (S) momentum fraction as a function of positron energy (a) and S as a function of W obtained by calculating the average value between positron energy 20 and 22 keV (b) in Fe samples before irradiation and after 1.5 MeV ^4He irradiation at different fluences in the range from 1.0×10^{13} to $1.0 \times 10^{17} \text{ cm}^{-2}$.

From the fluence of $5 \times 10^{14} \text{ cm}^{-2}$ ($5 \times 10^{-4} \text{ dpa}$), the S,W points begin to leave the single vacancy line. This indicates that the nature of vacancy defects has changed and larger vacancy-type defects are detected. When fluence increases, the shift from D1 characteristic line increases indicating that the size of vacancy clusters grows with the damage dose. Such clusters formation could be explained by the migration of single vacancies generated by irradiation that could agglomerate leading to the formation of larger clusters when their concentration increases that means with the fluence. Indeed, migration temperature of V in Fe is below the room temperature (-53 to 5°C) as suggested by Takaki *et al.* [14]. T. Iwai *et al.* [2] investigate accumulation of vacancy-type defects in iron produced by 1 MeV He irradiations at room temperature by positron beam both in-situ during irradiations and after irradiations. Vacancy clusters have been observed from $\sim 1 \times 10^{-2}$ to 0.1 dpa and the results suggest the S values are not likely to change under and after irradiation. Thus, the vacancy clusters in Fe by irradiation with ^4He 1.5 MeV (this study) could be considered to form during the irradiation by migration and agglomeration. Between $1 \times 10^{16} \text{ cm}^{-2}$ and the highest fluence of $1 \times 10^{17} \text{ cm}^{-2}$ the S and W values don't change too much indicating that saturation occurs. This saturation could correspond to a maximum size of clusters that can be formed in these irradiation conditions. We will define the straight line going from lattice S,W point and S,W measured in sample implanted at the highest

fluence as D2 characteristic of annihilation of positrons trapped in vacancy clusters.

Being given the migration energy of the single vacancies it is therefore unlikely that they survive in the irradiated sample at room temperature. They should either disappear by diffusion into sinks or agglomerate with other vacancies into clusters or form pairs with some impurities which stabilizes them. It has to be noted, the carbon concentration in these iron samples is about 56 appm. They could be bound to single vacancies and stabilize them as it has been shown in [13]. It suggests that D1 could be characteristic of the detection of single vacancies as proposed above.

3.3. 1.2 MeV Yttrium implanted iron samples

As shown in Figure 4(a), $S(E)$ increases fast from 0.5 to 4 keV and reaches a plateau value from 4 to 15 keV. From 15 keV, $S(E)$ begins to decrease. Figure 4(b) shows, from 0.5 to 4 keV, the corresponding S, W points go towards the D2 line which indicates vacancy clusters are formed in this region. S, W points remain superposed from 4 to 15 keV which means the size and the concentration of vacancy clusters do not change or that positrons are not able to detect this change. In the last positron energy range from 15 keV, S, W points evolve along the D2 line towards the $(S_{\text{ref}}, W_{\text{ref}})$ point. It indicates that the size of defects does not change but their concentration decreases in the probed depth range.

Compared to the result of 1.5 MeV ^4He irradiated at highest fluence $1.0 \times 10^{17} \text{ cm}^{-2}$ Fe sample, the $S(E)$ values of 1.2 MeV Yttrium implanted Fe are much higher and S, W points are further from the $(S_{\text{ref}}, W_{\text{ref}})$ point and slightly above the D2 line. It indicates that the vacancy cluster concentration is much higher in Y implanted sample. It could be explained by the higher damage dose accumulated with Y irradiation 0.242 dpa compared to 0.1 dpa for sample irradiated with ^4He at the highest fluence. Moreover their size is slightly larger. It could be due to the difference in PKA (Primary Knock-on Atom) energies for both projectile ions. Heavier ions produce more PKAs with high energy than lighter ions. PKAs which can induce collision cascades in which the displacement local density depends on their energy. Half of PKAs produced by 1.5 MeV ^4He in Fe have energies lower than 91.8 eV whereas this energy is 232 eV for 1.2 MeV Y. Molecular dynamics simulations show PKA with higher energy produce larger vacancy clusters [15].

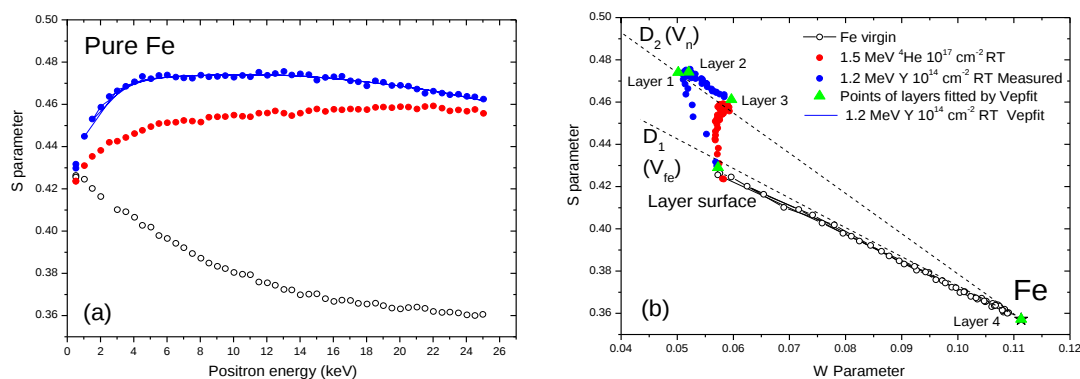


Figure 4. Low (S) momentum fraction as a function of positron energy (a) and S as a function of W (b) in Fe samples before implantation and after 1.2 MeV ^+Y implantation at fluence $1.0 \times 10^{14} \text{ cm}^{-2}$.

Another observation has attracted our attention. The $S(E)$ curve of 1.2 MeV ^+Y implantation begins to decrease from 15 keV instead of 10 keV energy from which a fraction of positron should annihilate in the defect-free Fe lattice as suggested by the depth damage profile calculated by SRIM (Figure 1.b). It indicates that positrons detect vacancy clusters deeper than expected by SRIM. A structure of four layers allows to fit the $S(E)$ and $W(E)$ experimental curves with Vepfit. The S and W values of each of these layers are plotted in Figure 4 and indicate that vacancy clusters are

detected in the first three layers. The first layer spreads from surface to 57.5 ± 6.4 nm, the second spreads up to 609.4 ± 11.3 nm and the third stops at the depth of 1661.80 ± 25.5 nm. For these three layers L^+ changes from 10 to 15 nm. In the fourth layer the S, W and L^+ correspond to the one of Fe lattice. So vacancy clusters are detected up to 1660 nm. Two phenomena which are not taken into account in SRIM code could explain this observation. First the migration of vacancies or vacancy clusters toward the bulk could lead to agglomeration of these defects deeper than the cascade region and the stopping depth of ions. Secondly channeling of ions in crystalline solids can occur. The Fe samples irradiated in this study are polycrystalline with mean size of the grains of 20 - 100 μm dramatically larger than the range of the Y ions. Calculations with Marlowe code in which the sample has been modeled as a polycrystal show that this effect is not negligible in these irradiation conditions. Kinomura *et al.* have observed this channeling phenomenon for 150 keV Ar implantation in polycrystalline Fe with grain sizes of the order of 10 μm [16].

4. Summary

Vacancy type defect formations in 1.5 MeV ^4He and 1.2 MeV Y irradiated bcc Fe samples are investigated by slow positron beam Doppler- broadening measurements. The positron diffusion length L^+ in Fe virgin sample and the reference value (S_{ref} , W_{ref}) for Fe lattice are determined to be 160 ± 2 nm and (0.357, 0.111). Single vacancies are observed in 1.5 MeV ^4He irradiated Fe samples at low fluences from 1×10^{13} to $2 \times 10^{14} \text{ cm}^{-2}$ and vacancy clusters are detected at high fluences from 5×10^{14} to $1 \times 10^{17} \text{ cm}^{-2}$. In 1.2 MeV Y implanted Fe sample, vacancy clusters are also formed even at low fluence $1 \times 10^{14} \text{ cm}^{-2}$. Their concentration and size are larger than that of 1.5 MeV ^4He irradiated sample at the highest fluence $1 \times 10^{17} \text{ cm}^{-2}$. In Y implanted Fe sample, defects profiles obtained by positron measurements are deeper than those calculated by SRIM calculation which could be explained by ion channelling predicted by Marlowe modelling.

Acknowledgments

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