

# Cavity nucleation and growth during helium implantation and neutron irradiation of Fe and steel

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**Abstract.** The present work concerns investigations of damage accumulation during helium implantation of pure iron and the reduced activation ferritic-martensitic steel ‘EUROFER 97’ at 323K and 623K as well as during neutron irradiation with or without prior helium implantation. The defect microstructure, in particular the cavities, was characterized using Positron Annihilation Lifetime Spectroscopy (PALS) and Transmission Electron Microscopy (TEM). The PALS investigations reveal a clear difference between the He implantation effects in Fe and EUROFER 97 at both temperatures. For both materials the mean positron lifetime increases with He dose in the range 1 – 100 appm, although the increase is stronger for Fe than for EUROFER 97 and for both materials stronger for implantation at 323K than at 623K.

## 1. Introduction

Effects of neutron irradiation on physical and mechanical properties of reduced activation ferritic-martensitic (RAFM) steels are being extensively studied since they are considered to be candidate materials for fusion reactors [1, 2].

It has previously been demonstrated that in pure iron as well as in the RAFM steel called F82H [2] irradiation with fission neutrons causes void formation already at temperatures as low as 323 - 373K [3-5]. This is in agreement with general experimental observations that voids in bcc metals and alloys are formed at relatively low temperatures, i.e. close to the recovery stage III [6]. In a fusion reactor these materials are expected to be exposed to 14 MeV neutrons that introduces concurrent generation of helium atoms at a relatively high He-to-displacement ratio ( $\sim 10$ -20 appm/dpa) and at damage rates of  $\sim 10^{-6}$  dpa/s in the form of cascades and sub-cascades. This is a matter of serious concern from the point of view of the application of these alloys in structural components.

The main objective of the present work was to obtain experimental results on cavity evolution already during the He-implantation stage and then to follow the evolution of these cavities after post-implantation neutron irradiation to different displacement doses.

Details of the present experimental work can be found in two Risø Reports [7, 8]. Reference [8] also describes the modeling of vacancy cluster and helium-vacancy cluster evolution under a variety of different conditions.

## 2. Materials and Experimental Procedure

Specimens of pure (99.99 wt%) iron and the RAFM steel called EUROFER 97 [2] were investigated in the present work. Details of the sample preparation procedure can be found in [7-9].



Strip specimens of both pure iron and EUROFER 97 steel were implanted with helium at Forschungszentrum Jülich (Germany). A beam of 28 MeV  $\alpha$ -particles, energy degraded by a rotating wheel equipped with Al foils, was used for implantation of the specimens to provide a homogeneous He concentration throughout a depth of about 100  $\mu\text{m}$  (i.e. the specimen thickness) over an area of about  $13 \times 12 \text{ mm}^2$ . The implantations were carried out at  $\sim 323\text{K}$  or at  $623\text{K}$  to concentration levels of 1, 10 and 100 appm of helium at implantation rates in the range of  $1.2 \times 10^{-3}$  to  $1.2 \times 10^{-2}$  appm He/s [9]. The implantation gives rise to displacement damage doses of  $1.5 \times 10^{-4}$ ,  $1.5 \times 10^{-3}$  and  $1.5 \times 10^{-2}$  dpa, respectively for the three concentrations of implanted helium, since on average, each implanted He atom gives rise to 154 atomic displacements [9]. (This is equivalent to  $\sim 6500$  appm He/dpa or roughly 400 times higher than expected in a fusion reactor. Thus, the present implantation-irradiation conditions do not simulate those in a fusion reactor). Both un-implanted and He-implanted specimens were irradiated with fission neutrons in the BR-2 reactor at Mol (Belgium) at  $323\text{K}$  or  $623\text{K}$  with a displacement dose rate of  $\sim 4.5 \times 10^{-8}$  dpa/s to doses in the range of 0.001 to 0.23 dpa.

Positron Annihilation Lifetime Spectroscopy (PALS) was used for measurements on a wide range of specimens and Transmission Electron Microscopy (TEM) to study fewer, selected specimens.

### 3. Results

#### 3.1 Positron Annihilation Lifetime Spectroscopy

A detailed analysis of the measured lifetime spectra was carried out: All spectra for implanted and/or irradiated specimens could be decomposed into two or three lifetime components using the PALSfit program [10]. In order to give a simple qualitative overview of the general trend of all the data obtained by PALS for iron and EUROFER 97 which have either been implanted with helium, neutron irradiated or neutron irradiated after helium implantation, Figure 1 shows mean positron lifetimes as functions of total displacement dose at both  $323\text{ K}$  and  $623\text{ K}$ .

**3.1.1 Iron.** For He implantation at both temperatures the mean lifetime increases with He concentration and hence displacement dose (Fig. 1a and b). Qualitatively, a mean-lifetime increase can be ascribed to an increase in density of defects and/or size of cavities. The difference in mean lifetime after implantation to the same He concentration at the two temperatures is mainly due to a much larger cavity density at  $323\text{ K}$  than at  $623\text{ K}$ . The figures also demonstrate that the influence of He implantation prior to neutron irradiation is quite different at the two temperatures. At  $323\text{ K}$  the presence of He has only a moderate influence on the result of neutron irradiation, while at  $623\text{ K}$  the presence of He has a clear effect on the cavity population after neutron irradiation.

**3.1.2 EUROFER 97.** Figures 1c and 1d show the mean positron lifetime as a function of total displacement dose for EUROFER 97 at  $323\text{ K}$  and  $623\text{ K}$ . The relatively small effect (compared to Fe) of neutron irradiation with or without prior He implantation is clearly displayed. As Fig. 1 shows, in the investigated He concentration range of 0 – 100 appm He, changes of the mean lifetime fall in the range up to 30 ps for EUROFER 97 at  $623\text{ K}$ , while for Fe the range of mean lifetime variations is about 60 ps at the same temperature. At  $323\text{ K}$  the changes of the mean lifetime are appreciably larger (about 95 ps and 135 ps, respectively).

The results of the detailed analyses of all measured lifetime spectra (lifetimes and intensities) showed an appreciable scatter for nominally identical specimens. This scatter makes it impossible to define any trend in these data that can be correlated with the He implantation rate. Also the mean lifetimes show a certain scatter. Thus, attempts to correlate mean lifetimes with implantation rates were also unsuccessful. On the other hand the mean lifetimes shown in Fig. 1 clearly correlate with total He concentration. In spite of the scatter of the data for the resolved lifetime components, it is possible to derive estimates of sizes and densities of the cavities in the irradiated and/or implanted samples (see details in [4, 7, 8]).

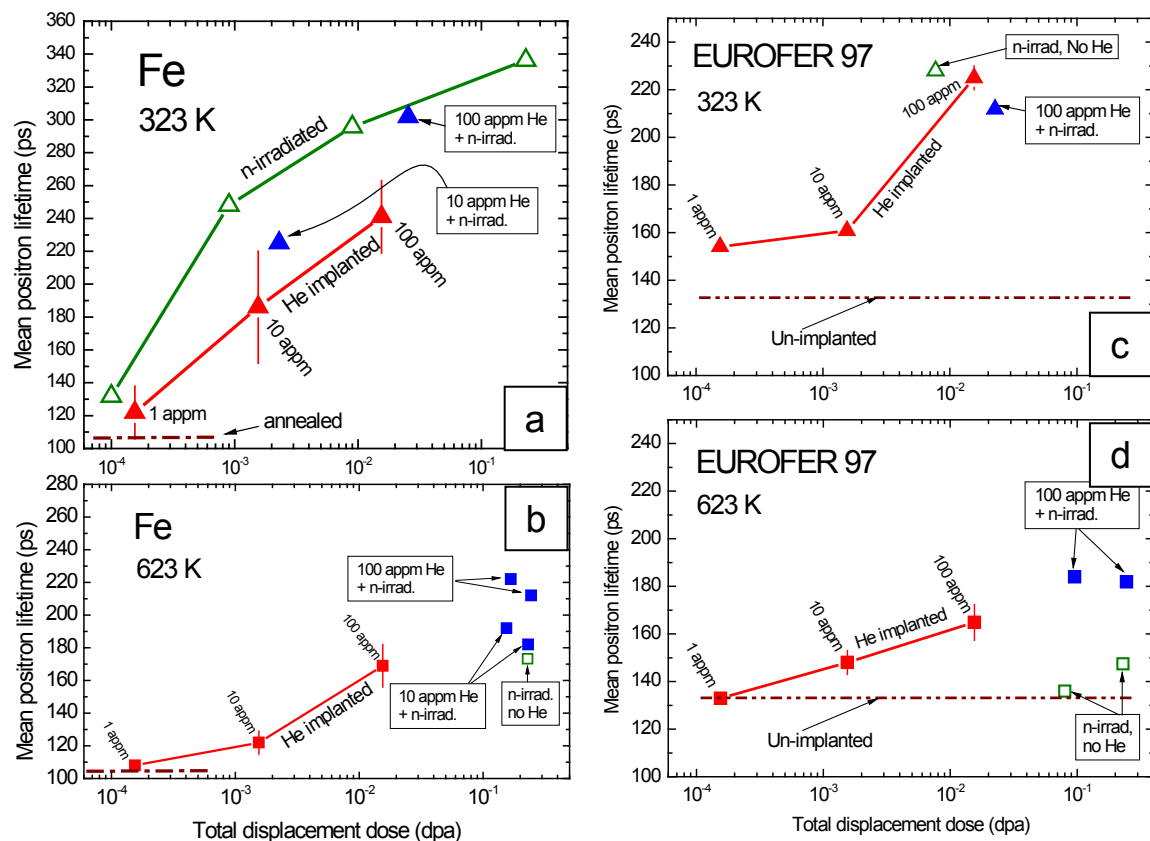


Figure 1. Mean positron lifetimes for iron (a, b) and EUROFER 97 (c, d) which have either been implanted with helium, neutron irradiated or neutron irradiated after helium implantation at 323K (upper frames: a and c) or at 623K (lower frames: b and d) [7, 8]. Implantation of 100 appm helium gives rise to a displacement dose of 0.015 dpa. The green open symbols are for neutron irradiation without helium implantation, while the blue filled symbols are results for neutron irradiation after implantation to a dose of 10 or 100 appm helium. An error bar associated with a red point indicates the scatter of results from several specimens implanted to the same nominal He dose.

### 3.2 Transmission Electron Microscopy

TEM could not resolve any cavities in Fe or EUROFER 97 after implantation of 100 appm He, neither at 323K nor at 623K. However, neutron irradiation of Fe at 623K to a dose level of 0.23 dpa leads to cavities (Fig. 2A). He implantation (100 appm) prior to neutron irradiation, results in a cavity density increase by a factor of  $\sim 7$  (Fig. 2B). In EUROFER 97 a very inhomogeneous cavity distribution, formed at dislocations and interfaces, was observed after He implantation with subsequent neutron irradiation. In addition, a very low density of very large voids has been observed in Fe neutron irradiated at 323K (no He), even at a dose level of as low as 0.036 dpa.

## 4. Conclusion

The present experiments have shown some rather interesting effects of helium implantation and post-implantation neutron irradiation on cavity formation and growth in pure iron. The results presented in Fig. 1a suggest, for example, that at 323K He implantation prior to neutron irradiation does not make any substantial addition of cavities compared to neutron irradiation without prior He implantation. This implies that at this temperature the homogeneous nucleation of cavities is very efficient even in the absence of implanted helium.

At 623 K (i.e. above the recovery stage V), on the other hand, the implanted helium does cause an increase in cavity nucleation and growth (Fig. 1b). This is because at this temperature, the implanted helium atoms help to stabilize the cavity nuclei against shrinkage by thermal evaporation of vacancies, which would take place in the absence of He. Consequently, both cavity nucleation and growth are enhanced.

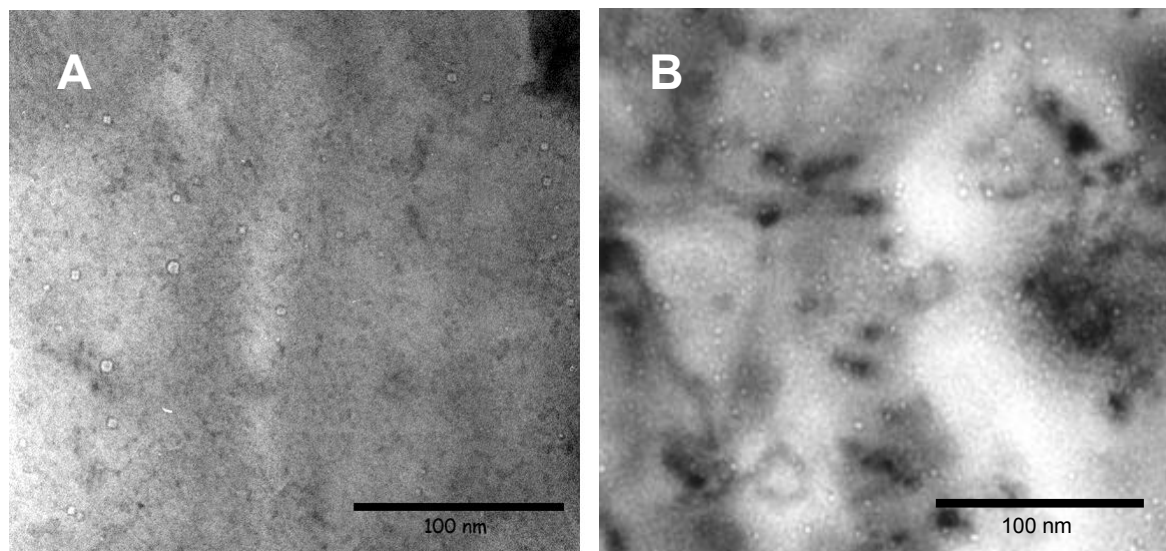


Figure 2. Transmission electron micrographs showing cavities in pure Fe after A) neutron irradiation to a dose of 0.23 dpa, giving an average void size of  $\sim 4.0$  nm and density of  $\sim 1.5 \times 10^{21} \text{ m}^{-3}$  and B) implantation of 100 appm He and subsequent neutron irradiation to a dose of 0.23 dpa, giving an average void size of  $\sim 3.5$  nm and density of  $\sim 1 \times 10^{22} \text{ m}^{-3}$ . Irradiation and implantation temperatures were 623 K.

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## References

- [1] Klueh R L, Ehrlich K and Abe F 1992 *J. Nucl. Mater.* **191** – **194** 116.
- [2] Baluc N, Gelles D S, Jitsukawa S, Kimura A, Klueh R L, Odette G R, van der Schaaf B and Yu Jinnan 2007 *J. Nucl. Mater.* **367-370** 33.
- [3] Eldrup M and Singh B N 2000 *J. Nucl. Mater.* **276** 269.
- [4] Eldrup M, Singh B N, Zinkle S J, Byun T S and Farrel K 2002 *J. Nucl. Mater.* **307** – **311** 912.
- [5] Eldrup M and Singh B N 2003 *Risø Report No. Risø-R-1241(EN)* pp. 21.  
[http://orbit.dtu.dk/fedora/objects/orbit:90110/datastreams/file\\_7712456/content](http://orbit.dtu.dk/fedora/objects/orbit:90110/datastreams/file_7712456/content)
- [6] Singh B N and Evans J H 1995 *J. Nucl. Mater.* **226** 29.
- [7] Singh B N, Eldrup M, Golubov S I, Edwards D J and P. Jung 2005 *Risø Report No. Risø-R-1496(EN)* pp. 38. [http://orbit.dtu.dk/fedora/objects/orbit:90096/datastreams/file\\_7710901/content](http://orbit.dtu.dk/fedora/objects/orbit:90096/datastreams/file_7710901/content)
- [8] Eldrup M, Singh B N and Golubov S I 2010 *Risø Report No. Risø-R-1619(EN)* pp. 64  
[http://orbit.dtu.dk/fedora/objects/orbit:82170/datastreams/file\\_4667852/content](http://orbit.dtu.dk/fedora/objects/orbit:82170/datastreams/file_4667852/content)
- [9] Jung P. 2004 *Helium implantation in pure iron and RAFM steels at <70 and 350 °C* (Task:TW4-TTMS-001a, EFDA Technology Programme).
- [10] Olsen J V, Kirkegaard P, Pedersen N J, Eldrup M 2007 *phys. stat. sol. (c)* **4** 4004 and [www.PALSfit.dk](http://www.PALSfit.dk)