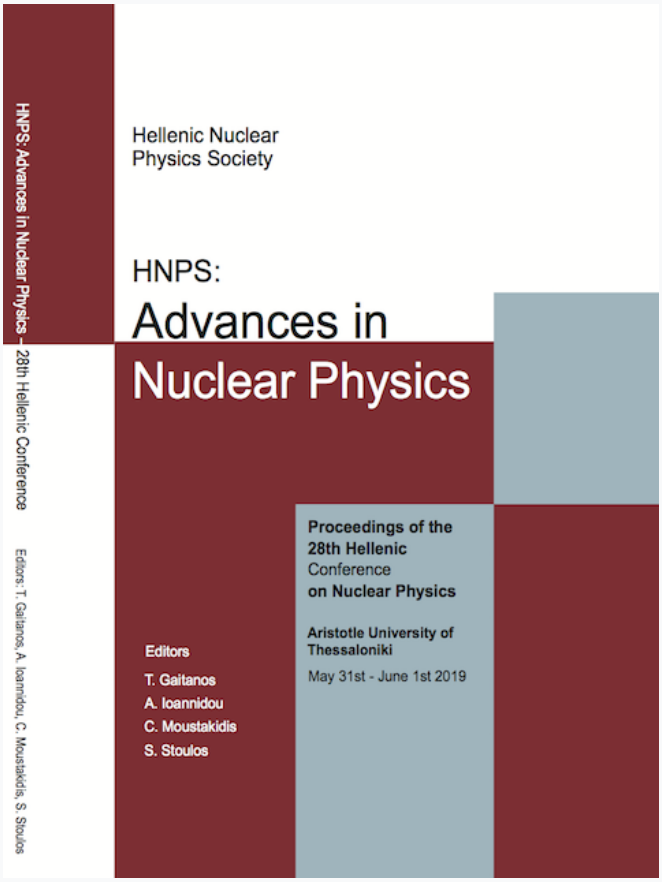
HNPS Advances in Nuclear Physics

Vol 27 (2019)

HNPS2019



Proton irradiation and resistivity recovery in stages I & II of pure and carbon-doped Fe

Andreas Theodorou, M. Syskaki, Z. Kotsina, M. Axiotis, G. Apostolopoulos

doi: 10.12681/hnps.3007

Proton irradiation and resistivity recovery in stages I & II of pure and carbon–doped Fe

A. Theodorou[1,2] , M. Syskaki[1,2], Z. Kotsina[2], M. Axiotis[3], G. Apostolopoulos[2]

[1] *Section of Solid State Physics, Department of Physics, National and Kapodistrian University of Athens, Panepistimiopolis, GR–15784,Zografos*
[2] *Institute of Nuclear and Radiological Science and Technology,Energy and Safety NCSR "Demokritos", 15310 Aghia Paraskevi Attikis, Greece*
[3] *Tandem Acceleration Laboratory, Institute of Nuclear Physics, NCSR "Demokritos", 15310 Aghia Paraskevi Attikis, Greece*

Abstract *Pure and C–doped Fe specimens were irradiated with 5 MeV protons at cryogenic temperature at the NCSR "Demokritos" TANDEM accelerator in order to investigate the interactions between carbon atoms and radiation defects. During the subsequent post–irradiation isochronal annealing up to 180 K the defects start to migrate and interact either mutually or with the C impurities. The defect evolution is observed by in situ electrical resistivity recovery measurements. Comparison of results from pure and C–doped Fe specimens reveals the effect of C solute atoms on the defect kinetics.*

Keywords *proton irradiation, resistivity recovery, radiation defects, C–doped Fe*

Corresponding author: A. Theodorou (theoda@ipta.demokrtitos.gr) | *Published online:* May 1st, 2020

INTRODUCTION

Reduced–activation ferritic/martensic (RAFM) steels are the primary choice for the structural materials of future fusion power plants due to their high resistance to radiation damage accumulation and swelling. Carbon is one of the most important alloying elements in steels playing a key role in the development of the microstructure and the mechanical properties. Furthermore, carbon has a great influence on the irradiation response due to its strong interaction with radiation defects. Previous studies [1,2] have shown the formation of carbon–defect complexes which exhibit reduced mobility and have a significant impact on the recovery of radiation damage.

In this work we investigate the interactions between carbon atoms and radiation defects by means of resistivity recovey measurements in C doped Fe compared to pure Fe as a model material for the behaviour of more complex RAFM steels. Specimens of these materials were irradiated with 5 MeV protons at the NCSR "Demokritos" TANDEM accelerator at a temperature of T=25 K where most of the defects created by the irradiating particles remain initially immobile in the lattice. The electrical resistivity was measured *in situ* during irradiation and resistivity recovery was recorded during post–irradiation isochronal annealing up to 180 K. The spectra of resistivity recovery rate as a function of annealing temperature give information on point defect migration and interaction. The most remarkable observation is the "trapping" of interstitial defects by carbon atoms and their "de–trapping" at higher temperatures.

EXPERIMENTAL DETAILS

Specimens

The starting materials were two high purity alloys obtained from the European Fusion Development Agreement (EFDA) as cylindrical bars of 10 mm diameter. The pure Fe material contained a residual C concentration of 20 at. ppm, whereas in the C doped sample the nominal concentration was 220 at. ppm. Specimens in the form of foils are needed for the resistivity recovery experiments. These were prepared by cold–rolling of thin wafers cut from the bars by means of a

diamond saw followed by electropolishing to their final thickness. The electrolytic solution was prepared by mixing 25 ml H_2SO_4 (96%) and 50 ml H_3PO_4 (85%). A platinum mesh was used as cathode, a DC voltage of 2.6 V was applied and the temperature of the solution was 85°C. After electropolishing the specimens exhibit a bright polished surface. The final thickness of the foil specimens was about 50 μm, which ensures full penetration by the 5 MeV protons. To relieve the effects of cold–working and to ensure complete dissolution of C, the Fe and Fe–C foils were annealed for 12 h at 720 °C under a hydrocarbon–free vacuum of 10^{-6} mbar. After annealing the samples were quickly removed from the furnace. Current and potential leads of pure Fe were spot–welded on the specimens for performing the electrical resistivity measurements according to the standard DC four–probe method. Current polarity reversal was used to eliminate thermal voltages.

Irradiation conditions

Irradiation was performed at the NCSR "Demokritos" 5MV TANDEM accelerator at the recently upgraded facility IR^2 [3]. A beam of 5 MeV protons was used with a flux of $9 \cdot 10^{11}$ cm^{-2}s^{-1}. The specimen temperature during irradiation was kept at 25 K by means of a cryocooler. The pure and carbon–doped Fe specimens were irradiated simoultaneously to a total fluence of $\Phi = 1.79 \times 10^{16}$ cm^{-2}. The total irradiation induced resistivity increase, $\Delta\varrho_0$, was 320 and 310 nΩ-cm for Fe and Fe–C, respectively. The total produced radiation damage is estimated by the ratio $\Delta\varrho_0/\varrho_{FP}$, where ϱ_{FP} is the specific Frenkel pair resistivity in Fe, $\varrho_{FP} = 3$ nΩ-cm / at. ppm [4]. In units of displacements–per–atom (dpa), the generated damage is in the ppm range and is equal to 110 and 100 displacements per million atoms for Fe and Fe–C, respectively.

ANNEALING & RESISTIVITY RECOVERY

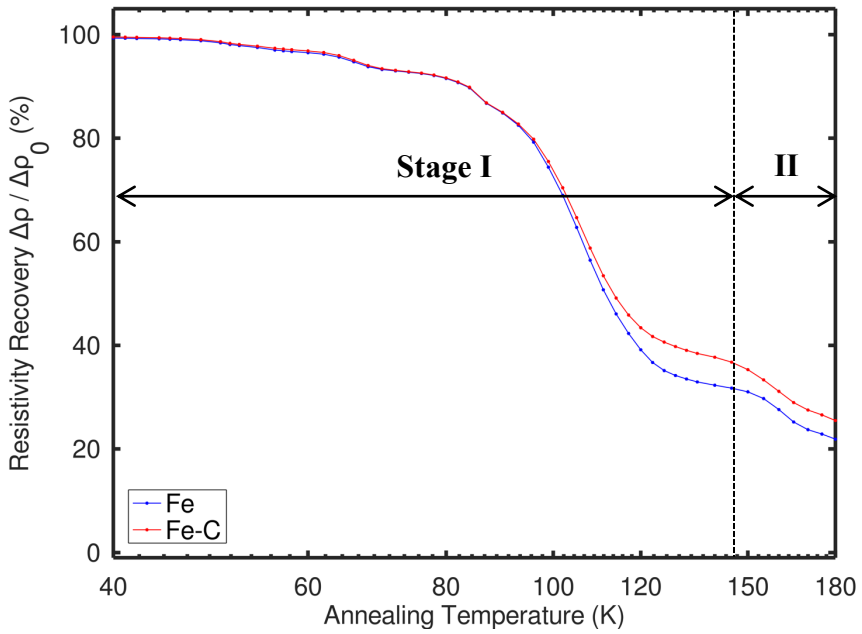


Figure 1. *Resistivity Recovery of proton irradiated pure Fe & Fe–C as a function of annealing temperature.*

The specimens were annealed together, *in situ* within the irradiation chamber up to 180 K. The annealing was performed isochronally with a time duration of 5 min and a temperature step $\Delta T/T \sim 0.03$ K. Fig. 1 shows the resistivity recovery as a function of annealing temperature which is the ratio of the remaing radiation induced resistivity after annealing at temperature (Ta) over the

initial resistivity increase immediately after the irradition. The quantity $\Delta\rho/\Delta\rho_0$ is proportional to the fraction of radiation defects that survive after annealing at a given temperature. During annealing the recovery of radiation defects occurs due to their migration and recombination. This is not a continuous process but occurs in temperature ranges which are called ‘”stages”, based on previous studies [1,2,4] of recovery processes in carbon doped Fe alloys. The various stages are due to different recovery reactions. As seen in the figure, the recovery is very similar in pure and C–doped Fe up to about 100 K, however, the recovery process slows down in Fe–C at higher temperature. This shows that the precence of C influences the defect reactions.

In Fig. 2 the resistivity recovery rate is shown as a function of annealing temperature. It is obtained by point–by–point differentiation of the data in Fig. 1. The recovery rate is particularly important because it reveals with great clarity the various features (“stages”) attributed to activation of different recovery processes. The recovery of Fe consists of two main stages up to 180 K [1]:

Stage I, 20 – 150 K.

This stage is separated into five substages I_A–I_E. The first substages I_A–I_C at 52, 70 and 90 K, respectively, are attributed to close–pair recovery. These are mostly unaffected by the presence of carbon as seen in the figure. In the substages I_D and I_E it is known that only self–interstitial atom (SIA) can migrate and either annihilate at vacancies or form small interstitials clusters like di–interstitials. Stage I_D, at 108 K, is due to SIA migration and correlated recombination with their respective vacancies while stage I_E, at 125 K, is attributed to uncorrelated recovery. The total defect recovery in the substages I_D and I_E amounts to 60% in pure Fe. In contrast, the carbon doped specimen exhibits a marked suppression of total defect recovery which amounts to 53%. This is attributed to trapping of mobile SIA defects by carbon solute atoms, creating the interstitial–carbon (IC) complex which prevents their annihilation with vacancies at this stage.

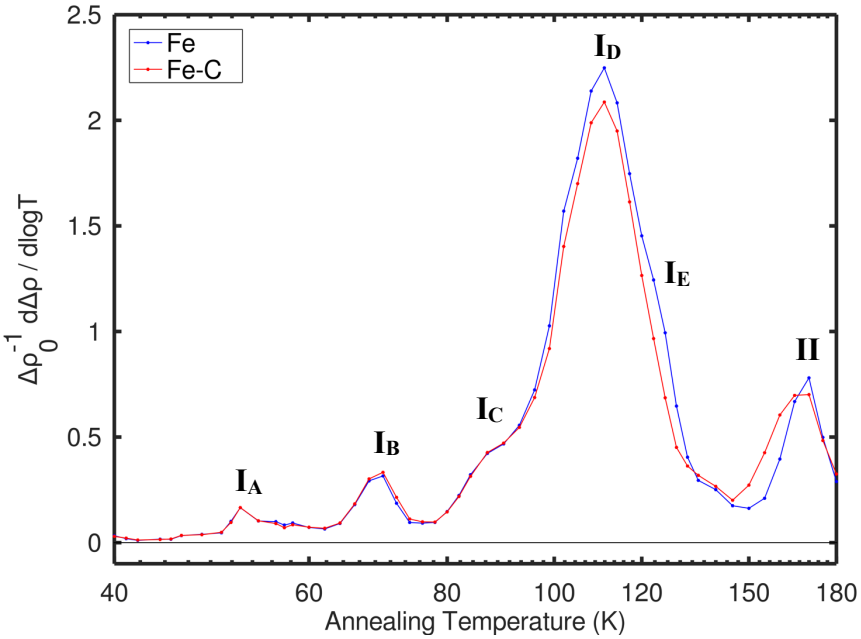


Figure 2. *Resistivity Recovery rate of proton irradiated Fe & Fe–C as a function of annealing temperature*

Stage II, 150 – 180 K

This stage is due to the migration of small SIA clusters (di–, tri–interstitials) created in stage I and their annihilation at vacancies. It is observed as a sharp peak at 165 K in the recovery rate of pure

Fe. This stage is also influenced by the presence of carbon, as can be seen from Fig. 2, however in this case the presence of carbon enhances the recovery. The peak of stage II in Fe–C specimen is broader and extends to lower temperatures. The total defect recovery in stage II inceases from 10% in pure Fe to 12% in carbon doped Fe. These observations may probably indicate the presence of a new substage which is due to the de–trapping of some of the SIA defects that were arrested by carbon atoms during stage I. However, the observed 2% increase of stage II recovery in Fe–C is smaller than the corresponding 7% decrease in stage I. Thus, the majority of the SIA defects that reacted with C in stage I remain in the sample after stage II. A possible interpretation is that these defects are in the form of higher order SIA–carbon complexes such as di–interstitial – carbon (I_2C) and tri–interstitial – carbon (I_3C) [2]. These complexes are immobile in this range of temperatures so they survive in the sample.

CONCLUSIONS

The generation, migration and recovery of defects produced in pure Fe and Fe–C by 5 MeV proton irradiation at T=25K, have been studied by measurements of the electrical resistivity in order to study the influence of carbon. The resistivity recovery was measured *in situ* up to 180 K. The resistivity recovery spectra of Fe and Fe–C are in general agreement with previous experiments conducted after electron irradiation [1,4]. The presence of carbon causes a reduction of the total recovery in stages I_D and I_E which are due to SIA migration. The reduction is attributed to interaction and trapping of SIA defects by carbon atoms. At the same time, the total recovery in stage II increases in Fe–C, indicating that some of the trapped SIA defects are released at this recovery stage. However, the majority of the trapped SIA defects remain stable up to 180 K. It is assumed that these defects are high order SIA–carbon complexes involving 2 or more SIAs.

Acknowledgments

This work has been carried out within the framework of the EUROfusion Consortium and has received funding from the Euratom research and training programme 2014–2018 and 2019–2020 under grant agreement No 633053. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

References

[1] S. Takaki et al., Radiat. Eff. Defects Solids 79, 87 (1983), doi: 10.1080/00337578308207398
[2] T. Jourdan et al., Phys. Scr. 2011, 014049 (2011), doi: 10.1088/0031-8949/2011/T145/014049
[3] A. Theodorou et al., HNPS Adv. Nucl. Phys. 26, 253 (2019), doi: 10.12681/hnps.1833
[4] F. Maury et al., Phys. Rev. B 14, 5303 (1976), doi: 10.1103/PhysRevB.14.5303