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Combined effect of injected interstitials and He implantation, and cavities inside dislocation loops in high purity Fe-Cr alloys

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Abstract

High purity Fe-Cr alloys with Cr content ranging from 3-14 wt.% were irradiated by self-ions at 500 °C in dual-beam mode up to 157 displacements per atom (dpa), 17 appm He/dpa. Transmission electron microscopy (TEM) on focused ion beam foils revealed the depth distribution of irradiation induced cavities and dislocation loops. A detailed quantitative analysis of cavity microstructure was performed on Fe14%Cr, while Fe(3, 5, 10, 12)%Cr were analyzed qualitatively. A homogeneous distribution of small cavities were observed up to the damage peak. From surface to the damage peak, cavity number density increased almost linearly. The cavity microstructure changed drastically at and after the damage peak, where the size increased and number density decreased. Most notably, the microstructure consisted of a striking mixture of heterogeneously nucleated cavities inside dislocation loops and freely nucleated cavities in the matrix. Further, the depth-dependent void swelling increased continuously along the damage depth. This contrasts with most ion irradiation results where suppression of void swelling occurs adjacent to the damage peak due the injected interstitial effect. We hypothesize a plausible mechanism of the observed swelling variation based on a combined effect of the injected interstitials and helium implantation near the damage peak by comparing the results with those in pure iron irradiated under same conditions. This synergistic effect may develop cavity microstructures which do not necessarily reflect microstructures expected due to injected interstitial effect and can be easily wrongly interpreted. The depth dependent dislocation loop microstructure was studied qualitatively in Fe14%Cr sample. Up to the damage peak, a complex dense network of dislocations formed due to the interaction/impingement of the dislocation loops. Individually resolvable loops were only observed after the damage peak, where their association with cavities was noted. Helium

also induced heterogeneous cavity nucleation on dislocations constituting a grain boundary and on pre-existing dislocation lines.

Keywords: Fe-Cr alloys; ion irradiation; transmission electron microscopy (TEM); void swelling; chromium.

1. Introduction

The irradiation conditions that in-core structural materials inside generation IV sodium cooled fast reactors (SFRs) and plasma facing first-wall components of future fusion reactors will be exposed to are unprecedented. Particularly for fusion reactors, the damage by 14 MeV neutrons in the first wall steel components can be as high as 150–200 displacements per atom (dpa) with high helium generation rates (10–12 appm He/dpa) [1]. Currently, owing to good thermo-mechanical properties and moderate void swelling under irradiation as compared to austenitic steels, high chromium (Cr) based reduced activation ferritic-martensitic (RAFM) steels and their nano-structured variant oxide dispersion strengthened (ODS) steels are envisaged as the most promising structural material candidates for these reactor concepts [1–3]. However, a thorough understanding of irradiation effects in these steels is still not well established, which is the key in predicting their long-term performance in irradiation environments. For this reason, significant international research is devoted to studying binary Fe-Cr model alloys, which are a simple representative of RAFM steels.

In Fe-Cr alloys, a large amount of research using fast neutrons and energetic ion irradiations exists on the study of cavity microstructure and void swelling behaviour [4–14]. It is known that addition of Cr to body-centered cubic (bcc) iron (Fe) matrix is beneficial to significantly reduce void swelling [4,5,7,8,11]. Thus, Fe-Cr alloys are highly swelling resistant as compared to bcc Fe. The variation of void swelling with Cr level is

non-monotonic, with a local swelling maxima appearing around 9 – 10 wt.%Cr [4,11]. Irradiation induced segregation/enrichment of Cr to defects such as dislocation loops is also known to occur [15–17]. Concerning the effect of helium, simultaneous dual beam irradiations have shown an early onset of swelling in these alloys owing to enhanced cavity nucleation [4,8], which is consistent with results on various other alloys [18–22]. Furthermore, for Cr content > 8-9 wt.%, these alloys are also well-known to suffer from irradiation induced phase separation into an embrittling Cr rich α' phase [23–27].

A grand challenge is that, at present, no experimental facility exists that can irradiate materials with 14 MeV fusion neutrons reaching high doses. Thus, most of the fundamental research is performed using ion irradiations or sometimes with fission/spallation neutrons. Particularly with ion irradiations, there are many associated challenges which hinder simulating neutron damage. For example, for decades α' phase formation in Fe-Cr alloys or RAFM steels was not reported after ion irradiations, and only recently a few low dose rate ($\leq \sim 10^{-4}$ dpa/s) ion irradiations have revealed their existence, thereby reflecting a potential role of ballistic dissolution due to high sink density at high dose rates attained using energetic ions [23,24,28]. Besides this, there are various other artefacts when studying irradiation effects using accelerated ions. The damage depths are considerably shorter (a few nm to a few microns) than neutrons because of smaller mean free path of charged ions compared to uncharged neutrons [29]. The irradiation damage phenomenon is heterogeneous along the target depth, due to the fundamental interaction of charged ions with the material as compared to neutral neutrons [29]. The effect of the free surface on microstructure evolution is significant which acts as a neutral point defect (PD) sink [30,31]. Due to the enhanced recombination at or adjacent to a free surface, areas devoid of any irradiation induced damage (denuded zones) are often observed

[32,33]. Most importantly, maximum damage occurs close to the ion projected range where the injected ions deposited into the material can artificially suppress cavities at and around the damage peak [30,31,33–40]. For example, recent ion irradiation experiments at 500 °C up to 157 dpa using self-ions on high purity bcc Fe observed nearly one-third reduction of void swelling at the damage peak as compared to mid-range [33]. Many previous ion irradiation experiments, some coupled with simulation studies on a variety of metallic materials, have also reported similar behaviour of highly reduced void swelling around the peak damage area [34,40–43] due to suppression of cavity nucleation rate [38,42] and cavity growth [43]. Additionally, recent ion irradiations in high purity Fe-Cr alloys by Tissot et al. have revealed a previously unknown role of the injected interstitials in suppressing irradiation-induced/enhanced precipitation at and around the damage peak [44], while in ceramics the implanted ions can also modify the irradiation induced amorphization phenomena [45]. Therefore, obtaining an irradiation representative of neutron irradiation by using ions can be challenging if the potential role of artefacts is ignored. Currently, it is largely being accepted that most reliable irradiation damage data is only obtainable from the mid-range regions, away from the surface and away from the damage peak [30,31,37,46]. But there is always a risk that radiation-enhanced diffusion (RED) of the injected ions may affect the microstructures even at mid-range depending upon the ion energy that governs the penetration depth [30,31]. Therefore, for obtaining a clear picture about a material and for the sake of obtaining all the information regarding the ion irradiated heterogenous damage microstructure, the entire depth distribution needs to be studied. Such studies can be performed on transverse focused ion beam (FIB) thin foil samples using transmission electron microscopy (TEM). In this context, we have explored the depth distribution of cavities and dislocation loops

in high purity Fe-Cr alloys with Cr content ranging from 3 to 14 wt.%, irradiated at 500 °C to high doses (> 100 dpa) and injected simultaneously with helium. We performed a thorough quantitative analysis of irradiation induced defects on Fe14%Cr, while the other samples were studied qualitatively. To explain the variation of cavity size and number density along the damage depth in the Fe-Cr alloys, the results were compared to those obtained in bcc Fe irradiated under same conditions. In addition, aspects of helium induced heterogeneous cavity nucleation inside the prismatic dislocation loops, on pre-existing dislocation lines and grain boundaries are reported.

2. Experimental details

The materials studied were high purity binary Fe-Cr alloys, with 3.04, 5.40, 10.10, 11.63 and 14.25 wt.%Cr. For the sake of simplicity, these will henceforth be referred as Fe3%Cr, Fe5%Cr, Fe10%Cr, Fe12%Cr and Fe14%Cr. Their nominal impurity composition, as provided in Refs. [33, 34], is given in Table 1. These materials were prepared by induction melting at Ecole des Mines, Saint Etienne, France, under the European Fusion Development Agreement (EFDA) programme (contract no. EFDA-06-1901). The alloys were received as cylindrical rods having 1.1-1.4 cm diameter in fully recrystallized state after a cold reduction of 70%, followed by heat treatment for 1 hour under argon flow. Fe(3, 5)%Cr were annealed at 750 °C, Fe10%Cr at 800 °C and Fe(12, 14)%Cr at 850 °C, followed by air cooling. The resulting grain sizes are also reported in the Table 1. The dislocation densities in all the materials were of the order of 10^{12} m^{-2} , which is typical of fully solution annealed metals and alloys.

For preparing samples for irradiation, circular discs of 1 mm thickness were cut from the as-received rods using a Struers cutting saw, followed by mechanical polishing to mirror finish by silicon carbide and diamond abrasives to 100 μm thickness, using a

Struers polishing machine. Then, discs of 3 mm diameter were punched out, which were jet-electropolished in a Tenupol-5 thinning device for a few seconds to obtain mirror finished dimpled discs.

The ion irradiations were performed at 500 °C using JANNuS (Joint Accelerators for Nano-science and Nuclear Simulation) multi-beam irradiation facility at CEA, Saclay, France [47]. The irradiation conditions and post-irradiation sample preparation methodologies were the same as those detailed in Refs. [4,33]. The specimens were irradiated in simultaneous dual-beam mode (damage by Fe self-ions coupled with co-implantation of He) on the 3 mm diameter dimpled discs. 2 MeV Fe²⁺ self-ions were used to reach 1.38×10^{21} ions.m⁻² fluence with a rastered beam, using 3 MV Épiméthée single-ended accelerator. The depth dependent displacement damage profile, estimated by SRIM in a Fe matrix, using a displacement threshold of 40 eV, and using Kinchin–Pease calculation, is shown in Fig. 1. Maximum dose at the damage peak, located at ~550-600 nm below the irradiated surface, was 157 dpa. Dose rate at the peak was $\sim 3.3 \times 10^{-3}$ dpa/s. For simultaneously implanting He, 2.5 MV Yvette Van-de-Graaf accelerator was used to generate a 2 MeV He⁺ rastered beam, to reach a fluence of 8.4×10^{20} ions.m⁻². To implant He at the damaged zone created by the Fe ions, a rotating energy degrader consisting of a combination of thin aluminium foils of different thicknesses was used to obtain a continuous energy degraded He beam, described in Ref. [8]. The He implantation profile was not homogeneous because of significant ion loss due to divergence, after scattering in the foils. The implantation profile was calculated after this correction and is given in Fig.1. The He implantation with respect to the cascade damage obtained at the damage peak was 17 appm He/dpa. The profile of the injected Fe ions is also given in Fig. 1. The irradiation conditions are detailed in Table 2. It is important to remember that when the

He beam was degraded using the degrader system, one slot was kept open without any Al foil. A geometry like this is necessary for performing beam current measurements. This means that even undegraded 2 MeV He⁺ ions, which pass through this window, were deposited in the sample. However, the depth of this He implanted zone was 3.34 μm below the implanted surface. It was, thus, much further away from the dual beam zone of interest which was around 600 nm depth. It is hence considered in the analysis that there is negligible influence of these two zones on each other. Since there are five Al windows and one empty window, the He implantation fluence at this depth is one-sixth of the net fluence given in Table 2 (i.e. 1.4×10^{16} ions.cm⁻²). These two zones are shown in a TEM image of transverse FIB thin foil of Fe10%Cr in Fig. 2. Maximum damage produced by this implantation was ~0.25 dpa which is largely lower than the damage produced by the heavier Fe ions at the main damage peak.

After the irradiation, focused ion beam (FIB) electron-transparent thin foils were extracted from the irradiated samples, to access the full depth distribution of damage. These were prepared using a FEI Helios 650 DualBeam™ FIB at Service de Recherches Métallurgiques Appliquées (SRMA), CEA-Saclay. The initial lift-out was performed with 30 keV gallium (Ga) ions. During thinning, the ion energy was progressively decreased up to 2 keV to minimise defects induced by the preparation method. All the FIB samples were analysed using a field emission gun (FEG) based 200 keV JEOL 2010F TEM, equipped with a Gatan double-tilt holder and a Gatan Orius high resolution CCD camera. The cavities in the microstructure were imaged using through-focal series technique [48], in which cavities appear bright with a dark Fresnel fringe around when the objective lens of the TEM is under-focused, and vice-versa. For the sake of simplicity, only under-focused images are presented in this article. Depth dependent dislocation loop

microstructure was imaged in two-beam kinematical bright field (KBF) imaging conditions [48]. Particularly for imaging dislocation loops, care was taken to ensure FIB samples were milled for long times at lower voltages (5 and 2 kV) to minimize FIB induced defects. Nevertheless, as will be seen in the results section, the loops were considerably larger (> 50 nm or entangled networks) than the size of typical milling induced defects (which are usually 2-3 nm) and therefore FIB defects, if any, had negligible effect on TEM image qualities.

Quantitative depth dependent void swelling trend was estimated for Fe14%Cr sample by extracting the cavity volume fraction from the TEM micrographs. The error bars on void swelling values were estimated by calculating the error in the foil thickness determination, statistical error in the number of cavities in the analysed zone and the error in cavity size measurement, as detailed previously in Refs. [33,46]. Specimen thickness was determined using electron energy loss spectroscopy (EELS) log-ratio model [49]. The error in thickness, obtained by performing repeated measurements on the same zone, was about $\pm 30\%$. The statistical error on the cavity count was between 6-9 %, and it was minimised by analysing a larger zone to have sufficient number of cavities to consider. For faceted cavities, the longest distance was considered as the size, while for spherical cavities, the size is represented by their diameter. Depth variation of cavity number density was also estimated. The error bar on number density was calculated using the error in the thickness measurement and the statistical error in cavity count.

3. Results

3.1 Irradiation induced damage in Fe14%Cr.

Cavities were observed in the entire damage depth in the FIB sample. Fig. 3 shows bright field (BF) TEM images of cavities detected at different depths beneath the

irradiated surface, up to the damage peak (around 600 nm depth). The first thing to notice was that there was almost no denuded zone close to the irradiated surface (0 - 100 nm). However, cavity number density appeared small at this depth. Concentration of visible cavities increased upon going deep in the material, implying an increase in the cavity number density. No notable change in their size (diameter) was detected in the TEM images at least up to the depth of 400 – 500 nm. In these areas, the cavities were homogeneously distributed in the matrix

Upon reaching the damage peak and the areas immediately after the damage peak, the microstructure changed drastically. The spatial distribution of cavities consisted of mostly heterogeneously nucleated cavities inside the dislocation loops, combined with a low fraction of freely nucleated cavities in the matrix. BF TEM micrograph of an area between 650 – 1000 nm depth below the irradiated surface, imaged not far from [001] zone axis, is shown in Fig. 4. In this image, cavity clusters can be seen on the plane of the loops which are in open configuration and linear arrangement of cavities on the loops which are on-edge. A high magnification image of the zone is shown in Fig. 4b where many relatively smaller cavities inside the loops can be seen along with freely nucleated cavities in the matrix adjacent to the loops. The freely nucleated cavities mostly appeared to be a bit larger in size as compared to those associated with loops and exhibited facets.

Quantitative information on cavity sizes and number density, estimated from these images, are plotted in Figs. 5a and 5b. It is evident that cavity number density increased continuously from near-surface areas to the depths of around 450 – 500 nm. For instance, cavity density at ~ 450 nm depth was almost six times higher than the value obtained in the area up to 100 nm below the surface. However, the average cavity size did not vary significantly and stayed lower than 2 nm. The size of the largest cavities detected along

depth is plotted as “maximum cavity size” in Fig. 5 which also stayed lower than 3 nm up to ~450 nm depth, while the size of the smallest cavities detected (denoted as “minimum cavity size” in Fig. 5) stayed close to 0.5 nm, which is very close to the typical resolution limit of a conventional TEM. The variation of void swelling along the damage depth is also plotted in Fig. 5b. Similar to the variation in number density, it showed an increasing trend in the areas before the damage peak. But the absolute swelling values remained low (less than 0.02%). In the areas after the damage peak where heterogeneous cavity nucleation was detected (see Fig. 4), cavity size increased sharply as compared to the areas before the damage peak (see Fig. 5a). However, cavity number density decreased substantially. The estimated void swelling continued to increase. The void swelling trend in these areas appeared to follow the trend of the average cavity size evolution, showing an increase just after the 600 nm zone. Another thing to note is that there was a significant difference between the minimum (~0.6-1 nm) and maximum cavity size (~8 nm), indicating that the size distribution had broadened in the areas at and after the damage peak. This is quantitatively shown in the depth dependent histogram of cavity sizes plotted in Fig. 6. As evident from the TEM images in Fig. 3, the histograms show a narrow size distribution up to ~ 400 nm depth in the sample, with sizes typically less than ~3 nm. The distribution began to broaden upon approaching the damage peak, with average and maximum size of 2 nm and ~8 nm respectively. Beyond the damage peak, the cavity size distribution stayed significantly broad due to a wide range of sizes present in the microstructure.

The dual beam irradiated Fe14%Cr sample was characterized to also understand the depth distribution of the dislocation loop microstructure induced by the irradiation. Kinematical bright field (KBF) TEM image of the entire damage depth, imaged in two-

beam conditions using a $\langle 011 \rangle$ type diffraction vector close to $[001]$ zone axis is shown in Fig. 7. The imaging revealed a complex dislocation network present all along the specimen damage depth, formed due to the interaction between growing dislocation loops. Formation of the network started from the irradiated surface, with no denuded zone. Individually resolvable dislocation loops were detected only at larger distances beyond the damage peak where the dpa level reduced rapidly. Such loops at the end of the damage depth are shown in a zoomed inset image marked as A in Fig. 7. Most of these loops were almost on-edge and aligned along $\langle 100 \rangle$ directions, which is a peculiarity of a $\langle 100 \rangle$ type loops. A zoomed image of the dislocation network at mid-range depth in the specimen is shown in the same figure, indicated by inset B. Due to the complexity of the microstructure, we have not attempted to estimate quantitative information (number density or Burgers vector) from the observed network. Fig. 7 also shows an inset C which corresponds to an area nearly at the end of the damage zone. In this image, two partially non-linear dislocations can be seen. The non-linear part, indicated by an arrow, is where the dislocation is perhaps trying to climb. TEM analysis revealed that these dislocations were decorated with small cavities. A zoomed image of one of the dislocations decorated with small cavities is shown in Fig. 7, indicated by D. These cavities appeared dark on a relatively bright background as the image was obtained with an over-focused objective lens.

3.2. Irradiation induced damage in Fe(3, 5, 10, 12)%Cr

A qualitative TEM study was performed on Fe(3, 5, 10, 12)%Cr alloys to observe the evolution of the depth dependant cavity microstructures. For Fe3%Cr and Fe5%Cr, the entire damage depth was imaged in the FIB specimens. BF TEM images of cavities detected at various depths below the irradiated surface are shown in Figs. 8 and 9

respectively. Both alloys revealed very similar evolution of the microstructure as was seen previously for Fe14%Cr. It is evident from the images that the cavity number density increased from near surface areas (100 - 200 nm depth) to deeper zones. Cavity sizes did not seem to vary significantly in areas prior to the damage peak. However, at and beyond the damage peak, a slight increase of cavity sizes accompanied with a decrease in the number density can be visually inferred from the TEM image of 600 – 700 nm zone in Figs. 8 and 9. Furthermore, similar to Fe14%Cr, the spatial distribution of cavities was homogeneous prior to the damage peak, followed by a mixed distribution beyond (i.e. heterogeneous nucleation inside dislocation loops plus freely nucleated cavities in the matrix). For both these samples, cavities in the areas beyond the damage peak are shown in Figs. 10a and 10b respectively. For Fe3%Cr, a cluster of cavities nucleated in a loop is marked in red dashed lines in Fig. 10a. The associated dislocation loop is not visible in the image because the imaging was performed away from Bragg condition. A linear arrangement of cavities, possibly nucleated on a dislocation line, can also be seen above this cavity cluster in Fig. 10a which is indicated using a yellow arrow. The zone corresponded to 800 - 1000 nm depth below the irradiated surface. Similar cavity clusters associated with dislocation loops were detected in Fe5%Cr sample. TEM image of a zone 700 – 1000 nm below the irradiated surface of Fe5%Cr is shown in Fig. 10b, where two such cavity clusters are highlighted by red arrows.

For Fe10%Cr and Fe12%Cr sample, cavities in midrange depths for same irradiated materials were previously reported [4] which are qualitatively similar to homogeneously nucleated cavities detected in the other alloys here. Therefore, in this study the area beyond the damage peak was observed. TEM images of cavities beyond the damage peak area in both these materials are shown in Figs. 10c and 10d respectively.

They revealed similar association of cavities with the dislocation loops as was seen previously for the other compositions. For Fe10%Cr, the cavity clusters can be seen linearly aligned (highlighted by red arrows in Fig. 10c), indicating that they were associated with on-edge dislocation loops imaged close to [001] zone axis. Many freely nucleated cavities were also observed in the microstructure. In Fe12%Cr, cavities can be seen inside the dislocation loops which are in open configuration in Fig. 10d. In this case, the imaging was performed close to [011] zone axis, not too far from Bragg condition in order to see both cavities and the dislocation loops at the same time.

3.3 Heterogeneous cavity nucleation on dislocation lines and grain boundaries

Beyond the peak damage area, apart from the nucleation of cavities inside the dislocation loops, linear arrangement of faceted cavities was regularly observed in the irradiated Fe-Cr alloys. Such examples are shown in the TEM image of Fe10%Cr sample in Figs. 11a and 11b. These are expected to be heterogeneously nucleated cavities on the pre-existing dislocations in the matrix. Such cavities can pin the dislocation motion, forcing them to climb before they could glide freely. This behaviour is depicted in Fig. 11b in the arrangements A, B and C. In this image, the cavity arrangement is indicated by filled arrows and the position of the associated dislocation is indicated by dashed arrows. For arrangement A and B in Fig. 11b, we can see that the dislocation has perhaps partially climbed, while a part of it is still pinned by the cavities. Arrangement C shows a situation where the dislocation seems to have climbed fully and then glided away, leaving the linearly arranged cavities behind. In these micrographs, the imaging was performed in out-of-focus conditions slightly far from Bragg condition. Due to this reason, the dislocation contrast is not strongly visible.

TEM analysis also revealed strong He induced cavitation on the grain boundaries in Fe12%Cr and Fe3%Cr. These were the only FIB specimens which had intercepted a grain boundary. The corresponding images are shown in Figs. 12a and 12b respectively. The way the cavities were nucleated was largely different in the intercepted grain boundaries in these two specimens. For instance, we can see in Fe12%Cr case that the cavities show an alignment across the width of the grain boundary (see Fig. 12a). It could be due to heterogeneous nucleation of cavities on the dislocations constituting a grain boundary. However, in Fe3%Cr, such alignment did not exist. In this case, cavities along the grain boundary were significantly larger in size as compared to those in the matrix (see Fig. 12b). We could argue that the nature of the grain boundary in this case was different from that in Fe12%Cr, which yielded a different style of cavity nucleation. Due to the complexity of the study, we have not attempted to perform a detailed analysis to identify the grain boundary misorientation in both the cases.

4. Discussion

4.1 Depth dependency of cavity microstructure: combined influence of injected interstitial effect and helium implantation around damage peak

The depth dependent cavity microstructure was studied in high purity Fe-Cr alloys by TEM, including a detailed quantitative analysis of Fe14%Cr alloy. The peculiar aspects of the cavity microstructures were: (i). A narrow size distribution of cavities in the areas prior to the damage peak (up to 550-600 nm depth, see Fig. 6), with average diameters less than ~2-3 nm with . the number density of cavities and void swelling increasing continuously from near-surface areas up to the damage peak. (ii) A sharp increase in the cavity sizes, reduction of cavity number density but a continuous increase of the void swelling due to larger cavity sizes at and after the damage peak and (iii) a

dramatic change from the homogeneously nucleated cavities before the damage peak to cavities mostly located inside the dislocation loops after the damage peak. For the areas before the damage peak, void swelling values remained low (less than 0.02%), much lower than swelling reported in high purity bcc Fe irradiated under same irradiation conditions [33]. This further highlights the importance of swelling reduction due to Cr addition in Fe. However, the observation that the estimated swelling values were higher at and beyond the damage peak as compared to areas before the damage peak is in striking contrast to many ion irradiation results on Fe based alloys and other metals cited in the introduction where strong swelling suppression because of the injected interstitials is usually reported adjacent to the damage peak. One such example is the recent ion irradiation experiments in high purity bcc Fe irradiated to same dose and temperature as in the present study reporting nearly one-third reduction of void swelling at the damage peak as compared to the mid-range [33]. Similar trend was noted by the authors when the irradiations were performed in dual beam mode where He was co-implanted and in both cases the reduction in swelling around the damage peak was attributed to an evidently large reduction of cavity size and cavity number density due to the injected interstitial effect. Therefore, the question arises why swelling suppression around damage peak was not observed in Fe14%Cr and in the other Fe-Cr alloys in our case? The areas after the damage peak (700-1000 nm depth) showing higher swelling magnitudes as compared to the damage peak itself is unphysical even in the absence of injected interstitials because the dose in these areas reduce rapidly (see Fig. 1). 700-1000 nm depth is also lying heavily within the SRIM collision broadened estimate of the injected Fe ion profile (Fig. 1). In other words, highest swelling in our case was noted in areas after the damage peak where the dpa level is lower than at the damage peak, where the dpa level is reducing rapidly

and where the injected ion contribution is expected to be extremely high. A combination of all these points suggest there is something we clearly don't understand because the result cannot be well-explained by conventional means. To address this, we compared the results obtained in bcc Fe by Bhattacharya et al. [33] to Fe14%Cr in the present study, because both these samples were irradiated simultaneously using the same experimental configuration to same dose, dose rate and He implantation level. BF TEM images of cavities from midrange region and after the damage peak reported in the pure Fe sample, adapted from Refs.[4,33,46], is given in the Supplementary Materials. This comparison is justified because high-purity Fe being a very simple material free from the effects of major alloying elements and therefore if there were any obvious artefacts or unknowns they are expected to be better revealed. Fig. 13 plots the variation of average cavity size along the specimen depth extracted from the results in bcc Fe and the average cavity size observed in Fe14%Cr in our study. In the zones before the damage peak (before 500 – 600 nm depth), we can see that size of cavities in Fe were much larger than in Fe14%Cr (see Fig. 13). This is not surprising because Fe-Cr alloys have much higher swelling resistance as compared to bcc Fe [4]. Upon reaching the damage peak, the cavity sizes reduced in Fe and increased in Fe14%Cr. But, unexpectedly the average cavity size in both these materials reached almost the same value at and beyond the damage peak region. This area is highlighted by a red circle in Fig. 13. Additionally, the number density of the cavities in these areas in both materials were comparable. This is unforeseen because Fe-Cr alloys are highly resistant to swelling and are not expected to show remarkably similar quantitative cavity microstructures to Fe if irradiated to same irradiation conditions. Based on this comparison of results, we propose a hypothesis suggesting that the observed microstructure developed because of a combined influence of the injected interstitial

effect and He implantation around the damage peak. In Fig. 1, it is evident that maximum He is also implanted in the specimen close to the damage peak. The results in bcc Fe from Bhattacharya et al. showed that cavities were strongly suppressed due to injected Fe ions for the same irradiation conditions. It is reasonable to expect that injected interstitial effect would also exist in Fe-Cr alloys and might not be strongly perturbed by different Cr levels of these materials. To the best of our knowledge, a direct evidence of injected interstitial effect in Fe-Cr alloys is not present in the literature, but it is indeed observed in Fe-Cr based steels [50,51], and hence there is no reason to believe that injected interstitial effect would not play a role in the Fe-Cr binaries. Hence, as expected, the injected interstitial effect would reduce the vacancy supersaturation in the Fe14%Cr that would be present otherwise around the damage peak, while the material continuously gets implanted by He with major portion lying around the damage peak. Consequentially, the assumption that the injected interstitial effect is always present and the co-existence of the injected interstitial effect with simultaneous He implantation synergistically puts the areas around the damage peak in a situation where He to vacancy ratio will artificially increase (to values higher than those obtained by SRIM). The irradiation conditions can be visualised as transitioning towards conditions more similar to pure He implantation experiments where vacancy concentrations are usually low (due to low dpa imparted by He). We therefore believe that the cavities which we experimentally see in our case in the Fe-Cr alloys beyond the damage peak are formed simply as an effect of the excess He implantation (He filled bubbles), with negligible contribution from the ballistic damage produced by the Fe ions (because of the injected interstitials). This hypothesis is supported by the observation that the average size of cavities were similar in Fe and Fe14%Cr beyond the damage peak area because Cr is not expected to drastically modify

He implantation induced bubbles (not to be confused with voids on which Cr has a strong suppression effect). As discussed in the next section, the peculiarities of the spatial distribution of the cavities in this region also strongly resemble microstructures observed with significantly high He/dpa ratios or under pure He implantation experiments and therefore further support our hypothesis. The observations are therefore an indirect consequence of the injected interstitial artefact. This highlights that unpredictable microstructure evolution could occur in dual beam irradiations around the damage peak, that may not necessarily resemble the classical injected interstitial effect and this is imperative to be taken into account when analysing a material. We also highlight the value in always adding a reference bcc Fe when studying Fe-Cr alloys or Fe-Cr based steels because if there were no complimentary results available in bcc Fe for this study, one would wrongly interpret Fig. 5 in saying that there appears to be no significant injected interstitial effect. We believe this observation is more critical for a highly swelling resistant alloy in dual beam conditions where cavities produced in the midrange due to ballistic damage maybe much smaller in size than those around the damage peak occurring primarily because of He implantation. For a swelling prone material like bcc Fe or face-centred cubic Fe-Ni-Cr based austenitic steels, we believe this synergistic effect will be easy to detect with less possibility of wrongly interpreting the data because in high dose experiments ($> 50-100$ dpa), midrange cavities are expected to be much larger in these materials compared to He implantation induced cavities at and after the damage peak, as seen in Ref. [33].

4.2 Spatial distribution of cavities: heterogeneous nucleation inside dislocation loops, dislocation lines and grain-boundaries

The spatial distribution of cavities in the Fe-Cr alloys was not the same in the areas before and after the damage peak. In the regions between the irradiated surface and the damage peak the cavities were homogeneously distributed in the matrix while the regions just after the damage peak consisted of a mixture of freely nucleated cavities in the matrix and heterogeneously nucleated *cavities inside the dislocation loops*. The cavities inside the loops were in relatively high proportion as evidenced from the TEM images in Figs. 3 and 9. We believe the difference in the observed cavity microstructures before and after the damage peak was highlighted by the presence of a different dislocation loop microstructure existing in these two regions and the presence of high concentration of He around the damage peak. In the zones after the damage peak, the dose level and dose rate decreases rapidly. Due to this reason, the density of dislocation loops in these areas was not very high to an extent that individually resolvable loops were present, as can be seen in Fig. 7 for Fe14%Cr and in Fig. 10d for Fe12%Cr. Thus, the correlation between the cavities and the dislocation loops was straightforward to see by TEM. On the other hand, in the regions before the damage peak in Fe14%Cr, the microstructure consisted of an entangled dislocation network (see Fig. 7, Inset B). Similar network dislocations were noted in Fe(3, 5, 12)%Cr studied under same irradiation conditions in [46]. Thus, if any correlation existed between the cavities and the dislocation loops, it could not be seen because individually resolvable loops did not exist any longer. Nevertheless, the striking observation is the presence of cavities inside the dislocation loops. Similar microstructure development was reported in dual beam irradiated bcc Fe at 500 °C by Brimbal et al. [52] and in the pure He implanted depth of 3.34 μm in the Fe-Cr alloys in the present study (which are reported in Ref. [46]). For the latter case, the corresponding TEM images from Fe10%Cr and Fe12%Cr are reproduced in Fig. 14 from

Ref. [46]. In the case of Brimbal et al., the irradiation was performed on TEM thin foils up to 0.92 dpa (~0.45 dpa by Kinchin Pease option), implanted simultaneously with ~540 appm He. This gives a He/dpa ratio of ~1200. From the dual beam results of Brimbal et al. and from only pure He implantation (Fig. 14), it therefore appears that this situation is usually observed in bcc Fe and Fe-Cr alloys when the He amount is rather high with respect to dpa (vacancies). It further strengthens the argument of the previous section that around damage peak, the microstructure was mostly He induced because of an indirect effect of injected ions. In accordance with the hypothesis formulated by Brimbal et al, we believe a potential mechanism for the existence of cavities inside the dislocation loops is as follows: it is well-known that He atoms can arrive at the core of the dislocation loops during irradiation where they can be trapped. Their trapping is expected based on theoretical results of positive binding energy between helium, helium-vacancy clusters with $\frac{1}{2}\langle 111 \rangle$ type dislocations [53–56] and $\frac{1}{2}\langle 111 \rangle$ type dislocation loops [57], The mobility of these trapped He atoms by “pipe-diffusion” can result in the formation of He-V clusters at the loop core [52]. As the loop grows by preferential absorption of interstitials, these He-V clusters are left behind. Since He is insoluble in Fe matrix [58,59], these clusters are stable which can grow by absorption of He atoms /vacancies, and are then observed in TEM as cavities on the loop plane. This behaviour is solely induced by the presence of He. Based on our result and those observed in bcc Fe, we conclude that the behaviour is similar between high purity bcc Fe and Fe-Cr alloys. One must note here that except for the present study and the results of Brimbal et al. [52], cavities inside dislocation loops were not reported in many decades of research on Fe, Fe-Cr alloys and Fe-Cr based steels. The present study and by Brimbal et al. also happens to be on materials having very high purity (in terms of elements like C, N, O, Si), which was generally not

the case for a plethora of published results. C, N, O are well-known to interact strongly with vacancies [60,61], consequentially modifying the cavity microstructures [62]. Therefore, apart from the presence of large He concentration and large individually resolvable dislocation loops (high irradiation temperature), it appears the purity level of the material might also play a role on this peculiar microstructure development. We believe this potential aspect of sample purity on irradiation effects demands dedicated research effort for the sake of a fundamental understanding.

Apart from heterogeneous nucleation inside dislocation loops, strong segregation of cavities was seen on the grain boundaries and on pre-existing dislocation lines much further away from the damage peak. For the grain boundaries, the segregation seemed dependent on its orientation, as reported theoretically [63]. For instance, in Fe12%Cr, the cavitation showed an alignment along the width of the grain boundary (Fig. 12a), reflecting that nucleation seems to have occurred heterogeneously on the dislocation lines constituting a grain boundary. On the other hand, no such alignment existed in the grain boundary intercepted in Fe3%Cr. Moreover, all the pre-existing dislocations beyond the peak damage zone were also decorated with cavities. Such phenomenon can block dislocation motion and, thereby, increase hardening of the materials. In many cases, the associated dislocation was seen perhaps trying to climb before it could glide freely (see Fig. 6 and Fig. 10b). These are expected results. It is well known that in metals irradiated at high temperature, nucleation of cavities take place at different interfaces and on dislocations due to He [59]. This is observed in many industrial F-M steels [42 - 44]. For example, Henry et al. [64] studied 9%Cr martensitic steel after implantation with 23 MeV α -particles at 550 °C up to a He concentration of 0.5 at.%. In this experiment, cavities were detected in many different defects in the materials: pre-austenitic grain boundaries,

lath boundaries, dislocations and matrix-carbide interfaces. It corresponds well with the fact that, in general, grain boundaries act as neutral sinks to point defects, but the presence of He makes them as preferred nucleation sites for He-V clusters.

Here, one must note that during ion-irradiation experiments, carbon contamination is a frequently encountered issue [65]. Carbon is known to interact with vacancies and in experimental situations this can induce changes in the irradiated microstructures [60,66]. In our case, carbon effect was minimized by working on very high purity alloys, a thorough cleaning of samples and the stage using pure ethanol prior to irradiations, and maintaining chamber pressures at very high vacuum (typically $\sim 10^{-7}$ torr). Nevertheless, some carbon pick-up is always expected. Recent work by Monterrosa et al. [66] in HT-9 steel has shown that the role of carbon is to reduce the cavity sizes and increase their number density, which caused reduced void swelling magnitudes. In our case, all the samples were irradiated simultaneously implying similar contamination, if any, across the samples. Therefore, although the absolute swelling values might get affected, the comparative swelling trends and magnitudes between bcc Fe and the FeCr alloys are robust. It is because if carbon were solely responsible for the results, one would have expected similar depth dependent swelling trend between Fe and FeCr alloys which is clearly not the case here.

5. Conclusions

The depth distribution of irradiation induced defects was studied after dual beam irradiations on high purity Fe-Cr alloys at 500 °C, up to 157 dpa, co-implanted with 17 appm He/dpa at the damage peak. Between the sample surface and up to the damage peak, a homogeneous distribution of small cavities was observed by TEM, whose number density increased along the damage depth. However, a drastic change in cavity

microstructure was detected at and after the damage peak. The cavity sizes increased and number density decreased. The microstructure consisted of a mixture of heterogeneously nucleated cavities inside the dislocation loops and freely nucleated cavities in the matrix. Because cavities inside dislocation loops have not been generally reported in studies on Fe, binary Fe-Cr alloys/Fe-Cr based steels except here and in a published result on high purity Fe, it appears that sample purity is also an essential ingredient (in conjunction with high He concentration and large dislocation loops) that is needed to observe this peculiar microstructure development.

Quantitative results revealed that cavity size and void swelling was higher at and beyond the damage peak as compared to regions before the damage peak. Such a behaviour is contradictory to most ion irradiation results in metals where a strong suppression of void swelling occurs adjacent to the damage peak due to the injected interstitial effect. By comparing the results in the literature in Fe irradiated to same irradiation conditions, a plausible mechanism of the observed swelling variation in Fe-Cr alloys is discussed reflecting a synergy between the injected interstitial effect and He implantation near the damage peak where injected interstitials suppress cavities/vacancy supersaturation, thereby artificially increasing the He to vacancy ratio and promoting purely He induced cavity nucleation. This highlights the possibility of unpredictable microstructure evolution in dual beam irradiations around the damage peak, which may not resemble the classical injected interstitial effect at first glance.

We also highlight that its always beneficial to add a reference bcc Fe when studying Fe-Cr alloys or Fe-Cr based steels because comparing results with Fe in most cases may help better identify any potential artefacts, as revealed in the present study.

The depth variation of the dislocation loop microstructure was also characterized. Up to the damage peak, a complex dense network of dislocations formed due to the interaction/impingement of the dislocation loops. Individually resolvable loops were only observed after the damage peak region, where their association with cavities was noted. A significant presence of He also induced heterogeneous cavity nucleation on the grain boundaries and pre-existing dislocation lines in regions much further away from the damage peak.

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Table 1: Nominal composition and mean grain size of the alloys.

Material	Cr wt. %	C wt. ppm	Si wt. ppm	O wt. ppm	N wt. ppm	P wt. ppm	Mean grain size μm
Fe3%Cr	3.05	4	2	6	2	X	390
Fe5%Cr	5.40	4	3	6	2	< 5	68
Fe10%Cr	10.10	4	4	4	3	< 5	82
Fe12%Cr	11.63	5	2	4	X	X	500
Fe14%Cr	14.25	5	7	4	5	< 10	141

Table 2: Details of the irradiation conditions using 2 MeV Fe²⁺ and energy degraded 2 MeV He⁺ ions at 500 °C.

Material	2 MeV Fe ²⁺		2 MeV He ⁺		Peak dose and dose rate	He/dpa (at peak)
	Fluence <i>ions.m⁻²</i>	Flux <i>ions.m⁻².s⁻¹</i>	Fluence <i>ions.m⁻²</i>	Flux <i>ions.m⁻².s⁻¹</i>		
Fe3%Cr						
Fe5%Cr					157 dpa	17appm
Fe10%Cr	1.38x10 ²¹	~3x10 ¹⁶	8.4x10 ²⁰	1.72x10 ¹⁶	3.3x10 ⁻³ dpa/s	He/dpa
Fe12%Cr						
Fe14%Cr						