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CHARACTERIZATION OF DISLOCATION TYPE DEFECTS FORMED AT LOW-ENERGY DEUTERIUM IRRADIATION OF SS316 STAINLESS STEEL

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The nature of small point defect clusters in SS316 austenitic stainless steel irradiated with deuterium ions at room temperature was identified from their behavior under annealing. In this analysis, the defect clusters which evolved due annealing were judged to be of interstitial (I)-type and of vacancy (V)-type. The standard technique for Burgers vector determinations was used and shown that the interstitial type defect clusters are Frank faulted dislocation loops having Burgers vector **b** = a/3<111> normal to the {111} plane. These results are discussed and compared with the results of studies of microstructural changes in austenitic stainless steels irradiated with fission neutrons.

INTRODUCTION

Austenitic stainless steels have been the subject of numerous investigations on radiation effects. Most of these studies have been driven by technological applications, ranging from light water and fast breeder fission reactors to fusion reactor first-wall structures. There are three major categories of transient behavior that can be identified in irradiated metals: point defect (vacancy and interstitial) supersaturation; microscopic nucleation of defect aggregates such as dislocation loops, voids and precipitates; and macroscopic changes such as void swelling and mechanical property [1]. A well understanding of the accumulation and evolution of point defects will allow us to better understand the mechanism of degradation and predict the mechanical properties of irradiated materials.

Simulation experiment in iron has proved the bias effect of dislocations that they absorb more interstitial atoms than vacancies [2, 3]. Dislocation loops are also considered as bias sinks and a main reason of irradiation swelling. The nature and Burgers vectors of the loops have an impact on their bias and thus influence the mechanical property. A computer simulation predicted that interstitial loops would contribute a larger effect on radiation hardening than vacancy loops [4], so it's of importance to partition the different types of dislocation loops to investigate the behavior of radiation damage in materials [5].

At elevated temperature and dose, both the defects created by irradiation and the helium and hydrogen gas produced by nuclear transmutations are likely to induce microstructural changes that could lead to swelling of the material, possibly resulting in its embrittlement. Hydrogen embrittlement is a severe environmental type of failure that affects almost all metals and their alloys. A good understanding of the dynamic behaviors of hydrogen isotopes introduced by fuel bombardment or formed by nuclear transmutation, such as permeability, diffusion, interaction with radiation defects and retention properties, is greatly important for considering the mechanism of degradation of material's properties.

Because of a very low solubility of hydrogen isotopes in stainless steels, the presence of various lattice defects (vacancies, vacancy clusters, voids, dislocations, grain boundaries, impurities) strongly influences hydrogen isotope retention. Therefore, parameters of trapping are essential for predicting hydrogen isotope transport in structural materials. The defects of dislocation type are often regarded by researchers as possible trapping sites of hydrogen isotopes in metals. In particular, Ono [6] for Fe -9Cr - 2W ferritic alloy clearly demonstrated deep correlation between the TDS of deuterium gas and dislocation loop annihilation. According to [6], deuterium ions irradiated on the specimen were trapped at dislocation loops and affects their thermal stability.

In our previous paper experimental investigations of the hydrogen-defect interaction are performed by thermal desorption spectroscopy (TDS), and the parameters of the interaction are obtained by fitting numerical calculations based on diffusion-trapping codes to experimental thermal desorption spectra. TEM results show that implantation of deuterium in SS316 steel is accompanied by the creation of dislocation type defects. System of dislocation defects have been observed within the depth layer corresponding approximately to the ions range profile. Nevertheless, microstructure remains almost unchanged throughout the temperature range where the gas desorption process is observed. The coalescence of dislocation defects began at annealing temperatures of about 1000 K and well-resolved loops of dislocations were observed, which finally disappeared at ~ 1200 K. Consequently, the presence of radiation induced dislocations in steel and thermally activated release of deuterium do not exhibit a distinct correlation [7].

The goal of the present study is to examine the defect structure produced in the near-surface region of SS316 austenitic stainless steel irradiated with low energy deuterium ions and to determine the reason of stability of the observed dislocation structure.

1. MATERIAL AND METHODS

Material investigated in this study is stainless steel of 300 series commonly used for core internals of PWR namely SS316. The material was solution annealed during 30 min at 1320 K. Their chemical composition is given in Tabl. 1.

Table 1

Chemical composition of SS316 steel, wt.%.

C	Si	Mn	P	S	Cr	Ni	Mo	Ti	Fe
0.056	0.68	1.6	0.034	0.014	16.68	12.03	2.40	<0.01	bal.

Samples for TEM studies were prepared as disks of 3 mm in diameter. Thin foils were obtained by mechanical thinning of the disks down to 130 μm followed by electropolishing and short-term annealing. The samples were implanted with 12 keV D_2 (6 keV/D) ions to the fluencies in the range of $(1…3)\cdot10^{20}$ D/m^2. The ion flux on the sample was in the range of $(1…2)\cdot10^{18}$ $D/(m^2\cdot s)$. Ion implantation was performed at normal ion incidence, and the sample temperature did not exceed 300 K [7].

After irradiation specimens of stainless steel 316 were thinned for transmission microscopy by electropolishing with a solution of 5% perchloric acid in ethanol from back side of sample until perforation.

Ion stopping distribution for 6 keV deuterium and irradiation damages (in dpa) in stainless steel have been calculated with the software The Stopping and Range of Ions in Matter (SRIM 2008) [8]. The dpa calculations are based on a displacement energy threshold of 40 eV and on the Kinchin-Pease formalism and Stoller recommendations [9]. According to SRIM calculations it is estimated that the center of the investigated TEM foils approximately coincides with the maximum of deuterium depth distribution profile as shown in Fig. 1. The thickness of the foils was not measured directly but assumed to be in the range of 100…150 nm according to their brightness in electron microscope.

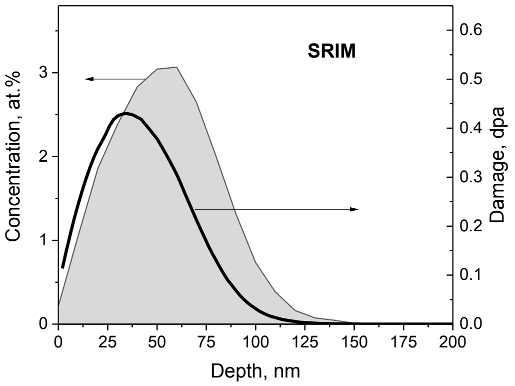


Fig. 1. Calculated 6 keV deuterium profiles of damage and concentration for irradiation fluence of $2\cdot10^{20}$ D/m^2

The dislocation loops were examined using bright field (BF) or dark field (DF) images in two-beam conditions. Electron microscope JEM-100CX was used to perform TEM characterization for all specimens.

2. RESULTS AND DISCUSSION

Fig. 2 shows the microstructure of initial and irradiated SS316. The initial structure consists of austenite grains with average size of about 30 μm. Precipitates of second phase (carbides and carbo-nitrides) and dislocations (~ 10^8 cm^{-2}) are seen in grains.

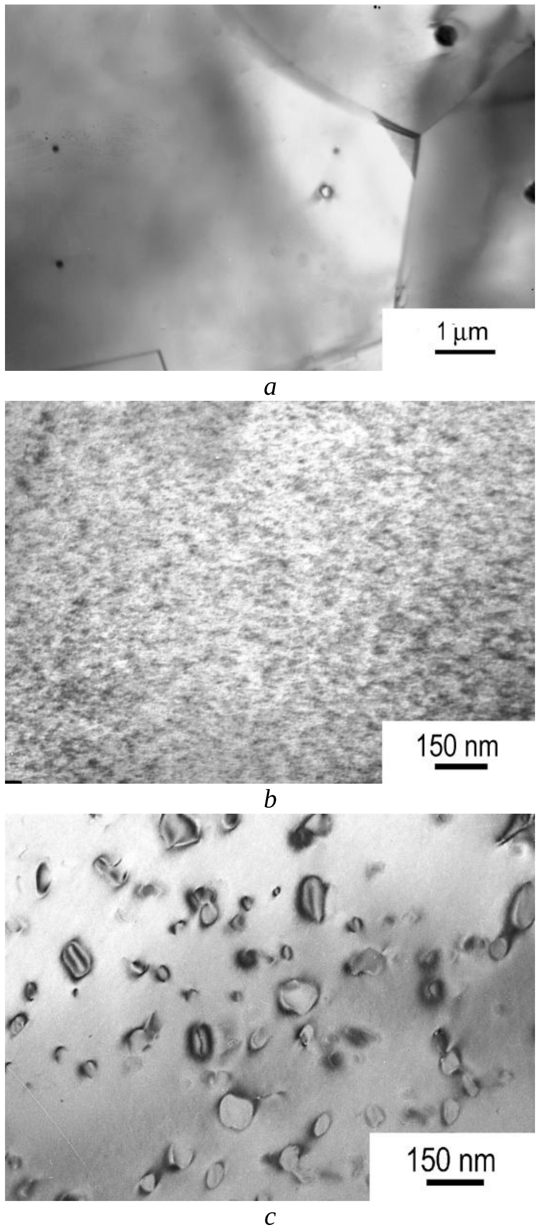


Fig. 2. Microstructure of SS316 steel un-irradiated (a) and irradiated with 12 keV D_2^+ ions to a dose of $2\cdot10^{20}$ D/m^2 at room temperatures (b) and annealed at 920 K (c)

High density of small clusters formed in SS316 specimens after deuterium ion implantation at room temperature, as seen from Fig. 2,b. These tiny defect clusters may be of interstitial type, or vacancy type, but are usually too small to be easily identified as a particular type of cluster. The density of these clusters depends on the irradiation temperature, beam flux and the final fluencies.

At low temperatures, interstitial atoms are mobile and can cluster or migrate to sinks, such as grain boundaries and surface, whereas vacancies are much less mobile and they can only annihilate or aggregate with neighboring vacancies to form vacancy clusters unresolvable by TEM. When annealed at higher temperature, interstitial clusters will migrate and coalesce to form dislocation loops, with low density and large loop size. Meanwhile, vacancy clusters also become mobile and start to migrate, resulting in the annihilation of interstitial clusters and the formation of vacancy loops [5].

Fig. 2,c shows the microstructure evolution after annealing of SS316 specimen at 920 K in high vacuum for 15 min. It is expected, that at such temperatures both interstitial and vacancy clusters achieve sufficient mobility for migration, that can result in the mutual annihilation or formation of interstitial and vacancy loops, respectively. Fig. 2,c shows that after annealing at 920 K small point defect clusters were grow resulting in formation of dislocation loops.

We have taken steps focused on the characterization of defects produced in the near-surface region of SS316 austenitic stainless steel irradiated with low energy deuterium ions in particular on the experimental determination of the nature of the observed loops and their Burgers vector.

The principle underlying the most popular technique for the determination of the Burgers vector of dislocation is as follows. Local bending of crystal planes around the dislocation change their diffraction condition. This produces a contrast in the image that can be used in the **g**·**b** analysis for Burgers vector. Planes parallel to the Burgers vector are not distorted by the dislocation, so these planes show no change in contrast – a condition corresponding to **g**·**b** = 0. This condition is valid for perfect dislocations in the implementation of the two-beam approximation, when only two electron beams are intense – a direct and one diffracted. According to the classical approach, for the determination of the direction of the Burgers vector of dislocation it is necessary to find two vectors $\mathbf{g_i}$ for which the dislocation contrast vanishes. Then take the cross product between the two **g**-vectors to find the direction of **b**. In practice, to determine the Burgers vector, a series of images of the same area is usually made at different slopes of the crystal with respect to the electron beam, i.e. for different **g**. It is necessary to obtain such a series of photographs, so that on one of them the dislocation was visible, and on others the extinction of contrast took place. Then, using the condition **g**·**b** = 0 and taking into account that the dislocation image is present for **g**·**b** ≠ 0, we can find the direction of the Burgers vector.

Fig. 3 shows a set of images, along with diffraction patterns, for the two-beam orientation. Two-beam conditions were obtained by tilting from the [011] zone axis orientation.

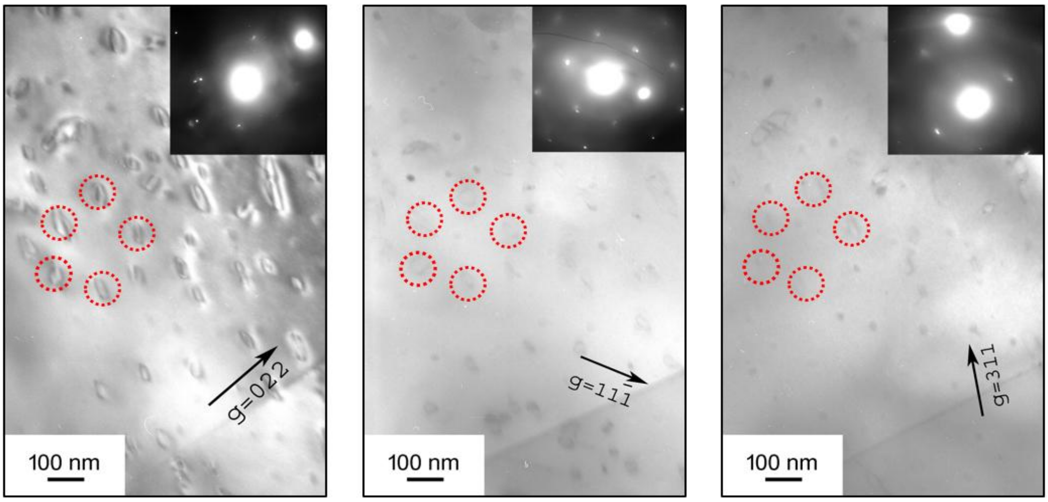


Fig. 3. Changes in the contrast of the image showing Frank loops by varying ***g*** *= between 022, $11\bar{1}$, and 311 for the specimen irradiated with 12 keV D_2^+ ions to a fluence of $2\cdot10^{20}$ D^+/m^2 at room temperature and heated up to 920 K*

The dislocation loops are faulted or perfect and generally form on closed packed planes in FCC materials (see Fig. 2,c).

Frank partial dislocations shown in Fig. 3 outlines a stacking fault formed by inserting or removing a region of close packed {111} plane. Frank partial has Burgers vector **b** = a/3<111> normal to the {111} plane.

In the discussion above, dislocations have been considered to be invisible if the product **g·b** = 0; those dislocations with **g·b** = 1 and **g·b** = 2 are have to be visible. The magnitude of the product of **g·b** for dislocations with small Burgers vectors, such as partial dislocations in FCC materials, can be greater than zero, but have a fractional value less than one. Computer simulations using the dynamical theory of electron diffraction have shown that dislocations with Burgers vectors such that **g·b** < 1/3 are essentially invisible [10].

Tabl. 2 lists the values of the products **g**·**b** for all four possible of the Frank loop variants.

Table 2

The values of the products **g**·**b**

b	**g**		
	(111)	(311)	(022)
a/3 [111]	1	5/3	4/3
a/3 [11$\bar{1}$]	1/3	4/3	0
a/3 [1$\bar{1}$1]	1/3	4/3	0
a/3 [$\bar{1}$11]	1/3	-1/3	4/3

According to Tabl. 2, the invisibility conditions are satisfied for the Burgers vector in the last entry. Thus, it was found that the Frank dislocation loops in specimens pre-irradiated with deuterium ions at room temperature and annealed at 920 K have $\mathbf{b} = a/3\,[\bar{1}11]$.

The Burgers vector and the nature of the dislocations determine the magnitude of the fluxes of point defects absorbed by the dislocation and, consequently, the stability of the dislocation structure. From the point of view of further microstructure evolution, as well as radiation stability, this is very important, since the conservative nature of Frank loops limits the rate of dislocation microstructure evolution, while other dislocation components can glide as easily as climb.

To identify the nature of dislocation loops, inside-outside method is often used from the contrast of dislocation loops in bright-field images, which is reliable for dislocation loops with size larger than 10…20 nm. This experimental method is based on changing the dimensions of loop image under different diffraction conditions [11]. The image of a loop on a microscopic picture can be located inside or outside its true contour created by the dislocation core in the crystal. The image must lie entirely inside the loop for the case $(\mathbf{gb})\,\mathbf{s} > 0$ ($\mathbf{s}$ – "the deviation error" that measures deviation from exact Bragg condition) and entirely outside for $(\mathbf{gb})\,\mathbf{s} < 0$. Additionally, it is necessary to determine the orientation of the loop in the foil.

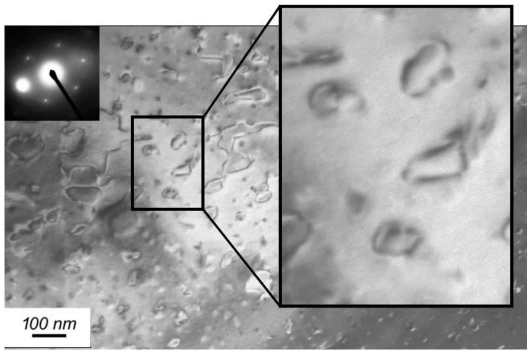
The intensity of the images of the defects located on opposite surfaces of the foil turns out to be different at $\mathbf{s} \neq 0$ in a dark field due to the effects of anomalous absorption. On a dark-field photograph, for $\mathbf{s} >> 0$, the image of the defect at the lower surface of the foil is more intense, and for $\mathbf{s} << 0$ – at the upper surface. Thus, if the loop orientation in the crystal is known, and also **g** and **s**, then the component **b** along **g** can be uniquely determined, noticing the change in the size of the loops when changing the sign of **g** or **s**.

Using above approach, photomicrographs were obtained (Fig. 4) with the conservation of $\mathbf{s} >> 0$ and diffraction vectors $\bar{2}00$ and 200. Some typical dislocation loops (an enlarged area on Fig. 4) were characterized to be interstitial-type, since they showed growth when changing the sign of **g**. The size of loops was measured as the length of transversal axis.

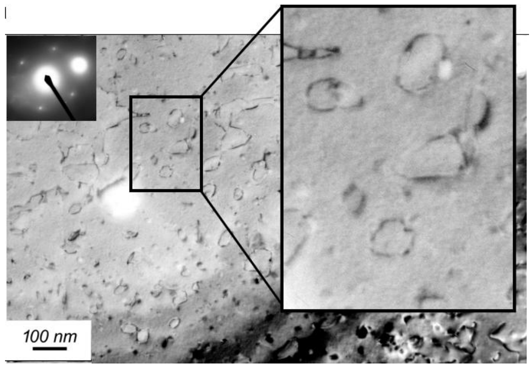
Loops showing shrinkage at the same diffraction conditions are also observed in irradiated specimens. However, their concentration is significantly lower under the current experimental circumstances. More detailed studies are needed to establish the ratio of interstitial and vacancy loops in hydrogen irradiated stainless steels.

The microstructural changes in austenitic stainless steels due to neutron irradiation have been reviewed in [1, 12, 13]. Their authors concluded that at temperatures below 570 K the radiation-induced microstructure consists of a mixture of unidentified "black spot" damage and Frank loops (faulted loops with $\mathbf{b} = a_o/3[111]$ with the density of Frank loops saturating at slightly less than 10^{23} m^{-3} at doses less than 1 dpa. The Frank loops are generally considered to be interstitial loops, whereas the "black spot" damage is thought to be vacancy in nature, perhaps stacking fault tetrahedra [14].

In more recent study Edwards et al. [15] characterized the microstructure in a 316SS and 304SS irradiated over a range of doses from 0.7 to 13 dpa at 550 K, showing that the Frank loops exist in sizes from 30 nm down to very small sizes of less than 1 nm. They concluded that the "black spots" are not a distinctly different component, but simply small Frank loops. They also found that the density of Frank loops was already saturated at ~ $1.5\cdot10^{23}$ m^{-3} in both alloys at the lowest dose measured. The loop size distribution continued to evolve up to several dpa as larger Frank loops appeared.



a



b

*Fig. 4. The change in the size of the loops when changing the sign of **g** with the conservation of **s**>>0 and diffraction vectors $\bar{2}00$ (a) and 200 (b)*

The defects formed in 316 stainless steel irradiated with low energy deuterium ions at room temperature, as shown in the present study, have a qualitatively similar character to neutron irradiation. Additional data on the influence of the irradiation temperature, ion energy and hydrogen ion flux will make it possible to perform a more detailed comparison of the microstructural changes caused by neutron and hydrogen-ion irradiation and to determine the role of hydrogen in the mechanisms of defect structure formation in stainless steels.

CONCLUSIONS

The nature of small point defect clusters produced in the near-surface region of SS316 austenitic stainless steel irradiated with low energy deuterium ions at room temperature was identified and the Burgers vector and the reason of stability of the observed dislocation structure were determined. The dislocation loops were examined using BF or DF images in two-beam conditions:

– the radiation induced defect clusters have evolved to dislocation loops at annealing;

– both interstitial-type and vacancy-type loops formed simultaneously in SS316 when annealed at temperatures 920 K;

– the conservative Frank fault loops have Burgers vector **b** = a/3<111> normal to the {111} plane.

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ХАРАКТЕР ДЕФЕКТОВ ДИСЛОКАЦИОННОГО ТИПА, ОБРАЗУЮЩИХСЯ ПРИ ОБЛУЧЕНИИ СТАЛИ SS316 НИЗКОЭНЕРГЕТИЧНЫМИ ИОНАМИ ДЕЙТЕРИЯ

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Определен характер дислокационных петель в аустенитной нержавеющей стали SS316, облученной ионами дейтерия при комнатной температуре. В этой работе изучена трансформация мелких дефектных кластеров, результатом которой стало образование дислокационных петель межузельного и вакансионного типов. Использовался стандартный метод определения векторов Бюргерса дислокаций. Показано, что дефектные петли Франка имеют вектор Бюргерса типа **b** = a/3 <111>, направленный перпендикулярно плоскостям {111}. Эти результаты обсуждаются и сравниваются с данными исследований микроструктурных изменений в аустенитных нержавеющих сталях при нейтронном облучении.

ХАРАКТЕР ДЕФЕКТІВ ДИСЛОКАЦІЙНОГО ТИПУ, ВИНИКАЮЧИХ ПРИ ОПРОМІНЕННІ СТАЛІ SS316 НИЗЬКОЕНЕРГЕТИЧНИМИ ІОНАМИ ДЕЙТЕРІЮ

Б.С. Сунгуров, Г.Д. Толстолуцька, С.О. Карпов, В.В. Ружицький, В.М. Воєводін

Визначено характер дислокаційних петель в аустенітній нержавіючій сталі SS316, опроміненої іонами дейтерію при кімнатній температурі. У цій роботі вивчена трансформація дрібних дефектних кластерів, результатом якої стало утворення дислокаційних петель міжвузельного та вакансійного типів. Використовувався стандартний метод визначення векторів Бюргерса дислокацій. Показано, що дефектні петлі Франка мають вектор Бюргерса типу **b** = a/3 <111>, спрямований перпендикулярно площинам {111}. Ці результати обговорюються і порівнюються з даними досліджень мікроструктурних змін в аустенітних нержавіючих сталях при нейтронному опроміненні.