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The effect of irradiation with inert gas and hydrogen ions on nanohardness of SS316 stainless steel

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The influence of gas ions irradiation (hydrogen, helium, argon) on nanohardness and microstructure changes in SS316 austenitic stainless steel has been studied. Samples were irradiated with 15 keV/D, 30 keV/He and 1400 keV/Ar ions at different temperatures. It has been found that irradiation at room temperature leads to the formation of dislocation structure in the steel, regardless of ion species. The formation of the bubble structure was observed after irradiation of SS316 steel with argon ions at 873 K. An increase of nanohardness of about two times was observed for ion irradiated steel. It was established that the main factor of hardening is the formation of radiation induced dislocation structure.
Keywords: ion irradiation, nanoindantation, hardness, microstructure, stainless steel.

Изучено влияние облучения газовыми ионами (водород, гелий, аргон) на изменение нанотвердости и микроструктуры аустенитной нержавеющей стали SS316. Образцы облучали ионами 15 кэВ/D, 30 кэВ/He и 1400 кэВ/Ar при различных температурах. Обнаружено, что облучение при комнатной температуре приводит к образованию в стали дислокационной структуры вне зависимости от сорта ионов. После облучения стали ионами аргона при 873 К наблюдается образование пузырьковой структуры. Обнаружено радиационно-стимулированное упрочнение стали более, чем в два раза. Установлено, что основным фактором упрочнения является формирование радиационно-индуцированной дислокационной структуры.
Ключевые слова: ионное облучение, наноиндентирование, твердость, микроструктура, нержавеющая сталь.

Вивчено вплив опромінення газовими іонами (водень, гелій, аргон) на зміну нанотвердості і мікроструктури аустенітної нержавіючої сталі SS316. Зразки опромінювали іонами 15 кеВ/D, 30 кеВ/He і 1400 кеВ/Ar при різних температурах. Виявлено, що опромінення при кімнатній температурі призводить до утворення в сталі дислокаційної структури незалежно від сорту іонів. Після опромінення стали іонами аргону при 873 К спостерігається утворення бульбашкової структури. Виявлено радіаційно-стимульоване зміцнення сталі більш, ніж в два рази. Встановлено, що основним чинником зміцнення є формування радіаційно-індукованої дислокаційної структури.
Ключові слова: іонне опромінення, наноіндентування, твердість, мікроструктура, нержавіюча сталь.

The rapid development of nuclear power has led to the elaboration of new generation reactors that are more safe, reliable and economical. Further implementation of these reactors requires the development of new radiation-resistant materials, because the behavior of structural materials in nuclear reactors determines the safeness of nuclear power station. An understanding of the effects of irradiation on the mechanical properties of the candidate materials is essential for such materials development initiatives.

In structural materials during their exploitation gas impurities, in particular, helium and hydrogen are accumulated contributing the appearance of helium and hydrogen embrittlement and gas swelling [1]. Typical concentrations of helium and hydrogen, which generated by the displacement of one atom (dpa) from its equilibrium position in the lattice (He/dpa, H/dpa) are << 1 for fast neutron reactors, about 10 for fusion reactors and ~100 for "spallation"-type installations [2,3]. So, it is needed to pay special attention to complex effects of hydrogen, helium and radiation defects on these materials.

Austenitic stainless steel SS316 is widely used as structural material in an II and III generation reactors due to the combination of its good creep resistance at high temperature and oxygen corrosion resistance. In addition, the vessel of «spallation» neutron sources is manufactured from SS316 steel. It is selected as a structural material for

ITER and considered as a candidate structural material for reactors with molten salt as the heat-transfer agent (MSR).

To simulate neutron irradiation damage in structural materials, heavy ion irradiation experiments have been performed because of the simplicity of use, easier control of irradiation parameters, reduction of cost, rapid damage production, the absence of induced radioactivity, and the occurrence of the co-implantation of helium/hydrogen [1–3]. On the other hand, ion irradiation has a significant drawback – shallow depth of damage layer that making it difficult to investigate the mechanical properties of materials. The solution of problem is possible by using nanoindentation method that provides a study of the mechanical properties of the samples in the near-surface region.

The goal of the present work is the investigation of changes of the hardening of SS316 steel and its microstructure after irradiation with deuterium, helium and argon ions.

Materials and methods

Specimens of SS316 steel with dimensions 27×7×0.1 mm were used for investigations. Before experiments the samples were annealed at 1340 K for one hour in a vacuum of ~10^{-4} Pa. Chemical composition of steel is shown in Table.

Samples were irradiated with 30 keV D_2^+ (15 keV/D^+) ions up to a dose of 3×10^{17} D/cm^{-2}, 30 keV helium ions to a dose of $5\cdot10^{16}$ cm^{-2} and 1.4 MeV argon ions to a dose of $1\cdot10^{17}$cm^{-2}. All irradiations were carried out with accelerating-measuring system “ESU-2” [4], consisting of Van de Graaf accelerator (high-energy argon irradiation) and two irradiation facilities for hydrogen and helium implantation. The irradiation with light ions was performed at room temperature, whereas argon irradiation was conducted at room temperature and at 873 K.

Studies of the steel microstructure were performed by transmission electron microscopy at room temperature, employing standard bright-field techniques on an EM-125 electron microscope at accelerating voltage 125 kV. Preparation of specimens to suitable for TEM thickness was performed using standard jet electro-polishing from unirradiated surface. The initial structure of SS316 steel is shown in Fig.1.

Nanohardness was measured by Nanoindenter G200 with a Berkovich type indentation tip. Tests were performed with a constant deformation rate of 0.05 s^{-1}. Each sample was applied at least 10 prints at a distance of 35 μm from each other. The methodology of Oliver and Pharr was used to find the hardness [5]. The details of nanoindentation tests have been presented elsewhere [6,7].

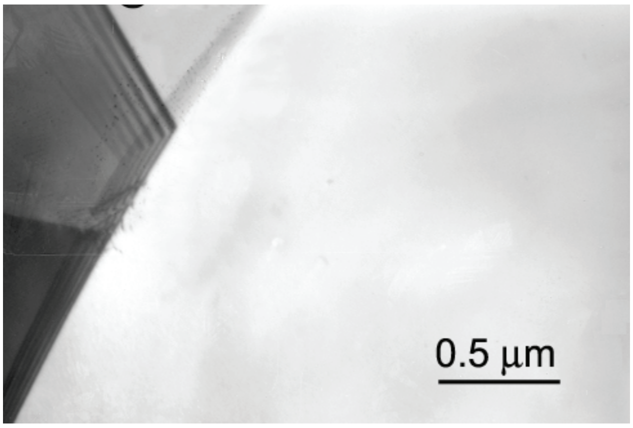


Fig. 1. The initial microstructure of SS316 steel after heat treatment at 1340K/0.5 h.

Results and discussion

In the present paper we have determined the nanohardness of SS316 steel in the initial state and after irradiation with deuterium and helium ions at room temperature, and argon ions at T_{room} and 873 K.

The depth distribution of gas atoms concentration and damage was calculated by SRIM 2008 [8] and shown in Fig. 2. The damage calculations are based on the Kinchin-Pease damage energy model, with a displacement energy of 40 eV for Fe and Cr, as recommended in ASTM E521-96 (2009) [9].

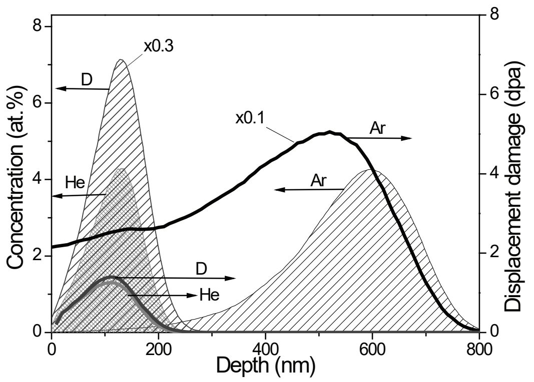


Fig. 2. Calculated profiles of damages and concentrations of D, He and Ar ions.

Calculated damage level of He and D was ~1.4 dpa. Herewith, the maximum of calculated deuterium

Table

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	бал.

concentration profile is almost 6 times higher than maximal concentration of helium – 25.3 and 4.22 at.%, respectively. On the other hand, at almost identical maximum concentrations of helium and argon, the ability of damaging of argon is more than 20 times higher in the depths 0-200 nm.

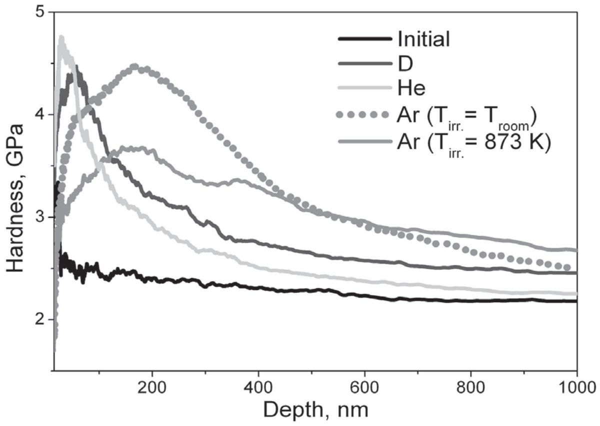


Fig. 3. Nanoindentation hardness of SS316 steel vs. indentation depth of the unirradiated sample, and samples irradiated with D_2^+, He^+, Ar^+ at T_{room} and 873 K.

Fig. 3 shows indentation-depth profiles of nanohardness of SS316 steel before and after ion irradiation with deuterium, helium and argon ions.

For the nonirradiated steel, the depth dependence of the hardness is relatively simple and close to constant. After the ion irradiation of speciments with D, He and Ar ions at T_{room} an increase of nanohardness of about two times is observed, independently of species of ions. It should be noted that G.S. Was et. al. analyzing the data of radiation induced segregation, irradiated microstructure, radiation hardening and IASCC susceptibility of the same heats of proton- and neutron-irradiated 304SS and 316SS have shown that the irradiation hardening of austenitic steels saturates at doses about a few dpa [10].

Irradiation of steel with Ar ions at 873 K also causes the increasing of nanohardness, but not such evident compared to irradiation at T_{room}.

Apparently, the observed increase in hardness of irradiated steel is associated with the hardening of the implanted layer. However, at hardness measuring by the nanoindentation technique should take into account the so called size effect, leading to deviations from the true hardness values. This effect consists in the increase of the hardness values in the region of small depths of indenter implementation while decreasing the pressing forces (Indentation size effect - ISE) [11].

To explain the ISE, Nix and Gao have developed a model based on the concept of geometrically necessary dislocation [12]. This model predicts the hardness depth profile as follows:

$$\frac{H}{H_0} = \sqrt{1+\frac{h^*}{h}} \quad , \qquad (1)$$

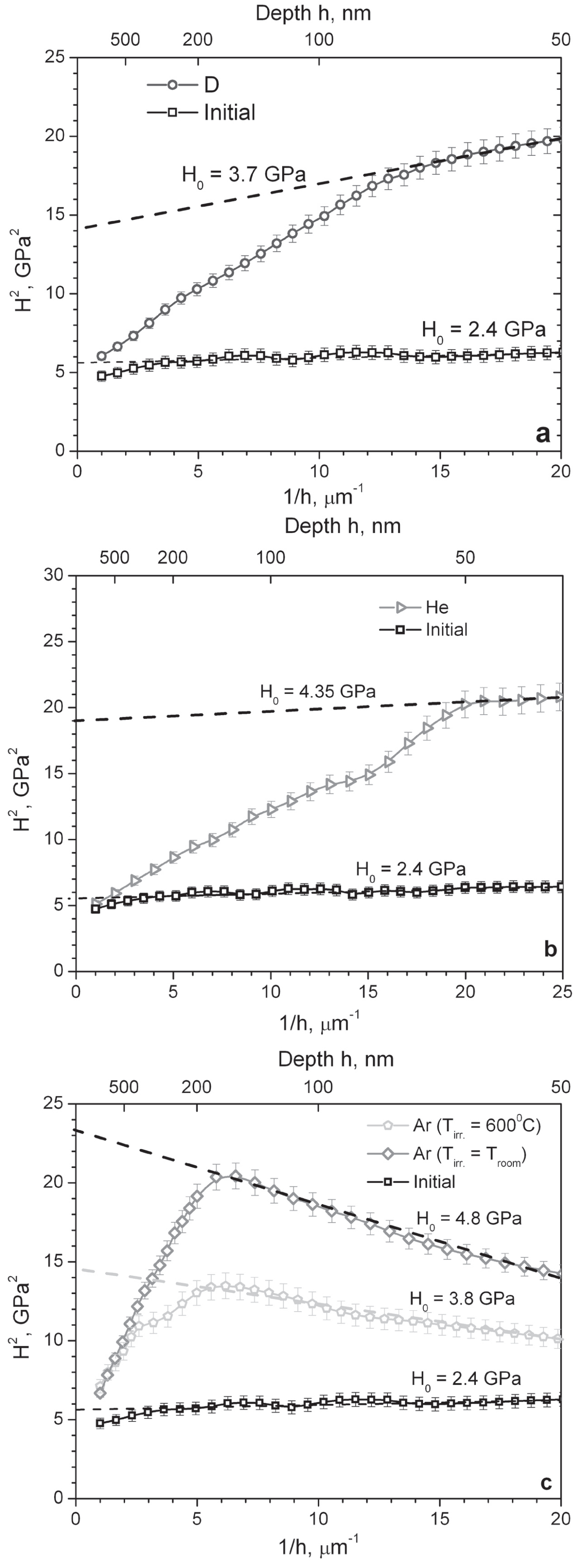
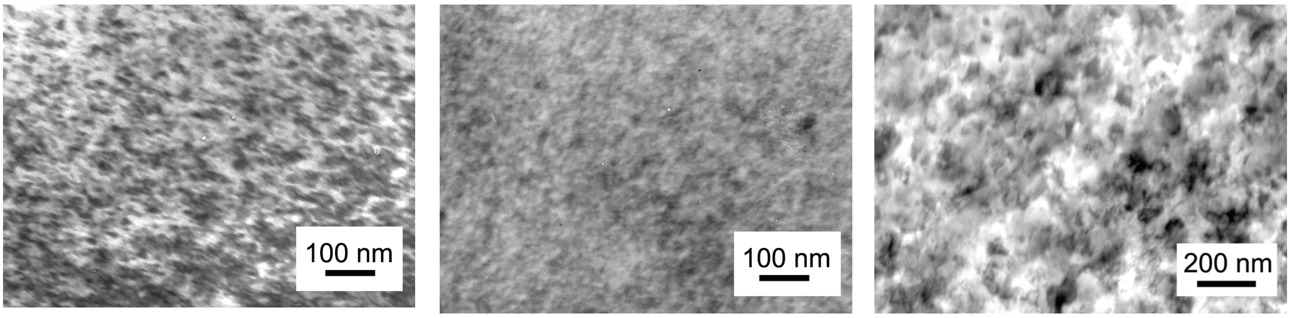


Fig. 4. The dependence of the square of the hardness on inverse depth introduction of the indenter for samples irradiated with deuterium (a), helium (b), argon (c) and the non-irradiated steel. The error bars show standard deviation.



a *b* *c*

Fig. 5. TEM images of the structure of SS316 steel irradiated by He (a), D (b) та Ar (c) at T_{room} .

where H is the measured hardness at the depth of h, H_0 is the hardness at infinite depth (i.e., macroscopic hardness), which will be referred to as the bulk hardness hereafter, h* is a characteristic length which depends on the material and the shape of indenter tip.

There are a few papers, in which radiation hardening is evaluated using the Nix and Gao model [13-16]. In particular, Kasada et al. have suggested a new model to extrapolate the experimentally obtained nanoindentation hardness to the bulk-equivalent hardness of ion-irradiated Fe-based binary model alloys [16]. This model is based on a combination of the Nix-Gao model [12] for the indentation size effect and a composite hardness model for the softer substrate effect (SSE) of the non-irradiated region beyond the irradiation range.

The hardness of bulk sample can be obtained if plot the graph H^2 (1/h). The square root of the value obtained by the intersection of the tangent to curve $H^2 = f(1/h)$ with the H^2 axis gives the value of H_0. [16]

Plots of H^2(1/h) for irradiated with D, He and Ar ions samples are shown at Fig. 4.

The unirradiated SS316 steel showed a good linearity, and the bulk hardness H_0, which is the square root of the intercept value, was estimated as 2.4 GPa.

In the case of deuterium and helium irradiation the data can be divided at least into two regions. In the region of shallow depth, the square of the nanoindentation hardness, H^2, was proportional to the reciprocal of the indentation depth, 1/h. The data in this region could be interpreted as the hardness of the region irradiated to 1.4 dpa (see Fig.2). The bulk hardness H_0 estimated from this irradiated region was 3.7 and 4.35 GPa for deuterium and helium irradiation, respectively.

The argon irradiated SS316 steel appear to have a bilinearity with a shoulder at around 160-180 nm. The bulk-equivalent hardness of argon ion-irradiated steel can be obtained from the shallower depth region before the shoulder: 4.8 GPa for room temperature irradiation and 3.8 GPa for irradiation at 873 K.

For specimens irradiated with argon ions (at T_{room}) a smooth decrease of hardness is observed, since the depth of ~200 nm. This is due to the fact that during nanoindentation, the plastic zone around the indenter tip extends well below it, and in an approximately hemispherical volume around it, with the depth of the plastic zone reaching about 5-7 times the depth of the tip in most cases (SSE).

The mismatch observed experimentally between the maximum of nanohardness and the maximum in the distribution of defects (see Fig. 2 and Fig. 4) can be also explained by the effect of a soft substrate [17]. Besides, the same effect may be due to the rapid decrease in nanohardness with depth.

Generally, the defects being responsible for the hardening in materials mainly are dislocation loops, which act as obstacles for dislocation glide [18,19].

Our TEM studies of irradiated samples have showed the formation of tangled dislocations at depths of 0-150 nm after irradiation of SS316 steel with He, D or Ar ions at T_{room} (Fig. 5). The formation of dislocation structure, as the main factor of hardening, is likely gives approximately similar growth of the nanohardness up to 4.3±0.5 GPa.

The hardening of steel implanted with inert gas (He, Ar) ions may results from two separate pinning centers: dislocation-like defects and gas-filled cavities (He or Ar bubbles) [18,19]. Some studies [20,21] have shown that dislocation structure and gas bubbles contribute to

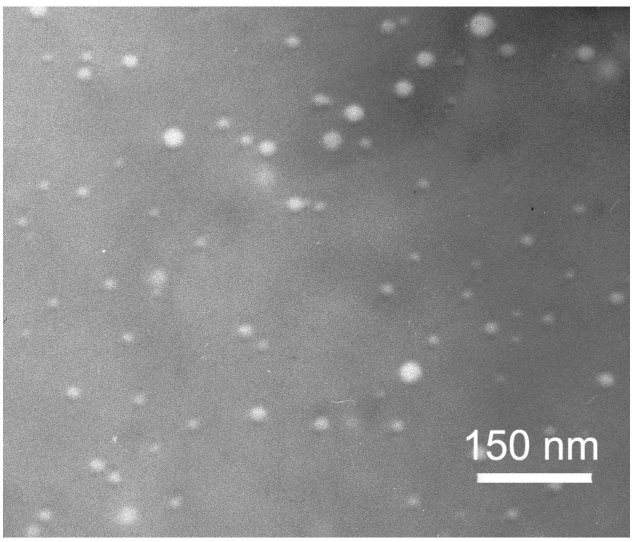


Fig. 6. TEM image of the structure of SS316 steel irradiated by Ar at T = 873 K.

hardening of stainless steel SS316.

In this study He ions were implanted only at room temperature, while Ar ions at room and at evaluated temperature. TEM studies of structure of irradiated at 873 K samples in the depths 500-600 nm from irradiated surface revealed the formation of the argon bubbles with mean size 20 nm (Fig. 6).

Nevertheless, it is obvious from Fig.4(c), that the formation of dislocation structure causes a more significant increase of the hardness of bulk material compared to the presence of the bubble structure.

Until recently it was believed that the contribution to the increase of yield strength of metals and alloys provides a spectrum of radiation-induced defects (point defects, clusters, dislocation loops of vacancy and interstitial types, the precipitation of a new phase, etc.). However, the present investigation with the use of modern high-resolution techniques allowed to show that the pinning of dislocations, causing hardening of steel SS316, occurs effectively only on the displacement damage rather than bubbles.

Conclusions

1. Radiation-induced nanohardness of SS316 steel increases about two times after bombardment with deuterium, helium or argon ions despite the difference in gas concentrations and levels of damage.

2. The main contribution to the hardening of the steel is due to the formation of radiation-induced dislocation structure, regardless of the type of bombarding ions.

3. The absence of correlation between nanohardness gain and irradiation dose in the range of 1-50 dpa may result from hardening saturation at ~1 dpa in austenitic stainless steels.

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