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Microstructural evolution in nickel alloy C-276 after Ar^+ ion irradiation

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ABSTRACT

Irradiation damage in nickel alloy C-276 irradiated by 115 keV argon ions at room temperature with irradiation doses from 0.28 to 82.5 dpa has been investigated by transmission electron microscopy. Nano-scale black spot damage appeared at a dose higher than 0.83 dpa. Large interstitial-type dislocation loops were observed at the dose of 8.25 dpa. Both the density of dislocation loops and the density of network dislocations grew significantly with the increase of irradiation dose. However, the density of network dislocations declined at the dose of 27.5 dpa. But the total of dislocation density (density of dislocation lines plus density of dislocation loops) kept increasing and no signs of saturation were seen in the dose range explored. The results showed that the nickel alloy C-276 had good performance in delaying the development of black spots, dislocations and dislocation loops. However, original grains have formed into sub-grains at the dose of 82.5 dpa, meaning that the grains in C-276 lost its structural integrity at doses higher than 82.5 dpa.

1. Introduction

As one of the six most promising Generation IV fission reactors, the supercritical water-cooled reactor (SCWR) has been attracting increasing attention [1–6]. The SCWR is cooled and moderated by supercritical water (SCW) above 500 °C, which undergoes no change of phase from liquid to gas above 22.1 MPa, and boiling does not take place in the supercritical-pressure region [7–10]. Thus, the SCWR could be a very aggressive oxidizing environment, and oxidation rates in such service condition are significantly enhanced especially at higher dissolved oxygen content. Therefore, there are more stringent demands on the fuel cladding materials [4], including good mechanical properties, resistance to corrosion and neutron irradiation, low susceptibility to stress corrosion cracking and microstructural stability [11]. Research and development of required materials has become a key issue in the development of the SCWR. Recently, numerous commercially available materials are qualified for service under the SCWR temperature and pressure, but those expected to be used in future commercial SCWR should have excellent resistance to both corrosion and irradiation.

The leading candidate materials for the SCWR are nickel-base alloys, austenitic stainless steels and ferritic/martensitic steels [11–20]. Because of their relatively higher creep strength compared to austenitic and ferritic/martensitic steels, nickel-base alloys have been proposed for Generation IV reactors in-core components that will experience temperatures higher than about 600 °C [21]. However, compared to austenitic stainless steels and ferritic/martensitic steels, the investigation concerned irradiation effects on the nickel alloys is rarely reported. Rowcliffe et al. drew some perspectives on radiation effects in nickel alloys for applications in advanced reactors and suggested some approaches to mitigate the effects of neutron irradiation on the ductility and dimensional stability of the present commercial nickel alloy [21]. Angeliu et al. have systematically studied radiation-induced swelling, creep and embrittlement on nickel-base alloys, and provided recommendations for further assessments of the material behavior and methods to minimize the effects of radiation damage through alloy design [22]. Irradiation-induced hardening in Inconel 718 implanted with Fe, He and H ions was reported by Hunn et al. [23]. Radiation-induced hardening for solution-annealed Inconel 718 was clearly evident, and it was associated with the production of ‘black dot’ defects. Microstructural development/evolution of alloy 718, irradiated with mixed spectra of high-energy protons, was shown by Sencer et al. [24,25]. They reported that black spot damage and larger loops whose size increased with dose were visible in irradiated samples.

Hastelloy C-276 is a high strength corrosion-resistant and heat-resistant nickel-base alloy with high contents of Cr and Mo, and has been widely used as pressure vessel steel at high temperatures [26]. Recently, some works evaluated the corrosion behavior of C-276 as the candidate material for the SCWR [6]. However, the

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investigation concerned its irradiation performance is rarely reported. Therefore, a systematic study on the behavior of this material under irradiation is needed to ensure its potential application in the SCWR.

This paper aims to study the microstructural changes of Hastelloy C-276 alloy under ion irradiation and represents a part of the ongoing research program in China for the future SCWR systems.

2. Experimental

Hastelloy C-276 is a face-centered cubic (FCC) nickel-base alloy with a nominal composition of Ni + 15.76 Cr, 15.55 Mo, 5.24 Fe, 3.47 W, 0.89 Co, 0.52 Mn, 0.02 Si, 0.01 V, 0.006 P, 0.002 C, 0.002 S in wt.%. The bulk material was first cut into 0.5 mm thick sheets and then thinned to a thickness of about 0.1 mm by mechanical polishing. Standard transmission electron microscopy (TEM) sample discs of diameter 3 mm were punched and then further thinned to a thickness of 50–100 μm using silicon carbide paper with the grade of 800–1200. The discs were electropolished to thin foils used for TEM observation using a MTPA-5 twin-jet electropolishing machine [27,28] (produced by Shanghai Jiaotong University, China). The electrolyte used for twin-jet thinning was a solution of 10% perchloric acid and 90% icy acetic acid solution, at room temperature.

A series of TEM specimens were irradiated with Ar^+ ion beams by an ion implanter at room temperature. The energy of the argon ions was 115 keV and the current density was approximately 1 μA/cm^2. The irradiation doses were 1×10^{14}, 3×10^{14}, 5×10^{14}, 1×10^{15}, 3×10^{15}, 5×10^{15}, 1×10^{16} and 3×10^{16} Ar^+/cm^2, corresponding to the maximum damage dose of 0.28, 0.83, 1.38, 2.75, 8.25, 13.75, 27.5 and 82.5 displacement per atom (dpa), respectively. The irradiation parameters are shown in Table 1. It should be noted that some of the specimens were irradiated twice, which was more conducive to observe the microstructural changes. The microstructural evolution of the specimens was observed by using JEM-2010 TEM. The accelerating voltage of the TEM was 200 kV, and the most often used image conditions were bright field.

3. Results and discussion

3.1. Micro-structural changes at low irradiation doses of 0.83 dpa

Fig. 1(a) shows TEM micrograph of the C-276 alloy before implantation. Almost no defects could be observed in the observation field, indicating that the unirradiated sample is defect-free. Black lines in Fig. 1(a) are equal thickness fringes, not dislocations. The diffraction patterns are typical bright and sharp diffraction spots.

The sample S1 was first irradiated to a dose of 0.28 dpa. Compared to the unirradiated sample, there is no obvious change in the microstructure of S1 (not shown). After an additional dose of 0.55 dpa, i.e., the total dose reached to 0.83 dpa, many black spots appeared, as shown in Fig. 1(b). The dots, such as A, B, C in Fig. 1(b), disappeared when the specimen was tilted by 2°, indicating that they are the defects instead of precipitates caused by irradiation. The size of these black spots is about 1–3 nm and the density is $3.0 \times 10^{10}/cm^2$. The implanting of Ar ion into the metal could knock out atoms in crystal lattice from its equilibrium position and cause collision cascades, inducing vacancies and interstitial atoms. The aggregation, formation and disappearance of these point defects may induce larger size defects such as black spot damage [Fig. 1(b)] and the secondary defects such as network dislocation and dislocation loop. Initial work based on ferritic Fe–Cr alloys indicated that the black spots were also remarkably effective traps for both point defects and helium atoms [21]. Helium is primarily produced in nickel-base alloys by high energy neutron reactions with Fe, Cr and Ni isotopes. Natural Ni contains 68% ^{58}Ni and 26% ^{60}Ni, and the two reactions $^{58}Ni(n,\alpha)^{55}Fe$ and $^{60}Ni(n,\alpha)^{57}Fe$ occur throughout the neutron energy spectrum [22]. The helium would be trapped by black spot damage, leading to helium embrittlement. The accumulation of helium could form bubbles, which would result in swelling. Therefore, the swelling and helium embrittlement would appear when nickel-base alloy C-276 used in the SCWR.

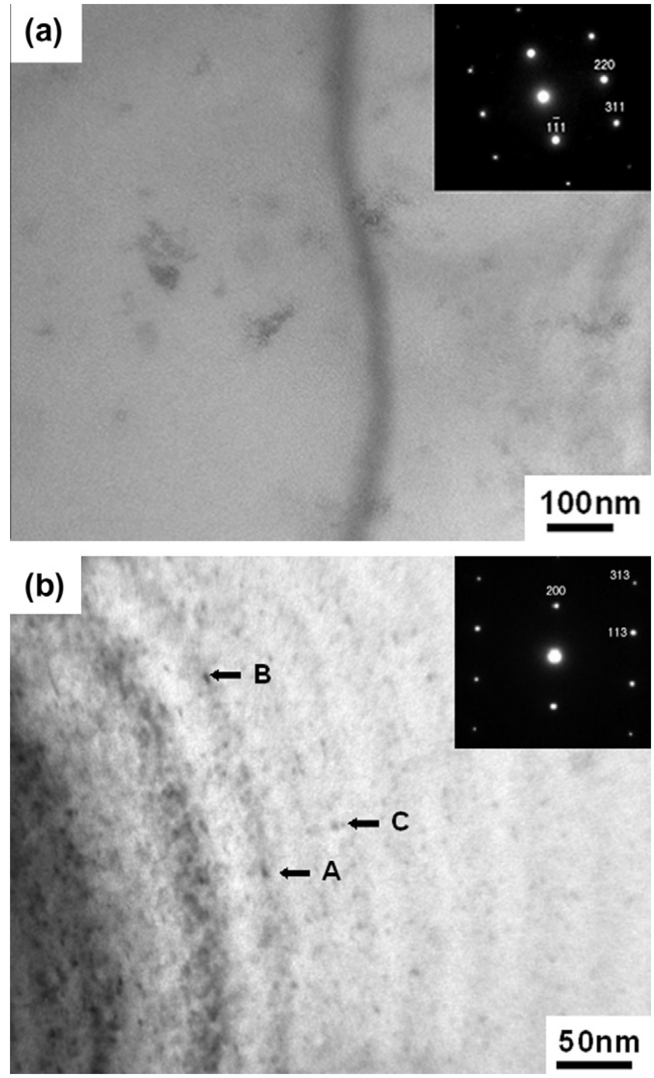

Fig. 1. The microstructure of (a) unirradiated C-276 sample and (b) irradiated to a dose of 0.83 dpa.

It was reported that black spots in nickel-base alloy 718 neutron-irradiated to 0.1 dpa could be visible [25]. In present investigation, black spots appeared at an irradiation dose greater than 0.1 dpa, implying that the C-276 alloy have better property in delaying the appearance of black spots than alloy 718.

Table 1
Ion implantation process of the Hastelloy C-276 alloy samples.

Sample code	Implantation times	The first implantation dose (dpa)	The sequential implantation dose (dpa)	Total dose (dpa)
S1	Twice	0.28	0.55	0.83
S2	Twice	1.38	1.38	2.75
S3	Once	8.25	–	8.25
S4	Once	13.75	–	13.75
S5	Twice	27.5	55	82.5