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Deformation modes of proton and neutron irradiated stainless steels

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Abstract

AISI 304 and 316 stainless steels of two purity levels that have been irradiated with high energy protons up to 0.3 dpa and neutrons in a high flux reactor up to 7.5 dpa were investigated in terms of irradiation induced mechanical properties and microstructural changes. The stress–strain relationships were obtained at room temperature. The deformation, grain, twinning and irradiation defect microstructures were investigated using both transmission and scanning electron microscopy. The results are discussed in terms of deformation mechanisms linked with the radiation induced defect microstructure.

1. Introduction

In stainless steels, the susceptibility for stress corrosion cracking (SCC) is found to rise above a fluence of $\sim 10^{20}$ n cm^{-2} ($E > 1$ MeV) equivalent to 0.15 dpa at 300°C. The cracking is intergranular and its susceptibility is influenced by the material properties, the chemical environment and the applied stress. It is a complicated multiparameter problem. The first problems of SCC under irradiation, the so-called irradiation assisted stress corrosion cracking (IASCC) phenomena, were found in light water reactors (LWR) in 1960s and at present, with the further ageing of the plants, different types of reactor present the same IASCC component susceptibility [1]. The search for the cause of the cracking is at the origin of an important database, developed in the last ten years, in areas such as microchemistry and radiation induced segregation [2,3]. However, the specific microstructural and microchemical components which promote IASCC remain unknown. In particular, the possible contribution of radiation hardening to the cracking susceptibility has not been elucidated. Therefore, an in-depth investigation of the IASCC causes in commonly used austenitic stainless steels in reactor engineering is needed to optimise the safety of reactors and the lifetime of its components.

The amount of data on the microstructure and deformation characteristics of stainless steel irradiated in the temperature region of light water reactor is sparse. Most of the studies were performed at higher temperatures and doses around 10 dpa. Furthermore, the intrinsic deformation mode found in pure FCC irradiated materials in this temperature region, namely the dislocation channelling mechanism, could lead to instabilities at grain boundaries, because of the large localised shear strains implied. The present work aims to describe and model the relationship that might exist between the localisation of the deformation and the radiation hardening. In this paper, first results on the microstructure and associated deformation modes found in 304 and 316 type stainless steel irradiated in the temperature region of 520–550 K, to doses below 7.5 dpa and tested at room temperature, are presented.

2. Experimental procedures

2.1. Materials

Four steels of technological interest were supplied by Asea Brown Bovery Sweden (ABB). The steels comprised 304L and 316L in commercial (CP) and high (HP) purity. The composition of these materials is given in

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Table 1
Chemical composition in wt% of the four steels provided by ABB; the balance is Fe

SS	C	Si	Mn	P	S	Cr	Mo	Ni	Co	N	B
304L CP	0.013	0.13	1.16	0.021	0.011	18.32	–	10.42	0.029	0.027	0.0005
304L HP	0.002	0.08	0.75	0.006	0.003	18.32	–	10.54	0.007	0.010	0.0005
316L CP	0.008	0.07	1.32	0.005	0.006	16.70	2.56	12.28	0.018	0.069	–
316L HP	0.009	0.05	1.30	0.004	0.006	16.66	2.54	12.28	0.020	0.068	0.0005

Table 1. The 316L CP and HP have almost identical compositions, but 304L CP has more C, P and S than the HP. The steels were hot rolled from ingots to a plate of 140×90 mm at 1180°C and further reduced at 1140–950°C to a final size of 145×10 mm. The steel plates were solution heat treated at 1050°C with a holding time of 30 min and water quenched in a carbon dioxide atmosphere.

2.2. Irradiations

The neutron irradiations have been performed by ABB in the light water reactor Barsebäck 1. The fast neutron flux was about $1.5–7.6\times10^{13}$ n cm^{-2} s^{-1} ($E>1$ MeV) and the irradiation temperature was 550 K. The four types of stainless steel were irradiated to two fluence levels of 2×10^{21} n cm^{-2} and 5×10^{21} n cm^{-2}, which corresponds approximately to 1.5 and 7.5 dpa. The as-received specimens were in the shape of ruptured constant elongation rate test (CERT) samples.

No low dose neutron irradiated specimen were available. Therefore, proton irradiation at low doses were performed at the proton irradiation (PIREX) facility located at the Paul Scherrer Institute, that allows the irradiation of tensile samples with 590 MeV protons at temperatures between 310 and 700 K, under helium gas flux cooling [4], at a typical dose rate of 5×10^{-7} dpa s^{-1}. Eighteen specimens, nine of each CP steel, were irradiated at a temperature of 520 K. The doses reached were 0.15 and 0.3 dpa.

Previous studies performed on pure materials have shown that at similar doses to those used in the present investigation, there is no drastic difference between the defect microstructure and tensile properties that are produced by neutron or by high energetic protons when compared in terms of dpa [5].

2.3. Mechanical testing

Two types of irradiated tensile samples were tested. The proton irradiated samples were in the shape of 20 mm flat tensile samples with a gauge length of $5.5\times2.5\times0.3$ mm^3. They were produced from archive plates of the same steels. The neutron irradiated samples which were cut from the non-deformed heads of the irradiated CERT samples, are in the shape of 10 mm flat tensile samples with a gauge length of $3\times2.5\times0.3$ mm^3. The size was optimised to fit the geometry of the as-received CERT sample heads.

The tensile tests were performed at room temperature. A strain rate of 1.5×10^{-4} s^{-1} was set for all tests. The uncertainties on the values obtained from the tensile tests are below 10%.

3. Results and discussion

3.1. Tensile tests

As mentioned above, the as-received samples were in CERT shape. In order to asses the specimen size effect on tensile tests results, a comparative study between the two specimen geometries was made. In Table 2, the values obtained are listed. It should be noticed at this point that, though the resulting values present some differences, no clear dependence of the strength either on the purity of the steel or the specimen geometry could be observed. The yield stresses measured are between 180 and 260 MPa. On the other hand, the ultimate elongation is always smaller in the 10 mm flat specimen geometry. The commercial and high purity specimens show comparable results.

No systematic effects of the impurities could be measured. Therefore, we assumed that the purity is not a relevant parameter in our case and the comparison is made rather between the two types of steels SS304 and SS316. The overall results are summarised in Table 3 for the unirradiated and irradiated specimens. Typical results of the tensile tests on the irradiated SS304 and SS316 are shown in Fig. 1. The two sets of samples of SS304 and SS316 at low doses exhibit very similar behaviour. The irradiated samples show already significant radiation hardening at low doses. The yield stress increase is about 80% for both the steels at 0.15 dpa.

The SS304 suffer a reduction of the ultimate elongation of about 40%, the SS316 seems to have less hardening and shows a reduction of the ultimate elongation of about 30%. Strong hardening is found at high doses for both steels. The yield stress presents an increase of around 100% for SS304 and 200% for SS316 at a dose of 1.5 dpa. The SS316 shows an increase of nearly

Table 2
Tensile properties of the stainless steels used before irradiation[a]

Specimen type	YS (MPa)	UTS (MPa)	UE (%)	Geometry
SS304L CP	190	800	47	20 mm flat tensile
SS316L CP	240	800	40	20 mm flat tensile
SS304L CP	204	769	34	10 mm flat tensile
SS316L CP	195	702	36	10 mm flat tensile
SS304L HP	260	719	30	10 mm flat tensile
SS316L HP	180	643	30	10 mm flat tensile

[a] The geometry of each sample is indicated in the last column. The tests have been performed at room temperature. The values listed are the average of 3–5 measurements. The standard deviation is above about 10%.

Table 3
Average tensile properties of the different irradiated specimens, tested at room temperature; the standard deviation is above about 10%

Specimen type	Dose or fluence	YS (MPa)	UTS (MPa)	UE (%)
SS304L	Unirradiated	190	800	47
SS304L	0.15 dpa	330	720	28
SS304L	0.3 dpa	335	820	31
SS316L	Unirradiated	240	800	40
SS316L	0.15 dpa	450	840	27
SS316L	0.3 dpa	394	828	26
SS304L	Unirradiated	260	719	30
SS304L	1.5 dpa	530	925	12
SS316L	Unirradiated	180	643	30
SS316L	1.5 dpa	578	803	17
SS316L	7.5 dpa	691	864	16

300% at a dose of 7.5 dpa. The SS304 presents a decrease of the ultimate elongation of about 70% at 1.5 dpa and the SS316 shows a decrease of approximately 50% at 1.5 dpa and 55% at 7.5 dpa. At low doses, the samples show approximately the same strain hardening as the unirradiated ones. At high doses, the samples show an even greater hardening, which increases with dose. An upper yield point appears and the strain hardening has decreased considerably.

The results of tensile testing at room temperature of both SS304 and SS316 have been summarised in Fig. 2, where the increase in yield strength has been plotted against the irradiation dose. It can be noticed that, at low doses, the hardening increases rapidly with dose to reach a maximum value at high doses indicating a saturation of the radiation hardening of the material. The saturation value is higher for SS316 than for SS304. However, due to the lack of data between 0.3 and 1.5 dpa, the transition to saturation of the radiation hardening can not be evaluated with confidence. Nevertheless, an estimation leads to a value of about 1 dpa, which is comparable to the one obtained by Zinkle et al. [6]. Furthermore, Heinisch et al. [7] have measured a linear dependence of the hardening in the region from 2×10^{-3} to 2×10^{-2} dpa with a similar slope to that found in the present investigation after proton irradiation, while a number of authors [8–11] have found comparable saturation values at higher doses, SS316L showing always a higher radiation hardening than SS304L.

3.2. Irradiation and deformation microstructure

3.2.1. Defect microstructure

The irradiation induced defect microstructure was investigated using transmission electron microscopy. In undeformed samples, following the 590 MeV proton irradiation to 0.15 dpa, the mean defect size is about 3 and 2 nm for SS304 and SS316, respectively. The defects are mostly unidentified ‘black dots’, believed to be small faulted interstitial loops, only about 30% of them have been identified as small Frank’s loops. At higher dose, Frank’s loop type defects became more frequent and a somewhat irregular and relatively broad loop size distribution is observed, indicating loop growth. The size distribution for neutron irradiated high purity SS304L and SS316L still peaks approximately at 4–6 nm, but there is an important number of loops with sizes going up to 25–30 nm.

No systematic identification of the character of the defect presents has been made yet. However, in the few cases where this analysis was performed, the loops were

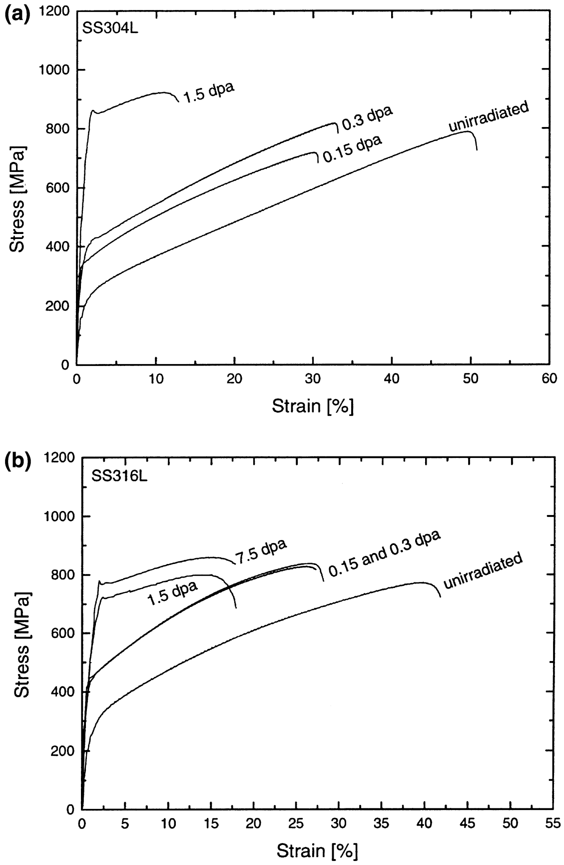

Fig. 1. Comparison between unirradiated and irradiated specimens, (a) SS304 and (b) SS316, tensile tested at room temperature. The doses and the type of irradiation are indicated.

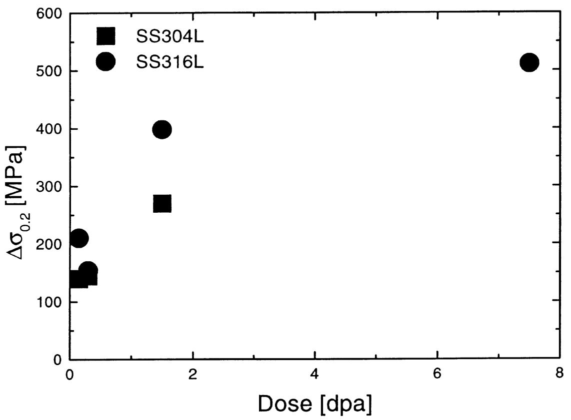

Fig. 2. Plot of the increase in yield strength $\Delta\sigma$ as a function of the dose for the proton and neutron irradiated 304L and 316L.

identified as interstitial loops lying on the {1 1 1} planes with a Burgers vector $b = (1/3)a0[1\,1\,1]$, in agreement with previous observations [12,13].

3.2.2. Deformation mode

The deformation mode of the irradiated and deformed tensile samples was analysed by performing TEM investigations. Both 304L and 316L show twinning as a deformation mode at doses below 1.5 dpa. Fig. 3 shows a typical micrograph of the observed twins. Their signature is demonstrated by the diffraction pattern inserted in the same figure. It can be seen that the twins, which are free of defects, are very narrow. The same deformation mechanism has been observed for both steels at any irradiation doses investigated. No difference has been observed between high energy proton or neutron irradiated specimen as well.

3.3. Microcracking investigation

It is expected that the strongly localised deformation by twinning of the irradiated samples could lead to cracking at the grain boundaries, as large shear strains are concentrated at the tip of the twins [14]. In order to check this hypothesis, the initially polished surfaces of the deformed flat tensile samples have been investigated using scanning electron microscopy (SEM). The tensile tests were interrupted at different strain levels in order to determine when the first microcracks appear at the surface. In the case of SS304L tensile tested up to necking, microcracking at grain boundaries, out of the necked zone, takes place at 0.15 dpa, as can be seen in Fig. 4. However, no systematic cracking was found when the specimen was deformed at 5% strain. In SS316L, no cracking was observed at the lowest dose namely 0.15 dpa, but after irradiation to 0.3 dpa, cracking at grain boundaries could be found. Deformation up to 11% strain shows induced intergranular cracks in a larger area around the necked zone.

At high doses, the samples showed microcracking along the whole gauge length. The cracks are already present at deformations as small as 5.5% strain. Fig. 5 shows an example of the microcracks observed in a SS316 specimen irradiated up to 1.5 dpa. The cracking tendency appears to increase with increasing dose. It reaches a very high level in the case of the high purity samples irradiated to a neutron dose of 7.5 dpa, where the entire gauge length was found strongly cracked at the end of the deformation.

The numerous microtwins formed contain a high density of dislocations, they can therefore initiate cracks at the intersection with grain boundaries. The twinning, enhanced by small defect sizes, results in a very localised deformation process [15]. Therefore, the microcracking

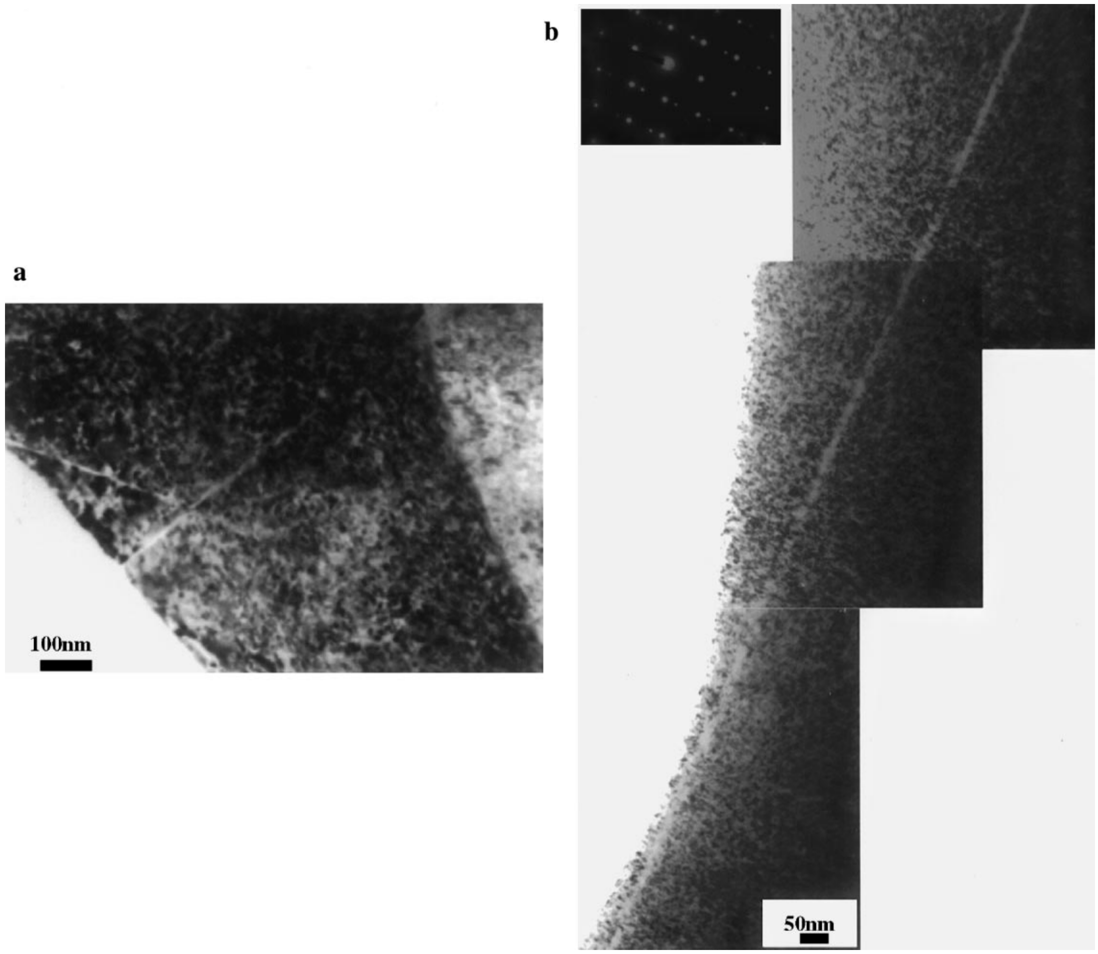

Fig. 3. 304L (a) and 316L (b) irradiated to 1.5 dpa, tensile tested at room temperature up to 5% strain. Bright field picture (a) shows a twin going towards a grain boundary and (b) a long microtwin free of irradiation defects with inserted diffraction pattern.

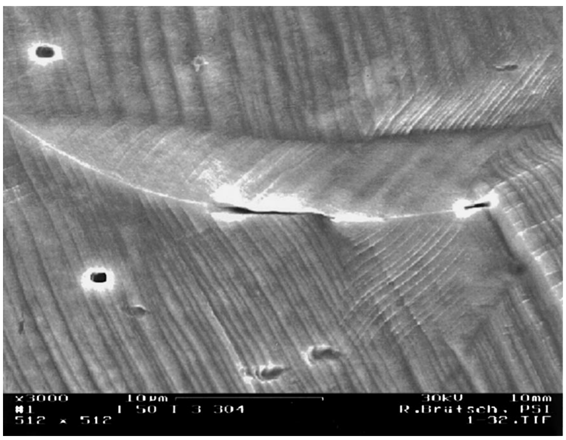

Fig. 4. SEM micrograph of proton irradiated 304L CP, to 0.15 dpa. Tensile test at room temperature interrupted just at necking.

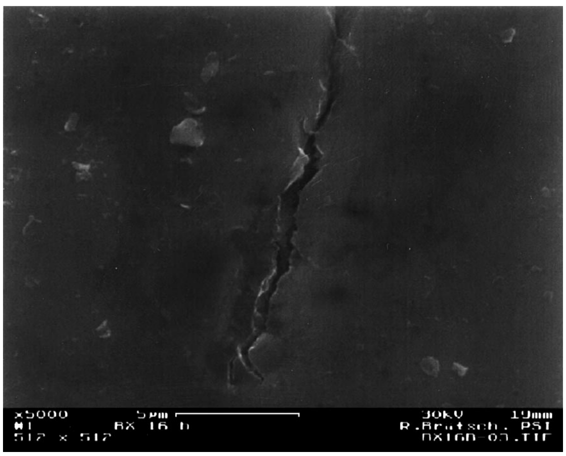

Fig. 5. SEM micrographs of neutron irradiated SS316L HP. The dose reached is 10^{21} n cm^{-2} at 550 K. The tensile test was interrupted at 5.5% strain.

observed is directly linked with this highly localised mode of deformation [16].

4. Conclusions

Irradiated stainless steels 304L and 316L show a strong radiation hardening. In comparison with SS304L, SS316L shows a higher radiation hardening at doses above 1.5 dpa.

The irradiation defect microstructure at 0.15 dpa consists mainly of black dots with a number of small Frank's loops being present. At 1.5 dpa, the black dots grow into a high density of Frank's loops.

Both 316L and 304L deform by twinning at room temperature, independently of the irradiation dose. The twins are very narrow (10–50 nm) and defect free.

Microcracking is observed at grain boundaries in both irradiated 304L and 316L. It is present already at low doses and seemed to increase with dose.

As indicated previously, the stainless steels are of first interest in an irradiating environment (fusion, fission, accelerators). A complete understanding of the effects of irradiation on structural components will increase the safety of a large number of installation. A problem of particular importance is the systematic investigation and modelling of the mode of deformation.

Acknowledgements

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