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Effects of Ta and C on irradiation hardening of austenitic stainless steel

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Type 304L austenitic stainless steels are used for reactor internal components of light-water reactors and the irradiation assisted stress corrosion cracking (IASCC) is one of the degradation issues. We have developed IASCC resistant stainless steels focusing on minor addition of Ta as an oversized element. In this study, the effect of Ta and C on the irradiation hardening of the alloys developed was investigated. 304L austenitic stainless steels with various Ta and C contents were fabricated and characterized. These alloys were irradiated by 1 MeV H^+ ion beam up to displacement damage of 0.1, 0.3 and 1 dpa at 473K, 573K, 623K and 673K. In order to investigate the irradiation hardening, Vickers hardness test was carried out and the microstructure was observed by transmission electron microscopy. The irradiation hardening of the alloys was almost the same as that of 304L. It is expected that Ta and C addition alloy increase IASCC resistance due to reducing irradiation damage.

Keywords: irradiation assisted stress corrosion cracking, austenitic stainless steel, Ta addition, proton irradiation, irradiation hardening, microstructure.

1. Introduction

Austenitic stainless steels are used for reactor internal components of light-water reactors (LWRs) because of its excellent corrosion resistance, workability and weldability. The reactor internal components play important roles in supporting the fuel assemblies and control rods, providing a flow path of cooling water and so on. These components are required to keep their soundness under the severe condition such as neutron irradiation in high temperature and high pressure water. Neutron irradiation causes various material property changes, such as irradiation hardening, embrittlement, swelling and irradiation assisted stress corrosion cracking (IASCC) [1, 2]. Especially, IASCC is one of the degradation issues of LWR. It is believed thatone of the IASCC causes is radiation induced segregation (RIS) [3]. In the case of IASCC, depletion of Cr in the vicinity of grain boundaries due to the vacancy diffusion is the key phenomenon. Vacancies introduced by neutron irradiation diffuse to grain boundaries, exchanging the position preferentially with oversized elements such as Cr or Mn. As a result, Cr is depleted at grain boundary, and the intergranular corrosion resistance decreases. For the purpose of preventing RIS, the oversized element addition such as Hf, Zr, Ta and Nb to austenitic stainless steels has been suggested because the additional oversized atoms dominantly trap vacancies and prevent Cr diffusion to grain boundary [4].

We have tried to develop IASCC resistant stainless steels focusing on Ta as an oversized element. In this study, the irradiation hardening of the steels under development are investigated by ion irradiation.

2. Experimental

Type 304L stainless steels with addition of Ta and C were used in this study: these were Low-Ta Low-C alloy, Middle-Ta Low-C alloy, Middle-Ta Middle-C alloy and High-Ta High-C alloy. The chemical compositions are shown in Table 1. The Ta/C ratio is respectively 0.86, 2.47, 0.90 and 0.88. Disks of 3mm in diameter were punched, and the disks were solution heat treated at 1323K for 1 h in vacuum and were quenched in water. Then, stabilizing heat treatment was carried out at 1173K for 1 h in vacuum, and the specimens were quenched in

Table 1 Chemical composition of specimens.: No unit for Ta/C.

	C [mass%]	Si [mass%]	P [mass%]	S [mass%]	Ni [mass%]	Cr [mass%]	Ta [mass%]	Fe [mass%]	Ta/C [-]
High-Ta High-C	0.046	0.01	0.004	0.003	10.7	19.2	0.61	Bal.	0.88
Middle-Ta Middlie-C	0.031	0.01	0.005	0.003	10.71	19.0	0.42	Bal.	0.90
Middle-Ta Low-C	0.011	<0.01	0.005	0.003	10.64	19.1	0.41	Bal.	2.47
Low-Ta Low-C	0.01	0.01	0.005	0.003	10.68	19.0	0.13	Bal.	0.86
304L	0.01	0.7	0.02	-	10.17	18.15	-	Bal.	-

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water. The average grain size after the heat treatment was about 27 μm. The specimen surface was mechanical polished and electropolished.

To introduce irradiation defects to the specimens, ion irradiation was carried out using Dynamitron accelerator at Tohoku University [5]. 1 MeV H^+ was used in this experiment. The displacement damage was 0.1, 0.3 and 1 dpa at the depth of 4 μm from the surface with the irradiation rate of 1.8×10^{-5} dpa/s. Fig.1 shows the displacement damage distribution calculated by SRIM2013 code [6]. Irradiation Temperature was 473K, 573K, 623K and 673K, which was measured by thermography. These irradiation experiments were conducted in a vacuum of less than 5.0×10^{-6} Torr.

Vickers hardness test was carried out to measure irradiation hardening, where the irradiation hardening is a difference between the hardness after and before the irradiation. Test load was 9.8×10^{-2} N, holding time was 15 sec, and test temperature was R.T.

In order to investigate effects of proton irradiation on microstructure evolution, transmission electron microscopy (TEM) and energy dispersive X-ray spectroscope (EDS) observation were conducted using JEM-2100F (JEOL) operating at 200kV. The thicknesses of the thin films were measured using convergent beam electron diffraction method.

3. Result and discussion

3.1 Vickers hardness test

Fig. 2 shows the displacement damage dependence of irradiation hardening of specimens irradiated at 573K. Irradiation hardening increased with increasing displacement damage for all specimens. Irradiation hardening behavior was almost the same between the Ta added alloys and 304L stainless steel. Fig. 3 shows the irradiation temperature dependence of irradiation hardening to 0.3 dpa. Irradiation hardening was decreased with increasing irradiation temperature.

The indentation depth of all tests ranged from 1.0 ~ 1.4 μm. If the deformation region was five times the indentation depth, the deformation region can be estimated to range from 5.0 ~ 7.2 μm. It is included in the irradiated region as shown in Fig. 1. Moreover, Yabuuchi et al. discussed on the relationship between Vickeres hardness and nanoindentation hardness with proton irradiated stainless steels [7], and the test condition of this study is the same. Therefore, the results of Vickeres hardness test in this study were reasonable.

3.2 TEM observation

Fig. 4 shows the images of TEM observation before irradiation. Precipitates were observed in High-Ta High-C and Middle-Ta Middle-C alloys. Fig. 5 shows result of EDS, (a) line analysis and (b)-(f) area analysis. As Ta was

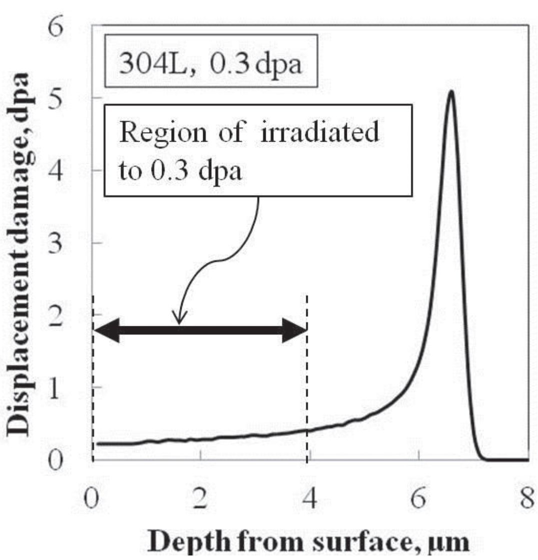

Fig. 1 Depth distribution of displacement damage.

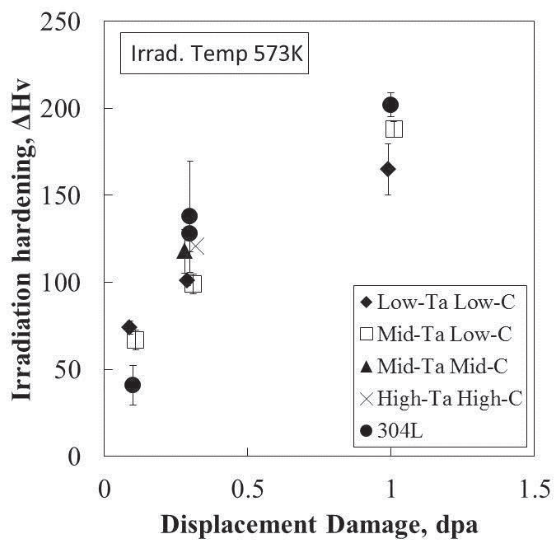

Fig. 2 Irradiation hardening as a function of displacement damage.

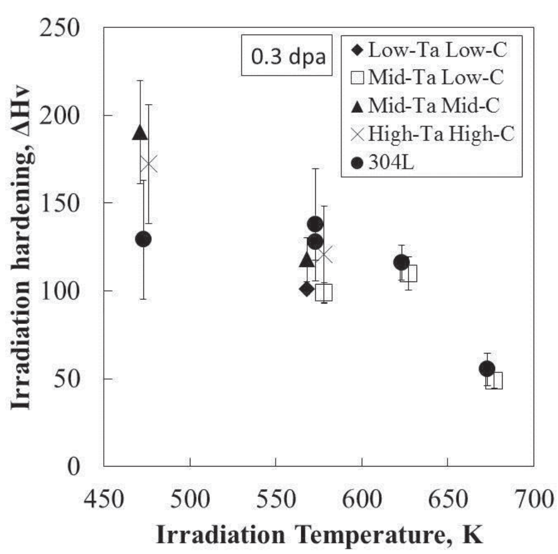

Fig. 3 Irradiation hardening as a function of irradiation temperature.

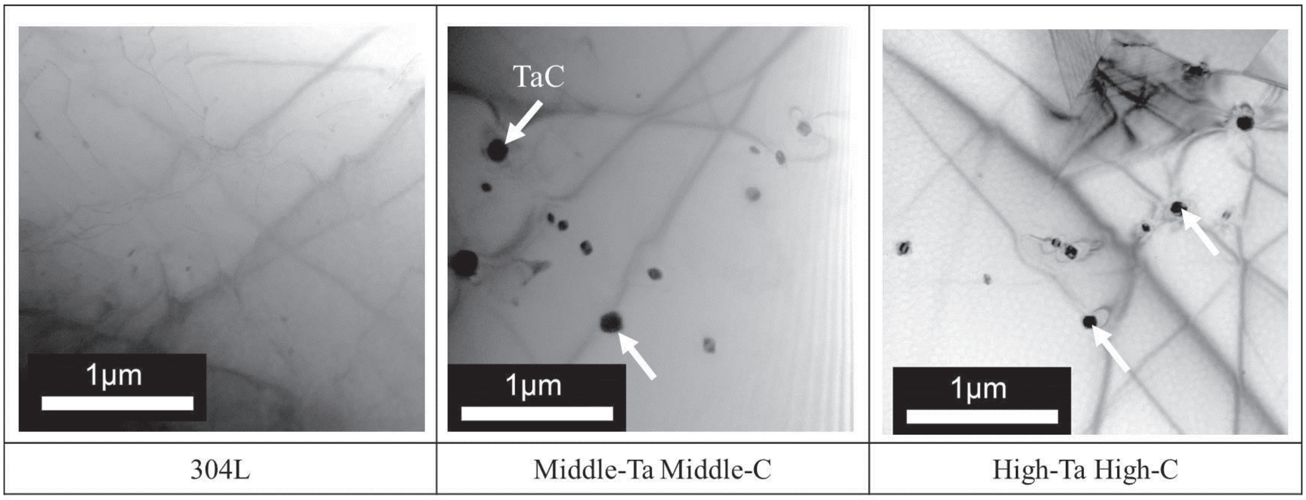

Fig. 4 The TEM images of unirradiated samples.

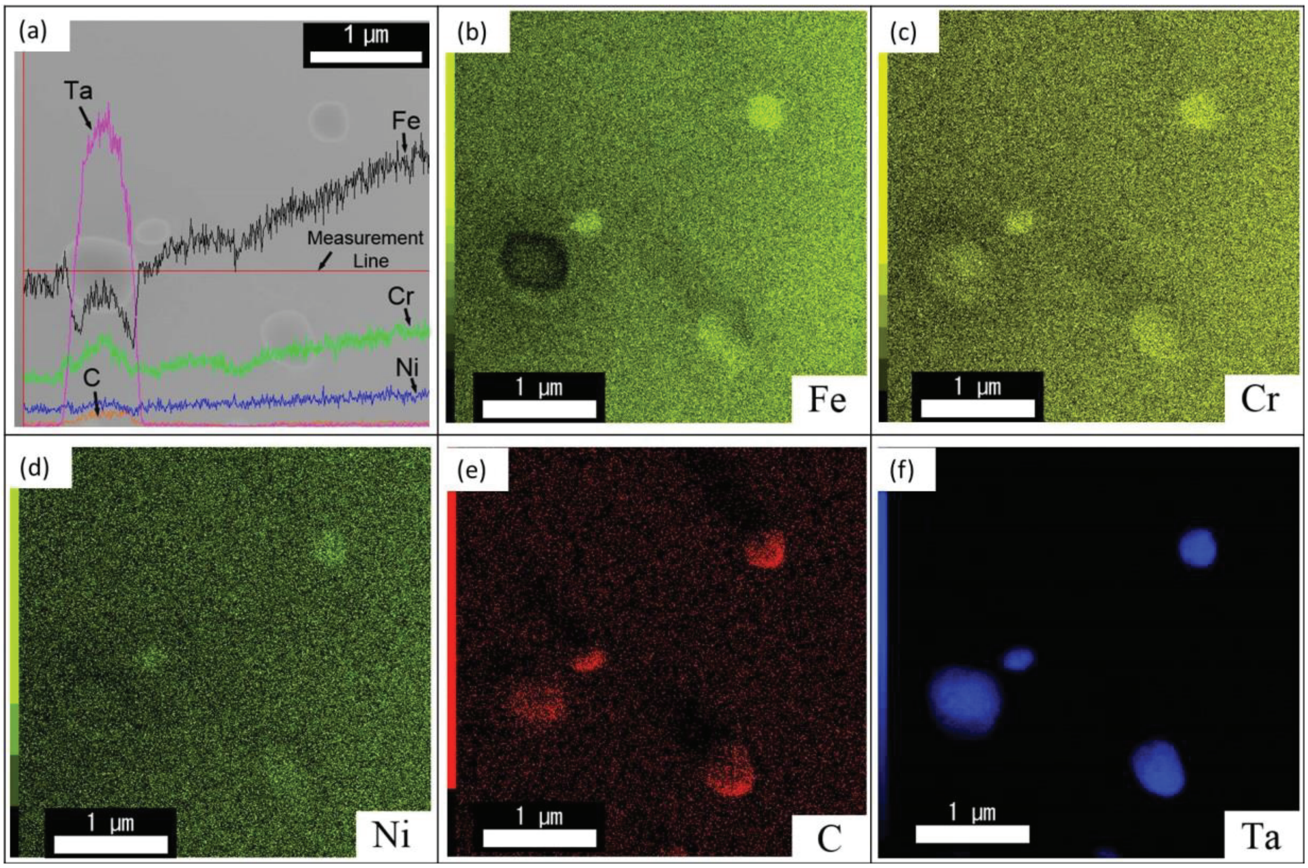

Fig. 5 The images of EDS (a) line analysis and (b)-(f) area analysis of unirradiated High-Ta High-C

mainly detected from precipitates as shown in Fig. 5, the precipitates were identified to be TaC.

TEM images of 304L stainless steel and Middle-Ta Low-C alloy unirradiated and irradiated to 0.3 dpa and 1dpa are shown in Fig. 6. Dislocation loops were observed in the specimens irradiated at 0.3 and 1 dpa. Fig.7 and Fig. 8 show dislocation loop size and number density dependence as a function of displacement damage. Evolution of dislocation loop in Middle-Ta Low-C and Low-Ta Low-C alloy was suppressed relative to 304L stainless steel. Fig. 9 shows TEM images of 304L stainless steel and Ta addition alloys irradiated at 473K to 673K. Dislocation loop size and number density as a function of irradiation temperature were shown in Fig. 10 and Fig. 11. The dislocation loop size increased with temperature.

3.3 Application of Orowan model

In order to clarify the contribution of dislocation loops to irradiation hardening, the increase in yield stress was estimated by Orowan model [8] using the following equation:

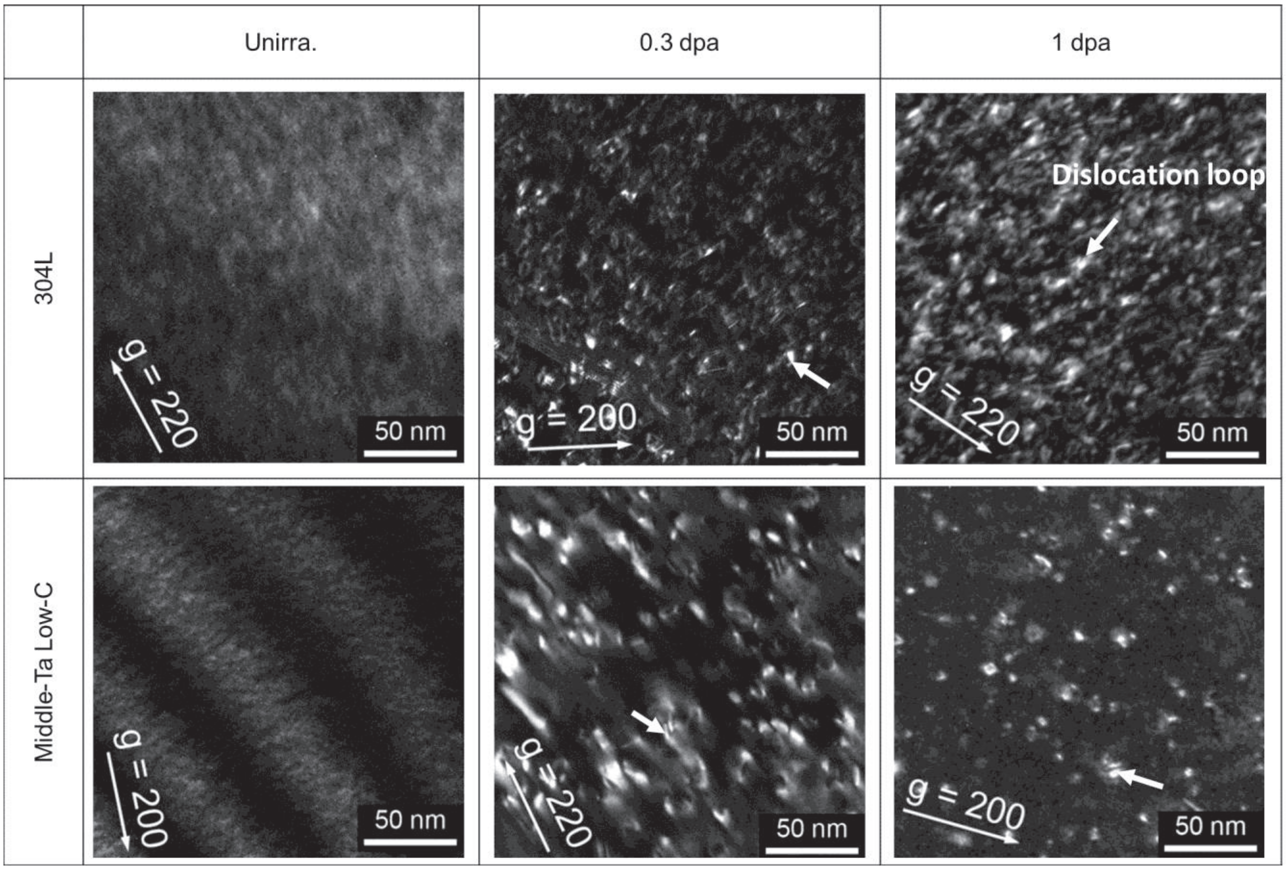

Fig. 6 TEM micrograph of 304L and Middle-Ta Low-C alloy irradiated up to 1 dpa.

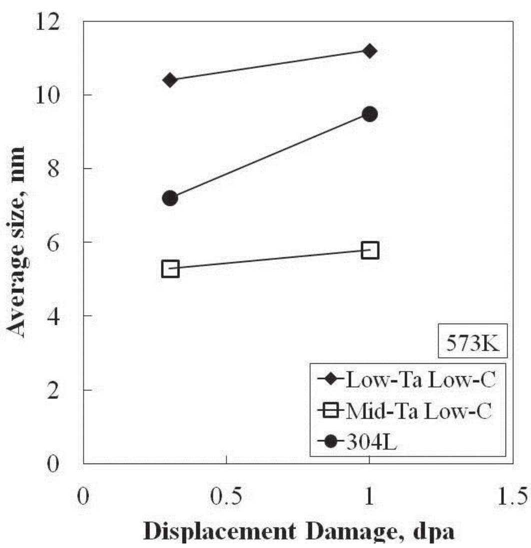

Fig. 7 Average dislocation loop size vs. displacement damage.

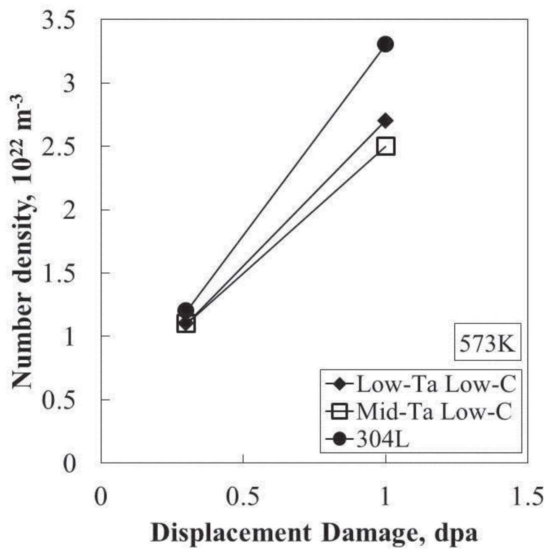

Fig. 8 Dislocation loop density vs. displacement damage.

$$\Delta\sigma_{calc} = M\alpha\mu b(Nd)^{0.5} \qquad (1)$$

In the above equation, $\Delta\sigma_{calc}$ is the increase in yield stress (MPa), M is Taylor factor (3.06), α is strength factor of dislocation loops (0.5) [9], μ is shear modulus (76 GPa), b is Burgers vector (2.5 × 10^{-8} cm), N is number density of dislocation loops, d is size of dislocation loops. The increase in Vickers hardness ΔH_V is converted into that of yield stress using the following equation [10]:

$$\Delta\sigma_{exp} = 3.03\Delta H_V \qquad (2)$$

The correlation between calculated yield stress $\Delta\sigma_{calc}$, and experimental value $\Delta\sigma_{exp}$ is shown in Fig. 12. As shown in Fig. 12, the calculated yield stress, $\Delta\sigma_{calc}$, is nearly equal to the experimental value, $\Delta\sigma_{exp}$, suggesting that the irradiation hardening was due to the dislocation loops.

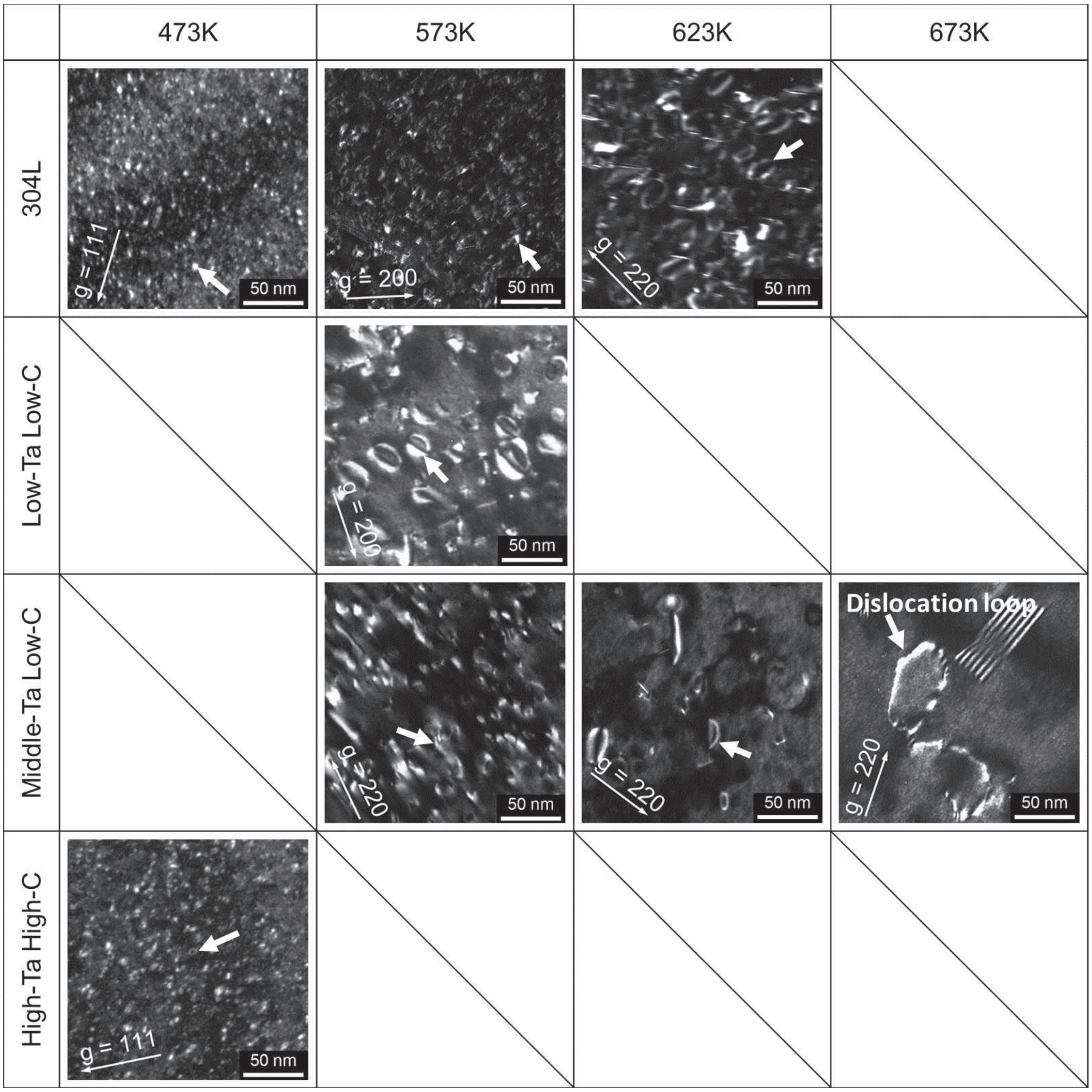

Fig. 9 The images of TEM irradiated at 473K to 673K.

3.4 Effect of Ta and C

The contribution of Ta and C to the hardening of specimens irradiated to 0.3 dpa at 573K is shown in Fig. 13. Tantalum suppresses the irradiation hardening of stainless steels as shown in Fig. 7 and Fig. 8.

Irradiation hardening increased with increasing C concentration. It is reported that dislocation loop density increased with increasing C concentration [11]. This means that the increase in irradiation hardening with C concentration observed in this study would be due to the increase in dislocation loop density. However, the irradiation hardening of the alloys is almost the same as that of 304L.

The effect of the difference of Ta/C ratio on the irradiation hardening was not observed in Fig.13.

4. Summary

In order to investigate the effect of Ta and C on irradiation hardening of stainless steels under development, Vickers hardness test and TEM observation were carried out after 1 MeV H^+ ion irradiation. The following results were obtained.

1. Irradiation hardening was observed in all alloys. The irradiation hardening of the alloys under development was almost the same as that of 304L, and the effect of Ta and C addition on irradiation hardening was not observed clearly within the investigated composition range. So it is expected that Ta and C addition alloy increase IASCC resistance without affecting

irradiation damage.

2. The irradiation hardening was explained by dislocation loop formation.

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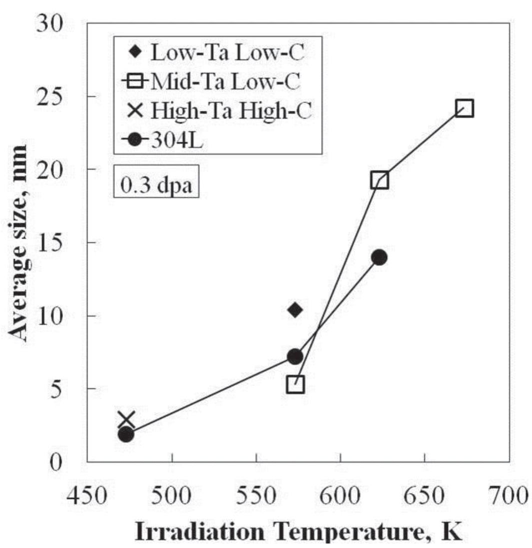

Fig. 10 Average dislocation loop size vs. irradiation temperature.

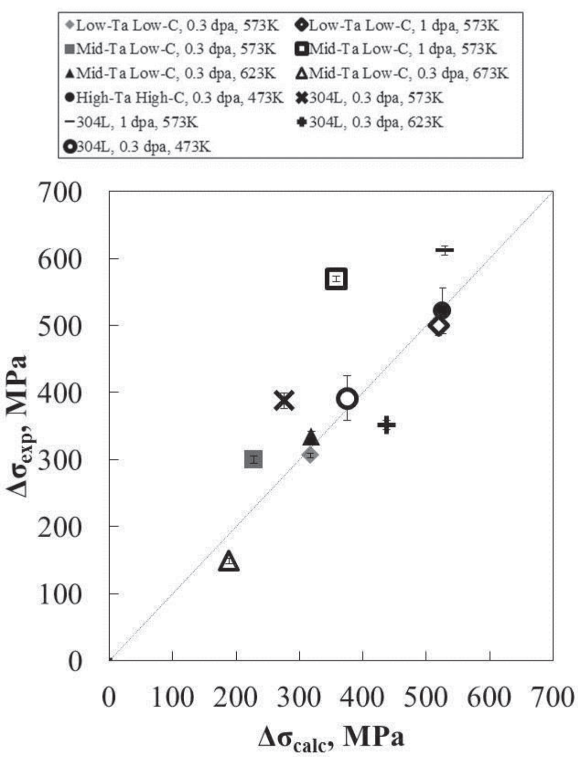

Fig. 12 Relationship between the yield stress calculated from microstructure and measured by Vickers hardness.

References

[1] C. Poker *et al.* , Journal of Nuclear Materials, **326**, 30-37, (2004)

[2] O. K. Chopra, A. S. Rao, Journal of Nuclear Materials, **409**, 235-256, (2011)

[3] S. M. Bruemmer *et al.* , Journal of Nuclear Materials, **274**, 299-314, (1999)

[4] T. Kato and H. Takahashi and M. Izumiya, Journal of Nuclear Materials, **189**, 167-174 (1992)

[5] S. Matsuyama *et al.* , Nuclear Instruments and Methods in Physics Research B, 267, 2060-2064 (2009)

[6] http://www.srim.org

[7] K. Yabuuchi *et al.*, Journal of Nuclear Materials, **446**, 142-147 (2014)

[8] E. Orowan, Symposium on Internal Stress in Metals Alloys, 451, 1948

[9] N. Hashimoto *et al.*, Journal of Nuclear Materials, **273**, 95-101 (1999)

[10] J. T. Busby *et al.*, Journal of Nuclear Materials, **336**, 267 (2005)

[11] F. Kano *et al.*, Journal of Nuclear Materials, **258-263**, 1713-1717 (1998)

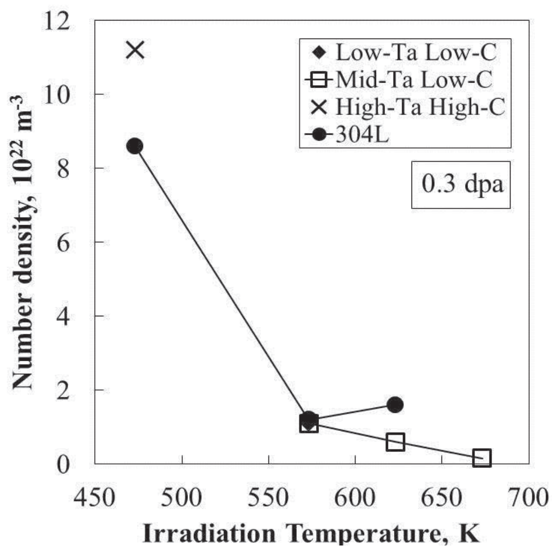

Fig. 11 Dislocation loop density vs. irradiation temperature.

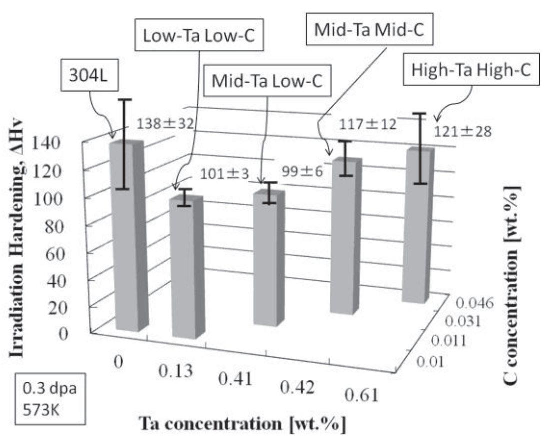

Fig. 13 Irradiation hardening as a function of Ta and C concentration.