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On Neutron Irradiation Resistance of Ni-Mo-Cr Reactor Pressure Vessel Steels

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1. Introduction

Influence of nickel on the neutron irradiation embrittlement of Ni-Mo-Cr reactor pressure vessel (RPV) steels was investigated in range of 0.9 ~ 3.5 wt.% of nickel content. The effects of alloying elements and impurities in RPV steels on the sensitivity to neutron irradiation have been extensively investigated [1-4]. It has been generally admitted that Cu and P are typical detrimental impurities, which lead to precipitation or segregation at grain boundaries during irradiation, so that they deteriorate the radiation stability. However, the effects of Ni, one of the key alloying elements in RPV steel, are still controversial; one claims that Ni alone has marked influence on irradiation damage while others report that Ni exhibits the synergism only with higher Cu concentration. Present work is to investigate and confirm the exclusive effect of Ni on the neutron irradiation hardening and embrittlement in modified Ni-Mo-Cr steels, where Ni content was increased up to 3.5 wt.%. Authors acknowledge that this report was re-edited based on manuscript already published in Metal and Materials International, 2013 [1].

2. Experiments and Results

In this section, experimental procedure including material, neutron irradiation test and crucial results are described.

2.1 Experimental procedure

Chemical compositions of the investigated steels are listed in Table 1. Impuries such as Cu, P and S was thoroughly controlled to examine the effect of Ni on irradiation embrittlement. Alloy 'S' has similar composition to SA508 Gr.3 and Ni was partially substituted for Mn in alloys 'A' and 'B'. Alloy 'C' contains higher Ni and Cr compared to the alloy 'S', similar to composition of SA508 Gr.4N steel. Heat treatment process of the steels followed a standard practice for RPV steels [5].

For the neutron irradiation test, 3 mm disc specimens were prepared. These discs were neutron irradiated in two different dose conditions in the JMTR (Japan Materials Testing Reactor). One dose condition was 4.5 $\times$ 10^{19} neutron/cm^2 at 290 °C and the other was 9.0 $\times$ 10^{19} neutron/cm^2 at 290 °C, which were higher than that nuclear pressure vessel experiences during the service life. Alloy 'S' and 'A' were irradiated under both conditions, and alloy 'B' and 'C' were under the second condition only.

Vickers hardness tests and small punch (SP) tests were performed on the disc specimens before and after irradiation. SP tests were carried out at temperatures between –196 °C and room temperature. DBTT in SP test were obtained using SP-fracture energy, which was defined as the total area below the SP load-deflection curve obtained at various test temperatures [6].

Microstructures of specimens were examined by optical and transmission electron microscopes (TEM).

Table 1. Chemical compositions of investigated alloys (wt.%)

Alloy	C	Mn	Ni	Mo	Cr	Si	Al	Cu	P	S
S	0.20	1.39	0.92	0.48	0.15	0.19	0.025	0.004	0.004	0.0011
A	0.20	0.70	1.91	0.49	0.15	0.18	0.017	0.006	0.002	0.002
B	0.20	0.31	3.52	0.49	0.19	0.20	0.007	0.002	0.001	0.0002
C	0.19	0.30	3.51	0.49	1.79	0.19	0.002	0.011	0.004	0.0013

2.2 Microstructure of the alloys

Microstructures of unirradiated alloys are given in Fig. 1. Optical micrograph of alloy 'S' shows typical tempered upper bainite structure in Fig. 1 (a). Detailed characterization using TEM with carbon extraction replica in Fig. 1 (b) reveals that long rod type cementites as well as spherical cementite particles are distributed in regions between bainitic ferrite laths. Fine needle-like precipitates in the ferrite laths are found to be molybdenum-rich M_2C carbides formed during tempering. Alloys 'A' and 'B', where Mn is partially replaced by Ni, showed similar microstructures to alloy 'S'. Fig. 1(c) and (d) demonstrate the microstructure of alloy 'C'. Due to the increased of Ni and Cr contents, it has higher hardenability and the microstructure shows a mixture of tempered lower bainite and martensite. Large cementite particles or cementite rods in alloy 'S' cannot be found but relatively fine cementite particles within the laths and Cr-rich $M_{23}C_6$ carbides were observed at grain boundaries.

Even after the irradiation, it was difficult to observe meaningful defects such as additional precipitates or clusters in all alloys, but dislocation loops and nanometer-sized defects regarded as vacancy cluster could be observed, which is comparable to those in the earlier literatures [7].

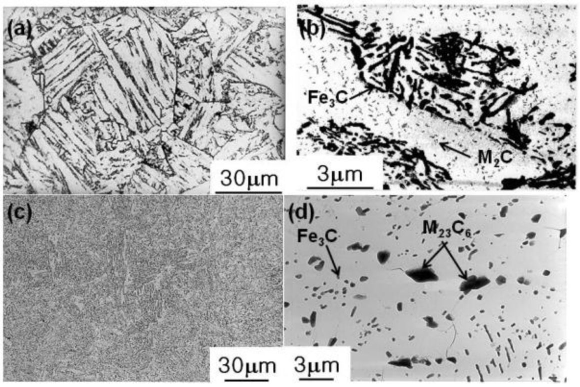

Fig. 1. Micrographs of alloy 'S' (a and b) and 'C' (c and d): Microstructures of alloy 'A' and 'B' where a part of Mn was replaced by Ni compared to the alloy 'S' showed little difference from that of alloy 'S', so that micrographs of them were omitted here.

2.3 Irradiation hardening

Fig. 2 shows the changes in Vickers hardness of the alloys before and after the neutron irradiation. The increase in hardness is about 25 $HV_{0.2}$ (0.2kg loading) after an irradiation dose of 4.5 × 10^{19} neutron/cm^2 and 65 $HV_{0.2}$ after an irradiation dose of 9.0 × 10^{19} neutron/cm^2. The hardening is affected by the irradiation dose, but almost independent of the alloy chemistry. Combined with the microstructural observation, it is thought that the neutron irradiation hardening was mainly caused by the defect structures like dislocation loops and vacancy clusters. It was confirmed that the Ni concentration did not have an exclusive influence on the irradiation hardening as long as the impurity elements such as Cu, P and S were strictly controlled to lower level.

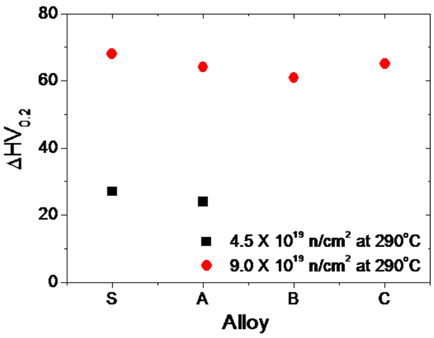

Fig. 2. Hardness change under two different neutron irradiation conditions.

2.4 Irradiation embrittlement

Figs. 3 (a) to (d) show the variation of SP-fracture energy with temperature. Under the irradiation dose of 4.5 × 10^{19} neutron/cm^2, the shift of DBTT was not remarkable in alloy 'S' and alloy 'A'. The shift of DBTT of alloy 'A' containing higher Ni was even less than that of the alloy 'S'. Subjected to the irradiation dose of 9.0 × 10^{19} neutron/cm^2, the shift of DBTT was about 40 °C in both alloys 'S' and 'A'. Even though full transition curve of absorbed energy could not be obtained by SP-test for unirradiated specimens in alloy 'B', the DBTT increase of 30 °C could be estimated. In alloy 'C', brittle transition was not observed at the tested temperatures as low as -196 °C for both unirradiated and irradiated conditions, so the DBTT shift could not be determined. Given that the DBTT shift after irradiation is around 30~40 °C even with the gradual replacement of Mn by Ni, the influence in Ni content on the DBTT increase by irradiation is thought to be not remarkable.

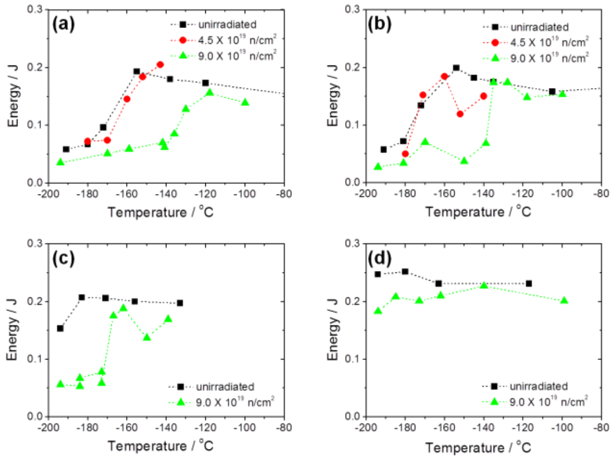

Fig. 3. The variations of SP-fracture energy with temperature before and after neutron irradiation: (a) alloy 'S' (b) alloy 'A' (c) alloy 'B' (d) alloy 'C'

Fig. 4 shows the variations of hardness and DBTT evaluated by SP-test as a function of irradiation dose. They increased with similar manner as irradiation dose increased. Based on Fig. 4, relationship between ΔDBTT by SP-test and ΔHV is illustrated in Fig. 5 for alloys 'S' and 'A', where the results† by Kasada et al. [7] are also plotted as reference. It draws following linear relationship in Fig. 5

$$\Delta DBTT_{SP} = 0.474\Delta H_V \qquad (1)$$

This result accords with the previous ones [2,7] also reporting a linear relationship between ΔDBTT measured by Charpy test and ΔHV after the irradiation. Even though the change of DBTT after neutron irradiation could not be detected in the tested temperature range in alloy 'C', similar increment of

†Actually the DBTT data were measured with 1/3 sized Charpy V-notch specimens. The results were converted into SP-DBTT using the relationship among SP-DBTT, 1/3CVN-DBTT and CVN-DBTT[6,7]

transition temperature accompanying the increase of hardness might be expected considering the linear relationship observed in other alloys. It suggests that irradiation embrittement is primarily attributed to the irradiation defects, which is increased with irradiation dose, while the increase in Ni concentration from 0.9 to 3.5 wt.% did not have significant influence on the neutron irradiation embrittlement under irradiation flux below ~10^{20} neutron/cm^2 at 290 °C.

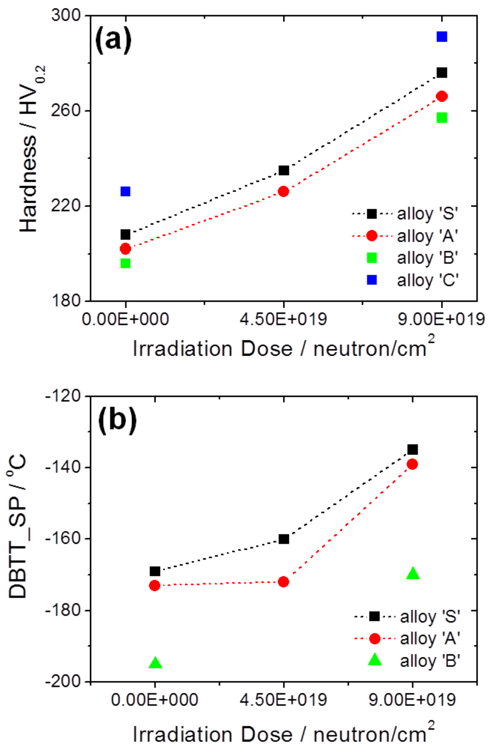

Fig. 4. (a) Hardness change, (b) DBTT change measured by SP-test after neutron irradiation.

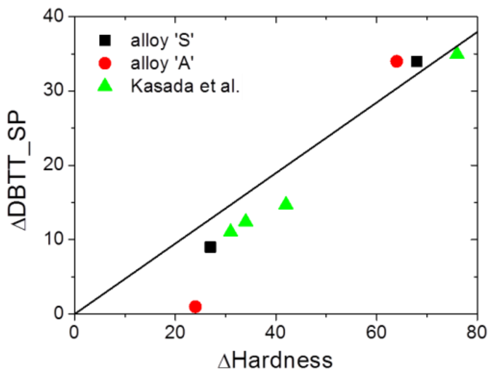

Fig. 5. Relationship between $\Delta HV_{0.2}$ and ΔDBTT measured by SP-test in alloys 'S' and 'A', compared with the results by Kasada et al. [7].

3. Conclusions

Ni-Mo-Cr steels for reactor pressure vessel containing different amount of Ni are irradiated under two dose conditions, one is 4.5×10^{19} neutron/cm^2 at 290 °C and the other 9.0×10^{19} neutron/cm^2 at 290 °C, to investigate the influence of Ni contents on the irradiation embrittlement. The irradiation embrittlement and hardening sensitivity are hardly affected by the Ni content in range of 0.9~3.5 wt.% when the concentration of Cu and other impurities are controlled to low level, but they are primarily influenced by the irradiation dose which is closely related with the generation of irradiation defects.

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