3D Atom Probe Tomography Analysis of Neutron-irradiated SA508 Gr.4N

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1. Introduction

The main lifetime limiting factor of reactor pressure vessels (RPV) of operating nuclear light water reactors (LWR) is radiation-induced embrittlement [1]. The embrittlement is induced by the formation of radiation defects (so-called 'matrix damage') and/or of radiation-induced precipitates. Cu-rich precipitates (CRP) have been observed in Cu-bearing RPV steels. In case of Cu-free, high Mn (and Ni and Si) containing RPV steels such as SA508 Gr.3 (1.4Mn-0.9Ni-0.15Cr-0.2C-0.002P in wt.%), Mn-Ni-rich precipitates (MNP) are reported to form at low temperature and high neutron fluence. These MNPs are believed to have long incubation time. But once nucleated, they rapidly grow to large volume fractions and mechanical properties deteriorate abruptly, e.g., increase in ductile-to-brittle transition temperature. For such reasons, MNPs are denoted as late blooming phases (LBP) [2].

MNPs are often observed in RPV steels with Mn content of ~1 wt.% and similar or lower Ni content. Atom probe experiments of neutron irradiated SA508 Gr.3, which has a composition of, showed Mn, Ni, Si, and P atoms are formed MNPs and segregated at dislocation lines [3].

Attempts have been made to improve fracture toughness and strength by increasing Ni and Cr, and decreasing Mn content. SA508 Gr. 4N is such an alloy, and the composition is 0.3Mn-3.64Ni-1.8Cr-0.2C-0.002P in wt.%. It is of interest what effect of increased Ni and decreased Mn content will have on the formation of MNPs. In this study, radiation-induced precipitates in neutron-irradiated SA508 Gr. 4N are evaluated using atom probe tomography (APT).

2. Methods and Results

2.1 Specimen and neutron irradiation

The chemical composition of the specimen used in this study is given in Table 1

Table 1. Chemical composition of SA508 Gr.4N

단위	Fe	Ni	Cr	Mo	Mn	Si	C	Cu	V	P	S
wt%	Base	3.6	1.8	0.49	0.3	0.24	0.21	0.03	0.006	0.003	0.002

Neutron irradiation was carried out at the High flux Advanced Neutron Application Reactor (HANARO). Before irradiation, specimen was mechanically polished using 1200-mesh SiC paper and 0.25- m-diamond suspension paste. In order to remove surface damage during mechanical polishing, the surface was gently polished using vibratory polishing machine. 3-mm-discs of 100- m-thickness were cut from the polished specimen. Irradiation was performed at 290 ℃ in OR5-irradiation hole for 53.6 days, which has fast neutron flux of 1.92x1013 n/cm2-sec. After irradiation, the irradiated samples were cooled down in Irradiation Material Examination Facility for 2.2 years. Dose rate was decreased from 40.4 to 27.8 μSv/hr after the cool-down period..

2.2 3D-APT sample preparation

Samples for 3D-APT were fabricated by using focused ion beam (FIB) machine at International Research Center for Nuclear Material Science in the Institute for Material Research (IMR, Tohoku University). FIB-milled specimen was lifted-out and attached on an integrated coupon array. Then final needle-shaped samples were made using annular ion milling [4]. Fig. 1 shows the sequence of 3D-APT sample preparation. Surface oxide layer on the neutron irradiated specimen was eliminated during FIB milling.

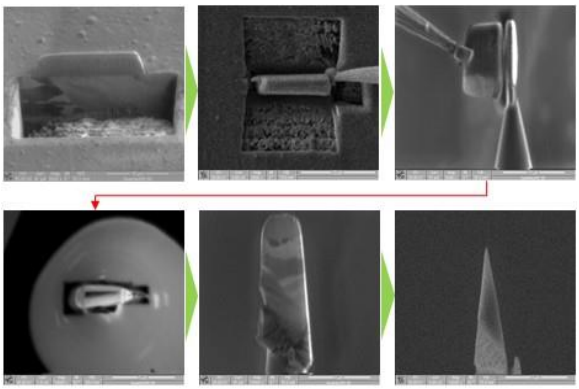

Fig. 1. 3D-APT specimen prepared by the FIB technique.

2.3 3D-APT experiment

3D-APT were performed at IMR by using the Local Electrode Atom Probes (LEAP 4000X HR, CAMECA Instruments shown in Fig. 2). Voltage pulse was applied on the apex of the sample (~10 kV, 200 kHz). The

experiment was performed at 60K in order to minimize heat effects.

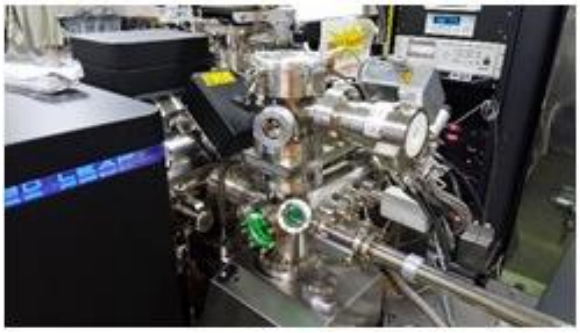

Fig. 2. LEAP 4000X HR.

2.4 3D-APT data analysis

3D-reconstruction of detected ion and Data analysis was performed using commercial IVAS 3.6.8 software program. Bulk composition of the data is posted on the table 2. Chemical composition are appropriate to the original chemical composition of SA508 Gr.4N.

Table 2. Bulk composition of 3D-APT analysis data.

단위	Fe	Ni	Cr	Si	Mo	Mn	C	P
AT%	94.4900	3.8571	0.6583	0.4486	0.2435	0.1989	0.0975	0.0061
WT%	94.4694	4.054	0.6128	0.4182	0.4182	0.1956	0.021	0.0034

3. Results and Discussion

Fig. 3 shows iso-surface image of Mn, Ni and Si elements. The contour surfaces correspond to 0.9, 6.45, and 1.35% for Mn, Ni and Si respectively.

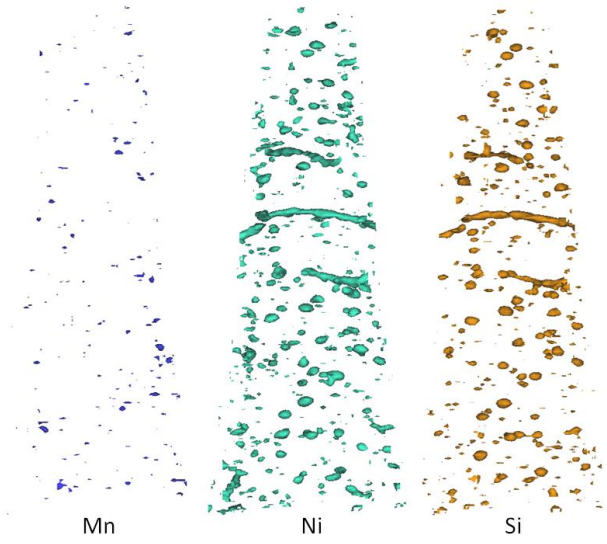

Fig. 3. ISO-surface image of Mn, Ni and Si.
(Mn: 0.9% Blue, Ni: 6.45% Green, Si: 1.35% Orange)

Small precipitates with a size of ~10 nm have been formed. These precipitates are rich in Ni and Si, i.e., the distributed positions of Ni and Si elements are coincident. Some of Ni and Si are segregated along lines, which are considered to be dislocations. The elements such as Ni, Si, Mn, and P are known to be dragged by vacancy. Vacancies formed by neutron irradiation are absorbed at sink sites, such as dislocations and grain boundaries. Vacancies attracted toward dislocations are thought to drag Ni and Si elements and formed the segregated lines shown in Fig. 3. It is of note that the distribution of Mn element has showed no clear coincidence with Ni. In high Mn steels, such as SA508 Gr. 3, MNPs were reported to form, in which both Mn and Ni elements are rich. The results in this study shows that Ni-Si rich precipitates without Mn could be formed by neutron irradiation.

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