# Anisotropic distribution of solute elements in ion – irradiated reactor pressure vessel model alloy


| | | |
|---|---|---|
| Nagaoka University of Technology | Ba Vu Chinh NGUYEN | Student – member |
| Nagaoka University of Technology | Kenta MURAKAMI | Member |
| Nagaoka University of Technology | Phongsakorn PRAK TOM | Non – member |
| Shianghai Jiao Tong University | Liang CHEN | Non – member |



Atom probe tomography (APT) has been performed on Fe-1.4Mn0.6Ni (at%) model alloy to study solute elements behavior under 2.8 MeV Fe ions at 400 $^0$C. Orientation imaging microscopy (OIM) analysis is applied on the detector hit map to determine the pole figure supporting for orientation calculation. The three-dimensional images show that Mn and Ni solute atoms are distributed close to some certain planes, namely <111> and <100> families. This arrangement of solute atoms on the habit planes of interstitial loop is directly evidence that the interstitial loops can trap solute elements during irradiation process.



Keywords: ion irradiation, atom probe tomography, solute elements, orientation calculation, dislocation loop.


## 1. Introduction

Because of the development of metallurgical techniques, current nuclear industry has switched to using low Cu RPV steels **[1]**. Therefore, manganese- and nickel-rich precipitates (MNP) have become a critical concern in the study of embrittlement of RPV materials. Numerous studies have observed MNPs in irradiated RPV steels **[2] [3] [4]**. Using atom probe tomography to investigate the WWER 1000 reactor weld under a total fluence of $1.4x10^{23}$ $nm^{-2}$ at 288 $^0$C, Miller and Russel **[5]** found that MNPs gathered to become small clusters with average size 1 – 2 nm in diameter. In 2014, Bonny **[6]**, after utilizing density functional theory as well as Monte Carlo simulations, suggested that MNP nucleation and growth is possible due to matrix defects. Furthermore, Meslin **[7]** has reported that the distribution of solute atoms was heterogeneously precipitated on the planar object, suggesting a dislocation loop or a grain boundary. Although there is evidence that solute atoms can be segregated to some certain planes, the question "what are these planes?" still has not been answered. To clarify the detail of anisotropic distribution of solute elements and to confirm the planar object containing solute elements whether is dislocation loop or is grain boundary, we have identified the orientation of solute elements distributed - planes based on lattice orientation of iron crystal.

## 2. Materials and methods

Fe-1.4Mn0.6Ni (wt.%) model alloy was prepared by solution annealing at 1050 $^0$C for 2h before quenching in water. Mechanical polishing as well as electrochemical method were utilized for surface treatment. 2.8 MeV ions $Fe^{2+}$ was irradiated to that bulk sample at 400 $^0$C up to 2 dpa at the High Fluence

Irradiation Facility of the University of Tokyo **[8]**.

SRIM 2013 **[9]** was used to calculate the irradiation dose with the displacement energy of 40 eV. The damage range from the surface to maximum damage at 800 nm in depth is considered as the interest region for APT study. Raw samples with triangular prism shape were preapared on the top of tungsten needle tips by using focus ion beam (FIB) system Hitachi FB2200 at Nagaoka University of Technology. The FIB technique continued with Hitachi NB5000 to prepare the final samples (needle - like shape, less than 50 nm diameter of the tip) for atom probe tomography.

APT measurements were conducted using the LEAP3000XHR by CAMECA at the Fugen Decomissioning Engineering Center of the Japan Atomic Energy Agency. Reconstruction parameters were carefully selected to optimize the length of z-axis. To know solute atom distribution, the iso-surfaces of solute elements were generated. Making the range of interests (ROI) corresponding to those of iso-surfaces, the certain planes containing solute clusters were built.

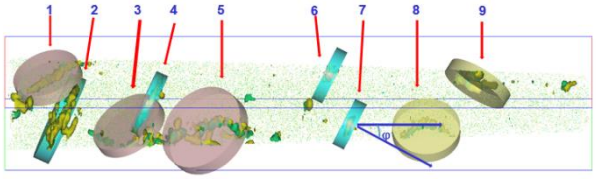


**Figure 1. Planes containing solute clusters**

The angle between the normal of the plane and z-direction was calculated **(table 1)**.

## 3. Results and discussion

### 3.1. Orientation determination

Based on the coordinates of the polar points from field ion microscpe (FIM) image, crystal orientation of specimen in z-direction was determined. From these calculations, the direction of z is very close to (2 1 -1).

**Table 1. The angle φ between the normal of the above planes and z direction**

| Plane | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 |
|---|---|---|---|---|---|---|---|---|---|
| Abs. φ (degree) | 75 | 20 | 62 | 18 | 61 | 24 | 24 | 84 | 85 |

Theoretical calculation of the interplanar angle φ' between <111> and <100> family orientations and (2 1 -1) direction for cubic system give us the angles as below:

**Table 2. Theorical calculation of interplanar angle between (hkl) plane and (21-1)-direction**

| (hkl) | (111) | (-111) | (1-11) | (11-1) | 100 | 010 | 001 |
|---|---|---|---|---|---|---|---|
| Angle (degree) | 61.9 | -61.9 | 90.0 | 19.5 | 35.3 | 65.9 | -65.9 |
| Matched | Plane 5 | X | Plane 8, 9 | Plane 2, 4, 6, 7 | X | Plane 3 | Plane 1 |

Firstly, based on the matching between calculated results and theoretical angles, the group of four parallel planes (number 2, 4, 6, 7) were pointed out as (11-1) lattice plane. In order to accurately determine other planes, 3D structure of the specimen was rotated around z-axis. The rotated angles are used to indicate details of those planes **(table 2).**

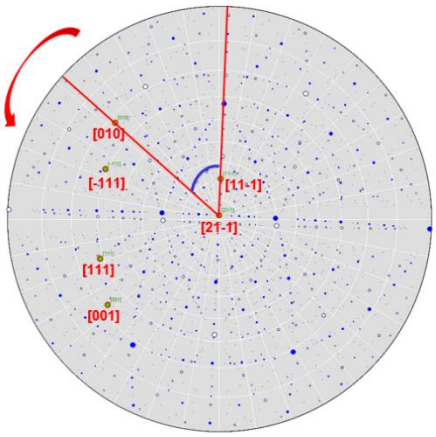


**Figure 2. Poles figure of bcc structure**

**In case pole [21-1] is close to the center**

The results **(table 2)** show that the solute clusters

are distributed in some certain planes, namely in <111> (for plane 2, plane 4, plane 6, plane 7, plane 8, plane 9) and <100> (plane 1 and plane 3) family orientations with the ring- or curve - shape. Interestingly, <111> and <100> orientation are two types of habit planes of dislocation loop in bcc structure **[10] [11]**. This matching suggests that the solute elements can be trapped by dislocation loop.

TEM results **[12]** have also indicated that both two types of loop were displaced in irradiated steels. Basically, in pure iron, <100> loop is more stable resulting the higher number density than 1/2<111> loop **[12]**.

## 3.2. Number density calculation

Because of the limited size of sample, there is only one fully loop appeared in the APT result; the others are some dislocation lines **(figure 3)**.

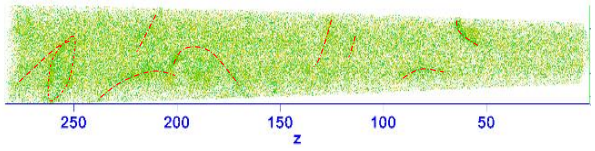


**Figure 3. Mn and Ni distribution in 50nm x 50nm x 280nm**

To calculate the number density of dislocation loop in 3-dimension, the fully loop (contained in plane 2) is used as reference; the total length of this loop (about 97 nm) has been normalized to 1 (loop). Total number of loops inside the volume of 50nm x 50nm x 280nm was calculated as: $N = \frac{\sum_1^9 l_i}{97}$ , then the number density is: $n = \frac{N}{V}$.

The order of $10^{21}$ ($m^{-3}$) in number density of dislocation loop is similar with the other reports using TEM observation **[13] [14]**. This is an addition evidence that anisotropic distribution of solute elements in bcc Fe alloy is decorated along with the dislocation loop.

**Table 3. Calculation of number density**

| Dislocation line number | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 |
|---|---|---|---|---|---|---|---|---|---|
| Dislocation length (nm) | 30 | 97 | 28 | 30 | 38 | 25 | 13 | 29 | 24 |
| Total length (nm) | 314 | | | | | | | | |
| No. density of <100> ($m^{-3}$) | $1.0 \times 10^{21}$ | | | | | | | | |
| No. density of <111> ($m^{-3}$) | $3.6 \times 10^{21}$ | | | | | | | | |
| Total number density ($m^{-3}$) | $4.6 \times 10^{21}$ | | | | | | | | |

## Conclusion

Our study aims to clarify the detail of anisotropic distribution of solute elements in manganese nickel steel model alloy. To reach that purpose, Fe-1.4Mn0.6Ni (at%) model alloy was irradiated up to 2 dpa with 2.8 MeV ion $Fe^{2+}$ at 400 $^0$C. The observation outcome points out that solute atoms Mn and Ni are not homogenously distributed but are segregated to both types of habit planes of dislocation loop. Number density of loops has also been calculated. The higher number density of 1/2<111> loops than that of <100> loop was found suggested that 1/2<111> may be less mobile due to segregation of solute atoms.

## References

**[1]** N. Yamashita, M. Iwasaki, K. Dozaki, N. Soneda, J. Eng. Gas Turbines Power 132 (2010) 102919 –102919.

**[2]** P. Pareige, J.C. Van Duysen, and P. Auger. Applied Surface Science. 67 (1993) 342-374.

**[3]** A. Wagner, A. Ulbricht, F. Bergner, and E. Altstadt. Nuclear Instruments and Methods in Physics Research B. 280 (2012) 98-102.

**[4]** S. Ortner. “Anomalous Hardening in RPV Surveillance and MTR Data.” National Nuclear

Laboratory Report 10804. Issue 2. 2010.

**[5]** M.K. Miller and K.F. Russell. Journal of Nuclear Materials. 371 (2007) 145-60.

**[6]** G. Bonny, D. Terentyev, A. Bakaev, E.E. Zhurkin, M. Hou, D. Van Neck, and L. Malerba. Journal of Nuclear Materials. 442 (2013) 282-291.

**[7]** E. Meslin et al, Radiation-induced precipitation in a ferritic model alloy: An experimental and theoretical study, Acta Materialia 61 (2013) 6246–6254.

**[8]** K. Murakami, T. Iwai, H. Abe, N. Sekimura, Nucl. Instrum. Methods Phys. Res. B, 381 (2016), pp. 67-71.

**[9]** Ziegler J.F., Ziegler M.D., Biersack J.P. Nuclear Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms, 268 (11-12) (2010) , pp. 1818-1823.

**[10]** W. Carrington et al., D. Arrangement of dislocations in iron. Proc. R. Soc. A 259, 203 (1960).

**[11]** B.C. Masters, Dislocation loops in irradiated iron. Nature 11, 254 (1963).

**[12]** J. C. Haley at al., Acta Materialia 136 (2017) 390 – 401.

**[13]** K. Yabuuchi et al., Journal of Nuclear Materials, 442 (2013).

**[14]** H. Watanabe et al,. Journal of Nuclear Materials, 439 (2013).