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Identification of ultra-fine Ti-rich precipitates in V–Cr–Ti alloys irradiated below 300 °C by using positron CDB technique

Ken-ichi Fukumoto [a,*], Hideki Matsui [b], Hideaki Ohkubo [c], Zheng Tang [b], Yasuyoshi Nagai [d], Masayuki Hasegawa [d]

[a] Graduate School of Nuclear Power and Energy Safety Engineering, University of Fukui, Fukui 910-8507, Japan
[b] Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
[c] Institute of Advanced Energy, Kyoto University, Uji 611-0011, Japan
[d] International Research Center of Nuclear Material Science, Tohoku University, Oarai 311-1313, Japan

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Abstract

Irradiation-induced Ti-rich precipitates in V–Ti and V–4Cr–4Ti alloys are studied by TEM and positron annihilation methods (positron lifetime, and coincidence Doppler broadening (CDB)). The characteristics of small defect clusters formed in V alloys containing Ti at irradiation temperatures below 300 °C have not been identified by TEM techniques. Strong interaction between vacancy and Ti solute atoms for irradiated V alloys containing Ti at irradiation temperatures from 220 to 350 °C are observed by positron lifetime measurement. The vacancy-multi Ti solute complexes in V-alloys containing Ti are definitely identified by using CDB measurement. It is suggested that ultra-fine Ti-rich precipitates or Ti segregation at periphery of dislocation loops are formed in V alloys containing Ti at irradiation temperatures below 300 °C.

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1. Introduction

Vanadium-base alloys are attractive candidates of low activation structural materials for future fusion reactors. The alloys containing 4–5% Cr and 4–5% Ti exhibit high radiation damage resistance above 430 °C [1]. The addition of Cr is effective for high temperature mechanical performance and the addition of Ti in vanadium is effective for the suppression of void swelling under neutron irradiation in vanadium alloys. However, significant radiation hardening and embrittlement occur in neutron-irradiated V–4Cr–4Ti alloys in the temperature regime below 400 °C even in low damage levels below a few displacement per atom (dpa) [2,3]. This is one of the key issues of research and development of vanadium alloys to define the lower temperature limit for design windows of advanced fusion reactors.

In general, irradiation embrittlement of metals and alloys at lower temperatures is caused by irradiation hardening due to the formation of high concentrations of defect clusters and precipitates during neutron irradiation. A marked transition in the microstructure of V–4Cr–4Ti irradiated to 0.1–0.5 dpa has been observed in the temperature regime around 400 °C, where the tensile properties show a corresponding large change. Only small dislocation loops were observed at irradiation temperatures below 290–300 °C. On the other hand, small Ti (OCN) precipitates on {0 0 1} habit planes were observed at temperatures above 300 °C, and these precipitates were the dominant microstructural feature at elevated temperatures. The large decrease in dislocation loop density with coarsening of the Ti (OCN) precipitates at temperatures above 350 °C may be responsible for the pronounced decrease in radiation

* Corresponding author. Fax: +81 776 27 9712.
E-mail address: fukumoto@mech.fukui-u.ac.jp (K.-i. Fukumoto).

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hardening observed in this temperature regime [3]. However, it has been reported that a high density of small faulted dislocation loops, $a/2[1\,1\,0]$ loops were observed at irradiation temperatures below 300 °C in V–4Cr–4Ti alloys [4,5]. These dislocation loops became unfaulted at temperatures above ~300 °C and the dislocation barrier strengths of the faulted dislocation loops and {0 0 1} defect clusters were estimated to be 0.4–0.5 and 0.25, respectively. Thus, the small defect cluster formed at irradiation temperatures below 300 °C has a two times higher value of dislocation barrier strength than {0 0 1} defect clusters [4]. In general, a faulted loop in a bcc structure has too high stacking fault energy to form such a faulted loop in vanadium alloy even in irradiation environment. We have an interest in the internal structure of the small cluster formed in V–Ti containing alloys at irradiation temperatures below 300 °C.

In order to reveal this effect, the precipitation processes of Ti-rich precipitates and Ti (OCN) should be well understood. For this purpose, we have irradiated vanadium alloys (V, V–5Ti, V–4Cr–4Ti) by neutrons to induce defect clusters due to displacement cascades and have studied the defect–solute interaction by using positron annihilation methods: positron lifetime and coincidence Doppler broadening (CDB) of positron annihilation radiation.

The CDB method is the useful tool for getting knowledge of the microstructures of ultra-fine sub-nanosized precipitate and the nature of vacancy-solute atoms complexes in irradiated materials. From this study, it is found that the Ti-rich precipitate and vacancy–Ti solute atoms are sensitive for the CDB method and it is the powerful tool for the detection of the Ti-rich precipitation sign in vanadium alloys containing Ti atoms irradiated at low temperature.

2. Experimental procedure

Vanadium alloys in this study were unalloyed vanadium (C: 16 wt ppm, N: <1 wt ppm, O: 300 wt ppm), V–5 wt%Ti (C: 43 wt ppm, N: 14 wt ppm, O: 390 wt ppm), and V–4 wt%Cr–4 wt%Ti (C: 22 wt ppm, N: 10 wt ppm, O: 380 wt ppm). They were made by arc-melting and cold rolling to a thickness of 0.25 mm. The samples were punched out to disks of 3 mm diameter and annealed at 1100 °C for 2 h in a vacuum of $<2\times10^{-4}$ Pa. Irradiation experiments were performed in Japan Materials Testing Reactor (JMTR) with an irradiation temperature controlled irradiation rig. In-core irradiation rigs that are capable of controlling the irradiation temperature and are not influenced by the reactor power was used [6,7]. A couple of irradiation conditions were selected for the experiments, 4.5×10^{23} n/m^2 ($E>1$ MeV) at 220 °C and 1.0×10^{24} n/m^2 ($E>1$ MeV) at 350 °C. The corresponding displacement damage doses were estimated to be 0.08 and 0.17 dpa (displacement per atoms), respectively. Some of irradiated specimens were fabricated into TEM specimens after positron annihilation measurements in order to investigate microstructural changes by irradiation. TEM observations were performed with accelerating voltage of 200 kV by JEOL-2010.

Positron lifetime measurements were carried out using a conventional fast–fast spectrometer with a time resolution of 190 ps in full width at half maximum (FWHM). About 4×10^6 coincidence events were accumulated for each measurement for 12 h. After subtracting the source component and background, the spectra after irradiation were decomposed into two components (τ_1 and τ_2). The details of the method are described in reference [8]. A positron lifetime of bulk vanadium is 117 ps, and the positron lifetime of mono-vacancy in vanadium and that of a mono-vacancy–oxygen complex has been reported to be about 190 ps and 160 ps, respectively [9].

The CDB enables us to identify the chemical element whose electron annihilates with the positron, [10–12] by measuring the electron momentum distribution in the high momentum region, given by the positron annihilation with the inner orbital electrons. We can identify the chemical environment, where the positron is trapped, because the inner orbital electrons are tightly bound to the nuclei and are almost unaffected by the chemical bonding and crystal structure. Thus, the CDB method is expected to prove the formation of vacancy-solute complexes. The positrons are also sensitively trapped at the positron affinitive ultra-fine precipitates. In the present systems, the nanosize Ti precipitates in V are the positron trapping sites [13,14]. Thus, the CDB can also detect the presence of ultra-fine precipitates and initiation of radiation-induced precipitation. The CDB spectra were measured with 2 Ge detectors in coincidence. The energies of the annihilating γ-ray pairs were simultaneously recorded with these detectors located at an angle of 180° relative to each other [11]. The overall energy resolution is ~1.1 keV (FWHM), which corresponds to a momentum resolution of; $\sim4.3\times10^{-3}m_0c$ (FWHM), where c is the speed of light and m_0 is the electron rest mass. The sample-detector distance is 20 cm, and the strength of the ^{22}Na positron source is; ~1 MBq. Total counts of more than 2×10^7 for each measurement was accumulated for 12 h. The CDB ratio spectrum was obtained by normalizing the momentum distribution of each spectrum to that of the well annealed (defect-free) highly purified V. The shape of the spectrum in the high-momentum region (typically $>10\times10^{-3}m_0c$) exhibits characteristic signals of the elements through the positron annihilation with their inner orbital electrons. The S and W parameters are defined as the ratios of low momentum ($|p_L|<4\times10^{-3}mc$) and high momentum ($14\times10^{-3}mc<|p_L|<25\times10^{-3}mc$) regions in the CDB spectrum to the total region, respectively. When the positron is trapped at a vacancy-type defect the S parameter increases, while the W parameter decreases. In addition, the W parameter strongly depends on the chemical environment of the positron-trapping site. From the reference of positron affinity of each element, the value of positron affinity for Ti and V is −4.06 eV and −3.44 eV, respectively. The energy difference of positron affinity between Ti and V is 0.62 eV and it corresponds to 7.9 Å for

a critical size of particle for capturing positron inside [15]. The value of positron affinity for Cr is −2.62 eV, thus the positron affinity for Cr is weaker than that for V, bulk material. It means that the existence of Cr in V–Cr–Ti alloys may not affect the measurement for the CDB in order to investigate the positron trapping behavior in Ti solute atoms in vanadium alloys. It is anticipated that a positron works as a probe for Ti-rich clusters in bulk V, like for Cu-rich clusters in bulk Fe in the successful previous works [16,17].

3. Results

In order to investigate the crystallographic microstructure, TEM observations were performed and some experimental results of TEM works in the previous studies are referred with the present data. Fig. 1 shows the tiny defect clusters in V–5Ti observed by TEM after low temperature irradiation with damage levels of <0.1 dpa. Only small defect clusters may be dislocation loops were observed at irradiation temperatures below 300 °C. They are considered to be dislocation loops. However, small Ti (OCN) precipitates on (0 0 1) habit planes were observed at temperatures above 300 °C, and these precipitates were the dominant microstructural feature at elevated temperatures. It appears the large decrease in dislocation loop density and concomitant coarsening of the Ti (OCN) precipitates at temperatures above 350 °C in V–5Ti. In this study, the nature of small defect cluster formed below 300 °C was not determined by TEM observation. The size of defect cluster was too small (a few nm) to make a $\vec{g}\cdot\vec{b}$ criterion technique and to observe a stacking fault fringe inside the defect clusters in order to identify whether the dislocation loop is faulted or unfaulted type.

In order to characterize the nature of defect clusters unidentified by TEM and determine the morphology of vacancies and vacancy clusters, the positron lifetime measurement and the CDB method were used. Fig. 2 shows the positron lifetime measurements in the pure V, V–4Cr–4Ti and V–5Ti alloys irradiated in JMTR. The calculated positron lifetimes in bulk state and mono-vacancy (V_1) in V are also shown by horizontal dash lines. In the case of pure V after irradiations, the longer life component (τ_2) of 450 ps, which corresponds to large vacancy cluster like a nanovoid, was observed. A similar result of positron annihilation measurement in V–Ti alloys has been reported and the tendency of positron lifetime was in agreement with this work [18]. In order to determine the correlation between the positron lifetime and size of vacancy clusters, the positron lifetime at microvoids was calculated for pure vanadium, by an improved superimposed-atom method [9]. Fig. 3 shows the calculated positron lifetime as a function of the number of vacancies in a microvoid. It is calculated that the radius of about 0.5 nm (about V_{27}) corresponds to about 430 ps of positron lifetime in pure V. Also voids were observed in pure V irradiated at 300 °C with 0.05 dpa from the previous TEM works [19], the vacancy clusters like microvoids were formed by the migration of vacancies during irradiation or direct formation of vacancy clusters due to damage cascades. From the results of both TEM work and positron annihilation measurement, it is suggested that invisible vacancy clusters for TEM remain with visible dislocation loops and microvoids in the irradiated pure V.

The τ_1 for the irradiated V–5Ti and V–4Cr–4Ti alloys are ranged from 130 ps to 180 ps. The lifetime of positron annihilation in unirradiated pure Ti is about 146 ps and the existence of Ti may not affect the long lifetime component for V alloys containing Ti. It means that it cannot be decomposed into more than three components in the positron lifetime analysis so that there are few defect components with long lifetime. This suggests that the irradiation induced vacancies are stabilized by forming vacancy-solute

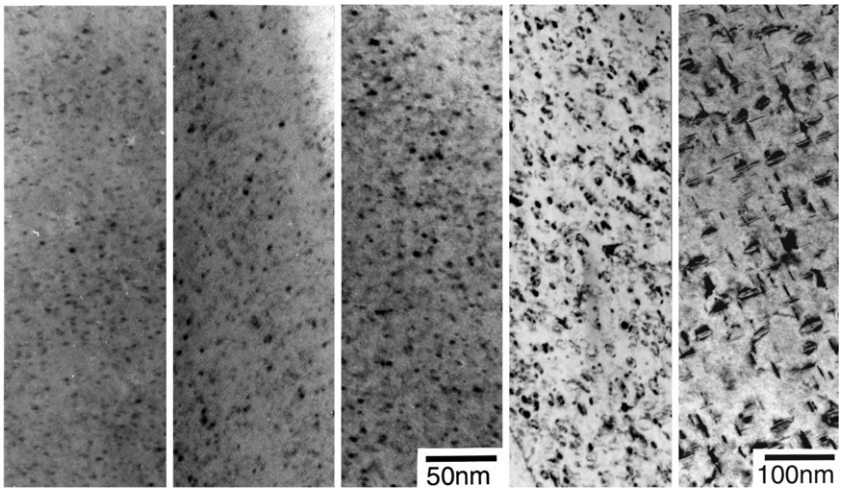

Fig. 1. A set of TEM micrographs for neutron-irradiated V–5Ti alloy in JMTR. Irradiation temperatures are ranged from 90 °C to 400 °C and damage levels are 0.05–0.17 dpa.

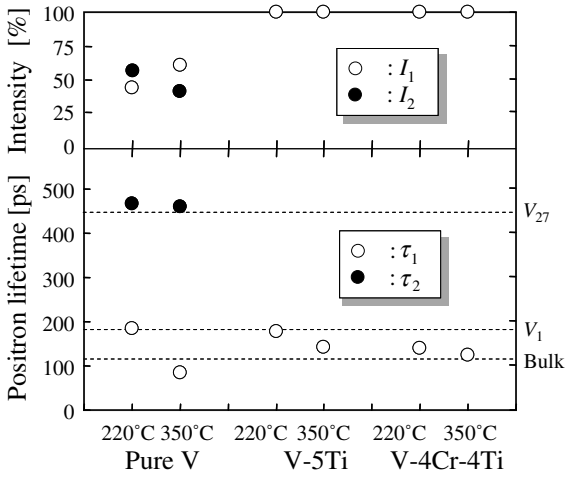

Fig. 2. Positron lifetimes in pure V, V–5Ti and V–4Cr–4Ti alloys irradiated at 220 °C and 350 °C. Calculated positron lifetimes for bulk state and V_1 in vanadium are shown by horizontal dashed lines.

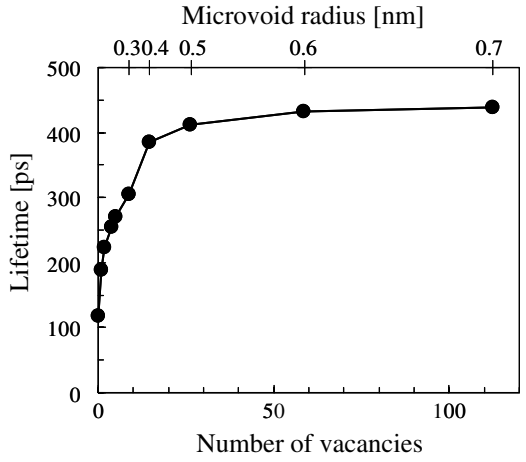

Fig. 3. Calculated positron lifetime as a function of nanovoid size in pure vanadium.

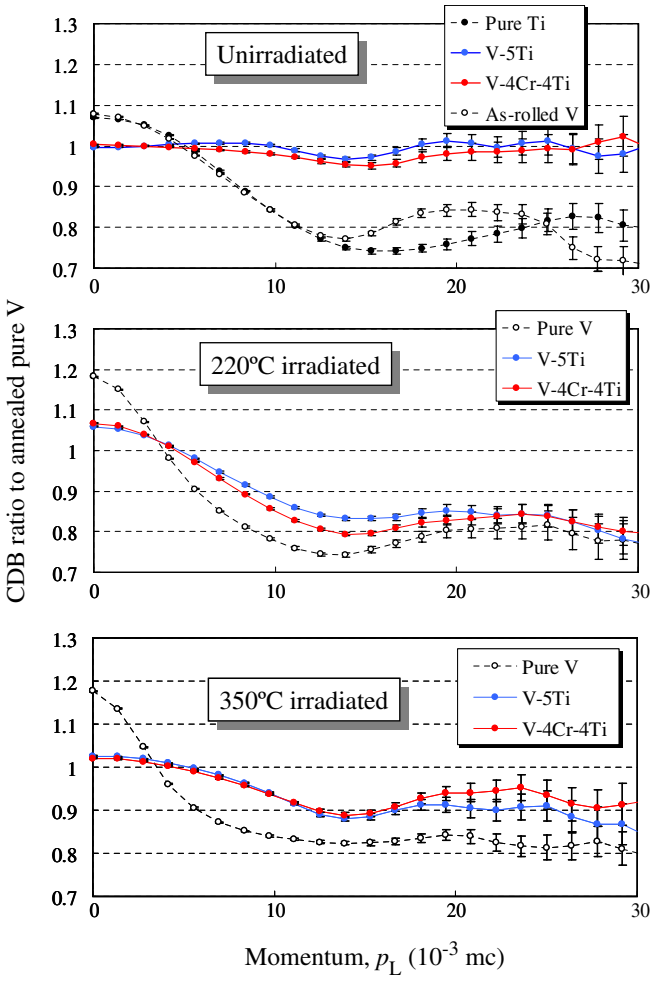

Fig. 4. Ratio curves of CDB spectra in unirradiated, 220 °C-irradiated and 350 °C-irradiated samples: pure V, V–5Ti, V–4Cr–4Ti and pure Ti. The ratio curves are normalized to the momentum distribution of well-annealed (defect free) V that contains quite low impurity levels (O: <20wt ppm, C, N: <5wt ppm).

complexes. In particular, the high I_2 values (~100%) for V–5Ti and V–4Cr–4Ti alloys indicate there is the only mono-vacancy of defect type in the bulk and the strong interaction between the vacancies and the solute Ti atoms. It is also suggested that Ti addition for vanadium alloys suppressed the nucleation of vacancy clusters and it is effective for the suppression of swelling under irradiation as mentioned in our previous work [20].

Fig. 4 shows the CDB ratio spectra for the unirradiated samples and irradiated samples. The CDB ratio spectra are normalized to the momentum distribution of well-annealed (defect free) pure V. The ratio spectrum for well-annealed pure Ti is also shown as a reference in Fig. 4. For pure Ti, the broad valley around $16\times10^{-3}mc$ in the spectrum shows that the positrons annihilate with inner electrons of Ti atoms. The enhancement in the low-momentum region (less than $5\times10^{-3}mc$) shows that the positrons are trapped in the vacancies, consistent with the results of positron lifetime mentioned above. Even though the enhancement in the low momentum region is also found in pure Ti, the peak height of the irradiated pure V is quite larger than unirradiated pure Ti and irradiated V alloys containing Ti.

4. Discussions

To see the shape of CDB ratio spectra more quantitatively, the S and W parameter correlation (S–W plot) is shown in Fig. 5. Reflecting the positron annihilation with the inner electrons of Ti, the W parameter of pure Ti is low. On the other hand, since positrons are trapped in vacancy clusters for the irradiated pure V, the S parameter of the irradiated pure V is high. In the case of pure V, vacancy clusters induced at low temperature show the large S parameter in (S, W) points. According to the small increase of W parameter and the small decrease of S parameter by increasing irradiation temperature for pure V, the (S, W) points moves to the point of unirradiated pure V. This is caused by the growth of vacancy clusters

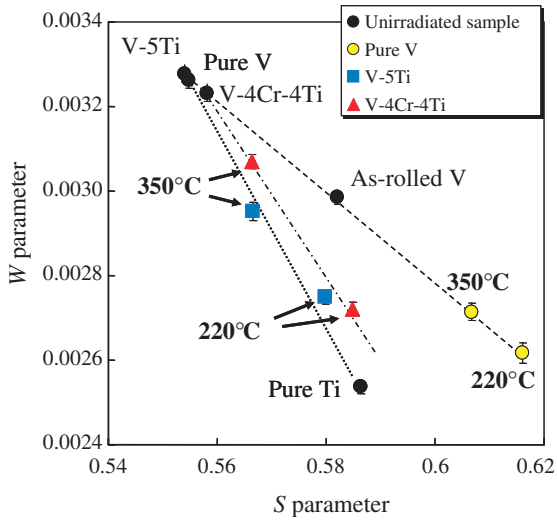

Fig. 5. *S–W* plots of unirradiated and irradiated samples: pure V, V–5Ti, V–4Cr–4Ti and pure Ti.

like nanovoids and the reduction of density of small vacancy clusters as increasing irradiation temperatures in pure V. A characteristic of a tendency line connecting the (*S*, *W*) points between unirradiated pure V and irradiated pure V can be elucidated by the position of (*S*, *W*) points for as-rolled pure V. The as-rolled pure V contains a lot of mono-vacancies formed in the matrix during rolling process and its (*S*, *W*) points show that the positrons are trapped in mono-vacancies for pure V. Furthermore, during rolling process, vacancy–oxygen complexes are dissociated into vacancies and oxygen atoms each other, and the signal of positron lifetime for as-rolled pure V does not shows the lifetime of vacancy–oxygen complexes but that of mono-vacancies. From the positron lifetime measurement, it was found that both mono-vacancies and large vacancies clusters such as voids and nanovoids invisible for TEM remained in pure V irradiated at 350 °C. Some of short lifetime component consist of dislocation core in dislocation loops, but the most part of short lifetime components should consist of mono-vacancies because the dislocation density in pure V irradiated at 350 °C was not so high in the order of 10^{12} m^{-2} from TEM works from previous study [19]. Consequently, the tendency of (*S*, *W*) points for pure V irradiated at 220 °C and 350 °C indicates that the positrons are trapped in mono-vacancies.

On the contrary to the behavior of (*S*, *W*) points for pure V, the (*S*, *W*) points of irradiated ones for V–5Ti and V–4Cr–4Ti alloys are on a straight line toward the point of pure Ti. As irradiation temperature increases, the (*S*, *W*) points move to the point of unirradiated original samples significantly. From the positron lifetime measurement, it was found that the vacancy clusters were not formed but vacancy–Ti solute complexes were formed in V–5Ti and V–4Cr–4Ti alloys. The difference from the line of vacancy defect clusters on the (*S*, *W*) plot is caused by the positron annihilation with the electron of Ti combined with a vacancy. It suggests that there is a strong vacancy–Ti solute interaction and vacancy–Ti solute complexes formed at low temperatures below 300 °C. When irradiation temperature increases, Ti aggregates by vacancy diffusion process and Ti-enriched clusters invisible for TEM grow up and turn into Ti (OCN) precipitates visible for TEM [21,22]. The transition from Ti-enriched clusters to Ti (OCN) decreases the site of vacancy-solute Ti atoms in the matrix and lowers the frequency of positron trapping in the core electrons in Ti at vacancy–Ti solute complexes. The return of the (*S*, *W*) points to unirradiated sample suggests that the conversion of Ti-enriched clusters into Ti (OCN) occurred when irradiation temperature increased from 220 °C to 350 °C. A Ti-rich precipitate consists of a lot of Ti–Ti bonding in the clusters but a Ti (OCN) consists of a mixture between Ti (OCN) bonding and Ti–Ti bonding. Thus, *(S, W)* position of V–Ti alloys containing Ti (OCN) may no longer lie on the line toward pure Ti. If the mono-vacancies are not associated with Ti, the (*S*, *W*) points should lie on the line for pure V. It is also suggested that the positrons were still trapped in vacancy–Ti solute complexes strongly and the formation of vacancy clusters did not occur even though the fraction of Ti–Ti bonding decreased at 350 °C due to the conversion of Ti-enriched clusters to Ti (OCN).

From the results of positron lifetime measurement and CDB measurement, there are apparent evidences to show that the characteristics of small defect cluster formed at irradiation temperatures below 300 °C is a type of Ti-rich precipitate. Otherwise, Ti solute atoms aggregate and concentrate around the expanded region of dislocation core for dislocation loops by diffusion process during irradiation, and it affects the phase contrast to show stacking fault fringes inside a loop. More or less, it is apparent that the Ti aggregation occurred around vacancies in V–Ti alloys and V–Cr–Ti alloys irradiated below 300 °C from this study. The enrichment of Ti solute in defect clusters formed at low irradiation temperatures will make the dislocation barrier factor increased and assist the significant irradiation hardening. Not only Ti (OCN) precipitate, but also the Ti-rich precipitate may work as a large barrier against dislocation movement during or after low temperature neutron irradiation. In this study, the role of Cr for precipitation is not explained at all, because the positron affinity of Cr is weaker than any other element in V–Cr–Ti alloys. The size factor of Cr to V is about 0.86 and the Cr atom is undersized for V, so that the Cr atom is not so attractive site for vacancy migration. Since there is no aggregation of Cr atoms during neutron irradiation in bulk and precipitation from TEM analysis, it is concluded that the effect of Cr addition for V is not effective for irradiation-induced precipitation process.

5. Conclusion

Pure V and V alloys containing Ti (V–5Ti and V–4Cr–4Ti) irradiated at low temperatures (100–400 °C) in JMTR are studied by the TEM, positron lifetime and the CDB methods. Large vacancy clusters like microvoids are

formed in pure V even irradiated at 220 °C with 0.08 dpa. Positron lifetime of mono-vacancy can be only found in V–Ti bearing alloys, but the CDB measurements show the existence of a strong vacancy–Ti solute interaction. Thus the vacancy multi-Ti complexes are formed in the matrix at low temperature below 300 °C. TEM observation cannot identify the nature of the small defect clusters formed at irradiation temperatures below 300 °C, but the results of CDB methods indicate an ultra-fine Ti-rich precipitate or Ti segregation at the periphery of dislocation loops. These results are important for understanding the fundamental precipitation processes of ultra-fine Ti-rich precipitate and Ti (OCN) precipitate and the mechanism of the significant irradiation hardening of the V–4Cr–4Ti alloys under low temperature irradiation.

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