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Plasma–surface interaction effects during high ion temperature long pulse experiments in TRIAM-1M

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Abstract

TRIAM-1M tokamak is very suitable for studies on plasma material interaction because of its very long duration plasma discharges. We have investigated the damage and modification of plasma facing materials by exposing to the plasma in the scrape-off layer (SOL) of this machine and review the recent results in this paper. Dense dislocation loops were formed in Mo and W by exposing to the plasma for a few 10 min. This damage was caused by energetic charge-exchange hydrogen neutrals generated mainly in the lower half of the plasma. The mean flux of responsible neutrals for the damage was estimated to be 3×10^{17} m^{-2} s^{-1} sr^{-1}. Plasma facing surfaces were covered by an impurity deposit consisting of very fine crystalline grains (about 1 nm in size), whose crystal structure is not normal body-centered-cubic (bcc) structure but face-centered-cubic (fcc) structure. It was found that retention of hydrogen in these peculiar materials is much stronger than 'normal' Mo. © 2001 Published by Elsevier Science B.V.

Keywords: Hydrogen; Hydrides; Plasma–materials interaction; Redeposition

1. Introduction

The plasma facing walls of fusion devices are bombarded not only with plasma particles but also with neutral hydrogen atoms created in charge exchange collision processes [1]. It has been reported that some part of the neutrals having a high energy cause significant surface sputtering as well as internal damage in the plasma facing materials [2–5]. The atoms sputtered from the surface, on the other hand, re-deposit on it. If the redeposition exceeds sputtering, impurities cover the wall surface and modify its properties [6–12]. One can say that the plasma facing surfaces of the actual machines are more or less covered by the deposited layer. Therefore, it is important to make clear the properties of the layer to understand the plasma wall interaction that happened in the actual devices.

TRIAM-1M, superconducting high field tokamak, is a very suitable machine for studies on plasma material interaction because of its very long duration plasma discharge. In this paper, we review our recent works on the effects of plasma surface interaction to the plasma facing materials in TRIAM-1M under high ion temperature mode long pulse discharge [5]. It will be emphasized in this paper that the energetic particle bombardment and impurity deposition strongly affect material performance and hydrogen recycling.

2. Experimental procedures

TRIAM-1M at Kyushu University is a superconductive high field tokamak with a vacuum vessel made of 304SS, divertor plates of Mo and limiters of Mo. Non-metallic materials such as graphite have not been used for in-vessel components. To examine the damage and surface modification due to the plasma wall interaction, a surface probe system, with which one can transfer the material samples in the plasma without breaking the vacuum of the torus, was installed (see Fig. 1). The head of the probe, on which the specimens

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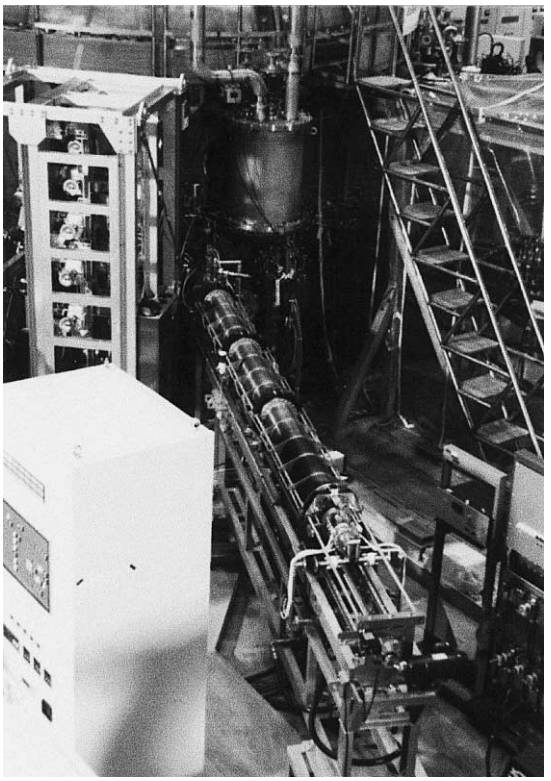

Fig. 1. Surface probe system connected to TRIAM-1M.

for material test are mounted, is actively cooled by flowing water and the temperature of the probe head can be monitored by thermocouples.

Various kinds of metal specimens mounted on the probe head were inserted into the scrape-off layer (SOL) through a horizontal port; 6 mm behind the poloidal limiter surface. In order to collimate the direction of the incident particles, the specimens were placed in the holes (4 mm in diameter and 4 mm in depth) at the plasma facing side (P-side). The specimen holes direct to the lower side $(-45°, -30°)$, the plasma center $(0°)$ and the upper side $(30°, 45°)$ with a semi-angle of 14°. Details are described elsewhere [13]. The pre-thinned Mo specimens placed at the bottom of the holes were exposed to successive high ion temperature discharge (hydrogen plasma, limiter configuration) sustained by lower hybrid current drive (2.45 GHz). Typical plasma parameters were as follows: $T_i = 1.5$–2.5 keV, $\overline{n_e} = 1.5 \times 10^{18}$ m^3, $I_P = 20$–25 kA. The duration time of each discharge was about 1 min and the total reached 31.5 min. The temperature of the probe head during exposure was almost constant at about 23°C. Pre-thinned 304SS and W specimens were mounted on the electron drift side (E-side) of the probe head to examine the impurity deposition. After the exposures to the plasma microscopic damage and the deposited layer were examined with a transmission electron microscope (TEM) with EDS.

According to the result that major metallic components of the impurity deposits in the TRIAM-1M were Mo [11,12], vacuum-deposition experiments of Mo were carried out in various atmospheres for a better understanding of the impurity deposits in TRIAM-1M. Taking into account that major gas component under a plasma discharge is hydrogen and some oxygen and water exist as residual impurities, the vacuum-depositions were carried in a high vacuum ($< 10^{-4}$ Pa) and low-pressure hydrogen or oxygen atmospheres. Microstructure of the deposits was also examined with a TEM.

Mo-deposits formed in oxygen atmosphere were implanted with 6 keV–D_3^+ ions (2 keV–D^+) at room temperature up to a fluence of 1×10^{22} ions m^{-2}. Thermal desorption of gas molecules composed of deuterium (DH, D_2, DHO and D_2O) was examined to estimate the impacts of the deposits on the hydrogen retention.

3. Experimental results and discussion

3.1. Radiation-induced damage

High-energy charge-exchange hydrogen neutrals escaping form the high-temperature core plasma will impinge on the first wall. Though the flux of the neutrals is much less than that of the ions, they can result in significant sputtering erosion as well as knock-on damage in the materials [4]. TEM observation made it clear that dense dislocation loops were formed even in Mo and W by exposing to the plasma for a few 10 min [2–5]. This fact directly indicates that plasma facing materials suffer radiation damage by plasma discharge and the formation of defects may change material properties such as thermal conductivity, mechanical properties and hydrogen retention.

Fig. 2 shows a dark field TEM image of radiation damage formed in the Mo specimen directing to the lower side of the torus plasma (−45°). Defects with a white dot image are interstitial type dislocation loops, which are aggregates of interstitial atoms formed by knock-on damage. The directional dependence of the area density of the dislocation loops is plotted in Fig. 3. As discussed in detail in [13], defect density is high in the specimens directing to the lower side and the plasma center, while almost no detectable defects for those directing to the upper side. These results imply that charge-exchange neutrals with energy enough to cause radiation damage in metals were mainly formed in the lower half of the plasma. According to the stereo-observation, the defects distribute up to 40–50 nm in depth, which corresponds to depth of the damage production by hydrogen particles ranging from 3 to 6 keV. This fact means that charge-exchange particles contributing to the defect formation have energy of several keV and correspond well with the result of the energy spectrum

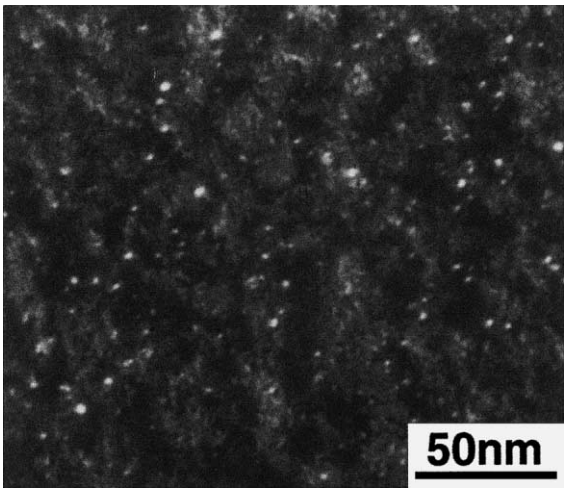

Fig. 2. TEM image (dark field) of radiation damage formed in Mo directing to the −45°.

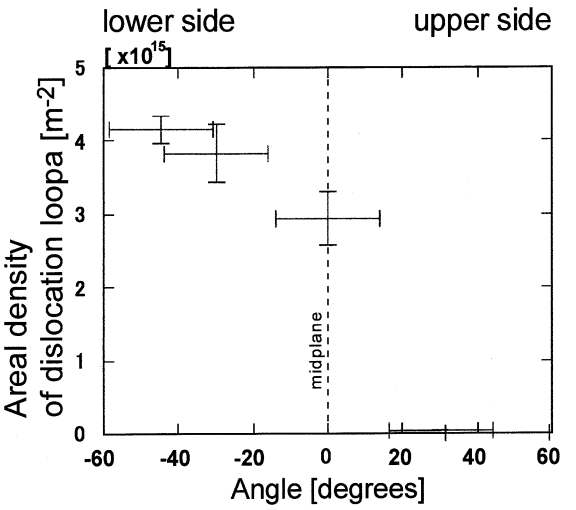

Fig. 3. Angular dependence of the area density of the dislocation loops formed in Mo by exposureto high ion temperature plasma.

measurement of neutral particles with a neutral particle energy analyzer [5]. From the detailed comparison of the dislocation loop density with that of the hydrogen ion irradiation, the flux of the charge exchange neutrals coming from the lower side and contributing to the damage was estimated to be 4.2×10^{17} particles m^{-2} s^{-1} sr^{-1} [13]. It is expected, therefore, that the total flux of the energetic hydrogen neutrals responsible for the damage at the bottom of the vacuum vessel is far above 10^{18} particles m^{-2} s^{-1}. One should note that this flux is not low for material damage. The amount of the dislocation loop density reaches the very high saturation level by the irradiation of only about 10^{22} ions/m^2 for several keV hydrogen ions [14] and results in strong surface hardening [15]. Due to strong interaction between hydrogen and the radiation-induced vacancies, formation of the dislocation loops is enhanced very much under keV-order hydrogen particle irradiation [16,17].

Fig. 4 shows an example of macroscopic damage of a pre-thinned W foil, which was exposed to the plasma for 72.3 min [5]. The specimen was cracked along the grain boundaries. It is likely that the energetic hydrogen atoms penetrated into the material through the surface diffuse deeply into the bulk and result in the hydrogen embrittlement. This fact indicates that the influences of the energetic hydrogen influx are not restricted in the narrow projected range of the incident neutrals but may change even the properties of thick bulk materials. This effect will be enhanced by the synergistic effects of neutron irradiation which uniformly form lattice defects acting as good trapping site for hydrogen.

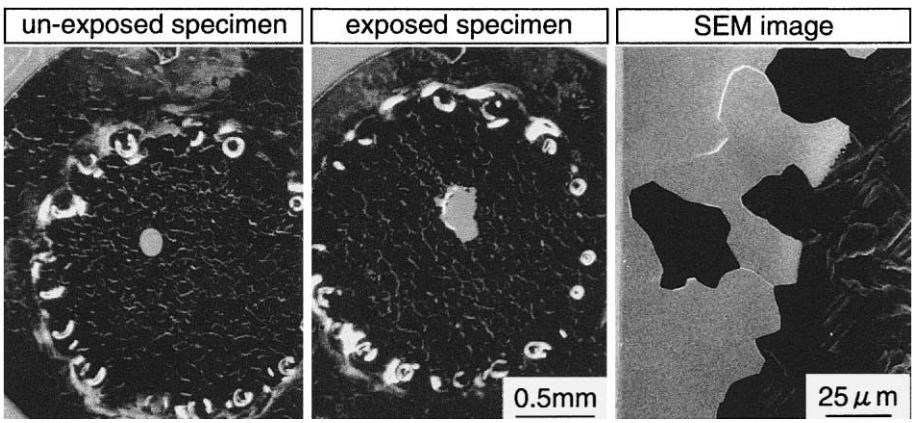

Fig. 4. Macroscopic damage of pre-thinned W specimens placed on the inner wall position of the vacuum vessel for 72.3 min.

Localization of the high ion temperature plasma at the lower part can be explained by the effect of gradient B drift of high-energy ions trapped in a toroidal ripple. The direction in the present experiments is toward the lower side. The localized formation of energetic charge-exchange neutrals at the lower half of the plasma indicates stronger sputtering and radiation damage at the bottom of the torus. In actual after installation of the Mo divertor plate at the bottom of the tours, the amount of Fe, Cr and Ni impurities, which are major elements of vacuum vessel material (304SS) decreased drastically and Mo becomes the major metallic impurity after that [11,12].

Accumulation of radiation-induced defects such as dislocation loops and bubbles is very temperature dependent. In the case of Mo, for example, the defects are formed up to 873 K [14], while up to 573 K for W [18]. These results indicate that the damage by hydrogen bombardment is important at relatively low temperatures.

3.2. Impurity deposition

3.2.1. Microstructure of the impurity deposits

Fig. 5 shows a dark field image and corresponding electron diffraction pattern of the impurity deposit on a thin 304SS probe specimen placed at the E-side. The image was obtained from a part of the first broad diffraction ring. Only the crystalline grains satisfying the Bragg diffraction condition have white image at this imaging condition. Numerous small dot images indicate that the deposit consisted of very fine crystalline grains around 1 nm in diameter. The diffraction rings indicate that the crystal structure is fcc, though its major metallic component is Mo, which normally forms the bcc structure. One should note that the crystal structure must be defective in such fine grains, because more than half of the constituting atoms are expected to locate on or near the grain boundaries where large disordering of atoms occurs.

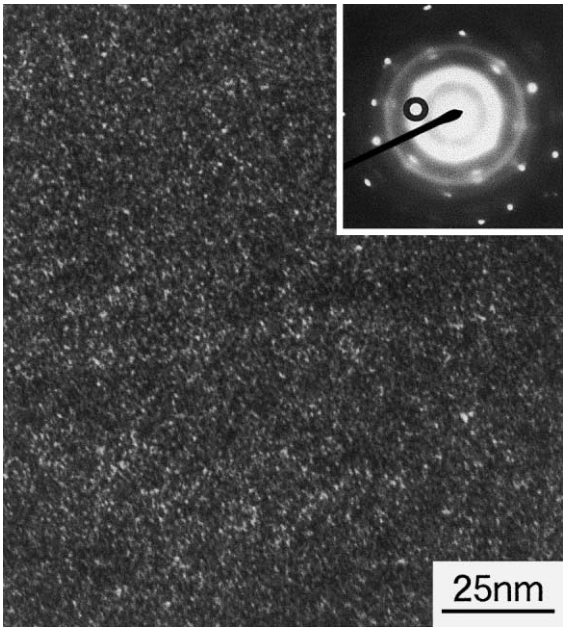

Fig. 5. Electron diffraction patterns and microstructures of the deposition on the stainless steel specimen exposed to tokamak. White image in these micrographs shows individual crystal grains.

As reported in [13] residual oxygen strongly affects the structure of vacuum-deposited Mo. If the ratio of the arrival rate of oxygen atoms and Mo atoms, A_O/A_{Mo}, is less than about 2 (partial pressure of oxygen $P_{O_2} = 5\times 10^{-4}$ Pa), its structure is normal large bcc grains while if the ratio is about 20 ($P_{O_2} = 5 \times 10^{-3}$ Pa), fine fcc grain structure similar to the deposit of TRIAM-1M appear. It is likely that oxygen atoms adsorbed on the surface suppress free migration and crystallization of the deposited Mo atoms and result in refinement of the crystal grains. Moreover, oxygen atoms involved in the Mo lattice may prevent to form the normal bcc structure and cause distortion of lattice due to its large atom size. Such peculiar structure is not special for Mo-deposits. Similar fine grain deposit with fcc structure was observed in W. In the case of Fe-deposits the structure changes successively from large bcc grains, fine fcc grains, fine FeO grains to fine Fe_2O_3–Fe_3O_4 grains, with increasing partial pressure of oxygen during the vacuum-deposition [19]. Electron diffraction pattern, however, indicates that no oxides are formed in the deposit in TRIAM-1M and the vacuum-deposited Mo formed in the oxygen atmosphere ($P_{O_2} = 5 \times 10^{-3}$ Pa).

Hydrogen atoms, in contrast, do not show strong effects on the structure of the vacuum-deposited Mo; the structure of the deposit is large bcc grains even if the ratio A_H/A_{Mo} is about 200 [12]. Weaker chemical interaction with Mo and its smaller atomic size must be the reason why the effects of hydrogen are not so strong as oxygen.

Though TRIAM-1M is an ultra-high vacuum machine, some oxygen exist as residual impurity. It is likely that co-deposition of the residual oxygen with Mo brought the peculiar defective structure.

3.2.2. Retention of hydrogen isotope

Due to their defective structure, it is anticipated that the deposit in TRIAM-1M will strongly affect the recycling process of hydrogen. To know the trapping of hydrogen in the deposits, thermal desorption of the implanted deuterium was measured for the vacuum-deposited Mo with similar defective fcc fine grain structure. Retention of deuterium in deuterium ion implanted Mo-deposit, bulk Mo and 304SS is plotted in Fig. 6

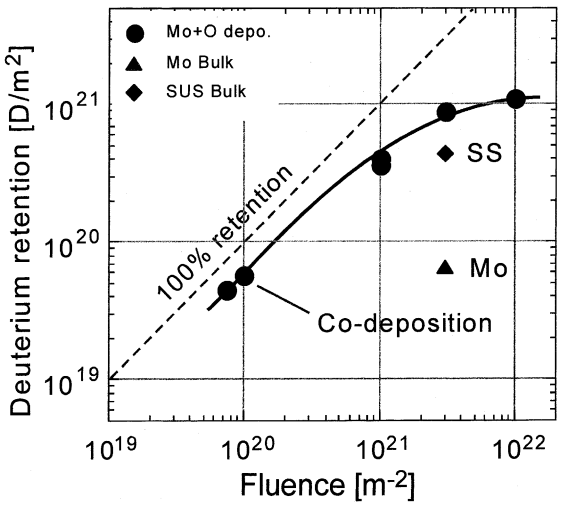

Fig. 6. Fluence dependence of the deuterium retention for Mo-deposit. Deuterium retention in bulk Mo and 304SS is also plotted. Specimens were implanted with 6 keV–D_3^+ at room temperature before the TDS experiments.

against deuterium ion fluence. Each data is the sum of the desorbed deuterium as DH and D_2, DHO and D_2O molecules. In contrast to the weak retention in bulk Mo, the defective Mo-deposit shows strong retention. It is worth noting that trapping of deuterium in the Mo-deposit is larger than stainless steel, which is the vacuum vessel material of TRIAM-1M. The ratios of D/Mo and O/Mo at the fluence of 1×10^{22} D m^{-2} were estimated to be 0.35 and 0.04, respectively. As shown in the micrograph of TEM in Fig. 7 dense micro-cavities, probably hydrogen bubbles, are formed in the Mo-deposit by the implantation of 4 keV–H_2^+ hydrogen ion at room temperature to a fluence of 7.2×10^{21} H m^{-2}. It is likely that the defective structure and the dense bubbles are responsible for large and strong retention of hydrogen in the Mo-deposits. Present work indicates that re-deposition of sputtered impurities on plasma facing surface increases retention of hydrogen isotopes even in metallic tokamak similar to carbon tokamak [7–9].

4. Conclusion

Material exposure experiments using a surface probe system have been carried out in TRIAM-1M to investigate the damage and surface modification of plasma facing materials. Long pulse of high ion temperature mode discharge of TRIAM-1M was very useful for such experiments, because the high accumulation of the plasma bombardment is essential to detect the effects.

Dense dislocation loops formed by the bombardment of charge-exchange hydrogen neutrals were observed only in the specimens directing to the lower half of the plasma. This result indicates that charge-exchange neutrals with energy enough to cause radiation damage are mainly created in the lower half of the plasma. This anisotropy of the neutral particle distribution can be explained by the effect of gradient B drift of high-energy ions trapped in a toroidal ripple. The mean flux of these energetic neutrals from lower side was estimated to be 4.2×10^{17} particles m^{-2} s^{-1} sr^{-1}. This value is never low from the standpoint of material damage. If the temperature of the plasma facing material is relatively low, the bombardment of the particles may result in the material hardening and increase of the hydrogen retention very quickly. One should pay more attention to the effects of diffused hydrogen in the matrix. In the hydrogen-sensitive materials such as W diffused hydrogen will bring a variety of effects such as embrittlement.

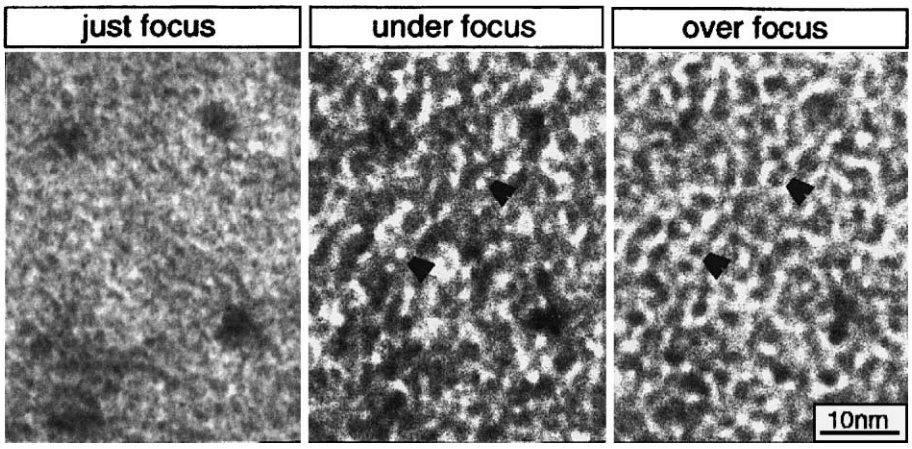

Fig. 7. Microstructure of Mo-deposit implanted with 4 keV–H_2^+ at room temperature to a fluence of 7.2×10^{21} H m^{-2}. Each micrograph was taken at the conditions of just-, under- and over-focus.

A major element of the impurity deposits in TRIAM-1M is Mo but its structure is peculiar; crystalline grain size is about 1nm and their crystal structure is not normal bcc but fcc due to the co-deposition with oxygen, which exists in plasma as a residual impurity. The Mo-deposits formed in the low-pressure oxygen atmosphere, which have the same structure of the deposit in TRIAM-1M, show very large and strong hydrogen isotope retention. Implanted deuterium is desorbed not only as D_2 and DH but also as D_2O and DHO. The defective structure and the micro-cavities contribute to the large hydrogen retention. The ratio of D/Mo reached 0.35 at high fluence. This result indicates that re-deposition of sputtered elements on plasma facing surface must be a serious problem even in a metallic tokamak. One should pay more attention to the modification of plasma facing surface by the impurity deposition and its effects on the hydrogen behavior. This is particularly important for long pulse and steady-state plasma.

References

[1] R.J. Goldston, P.H. Rutherford, Introduction to Plasma Physics, Institute of Physics Publishing, Bristol, Philadelphia, 1996, p. 156.
[2] N. Yoshida, A. Nagao, K. Tokunaga, K. Tawara, T. Muroga, T. Fujiwara, S. Itoh, The TRIAM group, Rad. Eff. Def. Solids 124 (1992) 99.
[3] K. Tokunaga, T. Muroga, T. Fujiwara, K. Tawara, N. Yoshida, S. Itoh, The TRIAM group, J. Nucl. Mater. 191–194 (1992) 449.
[4] N. Yoshida, Y. Hirooka, J. Nucl. Mater. 258–263 (1998) 173.
[5] S. Itoh, K.N. Sato, K. Nakamura, H. Zushi, M. Sakamoto, K. Hanada, E. Jotaki, K. Makino, S. Kawasaki, H. Nakashima, N. Yoshida, Nucl. Fus. 39 (1999) 1257.
[6] R. Behrisch, M. Mayer, C. Garcia-Rosales, J. Nucl. Mater. 233–237 (1996) 673.
[7] R. Behrisch, A.P. Martinelli, S. Grigull, R. Groetzschel, U. Kreissig, D. Hildebrandt, W. Sshneider, J. Nucl. Mater. 220–222 (1995) 590.
[8] K. Krieger, J. Roth, A. Annen, W. Jacob, C.S. Picher, J. Nucl. Mater. 241–243 (1997) 684.
[9] E. Gauthier, A. Grosman, J. Valter, J. Nucl. Mater. 220–222 (1995) 506.
[10] N. Yoshida, K. Tokunaga, T. Fujiwara, K. Tawara, T. Muroga, S. Itoh, The TRIAM group, J. Nucl. Mater. 196–198 (1998) 415.
[11] T. Hirai, K. Tokunaga, T. Fujiwara, N. Yoshida, S. Itoh, The TRIAM group, J. Nucl. Mater. 258–263 (1998) 1060.
[12] T. Hirai, T. Fujiwara, K. Tokunaga, N. Yoshida, A. Komori, O. Motojima, S. Itoh, The TRIAM group, J. Nucl. Mater. (ICFRM-9, 1999), to be published.
[13] T. Hirai, T. Fujiwara, T. Tokunaga, N. Yoshida, S. Itoh, The TRIAM group, these Proceedings.
[14] R. Sakamoto, T. Muroga, N. Yoshida, J. Nucl. Mater. 212–215 (1994) 1426.
[15] H. Iwakiri, H. Wakimoto, H. Watanabe, N. Yoshida, J. Nucl. Mater. 258–263 (1998) 873.
[16] N. Yoshida, R. Sakamoto, J. Nucl. Mater. 251 (1997) 284.
[17] N. Yoshida, Rad. Eff. Def. Solids 148 (1999) 535.
[18] R. Sakamoto, T. Muroga, N. Yoshida, J. Nucl. Mater. 220–222 (1995) 819.
[19] N. Yoshida, F.E. Fujita, J. Phys. F: Metal Phys. 2 (1972) 1009.