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Evaluation of residual tritium in stainless steel irradiated at SINQ target 3

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Abstract

A MW spallation target will need an accurate amount of tritium generation in materials for post irradiation experiments. A high-energy proton of 580 MeV generates a variety of constituents in the SINQ target mainly through spallation reaction, and these constituents included helium and hydrogen isotopes. For the SINQ target hydrogen and helium are generated to hundreds appm H and tens appm He, respectively. However, tritium gas production has not yet been reported. Evaluation of tritium gas generation in the specimens of target 3 was done by calculation. Results were compared with gamma spectrum measurement of austenitic and martensitic–ferritic steels three and a half years later after irradiation. Good agreement was obtained for activity of the main isotope. Residual tritium measurement was carried for the austenitic stainless steel specimens irradiated below 250 °C by the thermal desorption method four and a half years later after irradiation. An ion chamber monitored the tritium concentration. Water bubblers collected all tritium after oxidation of tritium by the oxidation catalysts. It is found that a release of tritium began over 250 °C and the ratio of residual tritium to generated one is estimated to be less than 20%.

1. Introduction

In order to establish spallation materials database for accelerator-driven nuclear transmutation systems and spallation neutron sources, post irradiation experiments (PIE) are needed for irradiated materials. High-energy protons of 580 MeV at SINQ target and 800 MeV at LANSCE target generate a variety of almost all constituents in the target mainly through spallation reaction process and these constituents included helium and hydrogen isotopes [1–4]. PIE requires accurate information on the residual tritium gas for samples. For the SINQ target hydrogen and helium are generated to hundreds appm H and tens appm He, respectively. However, tritium gas production has not been reported yet. Evaluation of tritium gas was done for STIP-I specimens irradiated at SINQ target 3. Material used in the experiment were three TEM disc samples of austenitic stainless steel 316F, which were irradiated below 250 °C. Before evaluating a total amount of tritium production by calculation, validation for reliability was done through analyses of decay gamma-ray measurement on JPCA and F82H. Those samples were cut from the tensile specimens. The amount of residual

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tritium was measured by a thermal desorption method [9]. And produced nuclides in the specimens during and after irradiation at SINQ target 3 were calculated.

2. Beam condition and material samples

Irradiation time for STIP-1 samples was two years in 1998 and 1999 and the total proton charge was 6.8 Ah. Incident proton beam current was 0.85 mA for the first 14 months and 1.04 mA for the last 2 months during 1998–1999. Beam profile was a truncated two-dimensional Gaussian distribution [1]. As the beam profile was not flat, radiation damage would be affected by the location of the specimens in the target. The specimens used for tritium measurement were three TEM discs of austenitic stainless steel 316F, 3 mm in diameter and 0.2 mm in thickness. The ID number of the austenitic stainless steel 316F is G in STIP-I [1]. The weight of the specimens is 13 mg. G^6 was irradiated to 5.9 dpa at 160–190 °C. G^2 and G^3 were irradiated to 5 dpa at 90–110 °C, respectively [1,2]. Before heating the samples, G^2 and G^6 were not polished but the surface of G^3 disc was polished by an emery paper in order to evaluate the effect of surface oxide.

The samples used for gamma-ray measurement were cut from two tensile specimens with 16 mm length, 2 mm width and 0.4 mm thickness. Materials used are JPCA and F82H, and their ID numbers are F08 and Q07. The weight of the samples for gamma-ray measurement was 2.9 and 3.3 mg, respectively. Sample Q07 was irradiated to 11.8 dpa at 320–380 °C. F08 was also irradiated under the similar condition with Q07. Chemical compositions of all materials are listed in Table 1.

3. Calculation model of SINQ target 3

SINQ target 3 consisted of Zircoloy-2 rods cooled by D_2O. The target was covered by a safety container made of $AlMg_3$. The safety container was also surrounded by D_2O moderator. Specimens, Q07, F08 and G^6, were put near the centerline but G^2 and G^3 were put at the edge of the second rod. As a result, Q07 and F08 received the highest proton flux, and G^2 and G^3 received less than a half of those flux.

Fig. 1 illustrates the target and moderator model used in the calculation. The target is assumed to be a homogeneous medium of Zircoloy-2 and D_2O. Ten detectors were put in the target, which occupies the same position with ten rods.

Above 20 MeV, the high-energy particle transport code NMTC/JAM [5] calculated nuclide yields produced by high-energy particles. MCNP-4C code [6] was used for neutron transport below 20 MeV. DCHAIN-SP code [7,8] was used for the calculation of produced nuclides in the specimens during and after irradiation with the considerations of both contribution due to high-energy particle and neutrons below 20 MeV.

4. Measurement of tritium

Fig. 2 shows the schematic drawing of residual tritium measurement apparatus. This apparatus mainly consists of a tritium extraction unit to extract tritium from the specimen, and a tritium measurement unit. Tritium extraction from the specimens was carried out with the thermal desorption method (TDS) under the constant ramp rate of 10 °C/min in flowing carrier gas with a flow rate of 250 cm^3/min and a mixed composition of nitrogen and hydrogen (<2% volume). The specimen was heated up to 800 °C and kept for 30 min. Then, the specimen was heated up again to 1000 °C and kept for 1 h. After that the specimen was cooled down. The released tritium from the specimen was measured continuously by an ion chamber (1.5 L volume) during the TDS experiment for the real time measurement. Water bubblers collected all tritium after the oxidation of tritium with oxidation

Table 1
Chemical compositions of specimens for gas production and gamma spectrum measurements

	Fe	Cr	Ni	Mo	Mn	Ti	Co	Cu	B	C	Si	P	S	N	Ta
316F G	Bal.	16.79	13.95	2.34	0.23	–	<0.001	<0.01	–	0.04	0.04	<0.003	0.002	0.011	–
JPCA F08	Bal.	14.14	15.87	2.29	1.54	0.22	0.028	–	0.004	0.058	0.50	0.026	0.004	0.003	–

	Fe	Cr	Ni	Mo	Mn	Ti	V	Nb	W	Ta	Cu	C	Si	P	S	N
F82H Q07	Bal.	7.87	0.02	0.003	0.1	0.004	0.19	0.0002	1.98	0.03	0.01	0.09	0.07	0.003	0.001	0.007

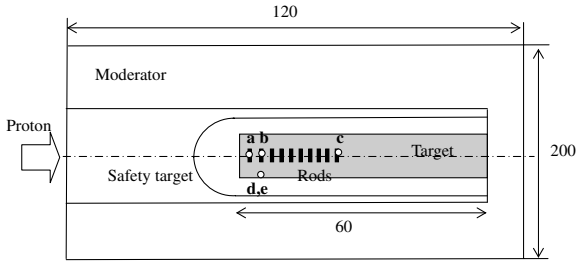

Fig. 1. Calculation model for SINQ target 3. Open circles suggested by letters a, b, c, d and e are locations of irradiated specimens Q07, F08, G^6, G^2 and G^3, respectively. G^2 and G^3 are located at almost same position. Unit:cm.

catalysts for quantitative measurements in the specimen.

5. Results

5.1. Measurement of activity

Fig. 3 shows activities of JPCA (F08) and F82H-TIG welded (Q07) samples. For F08 samples, main radioisotopes are Sc44, Sc46, Mn54, Co57 and Co60. For Q07 samples, main radioisotopes are Sc44, Sc46, Mn54, Lu172 and Lu173. The activity from Mn54 was over 10^9 Bq/g, highest among solid isotopes. The other isotopes have the order of 10^8 Bq/g or under 10^7 Bq/g.

5.2. Calculation of activity and gas production

The results were compared with the measurement, as shown in Fig. 3. Activity of Mn54 was realized within percents, but for other isotopes a good agreement could not be achieved. For F82H, the calculation underestimated an activity of Sc46 and overestimated an activity of Co57. For JPCA, the calculation showed realistic values comparative with F82H.

Table 2 shows the results of gas productions after irradiation with no cooling time. The amount of hydrogen was estimated considering incident protons. The amount of gas production was calculated for Q07, which received 11 dpa. At the center of tensile specimen Q07, the damage rate was reported to be 11.8 dpa [2]. Details will be mentioned in the discussion section. There were 72 appm tritium, 4720 appm hydrogen, 280 appm deuterium, 476 appm helium 4 and 16 appm helium 3. Gas productions in G samples were obtained from the result of Q07, assuming that those are proportional to dpa.

5.3. Measurement of tritium

Fig. 4 shows a typical tritium release behavior from G^6 specimen. The tritium release starts above 250 °C, and achieves a peak at about 400 °C. The second peak indicating additional tritium release is not observed above 400 °C. On the other hand, results of tritium measurement by the water bubbler for all specimens are summarized in Table 3. As to the effect of surface oxide on SS316, the tritium release behavior and the residual tritium amount from/in the G^3 specimen were almost similar to that from/in the G^2 specimen. Therefore, it is found that

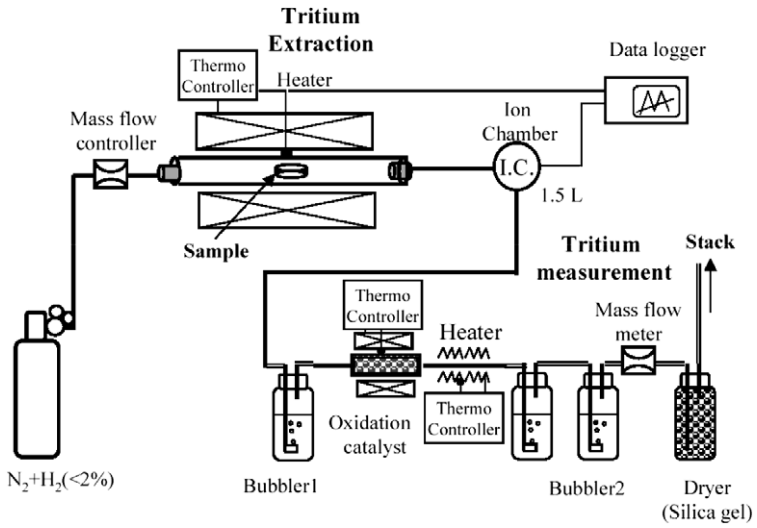

Fig. 2. Schematic drawing of tritium measurement apparatus.

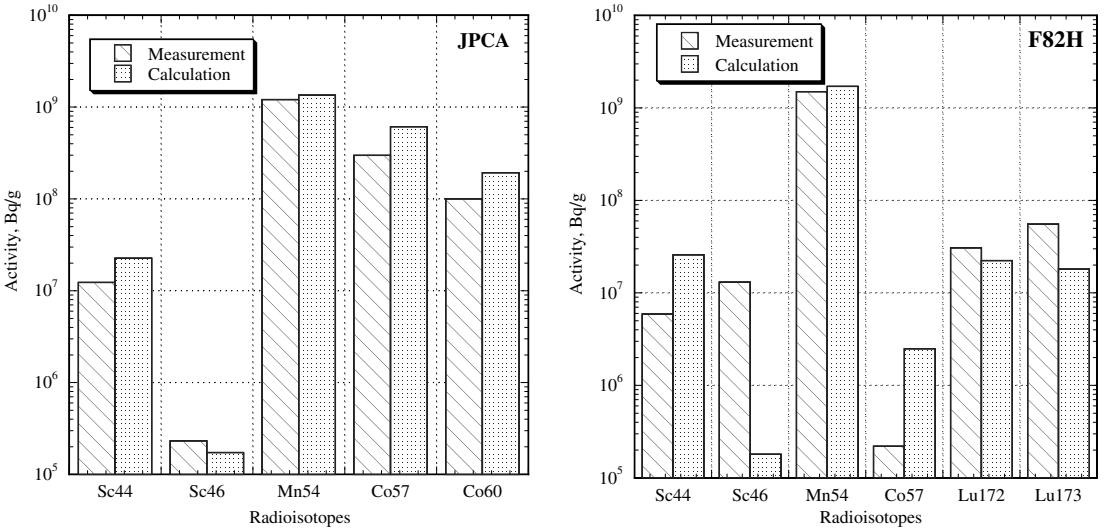

Fig. 3. Gamma spectrum measurement of JPCA and F82H-TIG samples.

Table 2
Analyses of radiation damage and gas productions of specimens

ID	G^6	G^2	G^3 (Polished)	Q07
DPA[(a)]	5.9	5.0	5.0	11(11.8)[(c)]
Irradiation temperature, °C[(b)]	160/190	90/110	90/110	320/380
Tritium, appm	39 (~8.6 MBq)	33 (~7.3 MBq)	33 (~7.3 MBq)	72 (~16 MBq)
Hydrogen, appm	2532	2145	2145	4720
Deuterium, appm	150	127	127	280
Helium 4, appm	255	216	216	476
Helium 3, appm	8.6	7.3	7.3	16

(a) and (b) were evaluated at PSI. (c) dpa was reduced to 11 after considerations of sampled position of tensile specimens.

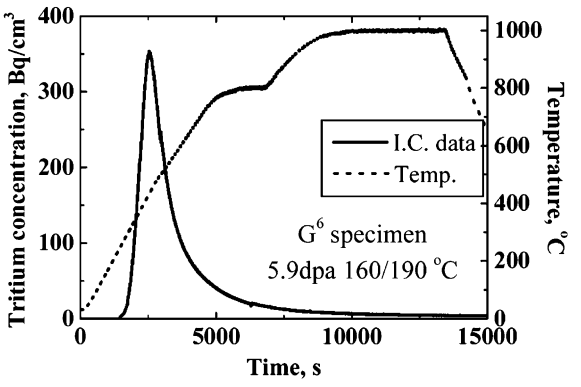

Fig. 4. TDS spectra of tritium from proton irradiated SS316 (G^6).

the surface oxide does not affect the tritium extraction and tritium does not maldistribute in the oxide layer in the austenitic stainless steel specimen.

Table 3
Results of tritium measurement of SS316

Specimen	Residual T[a] (MBq)	Residual T (R)[b] (MBq)	Generated T (G) (MBq)	R/G (%)
G^6	1.37	1.75	8.6	20
G^2	0.91	1.16	7.3	16
G^3	0.87	1.11	7.3	15

[a] Measured values at a time of 1280th cooling day.
[b] Estimated values at a time after irradiation.

6. Discussions

Detailed report on neutronics calculation, dosimetry analysis and gas measurements of the SINQ target 3 was reported by PSI [2]. Gas measurement was done for F82H TEM disc named P^{13} in rod 1 located at a position 7.5 mm from the centerline. Calculation showed that the sum of helium 4 and helium 3 was

920 appm, and hydrogen was 4960 appm. Rod 1 means the top position in the array of detectors, as shown in Fig. 1. The corresponding specimen with this study is Q07, located at a position 12.5 mm away from the centerline. The position of the sample from the tensile specimen with a total length of 12 mm must be taken into account. The position must be modified to 18 mm. Then it could be estimated that dpa and gas productions will be 11, 700 He appm and 5200 H appm, respectively.

Calculated values were 492 He appm and 4720 H appm, respectively. As mentioned in the results, the gas productions in G samples were calculated by multiplying the ratio of dpa. Dpa is estimated to be 11. Then the results indicate that the average amount of residual tritium in the austenitic stainless steel specimens is less than 17% compared to the generated tritium amount during the proton irradiation for each specimen.

In the measurement of tritium, it was found that tritium started to diffuse out in the sample over 250 °C. According to the results of hydrogen measurements for SINQ target 3 specimens [2], the measured amount of hydrogen is roughly less than a tenth of the calculated value for the specimens irradiated over 250 °C and less than a third for the specimens irradiated under 250 °C. The ratio of 16% is a half of that for hydrogen results. Decay of tritium produces He^3 with a half-life of 12.3 years. Tritium measurement was done at a time of 1600th cooling day. Estimated measured value after proton irradiation will be 1.28 times more. Then the residual tritium in G^6, G^2 and G^3 samples are 1.75, 1.16 and 1.11 Bq/g. And so the ratio of residual tritium to generated one is less than 20%. This value is not so far from that obtained in the previous report [2].

7. Conclusions

Residual tritium in the specimens irradiated at SINQ target 3 was measured and compared with the calculated results. Measurement was done by the thermal desorption method (TDS) under the constant ramp rate of 10 °C /min in flowing carrier gas with a flow rate of 250 cm^3/min and a mixed composition of nitrogen and hydrogen (<2% volume). Gamma measurement was done to verify the certainty of the target simulation model. The following conclusions were obtained. The residual tritium gas was 1.05–1.65 MBq/g after two years irradiation. Those are less than 20% of the calculated values, which is in reasonable agreement with the measurements and calculations reported in Ref. [2]. The release of residual tritium began at 250 °C and achieved a peak at 400 °C. It is also found that the surface oxide does not affect the tritium extraction and tritium does not maldistribute in the oxide layer in the austenitic stainless steel specimen.

Acknowledgement

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References

[1] Y. Dai, G.S. Bauer, J. Nucl. Mater. 296 (2001) 43.
[2] Y. Dai, Y. Foucher, M.R. James, B.M. Oliver, J. Nucl. Mater. 318 (2003) 167.
[3] F.A. Garner, B.M. Oliver, L.R. Greenwood, M.R. James, P.D. Ferguson, S.A. Maloy, W.F. Sommer, J. Nucl. Mater. 296 (2001) 66.
[4] B.M. Oliver, M.R. James, F.A. Garner, S.A. Maloy, J. Nucl. Mater. 307–311 (2002) 1471.
[5] K. Niita, S. Meigo, H. Takada, Y. Ikeda, High Energy Particle Transport Code NMTC/JAM: JAERI-Data/Code 2001-007 (2001).
[6] J.F. Briesmeister (Ed.), MCNP™– A general Monte Calro *N*-Particle Transport Code, Version 4C, LA-13709-M, LANL (2000).
[7] H. Takada, K. Kosako, Development of the DCHAIN-SP Code for Analyzing Decay and Build up Characteristics of Spallation products, JAERI-Data/Code 99-008, JAERI (2001).
[8] T. Kai, F. Maekawa, Y. Kasugai, K. Niita, H. Takada, S. Meigo, Y. Ikeda, Validation of Radioactivity Calculation Code System DCHAIN-SP, in: Proc. ICANS-XVI, 2003, p. 1041.
[9] P.A. Redhead, Vacuum 12 (1962) 203.