21pE-4

Tritium release from Inconel 625
インコネル625からのトリチウムの放出

Y. Torikai[1)], K. Kobayashi[2)], M. Saito[2)], N. Miya[2)], and Y. Ikeda[2)]
鳥養祐二[1)], 小林和容[2)], 齋藤真貴子[2)], 宮直之[2)], 池田佳隆[2)]

1) Hydrogen Isotope Research Center, University of Toyama, Gofuku 3190, Toyama 960-8555
1) 富山大学 水素同位体科学研究センター, 〒930-8555 富山市五福3190
2) Japan Atomic Energy Agency, Mukoyama, Naka-shi, Ibaraki-ken, 311-0193
2) 日本原子力研究開発機構 那珂研究所, 〒311-0193 那珂市向山

Disassembly of the JT-60U torus was started in 2010 after 18 years D_2 operations. In future the vacuum vessel that was made by Inconel625 will be treated as non-radioactive ones after the clearance procedure under the Japanese regulation depending on the tritium (T) contamination level. Therefore, it was very important to study the hydrogen isotope behavior in Inconel 625 from viewpoint of the clearance procedure. Inconel 625 specimen was exposed to the D_2 (92.8 %) – T_2 (7.2 %) gas mixture at 573 K for 5 hours. The tritium release from the specimen at 298 K was controlled for about 1 year. The contaminated specimen by tritium was released continuously the diffusible tritium under the ambient condition. From these results, the behavior of tritium in the vacuum vessel of the JT-60U torus will be evaluated and discussed.

1. Introduction

Disassembly of the JT-60U torus was started in 2010 after operations with deuterium (D) plasma for 18 years. The tritium was mainly accumulated in the graphite tiles. However, some amount of tritium was also accumulated in the JT-60U vacuum vessel manufactured from Inconel 625 steel. In future the vacuum vessel will be treated as non-radioactive ones after the clearance procedure under the Japanese regulation [1].

In this paper, in order to obtain data for clarifying tritium behavior in Inconel 625, especially release rates of tritium, a series of the loading experiment and the chronic tritium release experiment using D_2–T_2 mixture gas have been carried out.

2. Experimental

The experimental equipment and procedures used for loading samples with tritium have been described in detail in a previous paper [2].

Specimens, 15×15×0.5 mm and 15×15×0.1 mm in size, prepared from Inconel 625 derived from Nilaco Co. Ltd. Specimen was placed into a quartz tube vessel connected with an ultra-high vacuum system. Initially the specimen was annealed at 673 K under vacuum of 10^{-6} Pa for 3 hours. Then the specimen was loaded with the hydrogen isotope mixture consisting of D_2 (92.8 %) – T_2 (7.2 %). The loading conditions were 573 K at total gas pressure of 1.2 kPa for 5 hours.

After the D_2+T_2 loading, the tritium release at 298 K from the specimen was determined for long time under Ar gas flow using tritium release apparatus [2]. The apparatus consists of a gas supply system, a sample glass cell, and two water bubblers separated by a copper oxide bed held at 800 K. All parts are compactly connected to minimize distances. Commercial argon containing a small partial pressure of water vapor (0.073 Pa) was used as carrier gas. The flow rate was maintained at 50 ml/min. Oxidized tritium species released from the specimen were retained in the first bubbler; elemental species and hydrocarbons were oxidized over a copper oxide bed and retained in the second one. The concentration of tritium in the water bubblers was measured periodically using a liquid scintillation counter (LSC).

3. Results and discussion

Figure 1 shows the time dependence of tritium release rate from the 0.5 mm thick Inconel 625 specimen. The tritium release rate was 14.5 kBq/h for 10 minutes just after the tritium release experiment started. Thereafter, the release rate was lowered sharply by factors of about 200 after 1 hour and about 3000 after 1 day, and then finally the release rate decreased to less than 1 Bq/h after 600 hours passed.

It has been observed that tritium was released continuously throughout the year. More than 95 % of the tritium was released in the form of HTO molecules. Therefore, as shown in figure 5, it is believed that atomic tritium diffuses from the Inconel 625 matrix to the surface and desorbs from the surface into the gas phase via isotopic exchange with residual moisture present in the carrier gas [3].

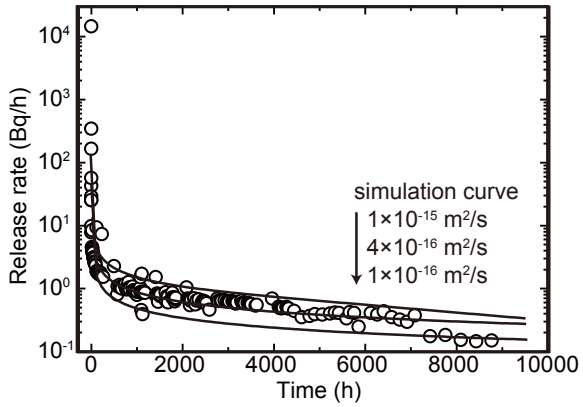

Fig. 1. The time dependence of the tritium release rate at 298 K from the 0.5 mm thick Inconel 625 specimen.

From the result, the one dimensional diffusion model, as reported by Naoe et al. [3], can be applied to describe the time dependence of the tritium release rate. The tritium release rate as a function of time (t) is calculated from equation (1), where Γ is the instantaneous flux, a is the half thickness of the specimen, t is the time, and D is the tritium diffusion coefficient.

$$\Gamma(t) = \frac{2c_0 D}{a} \sum_{n=0}^{\infty} \exp(-\frac{\pi^2}{a^2}(n+\frac{1}{2})^2 Dt) \qquad (1)$$

The results are presented as solid lines in figure 1. The simulation curves were given for the diffusion coefficients, D, in the range from 10^{-16} to 10^{-15} m^2/s. For D = 4×10^{-16} cm^2/s, the calculated time dependence of the tritium release rate fits well with the experimental data. This diffusion coefficient of tritium in the Inconel 625 specimen is agreed with a value of 1.4×10^{-16} m^2/s reported by Reiter et al. [4]. It is considered that the tritium release from Inconel 625 is limited rather by the diffusion in the matrix than the surface reaction.

Figure 2 shows the time dependence of tritium release rate at 298 and 313 K from 0.1 mm thick Inconel 625 specimen. The solid lines show the simulation results with the use of D = 4×10^{-16} and 8×10^{-16} m^2/s. It is found that a release of tritium from the specimen is governed by diffusion process.

Figure 3 shows experimental data on the fraction of tritium remained in the 0.1 mm thick Inconel 625 specimen during the tritium release experiment. More than 99 % of initially accumulated tritium is released at 313 K for 1500 hours. On the other hand, at 298 K about 14 % of tritium in the specimen is remained in the specimen after 2500 hours passed. .

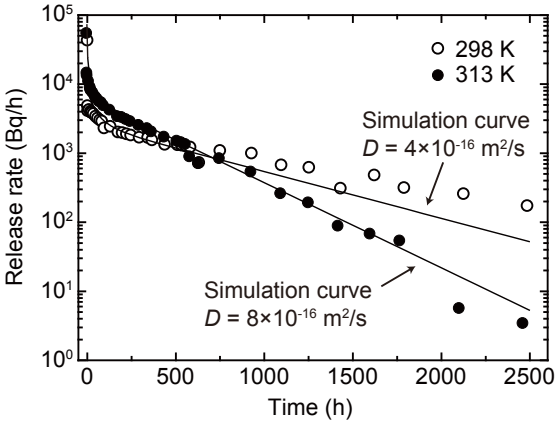

Fig. 2. The time dependence of tritium release rate at 298 and 313 K from the Inconel 625 of 0.1 mm thickness.

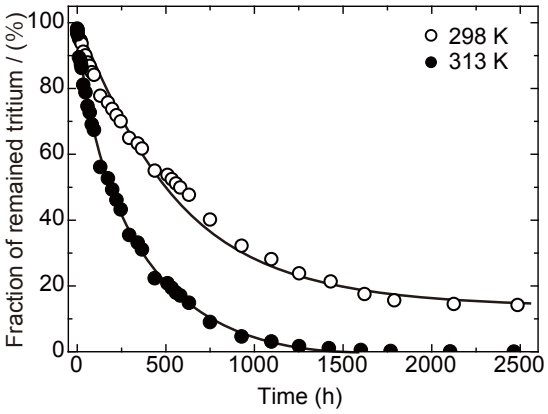

Fig. 3. The residual tritium in Inconel 625 of 0.1 mm thickness after the tritium release experiment

4. Conclusions

It has been found that:

(1) After exposure of Inconel 625 specimens to D_2-T_2 gas mixture at 572 K, tritium release from the specimens at 298 and 313 K was observed.

(2) The rate of the tritium release from the Inconel 625 is simulated with one-dimensional diffusion model.

(3) Tritium desorbed from the specimen surface into the gas phase in the form of HTO molecules via isotopic exchange with residual moisture present in the carrier gas (Ar).

References

[1] Y. Kamada, P. Barabaschi, S Ishida, Nucl. Fusion, **53**, 104010, 1-17 (2013).

[2] Y. Torikai, A.N. Perevezentsev, M. Matsuyama and K. Watanabe, Fusion Sci. and Technol., **41**, 736 (2002).

[3] S. Naoe, Y. Torikai, R-D. Penzhorm, K. Akaishi, K. Watanabe, M. Matsuyama, Fusion Sci. Technol., **54**, 515 (2008).

[4] F. Reiter, K. S. Forcey, G. Gervasini, EUR 1512 EN (1993).