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Release of tritium, protium, and helium from neutron-irradiated Li–Al alloy. II

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Abstract

Tritium gases (HT, T_2) and helium are released abruptly from highly neutron-irradiated Al–12.7 at.% ^{6}Li alloy around 570 K on isochronal heating, and H_2 gradually above the same temperature. In the previous study of the neutron-irradiated Al–0.1 at.% ^{6}Li alloy (J. Nucl. Mater. 139 (1986) 248), negligible release of helium at temperatures below the melting point of the alloy is attributed to trapping of helium at vacancies in the alloy. In this study, however, helium is released from the Al–12.7 at.% ^{6}Li alloy at temperatures below the melting point of the alloy same as tritium. The origin of helium release is assumed to be the rupture of helium bubbles. Furthermore, radiometric method shows that most of tritium produced by ^{6}Li(n, α)T reaction exists in the T^- state in the alloy. According to the above fact, an appropriate temperature to extract tritium from the alloy is suggested. © 1998 Elsevier Science B.V.

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1. Introduction

In Japan Atomic Energy Research Institute (JAERI), technology for tritium production by use of Li–Al alloy has been developed with the aim of furnishing fuel tritium for a controlled thermonuclear reactor (CTR) [1–3] and specimens of basic research such as the muon catalyzed fusion experiments [4,5]. The alloy was selected because of its high heat conductivity and low induced radioactivity.

Several studies on release of hydrogen gases from the neutron-irradiated alloy had been carried out [6–12]. However, in most studies the alloy was irradiated for a relatively short time, and H_2, HT, T_2, and helium released have not been examined individually, except for our previous work on the Al–0.1 at.% ^{6}Li alloy [13]. In the previous work published with the same title as the present paper, which focused on ^{6}Li burn-up of the alloy, the release of each species has been studied by heating the alloy isochronally and isothermally. In the present study, we have investigated the release behavior of tritium, protium, and helium from the neutron-irradiated Al–12.7 at.% ^{6}Li alloy on heating in comparison with the previous work [13]. The amounts of tritium and helium in the present study are 10^2 times larger than those in the previous one as mentioned in Section 2.3, owing to the Li content of the alloy. In particular, we discussed the difference between the helium release behavior from the Al–12.7 at.% ^{6}Li alloy and that from the Al–0.1 at.% ^{6}Li alloy. Furthermore, we have examined the relation between the released species of tritium and the valence state of tritium in the Al–12.7 at.% ^{6}Li alloy.

2. Experiment

2.1. Materials

The Al–12.7 at.% ^{6}Li alloy was prepared by dissolving lithium metal (99.9% in chemical purity and containing ^{6}Li of 95.5% in isotopic purity) in molten aluminum at 973 K in collaboration with Mitubishi Atomic Power Ind.

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[2,13]. The alloy was rolled in the form of a plate (12.0 mm (w) × 40 mm (l) × 0.5 mm (d)) and weighs 564 mg.

A NaOD/D_2O solution with 9.8 N was purchased from CEA. The isotopic purity was determined to be 97.5 at.% by infrared spectroscopy.

2.2. Neutron irradiation

The target was enclosed in the capsule of stainless steel with helium gas and irradiated in G-11 hole of the Japan Material Testing Reactor (JMTR) for 934 h. The thermal neutron flux was 4.0×10^{14} cm^{-2} s^{-1}. The fast neutron flux was 0.5×10^{14} cm^{-2} s^{-1}. Tritium (T) and ^{4}He atoms were produced by a nuclear reaction between thermal neutrons and ^{6}Li (cross-section: 953 b [14]) and T (half life: 12.26 yr [14]) decays to ^{3}He. The other nuclear reactions were negligibly small [15]. The irradiation temperature, which was assumed by the tritium release behavior as mentioned in Section 3.6, was below 473 K. The irradiated alloy was cooled for 2 years and 8 months at room temperature. In this work, the ^{6}Li burn-up of the neutron-irradiated Al–12.7 at.% ^{6}Li alloy was 38% as mentioned in Section 2.3. In the previous work, that of the typical Al–0.1 at.% ^{6}Li alloy, which was irradiated by the thermal neutron flux of 2.9×10^{14} cm^{-2} s^{-1} for 512 h at temperatures below 473 K in JMTR, was 35% [13]. Therefore, the amounts of tritium produced in the Al–12.7 at.% ^{6}Li alloy was 10^2 times larger than that of the Al–0.1 at.% ^{6}Li alloy with the same weights of the alloys. Here, we considered the decay of tritium during the cooling period.

2.3. Apparatus and procedure

The optical-microphotographs on the surface of unirradiated Al–12.7 at.% ^{6}Li alloy and neutron-irradiated Al–12.7 at.% ^{6}Li alloy were taken in the magnitude of 200 after mechanical polishing and electrolytic etching.

The surface of unirradiated Al–12.7 at.% ^{6}Li alloy was observed by the Rutherford backscattering (RBS) using 1.5 MeV H^+-ion of Van de Graaff accelerator. The RBS spectrum shows the depth profile of each element from the specimen surface.

The amount of tritium released in the capsule was measured to be 1.80×10^{10} Bq (485 mCi) by an ionization chamber. The amount of tritium contained in the target was estimated to be $(10.2 \pm 0.2) \times 10^{11}$ Bq $(27.6 \pm 0.6$ Ci) using calorimetry [16]. Correcting the decay of tritium, we estimated the value of ^{6}Li burn-up to be 38%. The apparatus consists mainly of an electric furnace, a cold trap cooled with liquid nitrogen, a gas chromatograph (GC), a small ionization chamber (IC), and a tritium removal equipment. The GC was furnished with a thermal conductivity detector (TCD) and a separation column. The flow speed of neon carrier-gas to separate helium was 60 ml min^{-1}. Details of the apparatus are described in a previous paper [13]. A small specimen of 1 mg was cut from the irradiated alloy. The specimen was inserted in the electric furnace and heated isochronally for 20 min at temperatures ranging from 473 to 1073 K with 100 K interval or heated isothermally at temperatures ranging from 593 to 693 K in a neon atmosphere. In the case of isochronal heating, the gas released during heating was analyzed with GC and IC; the amounts of H_2 and helium were determined with TCD, and those of HT and T_2 with IC. In the case of isothermal heating, the separation column and TCD were by-passed. The radioactivity of aqueous species (expressed by HTO) collected in the cold trap was measured by a liquid scintillation (LS) counter.

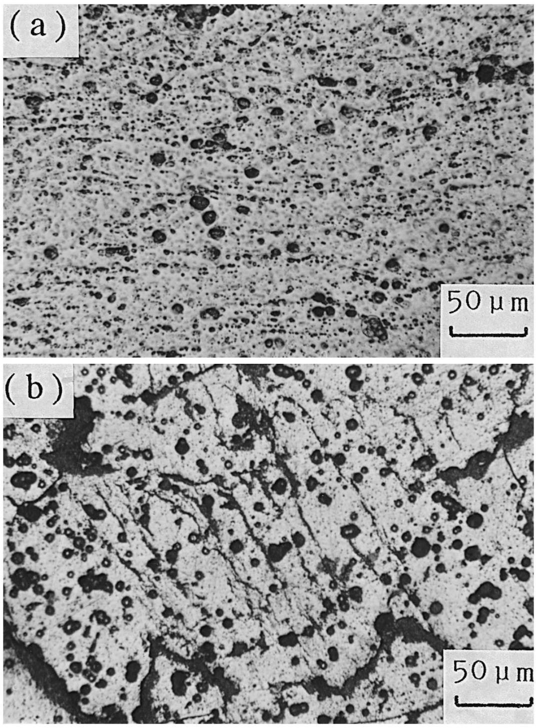

Fig. 1. Optical-microphotographs on the surface of unirradiated Al–12.7 at.% ^{6}Li alloy (a) and neutron-irradiated Al–12.7 at.% ^{6}Li alloy (b) after mechanical polishing and electrolytic etching.

The valence state of tritium in the alloy was estimated with the radiometric method [17]; the 1-mg specimen was dissolved by the NaOD/D_2O solution and the gaseous and the aqueous components in the tritiated solution were analyzed by the radiogas chromatography and LS.

3. Results and discussion

3.1. Optical-microscope observation of the surface

All the surface of both unirradiated and neutron-irradiated Al–12.7 at.% ^{6}Li alloys were covered with etch pits as shown in Fig. 1. The surface of the neutron-irradiated alloy was covered with the cracks as shown in Fig. 1b, but that of the unirradiated alloy is not as shown in Fig. 1a. Though we could not observe if the cracks were inside the grain or not in Fig. 1b, the lengths of the cracks were larger enough than the size of the β-phase particles (radius: 3.5 μm) and the distance between the particles (5 μm) were reported [18].

The origin of the cracks was attributed to the rupture of helium bubbles around the alloy surface as below. During irradiation (934 h) and cooling (2 years and 8 months), some amount of helium would migrate and bubbles, even if the environment was room temperature. Since helium bubbles near the surface are easy to rupture in the direction of the surface, we assumed that the rupture of helium bubbles induced the cracks of the surface, as shown in Fig. 1b. The amount of decayed helium-3 was one-twentieth of helium produced by the ^{6}Li(n, α)T reaction. The amount of helium released from the alloy during irradiation and cooling was a few percent of all helium produced in the alloy. The amount of tritium released from the alloy during irradiation and cooling was also a few percent of all tritium produced in the alloy.

3.2. Analysis of the oxide layer on the surface

The RBS spectrum of the unirradiated Al–12.7 at.% ^{6}Li alloy showed the existence of aluminum, oxygen, and lithium on the surface layer. That is, the surface of the specimen would be covered with the surface layer mainly composed of alumina and lithium oxide. The thickness of the layer on the alloy was estimated to be several hundreds angstrom, in comparison with that of the oxide layer of β-LiAl determined by the RBS experiment and the electron diffraction measurement [19].

The RBS can not detect the light element like hydrogen. However, the release behavior of H_2 from the alloy implies the existence of H_2O absorbed in the oxide layer of the alloy surface, as mentioned in Section 3.6 [13,20].

3.3. Valence states of tritium

We estimated the valence state of tritium in the alloy with the same method described in a previous paper [17], considering the isotopic purity of the NaOD/D_2O solution. The percentages of each valence states were 2% for T^+, 15% for T^0, and 83% for T^- in the neutron-irradiated Al–12.7 at.% ^{6}Li alloy. Here, the T^0 state means HT or T_2, and the T^+ state T_2O or HTO. The origin of the aqueous species was attributed to the oxide layer on the alloy, which was confirmed by the RBS experiment. The T^- state in the alloy is ascribed to the interstitial tritium ion in lattice which presents around Li^+-ion owing to the strong ionic affinity between the two ions. The strong affinity of lithium for tritium impeded the tritium diffusion in the Li–Al system [21] and the intermetallic compound β-LiAl [15,21,22]. The strong affinity originates from the fact that the electronegativity difference between hydrogen and lithium is larger than that between hydrogen and aluminum (for example, Ref. [23]). Hayashi et al. [21] have also reported that the T^- state in the neutron-irradiated Al–4.8 at.% Li alloy attained to 80.8% of the incorporated tritium. In their study, the percentages of other valence states were 15.2% for T^+ and 4.0% for T^0 in the neutron-irradiated Al–4.8 at.% ^{6}Li alloy of thermal neutron fluence 2.5×10^{16} cm^{-2}. In the present work, however, the percentage of the T^0 state attained to 15% owing to the high thermal neutron fluence 1.4×10^{21} cm^{-2} for the Al–12.7 at.% ^{6}Li alloy. In the both studies, most of tritium in the neutron-irradiated Li–Al alloys would diffuse in the T^- state and release in the form of HT or T_2 from the alloy on heating.

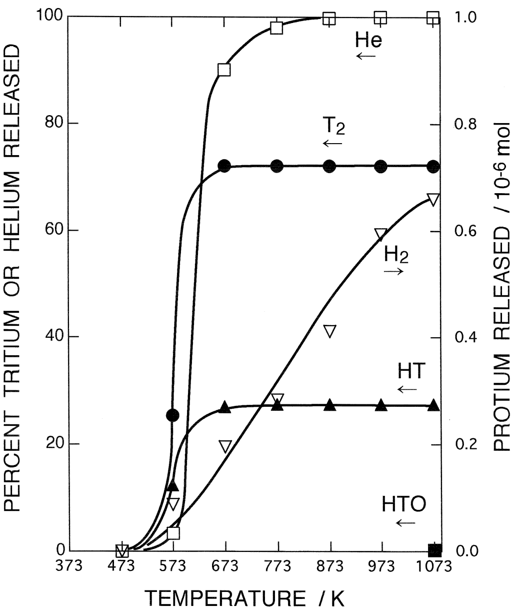

Fig. 2. Release of hydrogen gases (closed circle, T_2; closed triangle, HT; closed square, HTO; open inverse-triangle; H_2) and helium (open square) from the neutron-irradiated Al–12.7 at.% ^{6}Li alloy on isochronal heating.

3.4. Isochronal heating

Fig. 2 shows the release curves for H_2, HT, T_2, HTO, and helium obtained by heating the specimen isochronally. The quantities of H_2, HT, T_2, and HTO released from the specimen are expressed as percent fraction to the total tritium released. As shown in Fig. 2, HT and T_2 were released at temperatures between 473 and 673 K, and H_2 was released gradually at 1073 K. The behavior was almost the same as that of Al–0.1 at.% ^{6}Li alloy reported previously [13], except for the fact that the tritium (HT, T_2) was released from the Al–12.7 at.% ^{6}Li alloy at temperature 80 K lower than that of the Al–0.1 at.% ^{6}Li alloy.

In the case of Al–0.1 at.% ^{6}Li alloy, helium was not released at temperatures below the melting point of the alloy. In this study, however, helium was abruptly released around 570 K and gradually released to the melting point of the alloy (932 K).

The percentages of tritiated species released from Al–12.7 at.% ^{6}Li alloy at temperatures below 1073 K were 1% for HTO, 27% for HT and 72% for T_2. The origin of HTO was attributed to the oxygen atom contained in the oxide layer on the alloy and the fraction was less than 1% as that of the Al–0.1 at.% ^{6}Li alloy.

3.5. Isothermal heating

Fig. 3a shows the tritium release curves obtained by isothermally heating the specimen as a function of time. The isothermal release of tritium from Al–12.7 at.% ^{6}Li alloy was gradually enhanced by raising the heating temperature and it has no induction period. On the other hand, the isothermal release curve of tritium for the Al–0.1 at.% ^{6}Li alloy [13] is shown with an induction period of 60 min, as shown in Fig. 3b. In the two experiments, the fraction of HTO released was the same as that obtained on isochronal heating.

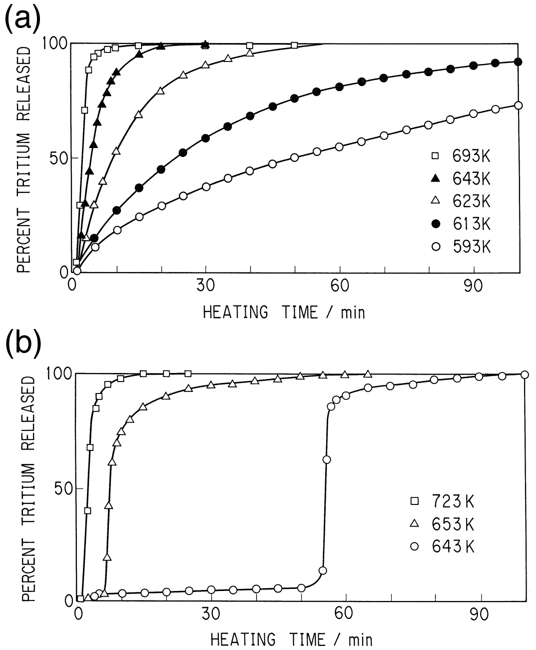

Fig. 3. Release of tritium from the neutron-irradiated Li–Al alloy on isothermal heating: (a) Al–12.7 at.% ^{6}Li alloy, (b) Al–0.1 at.% ^{6}Li alloy (Ref. [13]).

3.6. Release behavior of tritiated species and helium

In the previous work [13], we elucidated that the protium existing in the interior of the Al–0.1 at.% ^{6}Li alloy is released as HT after combination with tritium and most of protium is gradually evolved as H_2 above 570 K by the deoxidation reaction between aluminum and H_2O contained in the surface oxide [13,20]. That is, the discrepancy between the temperature dependence of tritium (HT, T_2) release in the isochronal heating and that of the protium (H_2) release originates from the difference of their position in the alloy. In this study, the temperature dependence of isochronally released H_2 of the Al–12.7 at.% ^{6}Li alloy is similar to that of the Al–0.1 at.% ^{6}Li alloy. Therefore, we concluded that most of the isochronally released H_2 of the Al–12.7 at.% ^{6}Li alloy originates from the surface oxide as same as the Al–0.1 at.% ^{6}Li alloy. It is reasonable that the amount of protium in the form of HT released from the Al–12.7 at.% ^{6}Li alloy is 10^2 magnitude larger than that from the Al–0.1 at.% ^{6}Li alloy with increasing lithium content in the alloy, since atoms like Li and Mg in aluminum alloy have a strong trapping effect to contain impurity atoms like hydrogen when the alloy is produced [24], as mentioned in Section 3.3 [15,21,22].

The amount of H_2 released from the Al–12.7 at.% ^{6}Li alloy is also 10^2 magnitude larger than that from the Al–0.1 at.% ^{6}Li alloy. Since we elucidated that most of protium is gradually evolved as H_2 above 570 K by the deoxidation reaction between aluminum and H_2O contained in the surface oxide [13,20], the above fact is reasonable. That is, H_2O contained in the surface oxide of the Al–12.7 at.% ^{6}Li alloy is much larger than that of the Al–0.1 at.% ^{6}Li alloy, because the thickness of oxide, which is composed of lithium, increases with the increase of the lithium content of the alloy.

The dose due to the ^{6}Li(n, α)T reaction in the Al–12.7 at.% ^{6}Li alloy was estimated to be 5.7 dpa by the NRT model [25] as mentioned in a previous paper [15]. The dose due to fast neutrons was estimated to be 0.8 dpa by the same way. Therefore, even though the effects of fast neutrons is not negligibly small, the dominant origin of the irradiation effects is the ^{6}Li(n, α)T reaction.

It is known that protium is trapped at some kinds of defects in metals; for example, in Fe at dislocations [26], and aluminum deformed 15% by cold work was recrystallized at 643 K after an induction period of 10 min [27]. As shown in Fig. 3b, the isothermal release of tritium from 0.1 at.% ^{6}Li–Al alloy, which had been deformed 20% by cold work, happened at 643 K after a longer induction period of 60 min. Thus, in the previous work [13], we concluded that tritium produced in the alloy is trapped at dislocations created by cold work and released when the trap sites disappear by recrystallization of the alloy. Though the Al–12.7 at.% ^{6}Li alloy used in this work was also deformed 20% by cold work, the isothermal release of tritium from the alloy has no induction period as shown in

Fig. 3a. In the present study, we could not understand the mechanism of irradiation effects in the neutron-irradiated Al–12.7 at.% ^{6}Li alloy in detail. However, the dose due to fast neutrons (0.8 dpa) in this study was as same as that in the previous study of the neutron-irradiated Al–0.1 at.% ^{6}Li alloy. On the other hand, the dose due to the ^{6}Li(n, α)T reaction in the Al–12.7 at.% ^{6}Li alloy reached 5.7 dpa, which is 10^2 times larger than that in the previous study, with increasing the ^{6}Li content of the alloy. Therefore, the difference in the isothermal release behaviors of tritium in Fig. 3 originates from the ^{6}Li(n, α)T reaction in the neutron-irradiated Al–12.7 at.% ^{6}Li alloy. The nuclear reaction changed the defect structure of the alloy by the knock-on due to the recoil atoms of the reaction, and produced tritium and helium whose doses are 10^2 times larger than that in the previous study of the Al–0.1 at.% ^{6}Li alloy. The ^{6}Li(n, α)T reaction in the neutron-irradiated Al–12.7 at.% ^{6}Li alloy did not only affect the isochronal release of tritium, but also affected the isothermal release, whose temperature was 80 K lower than that of the Al–0.1 at.% ^{6}Li alloy. Indeed, we demonstrated that the ^{6}Li(n, α)T reaction drastically affected the defect structure of β-LiAl [15].

The crystal structure of Al–0.1 at.% ^{6}Li alloy is f.c.c. as that of Al. On the other hand, the Al–12.7 at.% ^{6}Li alloy is composed of α- and β-phase as shown in a phase diagram of Li–Al system [28,29]. The crystal structure of β-phase is NaTl-type, which is composed of two interpenetrating diamond sublattices such that each atom has eight nearest neighbors: four like and four unlike atoms [30]. Considering the negligible release of tritium during irradiation and the isochronal release of tritium showed in Fig. 2, we assumed that the specimen temperature was less than 473 K during irradiation. Since the solubility of lithium in aluminum is below 2 at.% [18] at temperatures below 473 K, most of lithium is in the β-phase. However, the recoil range of tritium (37.5 μm in the β-phase, 52.3 μm in the α-phase) calculated by TRIM92 computer code [31] is much larger than the size of the β-phase particles (radius: 3.5 μm) which is determined by a chemical method [18], and the distance between the particles (5 μm) which is calculated by the volume fraction of the particles. Therefore, the newly formed tritium is distributed throughout both the α- and β-phases. Since the calculated helium recoil range (9.4 μm in the β-phase, 6.8 μm in the α-phase) is also larger than the distance between the particles, most of the newly formed helium is also distributed throughout both the α- and β-phases. Most of tritium and helium was not released at temperatures below 473 K as shown in Fig. 2, but the moderate heat treatment would promote the migration of both tritium and helium during irradiation. Tritium would be concentrated in the β-phase, which acts as a sink for it because of the chemical affinity between lithium and tritium as mentioned in Section 3.2 [15,21–23]. Helium tends to collect in grain boundaries that surrounds the β-phase particles and at the many vacancies produced by the ^{6}Li(n, α)T reaction, because helium atom has no strong affinity with the other element atom and becomes cluster or bubble even at small concentration in the crystal owing to the low solubilities [32–35].

Since the behavior of helium in Ni alloy is important in nuclear materials, many experimental and theoretical studies [32,33,36,37] were conducted.

For example, Bisson and Wilson [33] showed by a semi-empirical calculation that the interstitial cluster composed of more than several helium atoms around vacancy was deeply bounded in nickel, relating to the few release of helium-3 decayed from tritium at temperatures below 800 K in nickel [32].

Next, in the experimental work, Abell et al. [38] observed the decayed helium-3 release from palladium tritide (the helium-to-metal-atom ratio $[\mathrm{He}]/[\mathrm{Pd}] = 0.3$) at the lower temperatures (from 500 K) than that from palladium tritide ($[\mathrm{He}]/[\mathrm{Pd}] = 0.02$), and they summarized the mechanism of helium behavior as below [38]. The highly overpressurized gas bubbles grow by punching out dislocation loops, which in turn alter the mechanical properties and microstructures of the host material. Ultimately, helium release can occur when the chemical potential for helium atoms in bubbles approaches that for interstitial helium, or when stress from bubbles exceeds the tensile strength of the deformed host lattice to produce irreversible rupture of bubbles as they reach critical dimensions.

In the present study, the helium was released from the neutron-irradiated Al–12.7 at.% ^{6}Li alloy by isochronal heating around 600 K as shown in Fig. 2. Though the helium-to-metal-atom ratio ($[\mathrm{He}]/[\mathrm{Al\ and\ Li}] = 0.3 \times 10^{-3}$) is smaller than that of the palladium tritide mentioned above, the helium release behavior from the neutron-irradiated Al–12.7 at.% ^{6}Li alloy is similar to that from the palladium tritide. Moreover, the positron-lifetime technique [39] also shows that the He bubbles in aluminum grow through condensation of thermally produced vacancies, as well as bubble migration and coalescence above 600 K. Considering the behavior of helium bubble in the palladium tritide and in the aluminum mentioned above, we assume the helium behavior in the neutron-irradiated Al–12.7 at.% ^{6}Li alloy with heating as below. Since much amount of helium (10^2 times larger than in the case of Al–0.1 at.% ^{6}Li alloy [13]) and many vacancies were produced by the ^{6}Li(n, α)T reaction in the neutron-irradiated Al–12.7 at.% ^{6}Li alloy, helium would migrate and bubbles owing to the low solubilities [32–34], which causes the high pressure in the bubble [34]. Consequently, helium was released from the Al–12.7 at.% ^{6}Li alloy at temperatures below the melting point of the alloy, as shown in Fig. 2.

3.7. Application to tritium production

Target material used for the tritium production of 40 TBq per batch [2] was the Al–12.7 at.% ^{6}Li alloy plate clothed with the thin aluminum (0.75 mm thick) and irradiated in the cooling water of JMTR. In the tritium production [2], the neutron-irradiated Al–12.7 at.% ^{6}Li alloy was heated to 993 K over the melting point of alloy (932 K) to extract tritium. However, the tritium gas was released at temperatures below the melting point as shown in Fig. 2. Thus, in the recent tritium production of 40 TBq per batch, tritium is well extracted at temperature (763 K) below the melting point of alloy. It was also confirmed that the pressure in the vacuum furnace abruptly increases by increasing the temperatures in the range of 573 and 673 K [2], which is attributed to the release of tritiated gases and helium from the alloy target. It is preferable to extract tritium from the alloy at temperatures below the melting point (932 K) from a safety standpoint of view, because aluminum has an eutectic point (see for example, Ref. [40]) with the each component of stainless steel of the crucible at temperatures around 920 K.

4. Conclusion

In the neutron-irradiated Al–12.7 at.% ^{6}Li alloy, most of tritium produced by the ^{6}Li(n, α)T reaction existed in the T^- state and was released in the form of HT and T_2 at temperatures below the melting point of the alloy (around 570 K) on isochronal heating in the neon atmosphere. Protium (H_2) was gradually released at temperatures above 570 K and the origin was attributed to the deoxidation reaction between aluminum and H_2O contained in the surface oxide as that from the Al–0.1 at.% ^{6}Li alloy of the previous study. In the case of Al–0.1 at.% ^{6}Li alloy of the previous study, helium was not released below the melting point of the alloy owing to trapping of helium at vacancies in the alloy. However, in the case of Al–12.7 at.% ^{6}Li alloy of this study, much amount of helium (10^2 times larger) is not stable in the alloy even at the temperatures below the melting point of the alloy, because helium cluster grows to a bubble with many vacancies produced by the ^{6}Li(n, α)T reaction and it ruptures. The above facts result in the preferable procedure of extracting the tritium from the alloy at temperatures below the melting point (932 K) in the 40-TBq tritium production per batch.

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