Study on interaction of tritium with radiolysis products in Li_2O

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Abstract

The study of an influence of radiation defects on tritium release behavior from Li_2O was performed by the out-of-pile tritium release experiments. The linear temperature increase of the electron-irradiated samples showed two tritium release peaks with the maxima at $\sim$ 800 K and $\sim$ 1200 K. It is thought that the tritium release at high temperatures ($>$ 950 K) is due to the thermal decomposition of LiT. The mechanism of an interaction of tritium with radiolysis products in Li_2O is discussed.

1. Experimental technique

The out-of-pile tritium release experiments were performed using a polycrystalline Li_2O (99.9% pure) of the particle size of 2–4 mm. The specimens were sealed in quartz containers filled with air atmosphere and irradiated by 2.2 MeV electrons (8 kGy/s, $\sim$ 370 K) to absorbed dose up to 100 MGy at the DYNAMITRON facility of the University of Tokyo.

Afterwards, the specimens were exposed at $\sim$ 310 K to the thermal neutron fluence of 10^{17} n/m^2 at the research reactor "YAYOI" of the University of Tokyo. The samples without prior irradiation by electrons were exposed to the same fluence of thermal neutrons and used as reference ones. The out-of-pile tritium release experiments were carried out during the temperature transient from 293 to 1273 K with temperature ramp of 10 K per minute. The released tritium was removed by a sweep gas of N_2 and detected by an ionization chamber.

2. Results and discussion

Fig.1 shows a tritium release curve of the polycrystalline Li_2O specimen exposed at about 310 K to the thermal neutron fluence of 10^{17} n/m^2. A sharp peak of the released tritium can be seen at $\sim$ 750 K. The experimental data indicate that there is no a discernible influence of radiation defects on tritium release behavior. The absorbed dose for this specimen is estimated at $\sim$ 10 kGy. Thus, the formation of simple F-centers proceeds at such low absorbed dose [1, 2]. It seems likely that an interaction of F-centers with tritium at low irradiation dose is negligible.

The irradiation of Li_2O specimens by 2.2 MeV electrons to absorbed dose of 100 MGy followed with the thermal neutron fluence of 10^{17} n/m^2 caused the change of the tritium release kinetics (Fig.2). One can recognize two broad peaks in the tritium release curve: first starts at 600 K with the maximum at 800 K and second appears at 950 K with maximum peaked at $\sim$ 1200 K. The first broad peak is associated with (1) desorption of physically and chemically adsorbed tritium on the sample surface, (2) decomposition of LiOT, and (3) migration of tritium dissolved in the bulk. It seems that the tritium bonded in the form of LiT is not released below 850 K as a significant fraction. The second peak begins to appear at $\sim$ 950 K which is coincident with the phase transition of LiH (the 961.8 K corresponds to the crystal $\rightarrow$ liquid transition [3]). The second peak is thought to be due to the tritium bonded in Li–T bond, which was formed by the reaction of colloidal Li or F-center aggregates with tritium.

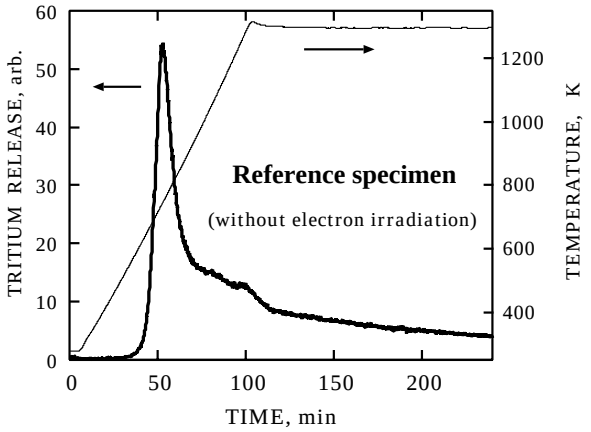

Figure 1: Out-of-pile tritium release from Li_2O specimen irradiated by thermal neutron fluence of 10^{17} n/m^2.

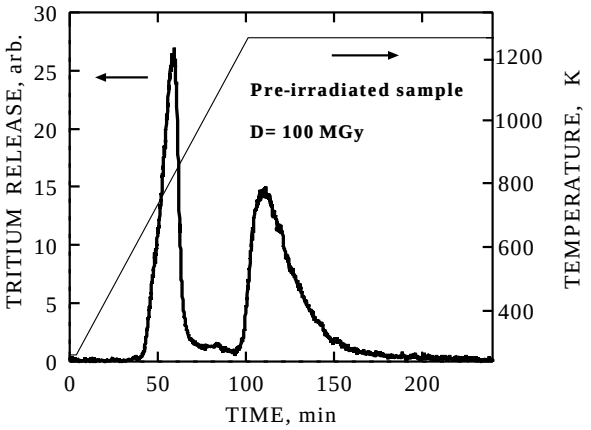

Figure 2: Out-of-pile tritium release from Li_2O pre-irradiated by electrons (2.2 MeV,100 MGy) and by thermal neutron fluence of 10^{17} n/m^2.

In the article by Vajda et al. [4], the recovery of metallic Li colloids and F^+ center agglomerates was reported to occur at around 673 and 923 K, respectively. F-center aggregates (in other words: clusters or agglomerates) are nothing more nor less than Li-atom aggregates which consist from the Li-atom ions surrounded by the F-centers. Thus, in any case the formation of Li-T bond can occur in Li_2O even at high temperatures. The problem is only one the amount of formed LiT. The concentration of F^+ center clusters determines the accumulation rate of LiT, and in its turn depends on temperature and irradiation dose. Thus, a certain region of temperature exists wherein the formation of Li-T bond is the most effective. In fact, the data of the ESR examination of Li_2O single crystals irradiated at 650 K by fast neutrons in the BEATRIX II [5] showed that the F^+ center aggregates is created in Li_2O. According to our investigations [6], the 650 K can be critical temperature for the formation of Li colloids. However, there is no clear understanding about the optimum temperature for the formation of F^+ center aggregates. Thus, the investigation of an influence of the radiolysis products on tritium release kinetics from Li_2O under the conditions expected in the thermonuclear reactor is desirable.

3. Conclusions

It has been shown that the radiation defects introduced by electron irradiation cause the retention of tritium due to its bonding in the thermally stable state. The interaction of tritium with colloidal Li produced by electron irradiation is thought to result in the formation of Li–T bond. The tritium release at high temperatures (> 950 K) indicates the decomposition of Li–T bond.

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