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**Deuterium Ion Irradiation for Tritium Breeding Material and Evaluation for
Tritium Inventories in Test Blanket Module and Blanket of a Demonstration
Reactor**

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Deuterium Ion Irradiation for Tritium Breeding Material and Evaluation for Tritium Inventories in Test Blanket Module and Blanket of a Demonstration Reactor

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Abstract

The tritium produced in a tritium breeder of blanket has to be recovered under the temperature distribution to reduce the tritium inventory. Lithium titanate pebbles were irradiated by deuterium ions with different temperatures and ion fluences. The deuterium retained in the pebbles desorbed in forms of HD, D₂, HDO and D₂O. The amount of retained deuterium decreased for the temperature higher than 473 K, and became to zero for the temperature higher than 773 K. If the temperature range is taken from 573 to 1173 K in the solid breeder TBM, the tritium inventory is lower than 1 gram. However, in the demonstration reactor, the total tritium inventory of blankets becomes a few kilograms. For the reduction of the tritium inventory, the region with the low temperature region has to be significantly reduced.

Keywords

test blanket module (TBM), blanket, tritium inventory, lithium titanate, deuterium ion irradiation,

1. Introduction

In-vessel tritium inventory in ITER is limited below a few hundreds grams from view points of safety and regulation. It is pointed out that the tritium inventory of co-deposited carbon dust/flake may become significantly large. The tritium inventory of Test Blanket Modules (TBMs) [1] also may become large if the design for TBM is not suitable. Tritium behavior in the tritium breeding material such as release behavior of tritium from Li_4SiO_4 [2], isotope exchange reaction in breeder materials [3] and adsorption characteristics of water vapor on Li_2ZrO_3 [4] has been investigated so far. However, the maximum amount of tritium inventory in TBM has not been evaluated yet.

In the fusion demonstration reactor, the in-vessel tritium inventory might be limited owing to the same reasons.

The tritium inventory of blankets in the fusion demonstration reactor becomes extremely large, so that the evaluation for the tritium inventory is quite important and the reduction of this inventory is required. In the present study, the tritium inventories of TBM and blanket in the demonstration reactor are estimated based on the results of the deuterium ion irradiation experiment, and the condition required for the reduction of the tritium inventory is suggested. The present approach for evaluation of tritium inventory can be applied for the different types of blankets.

In the water cooled solid breeder TBM, lithium titanate pebbles (Li_2TiO_3) are employed as the tritium breeder. As the solid breeder material, Li_2TiO_3 shows good chemical stability and tritium (T) release by thermal desorption [5, 6]. In the blanket of fusion reactors, the pebbles are irradiated by fusion neutrons and then tritium is produced by nuclear reactions of ${}^6\text{Li} (n, \alpha) \text{T}$ or ${}^7\text{Li} (n, n \alpha) \text{T}$. The temperature distribution of the solid breeding material in TBM has been estimated with taking account of nuclear heating to be in the range from 573 K to 1173 K [7]. Although it is desired for the tritium to be recovered completely in whole region of blanket during ITER operation, a small fraction of the tritium remains in low temperature region [8]. Therefore, in order to estimate the tritium inventory in Li_2TiO_3 pebbles in the TBM of ITER and the blanket of the demonstration reactors, the retention and desorption behavior of the tritium in the lithium titanate has to be investigated.

In the present study, the deuterium ion irradiation experiment was conducted for in Li_2TiO_3 pebbles to simulate the tritium retention and desorption behavior. The pebbles were irradiated by deuterium ions with energy of 1.7 keV at various irradiation temperatures (RT, 473 K, 573 K, 673 K, 773 K), and after that the retention and desorption behavior of retained deuterium was investigated by using a technique of thermal desorption spectroscopy (TDS). The tritium inventory depends on the burn-up ratio of the blanket. The tritium inventory in both the TBM and the blanket of the demonstration reactor was estimated based upon the obtained results.

2. Experiments

The diameter of tritium breeding material used for the TBM may be several mm [9]. In the present experiment, the pebbles with a diameter of 2 mm ϕ was employed. Totally 18 Li_2TiO_3 pebbles with diameter of were used for

the deuterium ion irradiation. These pebbles were heated at 973 K for an hour in a vacuum chamber to remove impurities, such as hydroxides and carbon oxides [9]. The amount of impurity gases was significantly reduced by this preheating. After the heating, the pebbles were installed to a sample holder in an electron cyclotron resonance heating (ECR) ion source (Fig.1) and irradiated by deuterium ions. The sample holder made of Ta plate and Mo mesh was used to fix these pebbles, and the sample was heated by indirect resistive heating during the ion irradiation. The irradiation temperature (surface temperature of the pebbles) was changed from RT to 773 K. The surface temperature of the pebble and the temperature of the sample holder were measured up to 1000 K, by using a thermocouple. The temperature difference between the surface of the pebble and the sample holder was approximately 100 - 200 K at each irradiation temperature.

The deuterium ion energy was 1.7 keV and the ion flux was approximately 9×10^{14} D/cm²·s. The deuterium retention saturate at the fluence of approximately 1×10^{18} D/cm². Then, the ion fluence was taken 5×10^{18} D/cm². The maximum amount of tritium concentration is needed to evaluate the maximum tritium inventory of the blanket. The maximum concentration of deuterium in the ion implantation depth (~10nm) can be known under this experimental condition. In the evaluation of tritium inventory in the blanket shown later, this concentration is assumed to the entire region of the tritium breeding layer irradiated by fusion neutrons. After the ion irradiation, the pebbles were extracted and quickly transferred to the TDS chamber. Then, the sample was heated by an infrared light furnace from RT to 973 K with a heating rate of 10 K/min. At the highest temperature, 973 K, the heating was conducted for 1h. The ultimate pressure before the TDS analysis was approximately 10^{-8} Pa. The change of the surface morphology and the atomic composition were also investigated. The lithium titanate is

electrically insulator, so that the surface has to be changed to electrically conductive for scanning electron microscope (SEM) and Auger electron spectroscopy (AES). For this purpose, the platinum coating was carried out on the surface of the lithium titanate.

3. Results

Figure 2 shows the thermal desorption spectra of gases containing the deuterium in the deuterium-irradiated Li_2TiO_3 pebbles with the irradiation temperatures of (a) RT, (b) 573 K and (c) 773 K. In these figures, the horizontal axis is the temperature of the lithium titanate pebbles. The deuterium retained in these pebbles desorbed in forms of HD, D_2 , HDO and D_2O . H and O were retained before the irradiation in lithium titanate as the impurities. In our previous study, the deuterium desorption behavior of the Li_2TiO_3 pebbles was very similar with that of the Li film, therefore most of retained deuterium is trapped in forms of Li-D and Li-O-D [8]. The deuterium retention saturated at the ion fluence of approximately $1 \times 10^{18} \text{ D/cm}^2$, and the deuterium concentration at RT in the atomic ratio became as high as $\text{D/Li} \sim 1$ [8].

The desorption rates of these gases decreased with increase of the irradiation temperature. At the irradiation temperature of 773 K, the retained deuterium completely desorbed during the irradiation. This result suggests that the tritium produced in Li_2TiO_3 pebbles at the temperature higher than 773 K completely desorbs during the blanket operation. Figure 3 shows the amounts of gases containing the deuterium, and total amount of deuterium desorbed from the pebbles can be obtained as a function of the irradiation temperature as shown later. In the cases of HD and D_2 , the amounts of desorbed gases decreased with increase of the irradiation temperature. On the

other hand, in the cases of HDO and D₂O, the amounts of desorbed gases did not decrease with the increase of the irradiation temperature in the range lower than 473 K, but decreased in the range higher than 473 K. This behavior in the irradiation temperature dependence of the desorption occurs due to the different desorption peak temperatures of these gas species. The amount of deuterium desorbed in form of HDO was largest in these gases at every irradiation temperature. The total amount of retained deuterium was almost the same in the temperature range lower than 473 K, while the total amount decreased sharply with increase of the irradiation temperature in the range higher than 473 K. At the temperature higher than 773 K, the desorption was not observed.

Figure 4 shows the desorption spectra of HD, D₂, HDO and D₂O after the irradiation at RT. In the spectra of HD and D₂, two peaks appeared at 500 K and 700 K. In the spectra of HDO and D₂O, three peaks appeared at 500 K, 600 K and 700 K. In our previous experiment for the Li film [8], two dominant peaks appeared at 500 K and 600 K. In addition, most of D was trapped by Li. Thus, it is presumed that the peaks at 500 K and 600 K are due to de-trapping from bonds of Li-D and Li-OD. In the case that the ion fluence increased, the peak intensity at 700 K relatively increased in the spectra of HD and D₂. Since the lithium content decreases by the selective sputtering, the titanium content relatively increases as shown later. In our previous experiment on deuterium ion irradiation for titanium [8], a single sharp peak at 700 K appeared. Then, the desorption at 700 K may be due to mainly de-trapping from titanium hydride, namely Ti-D.

Figure 5 shows the depth profiles of atomic composition of lithium titanate before deuterium ion irradiation (a), and after deuterium ion irradiation (b). The lithium content was high before the irradiation, but significantly decreased after the irradiation. The major contents at the surface were titanium and oxygen. Figure 6 shows the

surface morphologies before and after the ion irradiation. After the ion irradiation, the surface was eroded by the irradiation and the structure clearly changed. The surface structure consisted with the particles with a micron meter. The atomic concentration suggests that the major content of the particle is titanium oxide, TiO_2 .

Figure 7 shows the amount of retained deuterium (total amount of retained deuterium) versus the temperature of lithium titanate. In the design of water cooling solid breeder TBM, the tritium breeder (Li_2TiO_3) has a temperature range from 573 K to 1173 K. The dashed lines in Fig.7 show this temperature range. The retained deuterium completely desorbes in the region with temperature higher than 773 K, while a small fraction of the deuterium is retained in the region with temperature lower than 773 K. The trapping of deuterium takes place by destructions of the bonds in lithium titanate by energetic deuterium ion. The destructions of the bonds also occur by the fusion neutron irradiation, but the trapping state of deuterium is similar. So that, this conclusion on no-trapping of deuterium for temperature higher than 773 K remains the same even if the pebbles are irradiated by energetic neutrons. In the TBM, the region with temperature lower than 773 K exists. The amount of retained tritium in this region becomes the tritium inventory. Namely, the tritium is retained in the region with temperature of 573-773 K (shown as dot region in Fig.7). The amount of retained tritium in the TBM can be estimated using the present result. The amount of lithium in the TBM is approximately 7 kg [11]. It is assumed that each Li atom traps one atom of tritium, $T/\text{Li} \sim 1$. The tritium produced in the TBM becomes $(3/7 \times 7000 \text{ g} = 3000 \text{ g}) \times \text{burn up ratio}$. The fraction of the low temperature region (573-773 K) is 10 % of the entire region of the breeder [11]. In this low temperature region, approximately 10 % of the tritium produced remains in the blanket. Thus, the tritium inventory becomes $30 \text{ g} \times \text{burn up ratio}$. The burn up ratio in ITER is very small, 0.01 [10], so that the tritium inventory

becomes negligible small, 0.3 gram. However, in the case of the demonstration reactor, the tritium inventory becomes 6 g per blanket module if the burn up ratio is 0.2. The tritium inventory becomes approximately 3 kg if the number of blanket module is 500. Therefore, the tritium inventory has to be decreased by reducing the low temperature region in the tritium breeding region.

4. Conclusions

In the current design of the blanket including the TBM, the tritium breeder region has a temperature distribution. In order to evaluate the tritium inventory and quickly recover the tritium produced in the tritium breeder, the tritium has to be desorbed completely under the temperature distribution. In order to simulate this behavior, the pebbles of lithium titanate were irradiated by deuterium ions (1.7keV D^+) with different temperatures (RT-773 K). After the irradiation, the retention and desorption behavior of retained deuterium was investigated using a thermal desorption spectroscopy.

The deuterium retained in the lithium titanate pebbles desorbed in forms of HD, D_2 , HDO and D_2O . The amount of desorbed deuterium in form of HDO was largest in these gases at any irradiation temperatures. When the irradiation temperature was higher than 473K, the amount of retained deuterium decreased, and became to zero for the temperature higher than 773K. The spectra of HD and D_2 have two peaks at 500 K and 700 K. The spectra of HDO and D_2O have three peaks at 500 K, 600 K and 700 K. The peaks at 500 K and 600 K are owing to the bonds of Li-D and Li-OD. The peak at 700 K may be owing to the bonds of Ti-D and Ti-OD. In the deuterium irradiation for titanium containing oxygen After the irradiation, the atomic composition and the surface morphology were examined. The lithium at the surface disappeared owing to the selective sputtering.

Based on these data including the design parameters, the tritium inventory was estimated. If the ratio of low temperature region is 0.1 and the burn up ratio is 0.01, the tritium inventory may become lower than 1 g. However, in the demonstration reactor, the burn up ratio is high, 0.2, so that the inventory per blanket becomes a few grams. The total in-vessel inventory of blanket modules becomes a few kilograms. Thus, the low temperature region in the tritium breeding region has to be decreased by a suitable design for the temperature profile.

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Figure captions

Fig. 1 ECR ion source apparatus.

Fig. 2 Thermal desorption spectra of gases containing deuterium for deuterium ion irradiated Li_2TiO_3 pebbles at the irradiation temperatures of (a) RT, (b) 573 K and (c) 773 K.

Fig. 3 Amounts of gases containing deuterium desorbed from Li_2TiO_3 pebbles as a function of irradiation temperature.

Fig. 4 Desorption spectra at RT with peak separation for (a) HD, (b) D_2 , (c) HDO and (d) D_2O .

Fig. 5 Depth profiles of atomic composition of lithium titanate before deuterium ion irradiation (a) and after deuterium ion irradiation (b).

Fig. 6 Surface morphologies of lithium titanate before irradiation (a) and after irradiation (b).

Fig. 7 Amounts of retained deuterium in Li_2TiO_3 pebbles as a function of irradiation temperature.

Fig.1

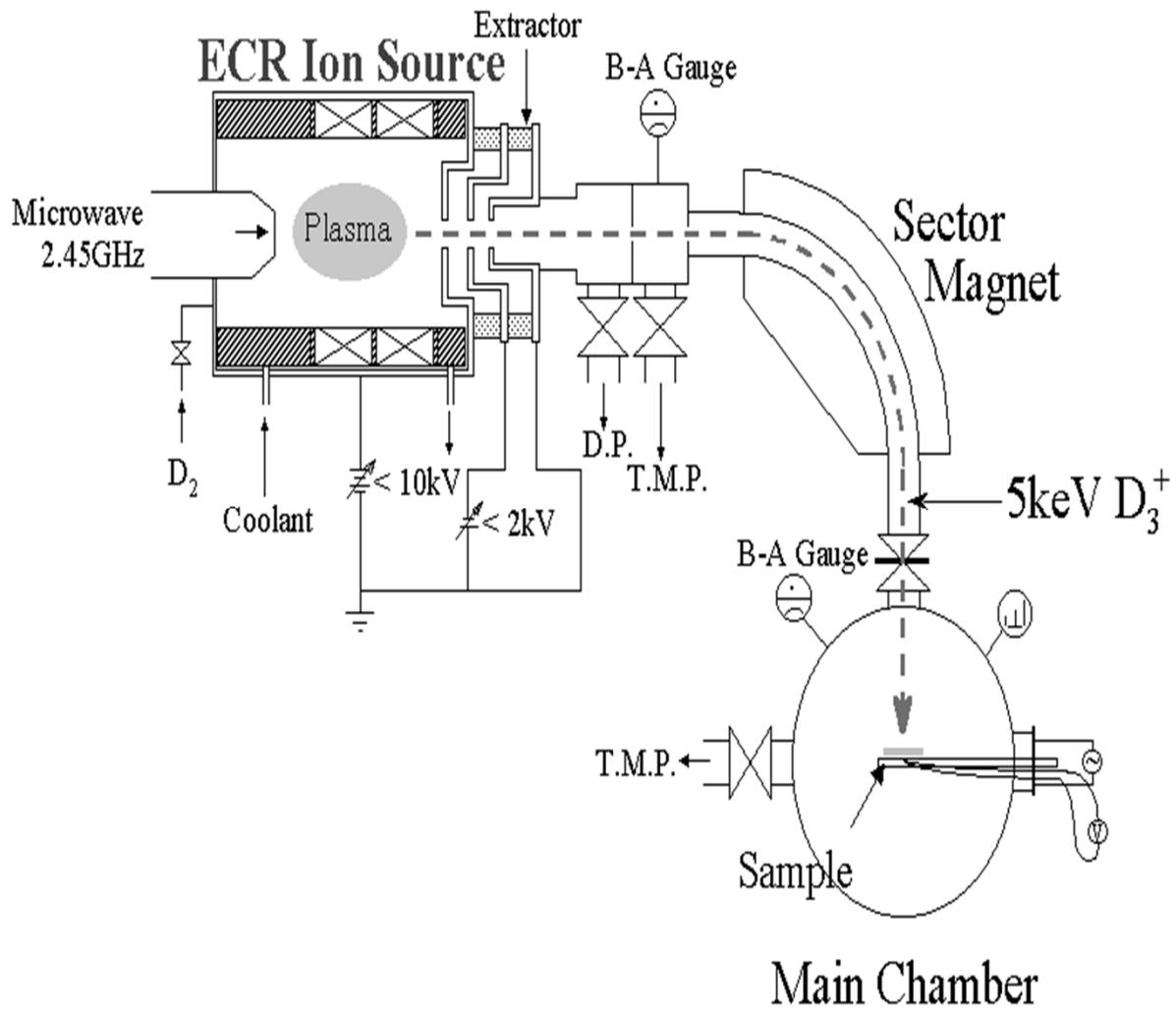


Fig.2

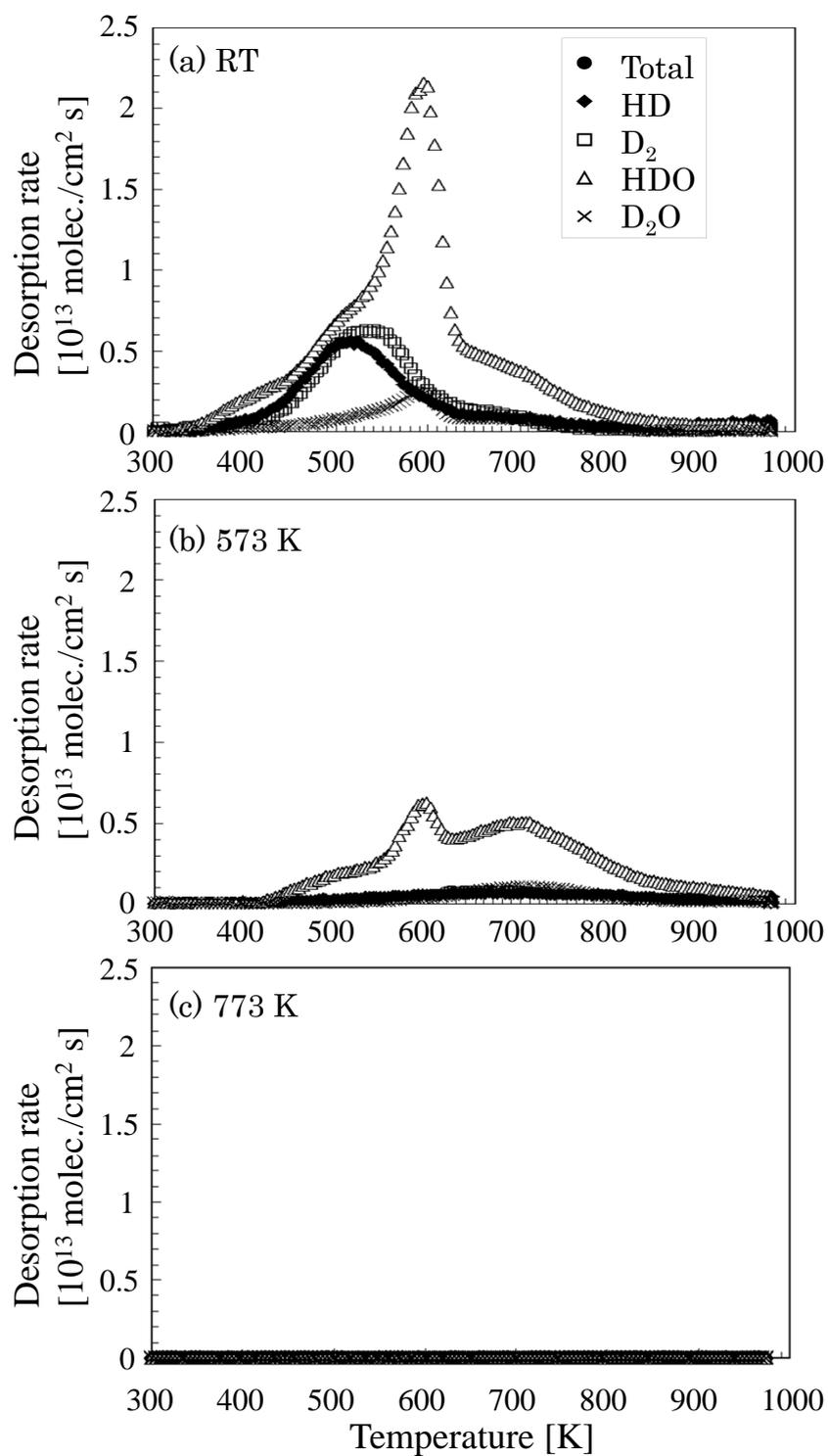


Fig.3

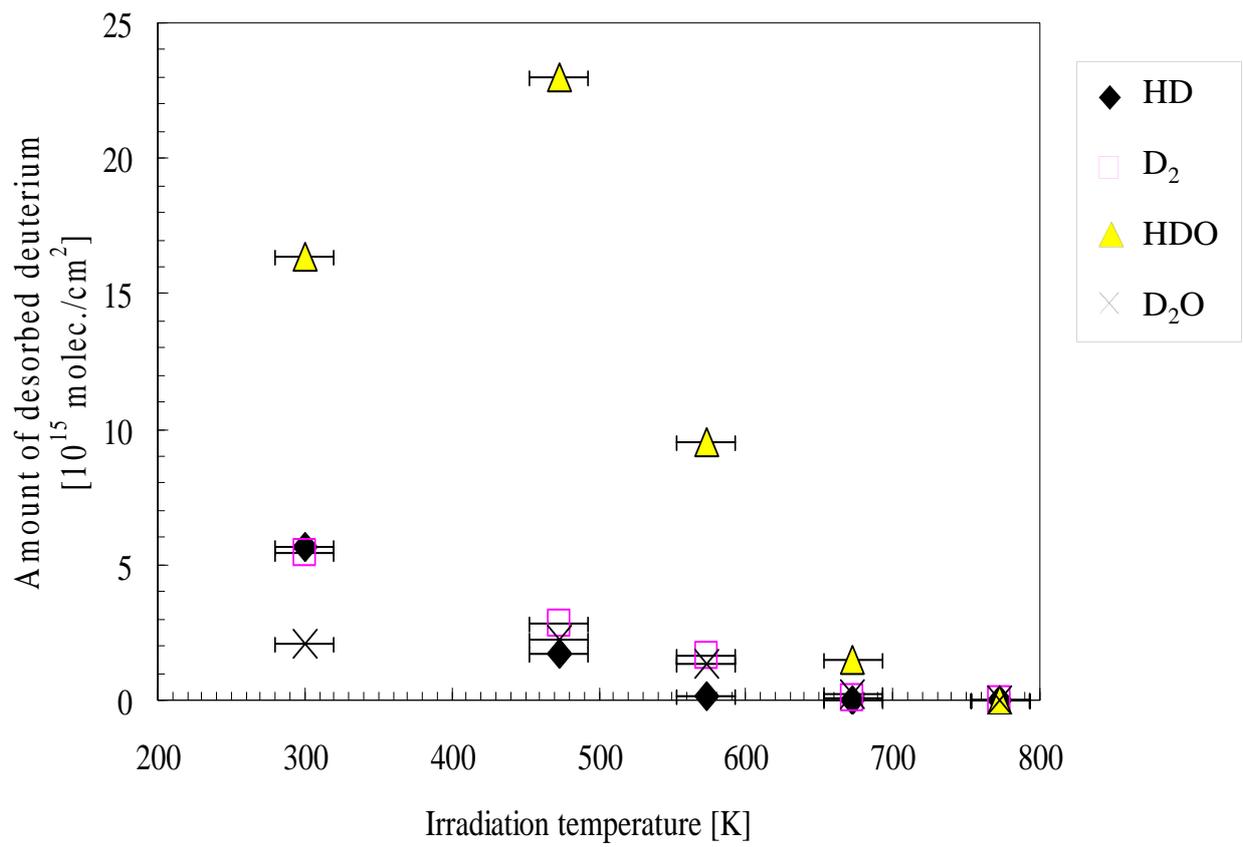


Fig. 4

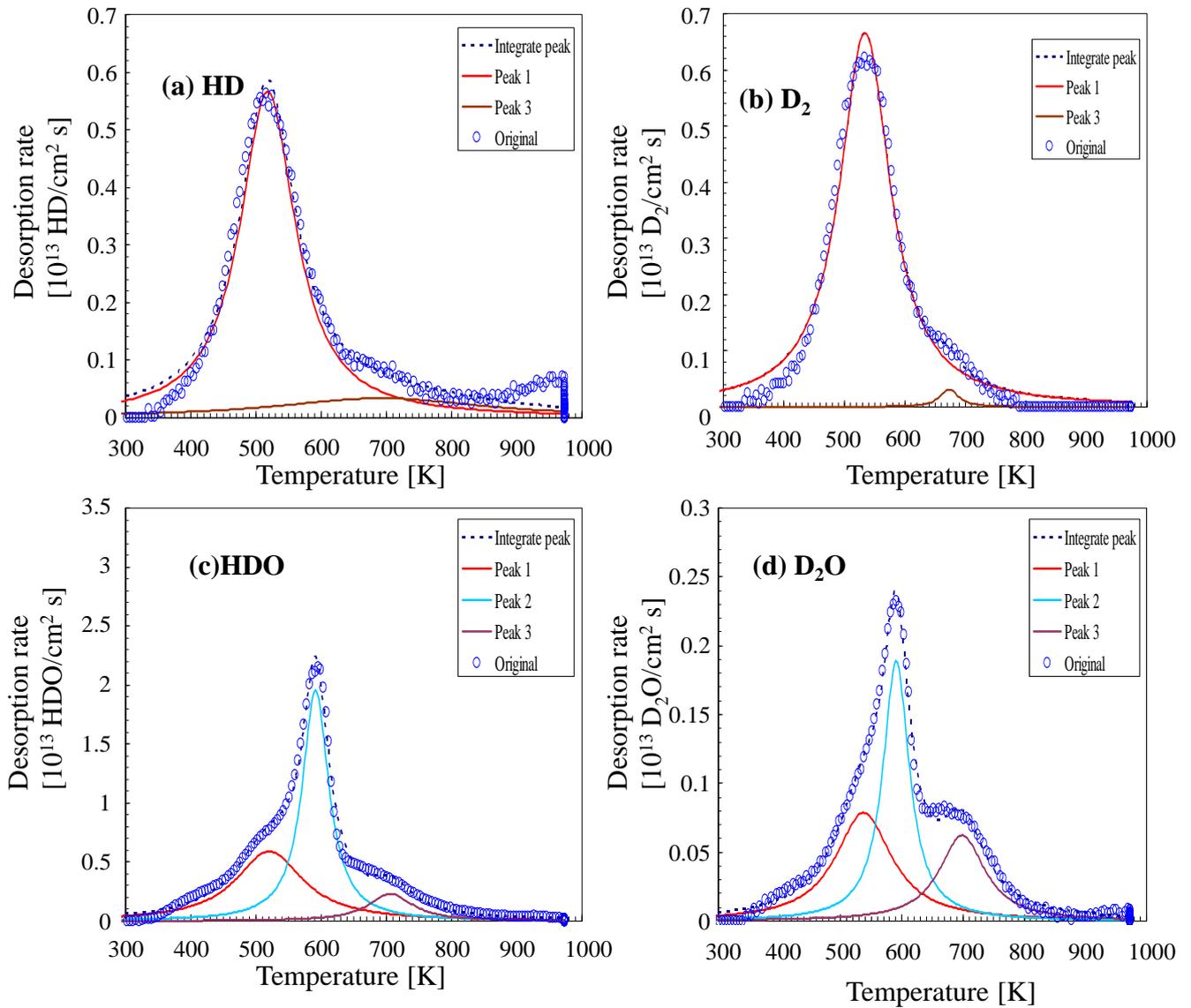


Fig.5

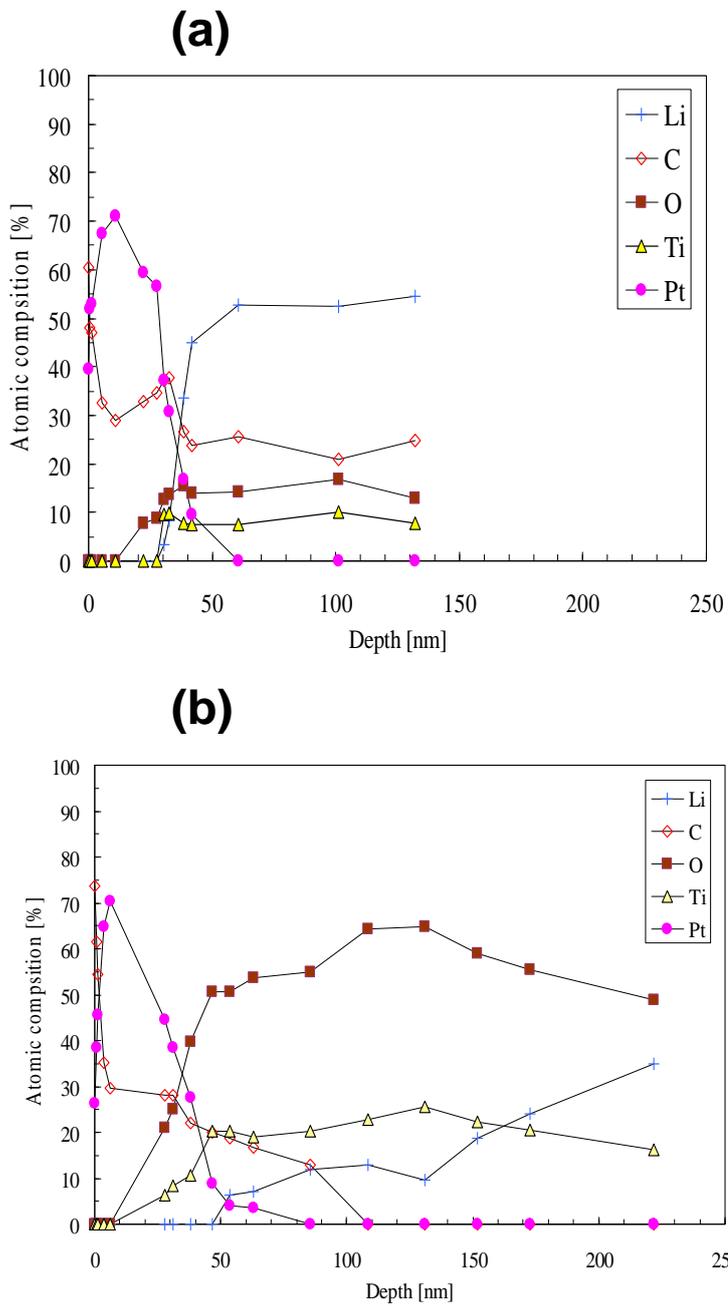
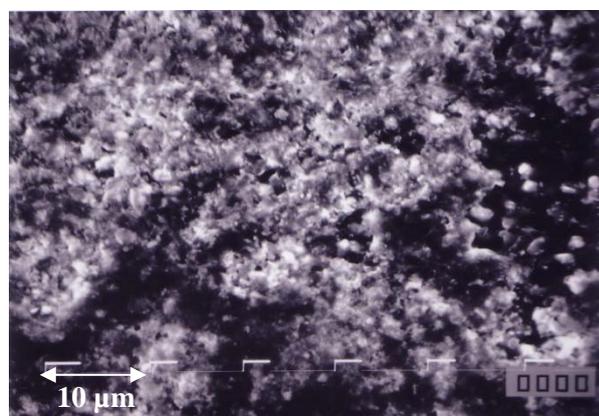


Fig.6

(a)



(b)

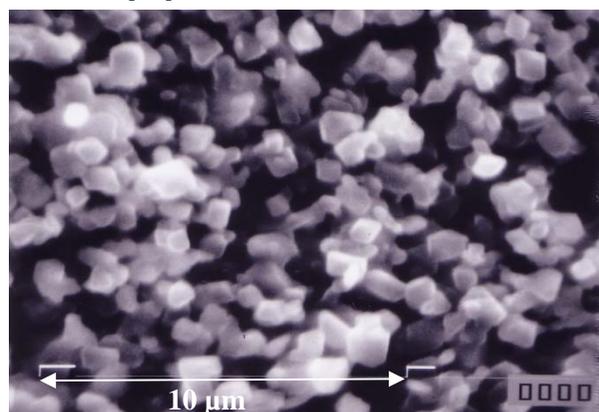


Fig. 7

