—Communication—

Determination of Tritium Species Produced in Neutron-Irradiated LiF-PbF₂ Salt

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Our strong concern in blanket materials of fusion reactor is to compile various data on neutron-irradiated LiF-PbF₂ in which Pb is selected as neutron multiplier under 14 MeV (fusion) neutron irradiation.

In the foregoing works on LiF-PbF₂ salt, the revision of LiF-PbF₂ phase diagram,¹⁾ an acquisition of thermal data of LiF-PbF₂ eutectic melt,²⁾ the specific suggestion for container material for the melt²⁾ and in-situ tritium recovery from neutron-irradiated LiF-PbF₂ melt³⁾ have been extensively elucidated.

Although some chemical forms of tritium recovered from neutron-irradiated LiF-PbF₂ salt, such as T⁰, T⁺ and T⁻, were determined by radio-gas-chromatography,⁴⁾ other tritium chemical form appeared in reports^{3, 6)} was based on the thermodynamic calculation,⁵⁾ not on experiment.

The principal objective of the present paper, therefore, is to determine the chemical form of tritium recovered from

neutron-irradiated LiF-PbF₂ by using a quadrupole mass spectrometer (Q-mass spectrometer: ANELVA Inc., Model AQA-100 MPX). Reagent grade LiF having natural abundance of ⁶Li and PbF₂ were mixed, melted and solidified under helium atmosphere. One gram of the solidified eutectic LiF-PbF₂ salt (60 mole per cent of PbF₂, m.p. 580 °C) was sealed in a silica ampoule, inserted in a vertical irradiation port of TRIGA MARK II at Musashi Institute of Technology and irradiated with a thermal neutron flux of 7×10^{11} n/cm²·sec for 3 minutes at room temperature. The amount of produced tritium was estimated to be about 0.11 Bq. In order to analyze the tritium species, the so-called dry vacuum system composed of a cryosorption pump with a liquid nitrogen cooled cryostat and a turbomolecular pump with a magnetically floated bearing was used for the Q-mass spectrometer. After the opened ampoule was set in the specimen chamber, the chamber was refrigerated with liquid nitrogen in order to hold the produced tritium species in the irradiated LiF-PbF₂ salt. The obtained Q-mass spectrum was shown in Fig. 1(a). The specimen chamber was then heated up to 400°C in order to introduce the released tritium species into the Q-mass spectrometer. Fig. 1(b) shows the Q-mass spectrum containing tritium species. A glance at the figure shows that the peak at the mass number 22 newly appeared, indicating the positive

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Fig.1 Q-mass Spectrum for (a) Irradiated Salt cooled with Liquid Nitrogen and (b) Irradiated Salt heated at 400 °C

evidence for the existence of tritium species, such as TF , T_2O , D_2^{18}O , TD^{17}O , CHT_3 and CD_2T_2 .

The intensity of the peak at the mass number 20 in Fig. 1(a), being the peak of THO formed by the reaction between T_2O and small amount of H_2O in the chamber, is comparable with that of Fig. 1(b). If the peak at the mass number 22 in Fig. 1(b) is that of T_2O formed by the fast reaction such as $2\text{THO} = \text{T}_2\text{O} + \text{H}_2\text{O}$, the intensity of the peak would be comparable with that in Fig. 1(a). The result that the peak at 22, however, increased in the intensity remarkably may exclude the existence of T_2O . Since the natural abundances of ^{18}O and ^{17}O in oxygen show the small values of 0.204 and 0.038 per cent, respectively, the existence of D_2^{18}O and TD^{17}O is improbable.

Furthermore, because the specimen was solidified directly from molten state and the vacuum system with the Q-mass spectrometer used here is oil free, no hydrocarbon such as CHT_3 and CD_2T_2 seem to be in the irradiated LiF-PbF_2 salt. As for the HF species, we should notice that no

peak at the mass number 20 was seen for LiF-PbF_2 salt which was sealed in the ampoule filled with hydrogen gas for one month, indicating that the existence of HF would also unfavour. From the above experimental fact, the release of chemical species F due to the heating of irradiated (that is, damaged) LiF-PbF_2 salt may result in the increase in the peak intensity at the mass number 19 although the definite answer for the dissociation of damaged LiF-PbF_2 salt cannot be given until the further experimental data become available. We, moreover, speculate that the small peak at the mass number 24 in Fig. 1(b) is more likely to be LiOH .

Thus, the peak at the mass number 22 can be attributed to the formation of TF in LiF-PbF_2 salt under neutron irradiation.

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