R&Ds on Li_2TiO_3 Pebble Bed for Test Blanket Module in JAEA

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Abstract In JAEA, the Test Blanket Module (TBM) with water-cooled solid breeder is being developing. This paper presents recent achievements of the research activities for the TBM, focusing on the pebble bed of the tritium breeder materials and tritium behaviour. For the breeder material, the chemical stability of Li_2TiO_3 has been improved by Li_2O additives. In order to analyze the pebble bed behaviour, thermo-mechanical properties of the Li_2TiO_3 pebble bed has been experimentally obtained. In order to verify nuclear properties of the pebble bed, the activation foil method has been proposed and a preliminary experiment has been conducted. For the tritium behaviour, the chemical densified coating method has been well developed and tritium recovery system has been modified taking account of the design change of the TBM.

1. Introduction

In JAEA, the Test Blanket Module (TBM) with water-cooled solid breeder is being developing for in-situ experiments in ITER. Design and test program of the TBM has being decided so that demonstrated engineering technology of the TBM is adequate to operate the DEMO blanket. In the current design of the TBM, pebbles of breeder and multiplier materials are packed into a container to form alternate layers of pebble beds. Lithium titanate (Li_2TiO_3) and Beryllium (Be) are our first candidates. This paper presents the key achievements of the latest research activities on the TBM, which includes development of the chemical stability of Li_2TiO_3, the measurement of thermo-mechanical properties of the Li_2TiO_3 pebble bed, the neutron field measurement in the TBM, the coating method for reduction of tritium permeation and the design of tritium recovery system.

2. Development of Breeder Material

Among the proposed solid breeder materials in fusion reactors, Li_2TiO_3 has been noticed as the most promising candidates because of its good tritium release and its low activation characteristics. Addition of H_2 to inert sweep gas has been proposed for enhancing the release of bred tritium from breeder material. However, Li_2TiO_3 is reduced in the H_2 atmosphere at the operating temperature above 900 K, which will lead to degradation of the tritium release characteristics. The reduction of Ti in Li_2TiO_3 from Ti^{4+} to Ti^{3+} is accompanied by mass decrease due to decrease in oxygen content of Li_2TiO_3. In order to control the mass-change at the time of high temperatures use, development of Li_2TiO_3 that contains excess Li_2O is necessary. In the present study, Li_2TiO_3 (L100) and $Li_{2.1}TiO_{3.05}$ (L105) were prepared by solid state reaction of Li_2CO_3 and TiO_2 powders at 1273 K.

In thermogravimetry at 1273 K, the masses of samples L100 and L105 were found to decrease with time in the hydrogen atmosphere, which was partly due to generation of oxygen defects in the samples, as well as due to evaporation of Li_2O. The calculation result of the molar fraction of oxygen deficient of L100 and L105 as compared to that of the previous work using L95 and L90 [1] is shown in FIG. 1. Sample L105 has fewer oxygen defects than the other kinds of Li_2TiO_3 samples. Sample L105 has the smallest mass of TiO_2 in Li_2TiO_3, so that the order of oxygen defects including the previous data was as follows, L105 < L100 < L95 < L90, which corresponds to the order of the molar ratio Li_2O/TiO_2 of the samples.

The atmosphere-controlled high-temperature mass spectrometry provides the vapor pressure data for samples L100 and L105 under the conditions of the D_2 atmospheres as shown in FIG. 2. The sum of the partial pressures of Li containing species (Li and LiOD) was calculated [2]. Its order under the D_2 atmospheres was as follows:

$P_{Li}^{total}(L105) > P_{Li}^{total}(L100)$ (1093 - 1473 K),

$P_{Li}^{total}(L105) = P_{Li}^{total}(L100)$ (< 1093 K).

The overall results of the present analyses indicate that the Li_2O additives are able to control not only the amount of oxygen defects but also the partial pressures of Li-containing species. Thus, the present study confirms the validity of Li_2O addition to Li_2TiO_3 for developing high-temperature resistant breeder materials.

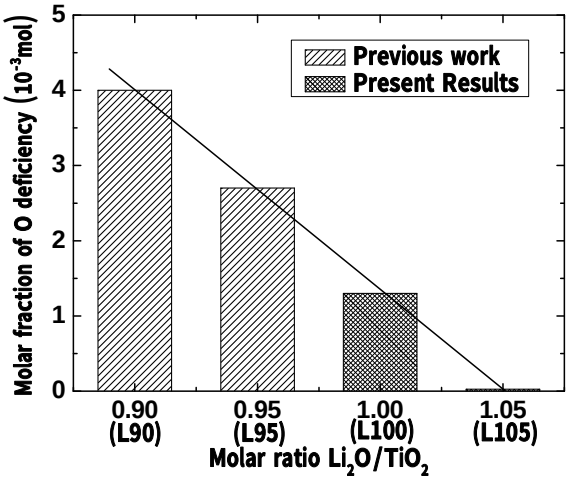
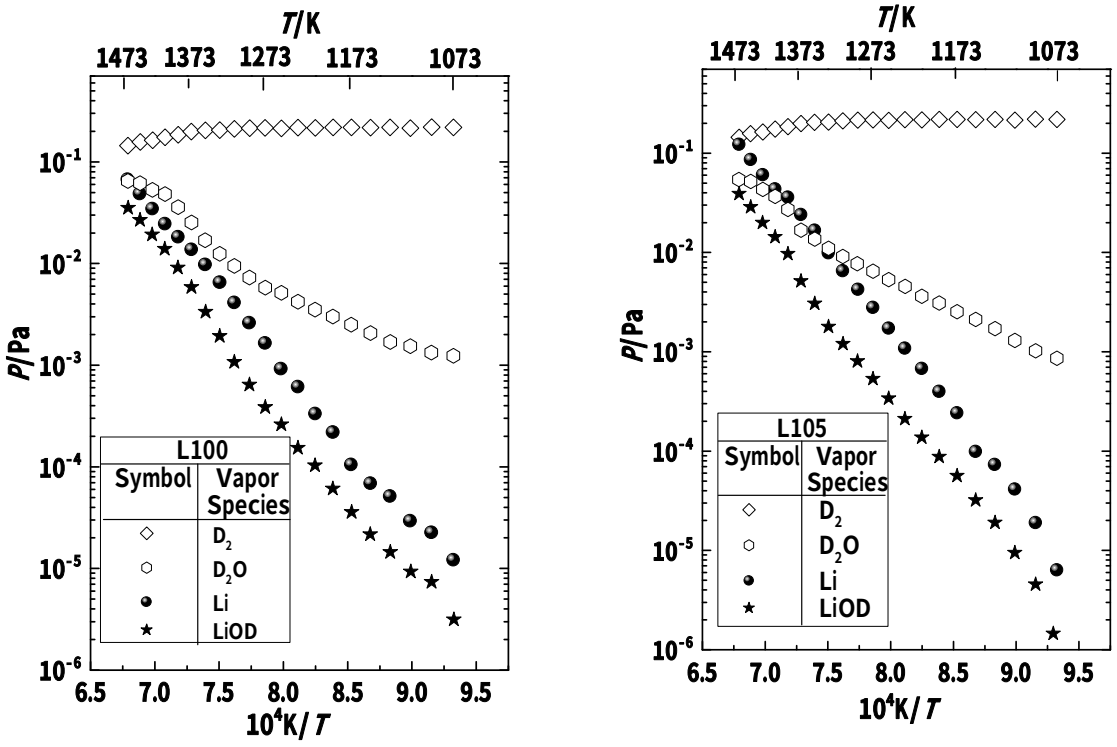


FIG. 1. Molar fraction of O deficiency of Li_2TiO_3 with several molar ratios Li_2O/TiO_2.



(a) L100 *(b) L105*

FIG. 2. Partial pressures of detected gaseous species over Li_2TiO_3 under conditions of D_2 atmospheres.

3. Thermo-Mechanical Characteristics of Li_2TiO_3 Pebble Bed

During operation, thermal stress arises from the temperature distribution in the blanket and difference in thermal expansions between the bed and structural materials. The resulting thermal expansion is the origin of thermal stress and is an important aspect of the thermo-mechanical analysis. Many groups have worldwide been studying thermo-mechanical properties, including the effective thermal conductivity, the heat conductance between the bed

and the wall and stress-strain property. However, there have been few studies of the thermal expansion of a packed bed. Authors investigated the thermal expansion of a Li_2TiO_3 pebble bed in the temperature range from room temperature to about 973 K.

Since external thermal and mechanical load conditions on the bed affect its thermal and mechanical properties, it is important to analyze these properties under controlled conditions. Using the apparatus developed by the authors (FIG. 3), the effective thermo-mechanical properties of the pebble bed, thermal conductivity, stress-strain property and thermal expansion, can be simultaneously measured under regulated temperature, compressive load, and atmospheric conditions. Details of the measurement apparatus were described in previous papers [3, 4].

In the temperature range from room temperature to about 973 K, the thermal expansion of Li_2TiO_3 pebble beds was measured under a compressive load of 0.1 MPa. For beds with different packing factors and loading histories, the average thermal expansion coefficient was $(1.4\pm0.2)\times10^{-5}$ K^{-1} as shown in FIG. 4. Here, the packing factor is defined as the volumetric ratio of packed pebbles to the container. In the present conditions, the thermal expansion coefficient of the pebble bed is equal to 78% of that for the bulk material. During these test campaigns, we found that a residual deformation in a preloaded pebble bed can be annealed when the pebble bed is heated without the load. This behaviour of the pebble bed can be explained by taking into account the progress of pebble compaction and its relaxation [4].

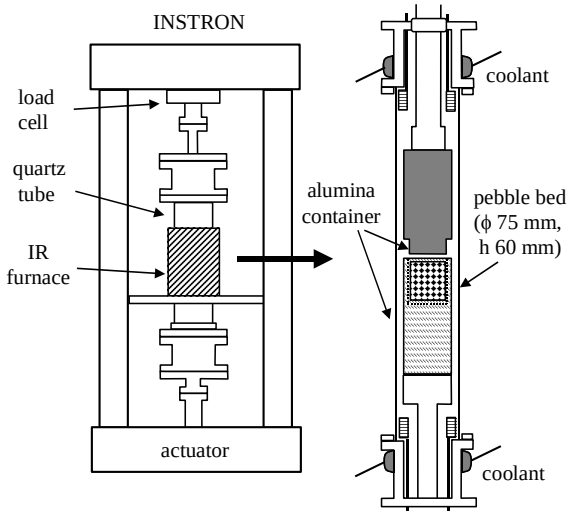


FIG. 3. Schematic diagram of apparatus for measurement of pebble bed properties.

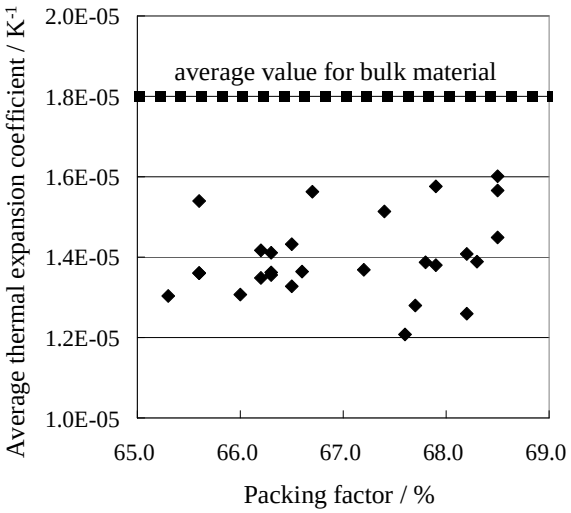


FIG. 4. Average thermal expansion coefficient of Li_2TiO_3 pebble beds.

3. Measurement of Neutron Field

Neutron field measurement is a key issue to evaluate the nuclear performances of the TBM. Because the inside of the TBM will be high temperature, neutron measurement systems that remain unaffected by high temperature are required. Therefore, JAEA has proposed a multi-foils activation method as the neutron measurement system in the TBM.

JAEA/FNS group has investigated the availability of the multi-foils activation method with DT neutron experiment. Figure 5 shows the experimental arrangement of the multi-foils activation method. The foils were inserted at the points of 0, 50.8, 101.6 and 152.4 mm in depth of the beryllium assembly and irradiated with DT neutrons. After suit cooling term,

authors measured gamma rays from $^{27}Al(n,\alpha)^{24}Na$, $^{48}Ti(n,p)^{48}Sc$, $^{58}Ni(n,p)^{58}Co$, $^{90}Zr(n,2n)^{89}Zr$, $^{93}Nb(n,2n)^{92m}Nb$ $^{115}In(n,n')^{115m}In$ and $^{197}Au(n,\gamma)^{198}Au$ reactions with a germanium detector, and each reaction rates were deduced. The neutron flux was estimated with the measured reaction rates and an initial guess neutron flux. The initial guess neutron flux was calculated with Monte Carlo calculation code (MCNP4C) and a nuclear data library (FENDL-2.1). JENDL Dosimetry file 99 was also used as the response function of the reaction rates.

Figure 6 shows obtained neutron flux at the point of 152.4 mm depth in the beryllium assembly. From our estimation, it was shown that the 14 MeV neutron flux at all measured point corresponds to the initial guess profile. However, estimated neutron flux between 3 MeV and 6 MeV had underestimation. The neutron flux at the energy range of 1 MeV to 2 MeV tended to overestimate. Same tendency also existed at other points with different depth. From the results, it is thought that the evaluation method and dosimetry data should be verified precisely. The calculation per experiment (C/E) of $^{197}Au(n,\gamma)^{198}Au$ reaction rate was near 1.2 and the estimated neutron flux below 1 eV showed to reflect the C/E.

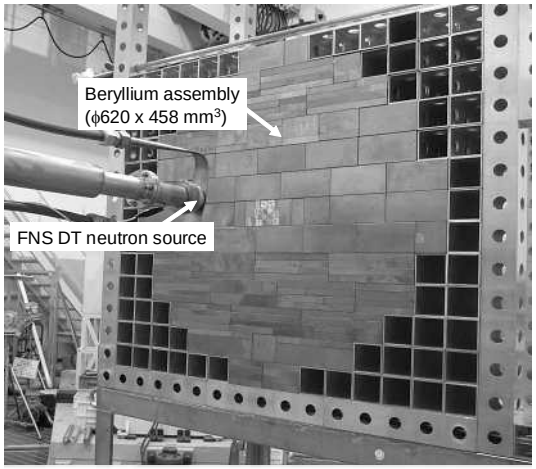


FIG. 5. Experimental arrangement for multi-foils activation method.

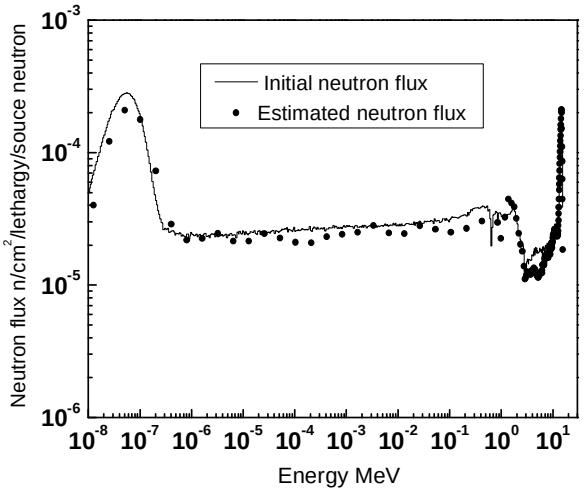


FIG. 6. Estimated neutron flux at the point of 152.4 mm depth in beryllium assembly.

4. Tritium Permeation Analysis

An effective coating for reduction of tritium permeation was developed by a chemical densified coating (CDC) method [5] to impregnate amorphous material in the coating. Compared with other coating methods, plasma spraying, pack-cementation and vapor-deposition techniques, for example, the CDC method has some advantage such as a capability of the coating on the outer and/or the inner surface of a tube or a container.

Figure 7 shows cross-sectional view of the obtained CDC coating of Cr_2O_3-SiO_2 including $CrPO_4$. The area around SiO_2 granules is filled with Cr_2O_3, and thin $CrPO_4$ layer about 2 μm thick is formed on the surface of the coating. It was confirmed from tritium permeation experiments that the permeation reduction factor of F82H steel with this coating reaches about 300 at 873 K [6]. Deuterium diffusion characteristic in the coating was studied by use of the nuclear reaction analysis [7]. Details of the experiment and analysis are given elsewhere [8]. To interpret the results, the diffusion coefficient of deuterium in this coating was calculated according to the following equation,

$D = 1.1\times10^{-10}\ \exp(-0.71\ \text{eV}/kT)$,

where D, k and T are diffusion coefficient (m^2/s), Boltzmann coefficient (eV/K) and temperature (K), respectively. The obtained diffusion coefficient is found to be smaller than that of SS304 by five orders of magnitude. This means that an effective diffusion barrier can be expected if this coating is used in the coolant tube in the TBM.

By using the obtained experimental diffusion properties, a preliminary tritium permeation analysis was carried out for the TBM using the Tritium Migration Analysis Program (TMAP) code [9]. The analysis result of the diffusion flux and the concentration of tritium into the cooling water are shown in FIG. 8 as a function of the elapsed time from the start of the plasma burning and the sweep gas flow [10]. The calculation results indicate that tritium permeation through the F82H cooling pipe with the CDC coating will be four orders of magnitude smaller than that without the coating.

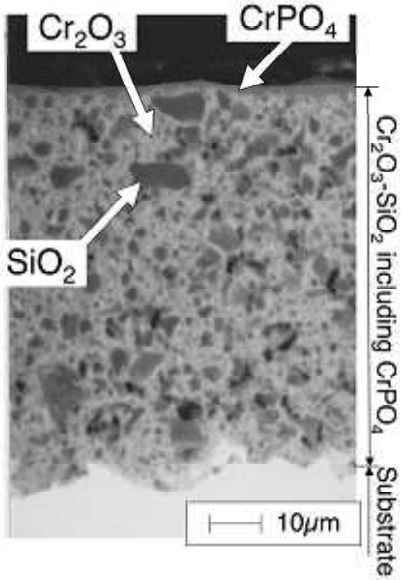


FIG. 7. Cross-sectional view of coating of Cr_2O_3-SiO_2 including $CrPO_4$ formed by CDC method.

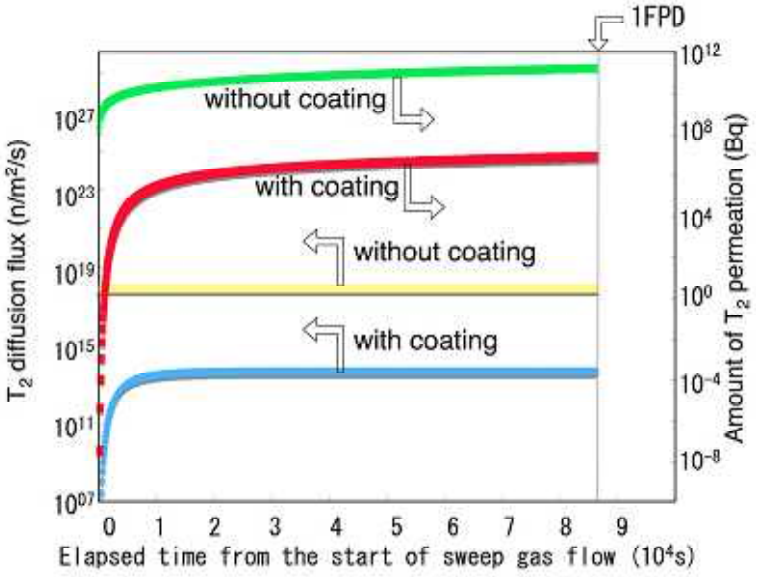


FIG. 8. Analysis result of diffusion flux and concentration of tritium into cooling water through F82H steel.

5. Tritium Recovery System

Figure 9 shows a schematic diagram of tritium recovery system (TRS) for the TBM. The main components of TRS are the dryers and the cryogenic molecular sieves beds (CMSB). The dryer and CMSB are used in the batch process. Therefore, two dryers and three CMSBs are prepared and are used one by one for the continuous processing. The palladium membrane diffuser (PD) is also an important component for the processing of the release gas from CMSB at the regeneration step. At first, the sweep gas out from the TBM is introduced to the dryer to remove the water form of tritium. Then, the sweep gas in which water vapor has been removed is sent to CMSB to remove other chemical forms of tritium (hydrogen and maybe organic forms), and then, the gas is returned to the TBM again. The roles of TRS in the TBM test plan are the recovery of the bred tritium from the sweep gas and the quantification of the recovered tritium. Therefore, the dryer and CMSB have a regeneration system. In the regeneration step, the dryer is heated up to 573 K, and the inert gas is passed through the dryer. This gas is sent to the condenser to collect water as liquid. Tritium concentration of this water is measured by a liquid scintillation counter or a calorimeter. In the case of CMSB, the bed is heated up to the room temperature after the liquid nitrogen draining. The released gas

from CMSB is circulated between CMSB and PD. From the viewpoint of the safety, the suppression tank is attached to this system. In the PD operated at 673 K, only hydrogen permeates through the palladium membrane and is collected in the measuring tank. The amount of tritium is measured by PVTC (pressure, volume, temperature, concentration) method. In accordance with recent design change of the TBM where the sweep gas flow rate decreases to 34.7 l/min (STP), the size of each component has been revised. In the current design, CMSB has become almost same size as the one that was used for the interlinked test with the main fuel cycle system [11]. The lithium hydroxide trap has been removed because of the doubt for the effectiveness. The numbers indicated in FIG. 9 are the measuring points of the analysis and measurement system (AMS) for the TBM. The equipments used in AMS are listed in Table 1. Figure 10 shows a schematic diagram of the effluent gas measurement system for the TBM. The effluent tritium from TRS and AMS are collected to the low pressure measuring tank via manifolds, and are measured periodically by PVTC method, and then, the collected gases are sent to the tritium plant of ITER. Finally, the whole gaseous tritium is measured here.

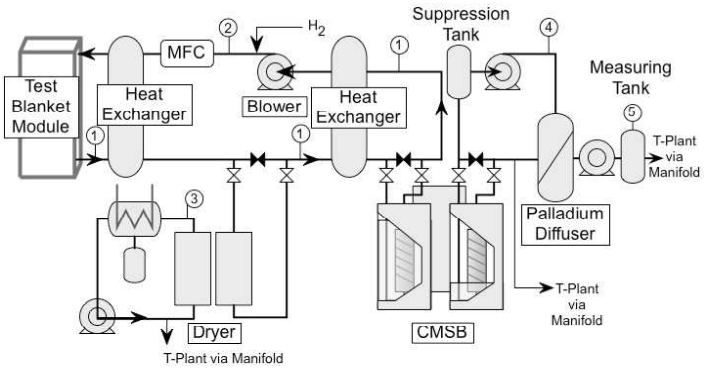


FIG. 9. Schematic diagram of tritium recovery system.

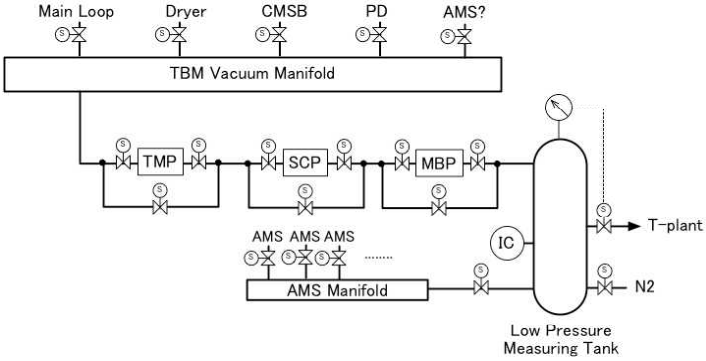


FIG. 10. Schematic diagram of effluent gas measurement system.

The TBM design is not fixed and the design of TRS has to be taking into account the operation scenario of ITER. Further design improvement is necessary hereafter.

TABLE 1: Equipment list of analysis and measurement system for TBM.

No.	Role	Equipment
1	Online Monitor	Ion Chamber / Hygrometer / Pressure Gauge
	Sampling Monitor	MicroGC / Ion Chamber / Mass Flow Controller / Metal Bellows Pump / Pressure Gauge
2	Sampling Monitor (H_2)	MicroGC
3	Online Monitor	Ion Chamber / Hygrometer / Pressure Gauge
4	Online Monitor	Ion Chamber / Hygrometer / Pressure Gauge
	Residual Gas Analysis	Mass Spectrometer
5	Tritium Measurement	Ion Chamber / Pressure Gauge

6. Conclusions

The achievements of key technology for ITER TBM in JAEA can be summarized as follows.

(1) The chemical stability of Li_2TiO_3 was improved by Li_2O additives, which enable us to use tritium breeder at higher temperature.

(2) The average thermal expansion coefficient of the Li_2TiO_3 pebble bed was experimentally obtained.
(3) The activation foil method was proposed to verify nuclear properties of the TBM and the preliminary experimental results were obtained.
(4) The chemical densified coating method was well developed on to the F82H steel and high permeation reduction factor was obtained.
(5) The design of tritium recovery system was developed taking account of the design change of the TBM.

These achievements in the elemental technologies is being integrated to establish detailed design and test program of the TBM, where demonstrated engineering technology is adequate to operate the DEMO blanket.

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