

Recent Studies of Production and Purification of Tritium in JAERI

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Using a little modified facility for producing tritium in 40 TBq per batch, 21 TBq and 30 TBq of gaseous tritium for utilization to μ -catalyzed fusion were extracted from neutron-irradiated ⁶Li-Al alloy targets by heating at 500 °C under vacuum and collected in a uranium getter. The recovery yields of tritium were about 100%. Through the production run, no leakage of tritium from the facility was observed. In order to use the tritium gas in the μ -catalyzed fusion experiments, hydrogen (H) in the tritium gas should be removed. Therefore, the tritium gas obtained was purified with an improved gas chromatograph, which enabled handling of about 5 TBq of tritium gas per batch using a separation column of 10 mm ϕ \times 9 m under the He flow of 300 ml/min. The old column had been 6 m in length and the flow rate 100 ml/min. About 60 TBq of tritium gas in a high purity was effectively obtained by the batch processing with the improved gas chromatograph. About 50 TBq of the purified tritium gas was transported to RIKEN-RAL Branch in England to be used for muon catalyzed fusion (μ CF) experiments. At the Branch, a tritium handling system (THS) was installed for removing ³He of a decay product of tritium. Before the fusion experiments, performances of U-getters, Ti-getters, Pd-filter, and all other parts were tested and the fusion experiments have been carried out.

1. Introduction

In the Japan Atomic Energy Research Institute, production of tritium from neutron-irradiated ⁶Li-Al alloy targets has been developed [1] and continued in 40 TBq level. Recently, the vacuum furnace in the facility was renewed. Using the facility, including the furnace, about 50 TBq of tritium to be utilized for μ -catalyzed fusion (μ CF) was produced in two runs. In this report, the new vacuum furnace has been outlined and the results of the production run described.

The products of tritium obtained in runs involve an inevitable impurity of hydrogen (H). Therefore, the product has to be purified isotopically when the product is used as a fuel material of μ CF. In order to purify the product, gas chromatography has been applied in JAERI [2]. The system was modified prior to purification in order to obtain the tritium more effectively; the column length was changed from 6 m to 9 m and the carrier-gas flow from 100 ml/min to 300 ml/min, respectively. The results obtained with the system are described in this paper.

About 50 TBq of the purified tritium gas was transferred to RIKEN-RAL (Rutherford Appleton Laboratory) Branch in England for μ CF experiments. For the experiments, a tritium handling system (THS), where a decay product of ^3He could be removed easily, was constructed at RAL. This paper describes the TRS and some results of the performance tests.

2. Experiments and Results

2.1 Production of tritium

A sketch of the renewed vacuum furnace is shown in Fig. 1. The vacuum furnace, which is made of stainless steel (316-ss), has a triple wall; it also has an argon and water jacket. A molybdenum heater is set in the innermost vessel of the vacuum furnace to heat the targets effectively. The main sections of the tritium production facility are a tritium extraction system (TRES) and a tritium removal system (TRS). The TRES consists of a vacuum furnace, two getter cartridges containing pulverized uranium, a circulation pump, a set of vacuum pumps and a tritium removal apparatus. These components are installed in an airtight lead cell or glove boxes.

In JAERI, tritium has been produced by irradiating ^6Li -Al alloy target with neutrons. The lithium content (^6Li enriched to 95.5%) of the ^6Li -Al alloy target is 3.2%. The sizes of the meat of the alloy are 80 mm long, 14 mm wide and 0.5 mm thick, and those of the aluminum sheath 100 mm, 20 mm and 2 mm, respectively. The ^6Li -Al alloy targets enclosed in aluminum capsules were irradiated for 60-70 days in the Japan Material Testing Reactor (JMTR) at a neutron flux of $2 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$. The targets were left aside for 10 years to lessen radioactive nuclei such as ^{65}Zn and ^{59}Fe induced there. Prior to the extraction of tritium, the amount of tritium in each target was measured by means of calorimetry to be 2-3 TBq. About 30% of ^6Li in the target was changed to tritium. Two sets of twelve targets containing 21 TBq and 30 TBq were used separately in each production run.

The targets were put in a stainless steel crucible. In the vacuum furnace, the targets were heated up to 700°C , while the pressure in the vacuum furnace was measured. The tritium gas released together with helium

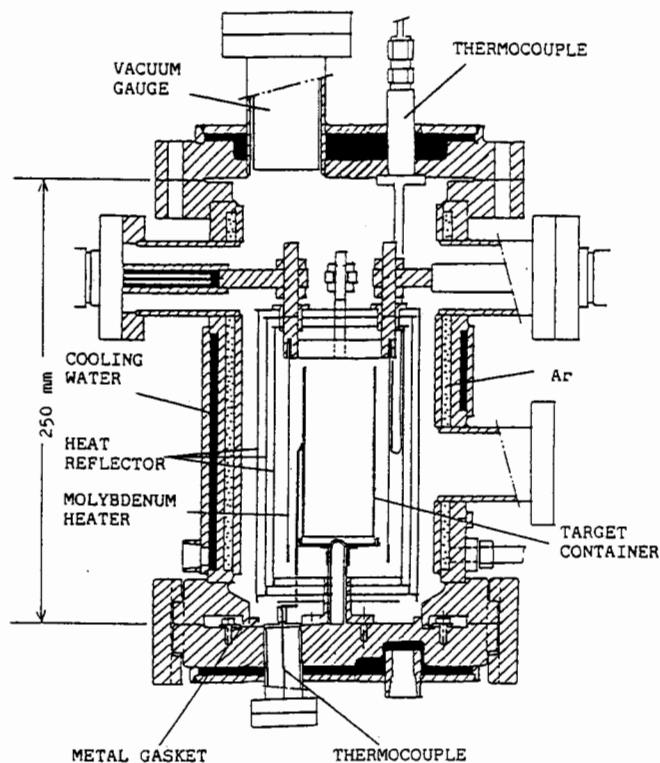


Fig. 1 A Vacuum Furnace

in 21 TBq run was gas-chromatographed as shown in Fig. 2, where the upper chromatogram was obtained by detecting radioactivity of tritium with an ionization chamber and the lower one by detecting hydrogen species with a thermal conductivity detector. Fig. 2 shows that the isotopical purity of tritium gas was 68.7%. The gas was circulated through the U-getter with the circulation pump to be trapped in the uranium at a room temperature. The trapping step was continued until more than 99.99% of tritium released was collected in U-getter.

2.2 Purification of tritium

As the tritium gas produced above contained an isotopic impurity of hydrogen (H), the tritium gas had to be purified isotopically in order to be used for μ CF experiments. The crude tritium gas which was also contaminated with deuterium (D)

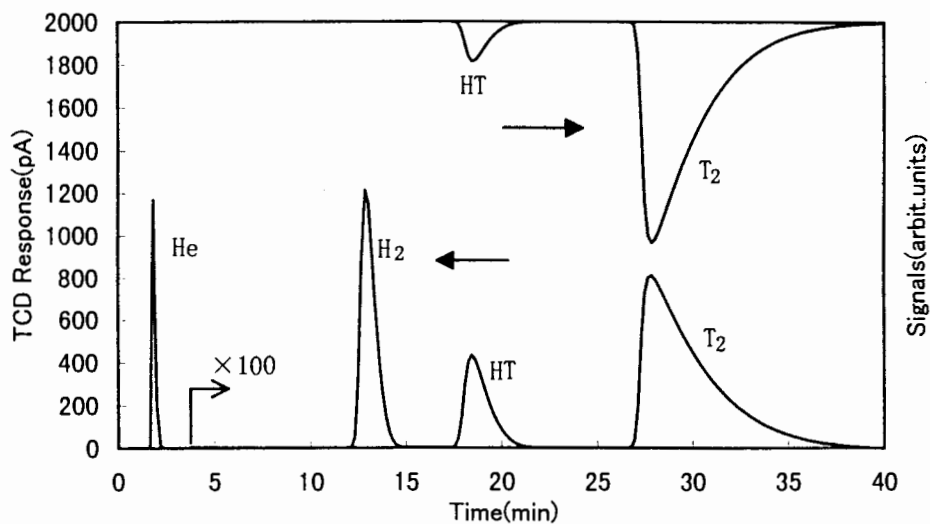


Fig. 2 A Gas Chromatogram of Tritium Gas Extracted from Target

was purified from hydrogen (H) with the purification system modified. We had already purified another crude tritium gas with the previous system [2]. In order to purify the present crude tritium gas more effectively, a separation column was enlarged from 6 m to 9 m and the flow rate of carrier gas was changed from 100 ml/min to 300 ml/min. The system finally assembled consists of a sampler (100 ml in volume), a separation column (10mm ϕ x 9 m) packed with 3 wt% MnCl₂-coated alumina adsorbents (60/80 mesh), a thermal conductivity detector, a specially designed ionization chamber (IC, 2.7 mL in volume), T₂- and HT-trap columns

(10mm ϕ x 3 m) packed with molecular sieve 5A, and two uranium getters. The exhaust of the system was connected to TRS. A portion of the crude T₂ gas, which had been stored in a U-getter of the TREX, was charged to the separation column immersed in liquid nitrogen with helium carrier gas at the flow rate of 300 mL/min. Fig. 3 shows the chromatograms detected by both the TCD and the IC. The appearance of the signals of HD, D₂ and DT is due to contamination with D₂ gas. The H₂ fraction was discarded through the TRS. The DT and T₂ fractions were collected in the cold molecular-sieve column and then stored in the uranium

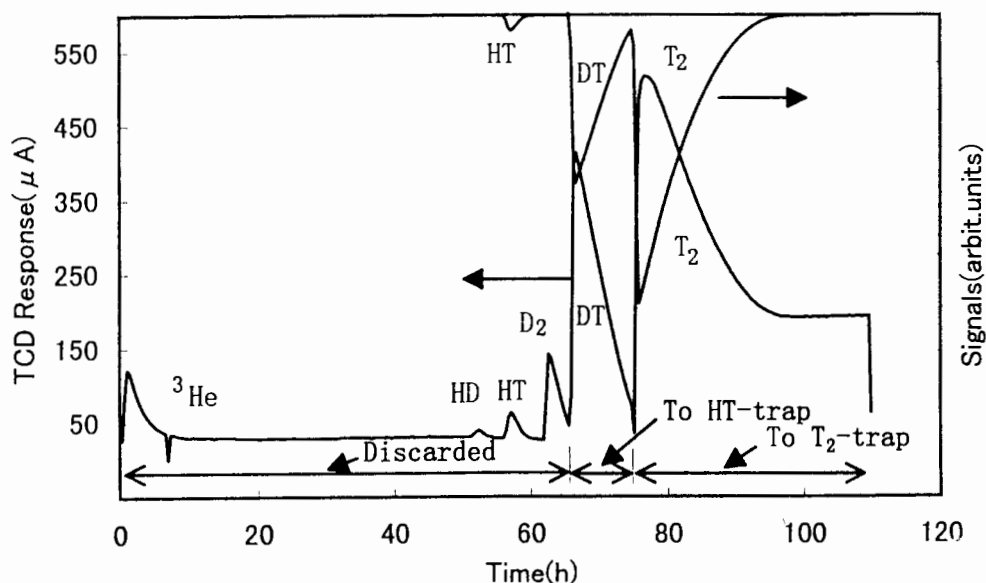


Fig. 3 A Gas Chromatogram of Tritium Gas Separated with Column

Table 1 Isotopic compositions of tritium gas and amount of tritium shipped to RAL

	H ₂	HD	HT	D ₂	DT	T ₂	Amount of T
Cylinder-1	0.00 %	0.00	2.54	0.00	1.76	95.7	27.4 TBq
Cylinder-2	0.05 %	0.22	2.84	0.54	12.9	82.7	26.4 TBq

getter. By a few tens of runs of above-mentioned processing, was first provided about 30 TBq of tritium gas including deuterium (D). The tritium gas was packed into cylinder-1 after the purity of tritium gas was determined. Another purification also provided about 25 TBq of tritium gas and the tritium gas was introduced into cylinder-2. The compositions of hydrogen gases in the cylinders were tabulated in Table 1 and the impurities of hydrogen (H) in D-T mixed gas were 1.27% (cylinder-1) and 1.67% (cylinder-2).

About 50 TBq of the tritium was carried to RAL for utilizing to μ CF experiments.

2.3 Utilization of tritium

As the μ CF experiments, Nagamine et al. have determined the α -sticking probability in a muon-catalyzed fusion (μ CF) cycle in the previous paper [3]. Further detailed experiments at RAL need extremely pure D-T gas in which the content of ³He, the decay product of tritium, is as low as possible.

To meet this requirement, we had developed a technique for preparing highly pure tritium gas and a tritium handling system (THS) was constructed at RAL. A schematic of the system is shown in Fig. 4.

The system consists mainly of T₂ cylinders (mentioned above), U-getters, Ti-getters, Pd-filter, a cryotrap, a cryopump, a radiogas chromatograph (Shimadzu Co. Ltd.) and a D-T target chamber. The T₂ cylinders are made of stainless steel with a volume about 1 L. The U-getters can store tritium, and Ti-getters can trap tritium released from both the vacuum system and the gas chromatograph. To remove ³He from tritium gas, a Pd-filter was furnished. A cryotrap and a cryopump were set to trap tritium gas temporarily. The mixed gases of tritium and deuterium are analyzed isotopically with the radiogas chromatograph and the amount of ³He in the mixed gas is determined with the chromatograph. The target chamber of 2cm³ was connected to the liquid He cryostat in order to liquefy the mixed gas. It has a Be window of 0.5 mm thick and 15 mm in diameter

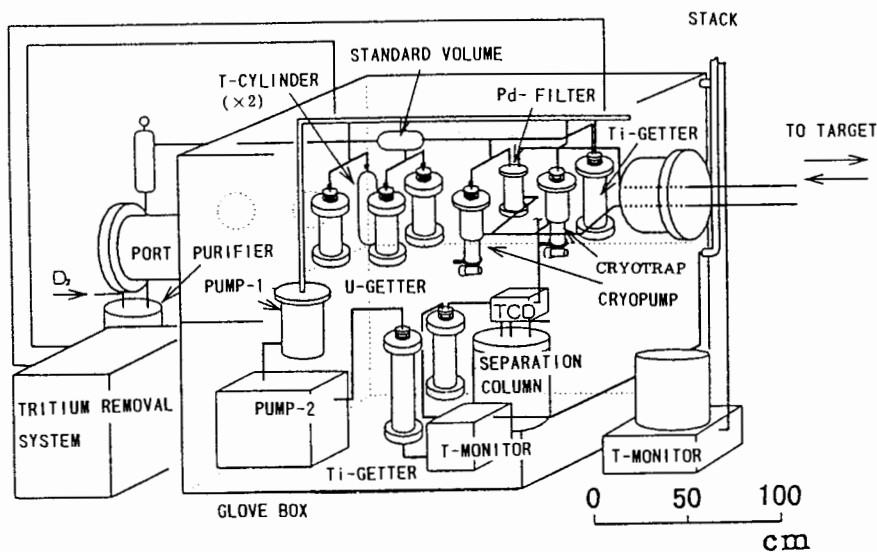


Fig. 4 A Schematic of THS

which μ ions can be introduced through. The THS equipments, except the target chamber, were installed in a glove box. The atmosphere in the glove box is replaced by Ar gas and the pressure kept over the normal one. A tritium clean-up system employing Zr-Fe getters was connected to the glove box to remove tritium gas in the Ar atmosphere.

The various operation tests for major components of U-getters, Ti-getters and a Pd-filter in the THS were conducted with and without tritium gas. And as a final test, the standard procedures described below were checked with 3 TBq and 30 TBq of tritium.

- (1) Firstly, transferring of tritium gas in the cylinder to the constant volume.
- (2) Trapping of tritium gas with U-getter at ambient temperature and evacuating of ^3He .
- (3) Transferring tritium gas to cryotrap through Pd-filter heated at 450°C .
- (4) Transferring tritium gas from the cryotrap to the target chamber.
- (5) Finally, transferring tritium gas from the target chamber to the U-getter at ambient temperature.

Through these tests, the whole experimental system was confirmed to be applicable to μCF experiments. The μCF experiments were begun on a full scale from this June.

3. Conclusion

In order to supply pure tritium gas for μCF experiments, 60 TBq of tritium was extracted from neutron-irradiated $^6\text{Li-Al}$ alloy targets and purified isotopically with an improved gas chromatograph. About 50 TBq of purified tritium gas was exported to RIKEN-RAL Branch in England. A tritium handling system (THS) was installed at the Branch. Main components in the THS were tested with tritium on their performances and the μCF experiments have been begun.

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