

## Comparison of release behavior of water vapor and tritiated water vapor from natural soil by heating



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### ABSTRACT

In a water-cooled nuclear fusion power plant, a large amount of tritiated water will be handled. Assuming severe accidents including the release of tritiated water to the surrounding environment, it is important to understand the tritium behavior in natural soil. In this study, two samples of natural soil were collected from different places in the Hakozaki campus of Kyushu University and the release behavior of water vapor and that of tritium by heating to 1000°C were separately investigated and compared. Three release peaks of water vapor appeared at 120, 270 and 450°C for both samples. Also, three release peaks of tritium appeared at the approximately same temperature as water vapor release from both samples of soil immersed in tritiated water. The chemical form of released tritium was mainly tritiated water vapor. These results suggest that tritium sorbed in soil during immersion in tritiated water exists with water originally contained in the soil. The amount of tritium released from each sample soil by heating to 1000°C was estimated to be only about 15% of that expected to be sorbed in the soil. Tritium remaining in the soil after heating is presumed to be retained in structural water stably present in clay minerals constituting natural soil.

## 1. Introduction

In a water-cooled fusion power plant, huge amount of water will contain tritium because hydrogen isotopes can permeate metal walls at high temperature conditions. Additionally, a certain amount of tritium leaking into the air in the buildings of the plant is continuously recovered as tritiated water by an air clean up system. Therefore, a large amount of tritium is handled as tritiated water in the plant. Here, tritiated water means the water containing tritium such as T<sub>2</sub>O, HTO or DTO. Assuming a severe accident such as tritiated water spills out from the tritium handling buildings, it is important to understand the tritium behavior in the surrounding environment.

In the previous study, the present authors performed the percolation experiment of tritiated water, which was a mixed solution of HTO and H<sub>2</sub>O, through a soil packed bed, focusing on tritium behavior in natural soil [1]. It was found that tritium is trapped in the soil during percolation process and the majority of trapped tritium cannot be recovered by water soaking [2]. Generally, it is said that clay minerals constituting natural soil contain three kinds of water such as adsorbed water, interlayer water and structural water. When soil is exposed to tritiated water, tritium is speculated to be retained in each water compartment. However, the relationship between tritium trapped in

soil and water originally contained in soil has not been understood sufficiently to date.

In the present study, two samples of natural soil were separately heated to 1000°C under dry Ar gas flow and the release behavior of water vapor was investigated. Also, the samples were separately immersed in tritiated water and the release behavior of tritium from each sample was investigated by heating similarly. From the comparison of the release curves of water vapor and tritium during elevating temperature, tritium sorption and desorption in natural soil exposed to tritiated water were discussed.

## 2. Experimental

### 2.1. Samples

Samples of soil used in this study were collected from the Hakozaki campus of Kyushu University. These samples were equal to sample 2 and sample 5 in the previous study reported by Honda et al. [1]. Sample 2 was collected from a grove in the garden beside main gate and it named “Soil 2”. Sample 5 was collected from a grove in the garden beside the center of clinical psychology and it was named “Soil 5”.

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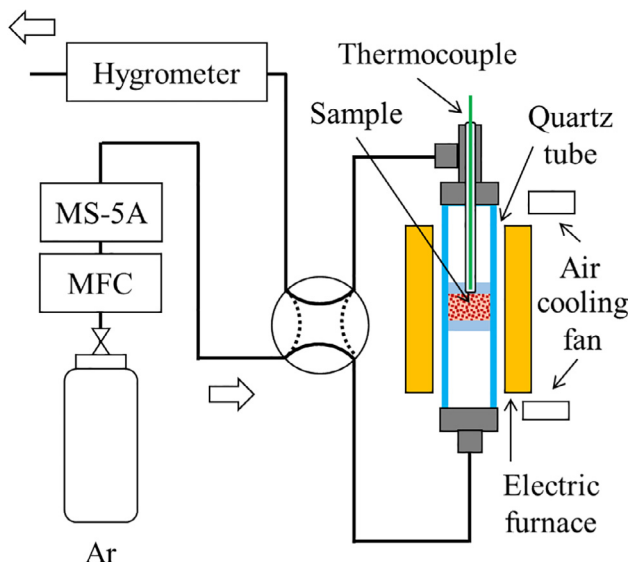


Fig. 1. Experimental apparatus for water vapor release.

2.2. Water vapor release experiment

The experimental apparatus for observation of water vapor release behavior is shown in Fig. 1. Soil 2 of 0.54 g and Soil 5 of 0.78 g were separately packed in a quartz tube, 400 mm in length and 8 mm in inner diameter. The soil packed bed was fixed at the center of the quartz tube by quartz wool. First, Ar gas was passed through the soil packed bed at room temperature for about 18 h to purge adsorbed water and then heated to 1000°C with a ramping rate of 5°C/min by an electric furnace. Then, the temperature of 1000°C was kept for 2 h. The temperature was controlled by a thermocouple inserted into a quartz tube with outer diameter of 3 mm which was inserted at the top of the soil packed bed. The Ar gas flow rate was controlled to be 300 cc/min by a mass flow controller. The impurity water vapor in the Ar gas was excluded by an adsorbents (MS-5A) bed. The concentration of released water vapor was measured by a hygrometer (QMA2030, Michell Instruments). To

stabilize the hygrometer, the Ar gas was input to the hygrometer for 1 h by bypassing the soil packed bed before the experiment.

To investigate an influence of water absorption on the release behavior of water vapor, Soil 5 was immersed in water for 2 days. Then the wet sample of 1.06 g was packed in the quartz tube and the water vapor release experiment was carried out.

2.3. Tritium release experiment

Soil 2 of 39.60 g and Soil 5 of 40.24 g were separately put in polyethylene containers. The tritiated water constituted of HTO and H<sub>2</sub>O was poured in each container. The weight of poured tritiated water was 40.46 g for Soil 2 and 42.18 g for Soil 5. These values were calculated by subtracting the weights of empty container and sample soil from the total weight of container containing sample soil and tritiated water. The tritium concentration in tritiated water just after pouring and mixing was 305 kBq/g for Soil 2 and 307 kBq/g for Soil 5. Then, the containers were stored at room temperature. The total weight of the container and the tritium concentration in tritiated water were measured some times during storage. A small amount of tritiated water was sampled from the container and its tritium concentration was measured by a liquid scintillation counter (LSC-5100, Aloka). Final measurement of tritium concentration was performed just before the tritium release experiment. The amount of tritium sorbed in the soil was estimated from the reduction of tritium concentration, considering the disappearance of tritium due to radioactive decay, the extraction of tritium due to the sampling of tritiated water to measure the tritium concentration, and the loss of tritiated water due to the leakage during storage and due to the failure in sampling operation.

The experimental apparatus for observation of tritiated water vapor release behavior is shown in Fig. 2. The wet samples of soil were scooped up from tritiated water in each container and separately packed in a quartz tube of the same size as used in the water vapor release experiment. The soil packed bed was fixed at the center of the quartz tube by quartz wool. A dry Ar gas and a wet Ar gas were used in this experiment. The dry Ar gas was formed by passing the gas into the adsorbent (MS-5A) bed, which can adsorb trace water vapor, at 100 cc/min. The wet Ar gas was formed by passing 1% H<sub>2</sub>/Ar at 80 cc/min and 40% O<sub>2</sub>/Ar at 20 cc/min through a Pt catalyst (Pt-SDB) bed. Because

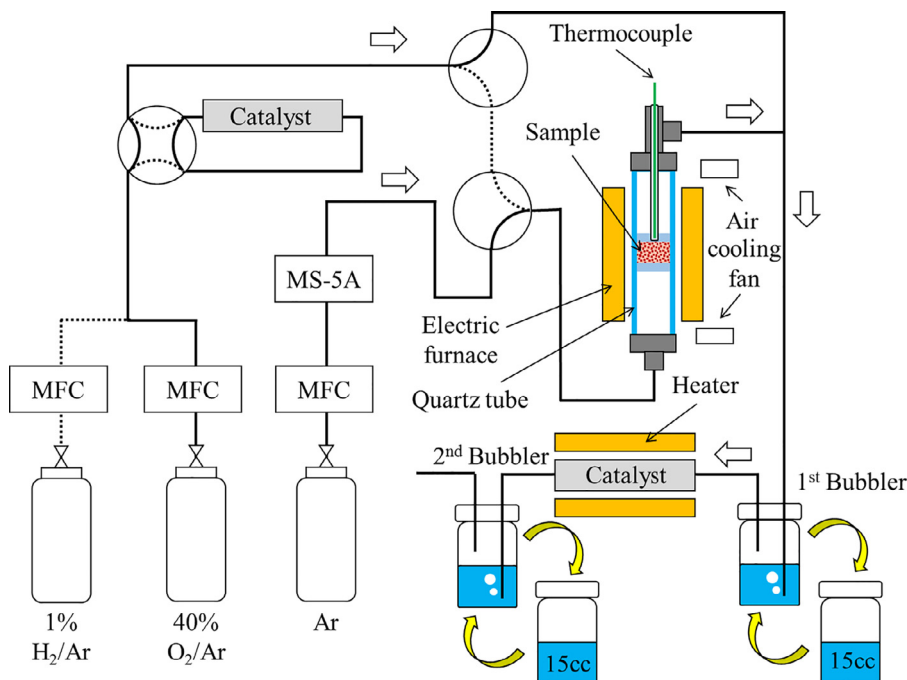


Fig. 2. Experimental apparatus for tritiated water vapor release.

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