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Penetration of tritiated water vapor through hydrophobic paints for concrete materials

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ABSTRACT

Behavior of tritium transfer through hydrophobic paints of epoxy and acrylic-silicon resin was investigated experimentally. The amounts of tritium permeating through their paint membranes were measured under the HTO concentration condition of 2–96 Bq/cm^3. Most of tritium permeated through the paints as a molecular form of HTO at room temperature. The rate of tritium permeating through the acrylic-silicon paint was correlated in terms of a linear sorption/release model, and that through the epoxy paint was controlled by a diffusion model. Although effective diffusivity estimated by a diffusion model was obtained 1.1×10^{-13}–1.8×10^{-13} m^2/s for epoxy membranes at the temperature of 21–26 °C, its value was found to be hundreds times larger than that for cement-paste coated with epoxy paint. Hence, resistance of tritium diffusion through interface between cement-paste and the epoxy paint was considered to be the most effective in the overall tritium transfer process. Clarification of tritium transfer behavior at the interface is important to understand the mechanism of tritium transfer in concrete walls coated with various paints.

1. Introduction

Behavior of tritium transfer in concrete materials has been investigated in order to obtain fundamental data for contamination and decontamination of radioactive tritium in the case where a large amount of tritium leaks through tritium handling facilities and buildings of fusion reactors [1–4]. It is important to understand tritium behavior not only in concrete materials but also in hydrophobic paint coating for estimation of tritium contamination and decontamination. Most of previous studies on hydrophobic paints focused on performance of tritium penetration barriers, but not on phenomena of tritium transport through various paints into concrete structures. Although several data for concrete materials are reported on tritium adsorption and desorption on various hydrophobic paints, such as epoxy resin paint, silicic resin paint, acrylic resin paint, fluoric resin paint and so on [5–7], the amount of data on tritium transfer rates of their paints is limited. Unfortunately, there is no paint which is able to prevent tritium from penetrating completely. It is inevitable that tritium penetrates through concrete walls and contaminates the walls in case of accidental tritium release inside concrete buildings which walls are coated with a hydrophobic paint. However, it is possible to delay tritium penetration because a tritium penetration rate through hydrophobic paint is lower than that through a concrete material [4,8–10].

In our previous studies, we investigated the amounts of tritium trapped in concrete pellets coated with several kinds of paints and the rates of tritium penetration and release [8–10] by means of a tritium exposure method and a water dissolution method. Their concrete pellets coated with various paints of 0.1 mm in thickness were exposed to tritium water vapor (HTO) atmosphere, and HTO penetration and release from the pellets were measured by soaking into distilled water. The previous studies showed that epoxy resin paint was the most effective to prevent tritium from penetrating through concrete pellets, although the paint comparatively trapped a large amount of tritium in itself for two months of HTO exposure period. We have obtained overall behavior of tritium transfer in a conjugated system composed of concrete materials and paints coating. However, elementary processes such as tritium diffusion in hydrophobic paints, tritium transfer at an interface between a concrete material and paint and surface reactions have not been understand clearly yet.

Therefore, we measured tritium transfer rates through hydrophobic paints by means of an unsteady-state permeation method, which makes it possible to measure permeation rates through thin membranes accurately. Discussion is made based on

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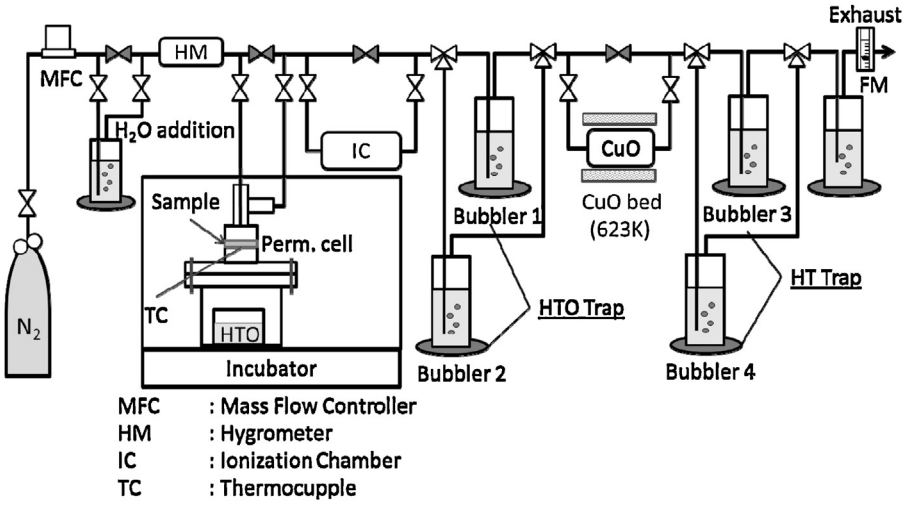

Fig. 1. Experimental apparatus of tritium permeation through a paint membrane.

our analysis of numerical calculation and experiment that were given in this paper.

2. Experimental

2.1. Preparation of paint membrane samples

Two kinds of paints, an epoxy resin paint (2-part Epoxy Waterproof, Asahipen Co., Japan) and an acrylic-silicon resin paint (Water Base Supercoat, Asahipen Co., Japan) were selected for comparison with our previous experiments [8–10]. The samples of paint membranes were manufactured to a form of thin disks by cutting out a thin paint film after a liquid paint were spread thinly on a plastic sheet having a smooth surface and dried off. The membrane diameter was 200 mm, and the thickness was 0.12 ± 0.2 mm. The samples conditions are shown in Table 1.

2.2. Experiment of tritium permeation through paint membrane

A tritium permeation experiment was performed using an experimental apparatus placed in a fume food as shown in Fig. 1. A container filled with tritiated water was put in a permeation cell, and HTO vapor atmosphere was generated in the cell. The HTO vapor concentration in the upstream side of permeation was controlled to concentration of tritiated water in the range from 0.2 MBq/cm^3 to 10.3 MBq/cm^3, and then the saturated concentrations of HTO vapor in the upstream side of permeation were from 2 Bq/cm^3 to 96 Bq/cm^3 at room temperature. The permeation cell was housed in an incubator to keep the temperature constant. A paint membrane was fixed in a couple of VCR connectors by holding with two metal gaskets. The permeation area of a membrane was 0.98 cm^2. Tritium permeated through a paint membrane was purged away by pure N_2 gas with 1% H_2O vapor to remove tritium adsorbing and retaining on the surface of piping lines. A flow rate of the purge gas was 50 cm^3/min (STP). The purge gas containing tritium passed through the water bubblers, and then tritiated water vapor (HTO) was trapped by the bubbler 1 or 2. Gaseous tritium (HT) was trapped by the bubbler 3 or 4 after HT was change to HTO by the copper oxide (CuO) bed. The bubblers were exchanged with other ones after a certain time interval to obtain variations of the amount of tritium permeation with time elapsed. The amount of tritium collected by the water bubblers was measured by a liquid scintillation counter (Quantulus 1220, PerkinElmer Japan Co., Ltd). After a permeation rate reach to be steady-state, the paint membrane trapping tritium was soaked in distilled water. The amount of tritium trapped in the membrane and the tritium release rate to water were measured by sampling the water at regular intervals, which is called a water dissolution method.

Table 1
Experimental conditions of samples.

Sample	Epoxy	Silicon
Weight [mg]	46–52	48
Thickness [mm]	0.1–1.4	0.1
Permeation area [cm^2]	0.98	
Temperature [°C]	21–26	25–26
HTO conc. [Bq/cm^3]	2–96	50

3. Results and discussion

3.1. Tritium permeation through paint membranes

Fig. 2 shows an example of experimental results of tritium permeation through an epoxy membrane. Temperature variations were within 2–3 °C in the region of 20–26 °C through permeation experiments although the permeation cell was housed in the incubator. Most of tritium permeating through paints was trapped as a chemical form of HTO by a water bubbler regardless of purge gas with/without H_2O vapor. The ratio of HTO/(HTO + HT) in the trap was higher than 99.9%.

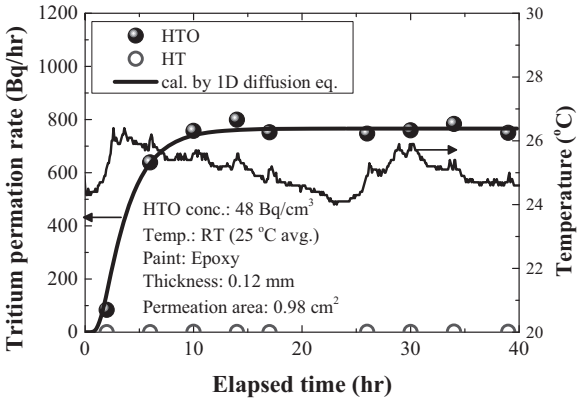

Fig. 2. An example of experimental results of tritium permeation through an epoxy paint membrane. Permeated tritium was purged away by N_2 with 1% H_2O vapor.