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Evaluation of tritium behavior in concrete

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Abstract

Concrete walls used in a fusion reactor or a tritium handling facility have the important role of separating the atmosphere in a facility from environmental air, since substantial tritium leakage is possible from a tritium system to the atmosphere of a tritium handling facility. Therefore, it is necessary to understand the transfer behavior of tritium through concrete. The experiments of this study were performed to observe the change of the tritium distribution with time in columns made of cement paste, mortar or concrete. It was found that exposure of the surface of concrete materials to HTO vapor for two months produced tritium permeation to the depth of 30 mm. Estimation of tritium penetration behavior was also performed in this study by applying adsorption, desorption and isotope exchange reactions and the estimated tritium profile was in good agreement with observed values.

1. Introduction

Research on tritium behavior in concrete materials has focused on the behavior of tritiated water in concrete materials that were placed in water to clarify tritium behavior in low-level solidified waste [1,2]. However, there has been little research on the behavior of tritiated water in concrete materials placed in air with tritiated water vapor despite the possibility of tritium release into the atmosphere of a fusion reactor or tritium handling facility. The authors have found that there is a lot of structural water in cement paste, which can trap tritium through the isotope exchange reaction, and that it is difficult to eliminate the trapped tritium from the cement paste [3,4]. Tritium-exposure experiments with mini and small size specimens were carried out to observe the tritium penetration behavior in cement paste and mortar in this study. Tritium-exposure experiments with large size specimens were also carried out in order to examine tritium behavior in concrete including large aggregate.

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2. Experimental

2.1. Materials

The specimens of concrete materials, which have a columnar shape, were made using ordinary Portland cement with the water-to-cement ratio (W/C) of 0.5 by weight. The W/C of cement paste specimens was 0.6. After casting in molds, all specimens were cured over one month at 20.5 °C before the experiment. The small size specimens were 13 mm in diameter and 55–100 mm in length, and the large size specimens are 100 mm in diameter and 100 mm in length. The other proportions of specimens are shown in Table 1.

2.2. Measurement of isotope exchange capacity using mini size specimens

The flowchart of the experiment is shown in Fig. 1. Columns of specimens made of cement paste or mortar, which were 13 mm in diameter and 10 mm in length, were exposed to the tritiated water vapor for several weeks. The sand particles used in the concrete material in this study were also separately exposed to tritiated water vapor. The exposed sand particles were almost spherical in shape. The concentration of tritiated water vapor in the air was 410 Bq/cm^3, and the partial pressure of water vapor (H_2O) was 950.1 Pa. Thus the T/H ratio of this tritium vapor is 1.51×10^6. This experiment

Table 1
The proportion of concrete materials

	Mixing proportion (by weight)				Porosity (cm^3/cm^3)
	Water	Cement	Sand	Aggregate	
Cement paste	0.6	1.0	–	–	–
Mortar-1	0.6	1.0	2.00	–	–
Mortar-2	0.5	1.0	2.26	–	0.05
Concrete 1	0.5	1.0	2.26	2.54[a]	0.04

[a] Largest stone size: 15 mm diameter.

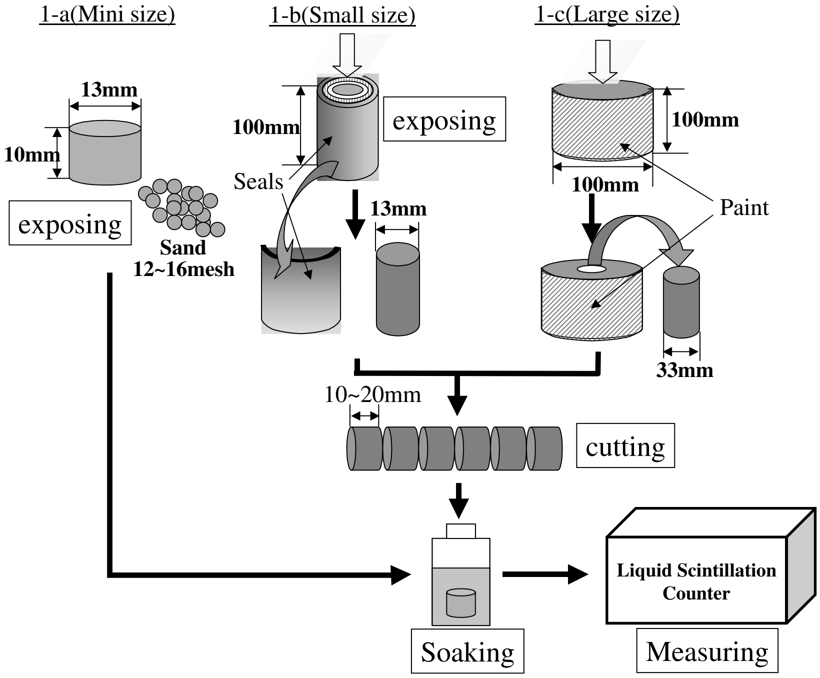

Fig. 1. The flowchart of experiment.

was continued until the tritium concentration trapped in the column reached a state of equilibrium.

2.3. Tritium exposure experiment with small size specimens

The columns of specimens were made of cement paste. The flowchart of the experiment is also shown in Fig. 1. Only one face of a specimen column was exposed to the tritiated water vapor, and the other surfaces of the column were sealed with the waterproofing paint and teflon-tape. The concentration of tritiated water vapor was 407 Bq/cm^3, and the partial pressure of water vapor (H_2O) was 950.1 Pa. After the specimens were exposed to the tritiated water vapor at room temperature for the scheduled duration time, the waterproofing paint and teflon-tape were removed from specimens. Then, the specimen columns were cut to produce several disc samples along the axial direction of the column, with each cut off specimen piece having about 1 cm length. Each specimen piece was soaked in distilled water to remove the trapped tritium in specimens, and the tritium concentration in the water was measured using a liquid scintillation counter.

2.4. Tritium exposure experiment with large size specimen

The flowchart of experiment is shown in Fig. 1. Only one face of a specimen column was exposed to the tritiated water vapor, and the other surfaces of the column were painted with epoxy paint to prevent the tritium ingress. The concentration of tritiated water vapor was 740–1110 Bq/cm^3, and the partial pressure of water vapor (H_2O) was 1000 Pa for several weeks. The temperature of the experiment was 288–298 K. The core specimen, which is 33 mm in diameter and 100 mm in length, was taken from the large size specimen using a drill. The distribution of tritium concentration in the core-specimens was measured using the same procedure as the small size specimen. The thickness of each specimen was about 15 mm.

3. Results and discussion

3.1. Estimation of isotope exchange capacity

The experimental results obtained from mini size specimens are shown in Table 2 after consideration of H/T ratio. The isotope exchange capacity in this study is based on the total amount of water used in the isotope exchange reaction with concrete materials. The tritium trapping capacity of the cement paste was much larger than the trapping capacity of metal materials to be used in the construction of fusion facilities [5].

Table 2
Isotope exchange capacity in concrete materials

	Isotope exchange capacity (Bq/g)
Cement paste	3.97×10^{13}
Mortar-1	1.50×10^{13}
Sand	6.12×10^{11}

Ratio of H/T: 1.51×10^6.

The isotope exchange capacity observed for the cement paste sample in this study is about two times larger than that of mortar and about 10 times larger than that of sand. This means that the isotope exchange capacity of mortar is mainly determined by the cement paste and the trapping capacity of the mortar is estimated by the following equation.

$$S_{\text{mortar}} = S_{\text{cement}} \times x_{\text{cement}} + S_{\text{sand}} \times x_{\text{sand}}, \qquad (1)$$

where S is the isotope exchange capacity, x is the fraction of the material contained in mortar. The isotope exchange capacity estimated from Eq. (1) gives

$$S_{\text{mortar}} = 1.80 \times 10^{13}\ \text{Bq/g}. \qquad (2)$$

This value shows close agreement with the observed value in Table 2. There was a previous report on the amount of trapped tritium in cement paste by Numata et al. [6]. The tritium trapping capacity estimated in that study is too small because the effect of isotope exchange capacity was not taken into account.

3.2. Tritium exposed experiment with small size specimens

The changes of the tritium distribution in the cement paste column with time are shown in Fig. 2. The tritium concentration profile in a small size cement paste column was estimated using mass transfer properties reported elsewhere by the present authors [3,4] and is shown for each exposure time as curves (1)–(3) in Fig. 2. The isotope exchange reaction may give a large effect on tritium transfer as shown in Fig. 3 because cement paste has a large amount of isotope exchange capacity as stated above. The comparison in Fig. 2 obviously indicates

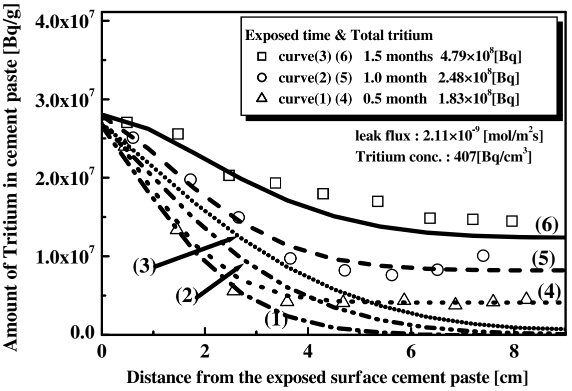

Fig. 2. Change of tritium concentration profile in cement paste.

that a certain amount of tritium penetrated into the cement paste column even through the side and bottom surfaces though these surfaces were supposed to be protected from tritium. Then, a second estimate of the tritium concentration profile was made that took the tritium penetration through the side wall into account. As shown by curves (4)–(6) in Fig. 2, the modified estimate gives good agreement with observed profiles when the tritium leak rate is assumed to be

$$Q_{\text{leak}} = 2.11 \times 10^{-9}\ (\text{mol/m}^2\ \text{s}). \tag{3}$$

In addition, more than 1.0×10^7 Bq/g-cement was trapped in the column of specimens exposed over one month in the condition of this work.

3.3. Tritium exposed experiment with large size specimens

Fig. 4 shows a comparison of tritium distribution observed in the core part of large size samples of cement paste, mortar and concrete. The histogram in Fig. 4 expresses the average amount of tritium trapped in a disc piece cut from the column specimen. The physically adsorbed water, chemically adsorbed water and structural water in Fig. 4 are defined elsewhere by the present authors [3]. It is known from this figure that tritium penetration from the side wall has no effect on the tritium profile in the core part of the large size specimen though some penetration is observed from the bottom surface. Mortar and concrete have the same transfer rate of tritium as cement paste, even though the isotope exchange capacity of mortar or concrete is less than half of cement paste. Because sand or aggregate has a smaller exchange capacity, the specific surface area of mortar and concrete effective for adsorption and isotope exchange reaction decreases by addition of sand or aggregate. The change of chemical composition with time may affect the tritium behavior because Terashima et al. reported that the diffusion coefficient of tritiated water in concrete is smaller than that of mortar and cement paste [1,7]. It is known from Fig. 4 that the penetration length of tritium in two months is only 30 mm from the exposure surface. This observation indicates that the tritium penetration along the core axis

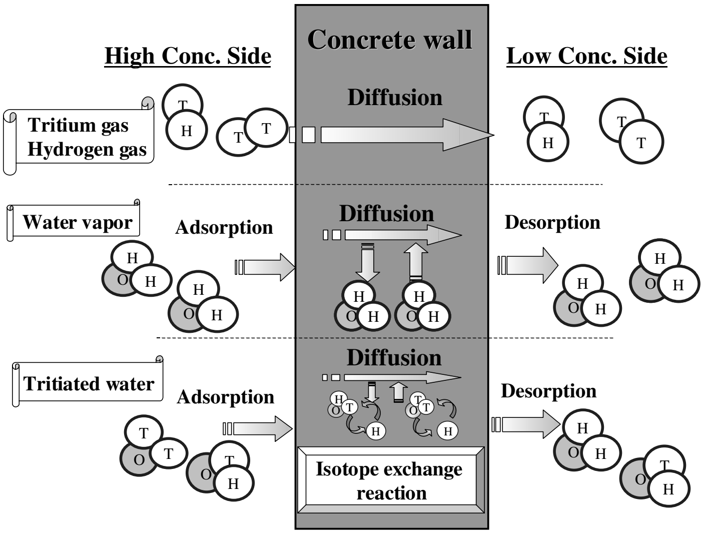

Fig. 3. Permeation model.

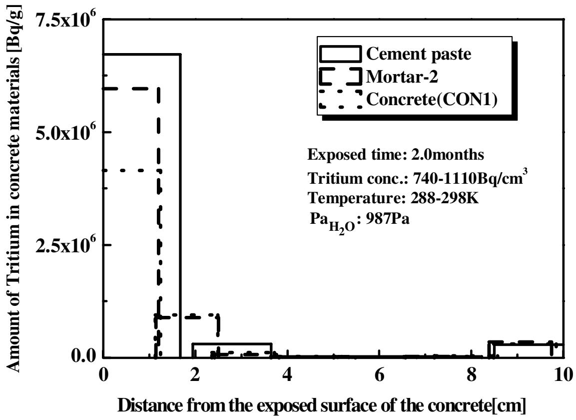

Fig. 4. Change of tritium concentration profile in core part of concrete materials.

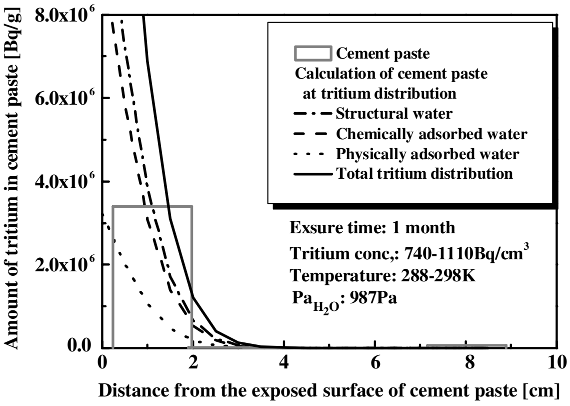

Fig. 5. Comparison of experimental results and calculated tritium concentration profile in cement paste.

is so slow because cement materials have a rather large isotope exchange capacity. It is also known from Fig. 4 that some penetration of tritium proceeds through epoxy paint, and the permeability and retention of tritium in epoxy paint was reported by Holland and Jalbert [8].

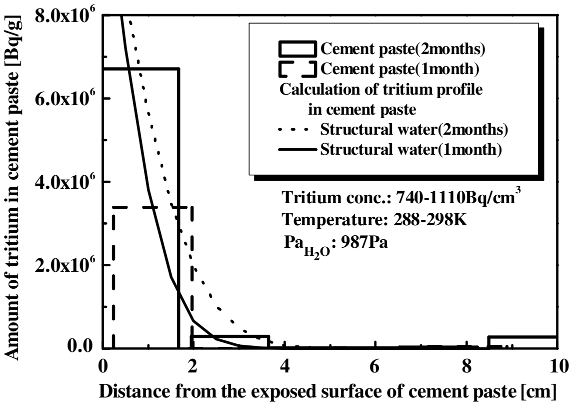

Fig. 6. Comparison of experimental results and calculated tritium concentration profile trapped in structural water.

Fig. 5 shows a comparison between experimental results for cement paste with large size specimens and a calculation for cement paste using the same mass transfer properties as the calculation of small size specimen. It is found from Fig. 4 that the estimated value for total tritium trapped in the large size cement paste does not agree with experimental results, while the estimated value for of tritium trapped in structural water gives good agreement with the experimental value. It is considered that a temperature rise during the drilling operation may release tritium trapped as chemically adsorbed water. Effect of exposure time on tritium penetration is compared in Fig. 6. This figure also shows that the estimated amount of tritium trapped by structural water gives good agreement with the observed value.

4. Conclusion

(1) The isotope exchange capacity of cement paste plays an important role in tritium trapping and penetration in concrete materials when concrete is exposed to tritiated water vapor.
(2) The calculation using mass transfer properties such as adsorption, diffusion and isotope exchange reaction of tritium with water in cement paste gives good agreement with experimental results.
(3) The Teflon seal and epoxy paint used in this study did not give perfect obstruction against the penetration of tritiated water into concrete materials.

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