Evaluation of Tritium Sorption Rate in Soil Packed Bed by Numerical Analysis[*]

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It is required that the behavior of tritium in the natural soil be clarified to evaluate the impact of the accident for fusion reactor. However, there has been little report on the behavior of tritiated water in the soil. In this study, top face of a soil packed bed was filled by tritiated water and subsequently distilled water was poured in the bed to recover tritium held up in the soil at room temperature. A tritium transport model in the soil has also been established and developed by this study. The tritium transfer phenomena in the soil is considered as percolation and isotope exchange reaction. The tritium transfer model in this study gives good agreement with experimental data for two samples by adjusting overall mass transfer coefficient representing percolation and isotope exchange reaction. It is also found that the value of overall mass transfer coefficient has a proper range for best fitting curve of experimental data of each soil sample.

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1. Introduction

In a fusion plant, huge tritiated water is handled and it is reported that tritium permeates metal walls at high temperature. The accidental spill of a large amount of tritiated water to the environment should be considered for the study of the environmental impact. The soil is the key material of underground condition.

When the tritiated water is percolating into the ground, a part of tritium is considered to be sorbed in the soil particles by adsorption, absorption and isotope exchange reaction. In our previous study, the percolation experiment of tritiated water into the soil packed bed were performed. It was supposed that a part of tritium is sorbed by the isotope exchange reaction between tritium in percolating water and hydrogen in soil particles [1]. From the analysis of the change of tritium concentration in the percolated water, it was confirmed that a proposed transfer model for tritium in a soil packed bed presented good agreement with experimental data approximately [2]. In this study, water behavior and tritium behavior in the percolation of tritiated water through natural soils were analyzed and the overall mass transfer coefficients representing isotope exchange reaction between tritium in percolating water and hydrogen in natural soil were obtained.

2. Experimental

One soil (sample 1) was collected from unpaved walk-way beside the swimming facility and another soil (sample 2) was collected from grove in garden beside main gate in the Hakozaki campus of Kyushu University [1]. Figure 1 shows experimental system for this study. The soils were separately packed in the fluoroplastic tube and tritiated water was poured to the top of the packed bed. The percolated water was drained into a tray which was put on an electric balance. Weight change of the tray was monitored to measure the weight of the effluent water. The effluent water was periodically sampled and tritium concentration was quantified by a liquid scintillation counter. The water height was monitored by a video camera during
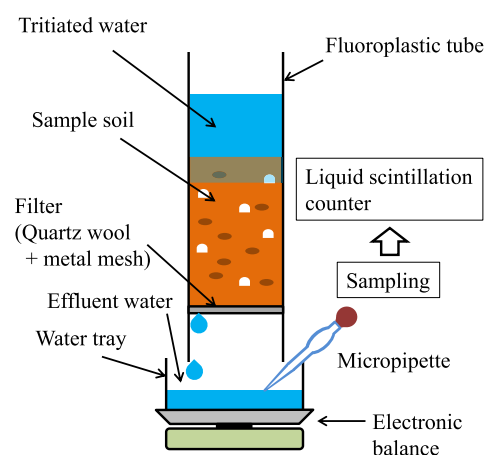

Fig. 1 The experimental system for tritiated water permeation in the soil packed bed.

Table 1   Soil property and experimental conditions.

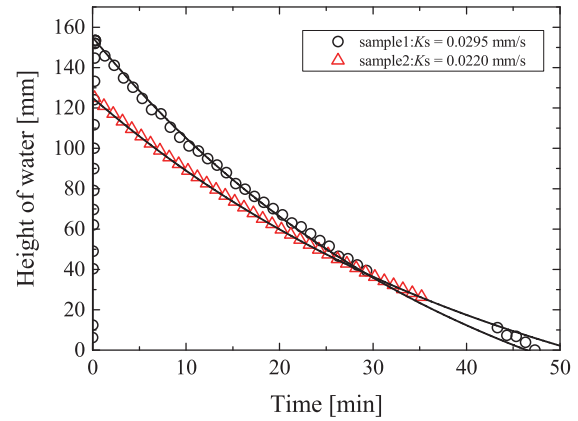| Soil property		
	Sample1	Sample2
Soil particle density [g/cm^3]	2.70	2.56
Moisture content [wt%]	1.07	5.47
BET surface area [m^2/g]	1.04	4.51
HTO percolation experiment		
Tube inner diameter [cm]	22.0	
Tube length [cm]	30.0	
Sample weight [g]	49.3	28.5
Height of the packed bed [cm]	7.09	6.13
Packing density [g/cm^3]	1.86	1.24
Packing factor [%]	61	49
Input HTO [MBq/cc]	0.871	0.581
Input HTO amount [cc]	58.2	50.7

Fig. 2   The changes of the height of water at the top of the packed bed with time.

Fig. 3   The changes of the effluent water amount through the packed bed with time.

the experiment. The experiments were carried out in the glove box. Soil property [1] and experimental conditions were summarized in Table 1.

3. Results and Discussion

3.1   Water percolation rate

The changes of water height at the top of the packed bed with time are shown in Fig. 2. Open symbols are experimental values and solid curves are calculated curves in this figure. Because the water surface overlapped with the instrument which supports the fluoroplastic tube, the position of the water surface could not be recognized by the video camera from 29 min to 43 min for sample 1 and after 35 min for sample 2. The water percolation rate was expressed as the value of saturated hydraulic conductivity for each sample. The profiles of water height at the top of the packed bed was estimated by the fitting method. Saturated hydraulic conductivity, K_s (mm/s) are varied as a parameter for each soil sample. The theory of water percolation model and the following equation were reported elsewhere by the present authors [2] based on Darcy's law.

$$b_{(t)} = (b_0 + L) \exp\left(-\frac{K_s}{L}t\right) - L, \tag{1}$$

where $b_{(t)}$ is the water height at the time "t" (s), b_0 is the initial water height (mm), L is the height of the packed bed (mm), t is time (s). The following saturated hydraulic conductivities were obtained,

$$K_s = 0.0295 \text{ mm/s for sample 1}, \tag{2}$$

$$K_s = 0.0220 \text{ mm/s for sample 2}. \tag{3}$$

Although the value of K_s for sample 1 has been already reported in [1], it was shown again in this paper for comparing to sample 2. By using obtained saturated hydraulic conductivities, the change of effluent water amount from the soil packed bed was calculated. The experimental values and the calculated covers are compared in Fig. 3. Non-adjusted curves indicates the calculated curves for the

packing condition of each sample before the water pouring. Non-adjusted curves were not agreement with experimental curves. The packing conditions such as packing height and porosity are modified by saturating with water. Additionally, $t = 0$ means the beginning time of water pouring but the beginning time of water effluent delays by water saturating process. Therefore, the packing height or the porosity and the beginning time of water effluent were adjusted. The adjusted curves was good agreement with experimental curves.

3.2   Isotope exchange reaction

The change of tritium concentration in the effluent water with time for sample 1 and sample 2 presented in Fig. 4 and Fig. 5, respectively. Tritium concentration in the initial stage was lower than that in the input water. This indicates that a certain amount of tritium was sorbed in the soil. The overall mass transfer coefficient representing the isotope exchange reaction between tritium in water and hydrogen in soil, $K_{f,ex}$ (m/s) and the isotope exchange capacity, Q_{ex} (mol-T$_2$O/m^2) were obtained for each sample by the curve fitting method as follows,
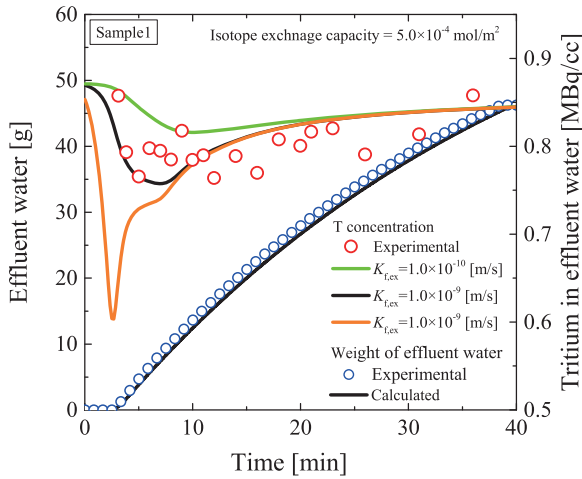
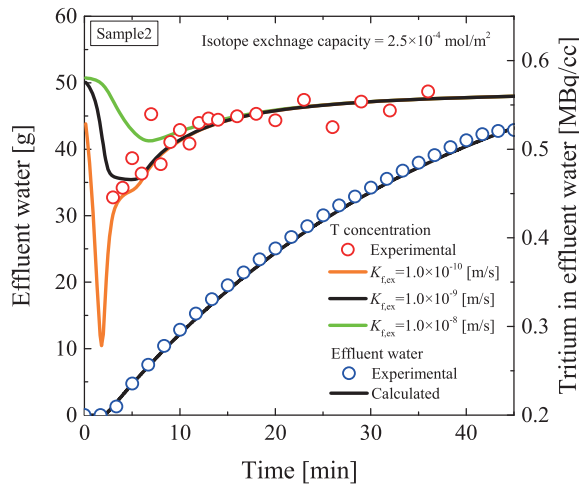Fig. 4   The changes of effluent water and tritium concentration for sample 1.

Fig. 5   The changes of effluent water and tritium concentration for sample 2.

$$K_{f,ex} = 1.0 \times 10^{-9} \, \text{m/s} \quad (4)$$

$$Q_{ex} = 5.0 \times 10^{-4} \, \text{mol/m}^2 \text{ for sample 1,} \quad (5)$$

$$K_{f,ex} = 1.0 \times 10^{-9} \, \text{m/s} \quad (6)$$

$$Q_{ex} = 2.5 \times 10^{-4} \, \text{mol/m}^2 \text{ for sample 2.} \quad (7)$$

The values of $K_{f,ex}$ for sample 1 and sample were same and the values of Q_{ex} were also close to each other. BET surface areas measured by micromeritics automatic surface area analyzer (TriStar-II3020, SHIMAZU Co.) is 1.04 m^2/g for sample 1 and 4.51 m^2/g. Therefore the isotope exchange capacity per weight is 5.2×10^{-4} mol/g for sample 1 and 1.13×10^{-3} mol/g for sample 2.

The obtained isotope exchange capacities are compared with the capacity on Sand, Mortar, cement paste [3] and Pt-MS5A [4] in Table 2. The isotope exchange capacity on soil was close to that on sand and Pt-MS5A. The mass transfer coefficients obtained in this work for soil are compared the mass transfer coefficient represent-

Table 2   Comparison of isotope exchange capacity for various materials.

Material	Isotope exchange capacity [mol-T$_2$O/g]	Ref.
Sample 1	5.20×10^{-4}	-
Sample 2	1.13×10^{-3}	-
Sand	2.85×10^{-4}	[3]
Mortar	6.98×10^{-3}	[3]
Cement paste	1.85×10^{-2}	[3]
Pt-MS5A	7.0×10^{-4}	[4]

Table 3   Comparison of overall mass transfer coefficient representing isotope exchange reaction for various materials.

Material	$K_{f,ex}$ [m /s] at room temperature	Ref.
Sample 1	1.0×10^{-9}	-
Sample 2	1.0×10^{-9}	-
Li$_2$ZrO$_3$	8.5×10^{-6}	[5]
Li$_2$TiO$_3$	3.0×10^{-6}	[6]
Stainless steel	1.6×10^{-4}	[7]
Copper	2.0×10^{-4}	[7]
Graphite	3.5×10^{-6}	[7]
SiC	3.5×10^{-5}	[8]

ing the isotope exchange reaction between tritiated water vapor and water vapor on various materials in Table 3. The mass transfer coefficients representing isotope exchange reaction in water for soil were smaller than that in water vapor for various materials.

4. Conclusion

1. Saturated hydraulic conductivity for two natural soils were obtained as follows,
   $K_s = 0.0295$ mm/s for sample 1,
   $K_s = 0.0220$ mm/s for sample 2.
2. Tritium transfer/sorption model based on isotope exchange reaction proposed and gives good agreement with experimental results.
3. The isotope exchange capacity for two natural soils were obtained as follows,
   $Q_{ex} = 5.0 \times 10^{-4}$ mol/m^2 for sample 1,
   $Q_{ex} = 2.5 \times 10^{-4}$ mol/m^2 for sample 2.
4. The overall mass transfer coefficients representing isotope exchange reaction was obtained as follows,
   $K_{f,ex} = 1.0 \times 10^{-9}$ m/s.

Acknowledgments

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