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# Transfer of tritium in concrete coated with hydrophobic paints

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## ABSTRACT

An experimental study on tritium (T) transfer in porous concrete for the tertiary T safety containment is performed to investigate (i) how fast HTO penetrates through concrete walls, (ii) how well concrete walls contaminated with water-soluble T are decontaminated by a solution-in-water technique, and (iii) how well hydrophobic paint coating works as a protecting film against HTO migrating through concrete walls. The experiment is comparatively carried out using disks of cement paste which W(water)/C(cement) weight ratio is 0.6:1 with or without hydrophobic paints, and mortar disks which W/C/sand ratio is 0.6:1:2 with or without the paints. The hydrophobic paints tested in the present study are an epoxy polymer resin paint and an acrylic-silicon polymer resin one. After T exposure during specified time under a constant HTO vapor pressure in an acrylic box, the amount of water-soluble HTO on/in the disks is determined using a technique of $H_2O$ dissolution during specified time. The results obtained here are summarized as follows: (1) HTO penetration in porous concrete can be correlated in terms of the effective diffusivity. (2) Its value in porous cement without coating is $1.2 \times 10^{-11}$ m$^2$/s at 25 °C. (3) HTO penetrates only through pores in cement, and there is no path for HTO transfer in non-porous sand. (4) Rates of sorption and dissolution of HTO in disks of cement and mortar coated with the epoxy resin paint are correlated in terms of the effective diffusivity through the paint film which value is $D_T = 1.0 \times 10^{-16}$ m$^2$/s. The rate-determining step is diffusion through the paint. (5) The epoxy resin paint works more effectively as an anti-HTO diffusion coating. (6) Another acrylic-silicon resin paint does not work well as anti-HTO diffusion coating. This may be because the hydrophobic property of the silicon resin paint is deteriorated with elongating the contact time with $H_2O$ vapor or liquid. (7) The HTO uptake inside the epoxy paint is greater than that of the silicon one. (8) The permeation reduction factor (PRF) of HTO for the epoxy paint at steady-state is expected large, if HTO vapor only contributes to diffusion. However, when concrete surfaces coated with the epoxy paint are under wet conditions, the PRF value becomes smaller. All those results can be used to estimate the effect of HTO soaking in concrete walls in case of accidental T release in a fusion reactor room and to decontaminate wastes of tritiated concrete.

## 1. Introduction

A large amount of radioactive tritium (T) in a hot laboratory or a future fusion reactor system is handled in triple enclosures which are composed of the primary enclosure of an experimental apparatus, the air-tight secondary enclosure of a glovebox or a caisson and the tertiary enclosure of a specially designed laboratory or building bounded by concrete walls [1–3]. Usually, two detritiation systems are attached in the secondary and tertiary enclosure systems. The first detritiation system for the secondary enclosure is always operated when a large amount of T is handled in laboratory rooms. This is not only because there is some possibility of inevitable T release such as permeation through various boundaries but also because the primary enclosure sometimes has open ends or pump exhausts inside experimental apparatuses. Therefore, chronic release of a small amount of T is inevitable. On the other hand, the air detritiation system (ADS) for the tertiary enclosure is not always operated, because the secondary enclosure system is expected to work well in normal operation. The ADS is launched in case of emergent accident such as unexpected T release. Operators or researchers usually work in laboratory rooms inside the tertiary enclosure. They are evacuated from the room in case of the accidental T release, and ADS is operated. The radioactivity of T in air of the laboratory room is regularly monitored. The ADS is

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being operated until time when its value is lower than the maximum allowable concentration (MAC) in laboratory rooms for each T molecular species.

Even though ADS starts operated in the tertiary enclosure, the flow rate to circulate contaminated air is limited. Therefore, the T concentration in room air remains much higher than the MAC for short time immediately after the accidental T release. Then, concrete is exposed to high-radioactive HTO vapor, and it can permeate through concrete walls [4–8]. In addition, there is also possibility that some HTO remains inside concrete walls because of chronic contact with HTO vapor for a long time [7]. This is because porous concrete walls adsorb/absorb HTO on/in its porous surfaces and HTO migrates inside concrete due to difference in the HTO concentration. Even when only $T_2$ or HT is handled in laboratories, the isotopic exchange reaction such as $T_2$ (in gas) + $H_2O$ (on concrete) = HT (in gas) + HTO (on concrete) can proceed.

So-called soaking effect is the phenomena that high-concentration T remains in laboratory air even after ADS has launched [7–10]. The history of the T concentration in air sometimes shows a two-step desorption curve for a long time when *ADS* start operating. In order to make clear the T soaking effect, adsorption/desorption, solution/diffusion and isotopic exchange on/in various materials and boundaries have to be understood quantitatively. Concrete to constitute the tertiary containment consists of cement, sand, aggregates and structural water. In order to avoid direct contact with radioactive HTO or HT and in order to avoid corrosion of concrete structures due to $H_2O$, concrete walls are usually coated with hydrophobic paints [11,12]. However, T is trapped on/in concrete walls or interstitials between paints and concrete, and it can permeate into deep position.

There were several researches on T migration in concrete. T transfer in concrete has been correlated in terms of effective diffusivity [4–6,13–18]. However, there are fewer researches on behavior of T transfer in concrete coated with hydrophobic paints [11,12,19]. In addition, it is important to develop a simplified technique to decontaminate T from concrete walls covered with or without paints. This can lead to large advantage to decrease the amount of radioactive wastes in case of decommission of fusion-related facilities [20,21]. It is difficult to decontaminate T trapped deeply in structural concrete walls. So that, a large amount of T wastes composed of broken concrete pieces are generated. In order to eliminate the amount of solid wastes composed of contaminated concrete, it is important to decontaminate T trapped inside concrete walls. In this meaning, it is also important to develop a simplified technique to decontaminate tritiated concrete.

Previously, there were several ways to remove radionuclides from materials surfaces such as smearing, gas purge, heating, etching, laser exposure, ultraviolet rays exposure and so on [4,7,20,21]. However, those are not efficient to decontaminate T that is trapped at deep positions inside materials. Incinerating is possible but not economical for concrete, because it needs a large amount of energy [5,6]. So, water dissolution is tested in the present study as a simplified technique to decontaminate water-soluble T species from porous concrete.

In the present study, experiment on HTO penetration in porous concrete materials for the tertiary enclosure is here performed to clarify the following things; (i) how fast HTO penetrates through disks of cement or mortar with porous and composite structure, (ii) how well disks of cement or mortar contaminated with T are decontaminated by a water-dissolution technique, and (iii) how well hydrophobic paint coating works as a protective film against HTO permeating through concrete. All those results can be utilized to estimate the amount of T that remains in concrete walls in case of accidental T release in a fusion reactor room.

## 2. Experimental

### 2.1. Manufacture of cement and mortar disks with or without painting

Experiment of HTO transfer is carried out using cement or mortar disks of 35 mm in average diameter and 7.84 mm in average thickness. Six kinds of disks are made up as follows: (i) disks of cement pastes which W(water)/C(cement) weight ratio is 0.6:1 with or without any of the two kinds of hydrophobic paints, and (ii) mortar disks which W/C/sand weight ratio is 0.6:1:2 with or without any of the two paints. In addition, three kinds of plates are made up using a 316 stainless-steel plate coated with or without any of the two paints to compare the effect of hydrophobic property. The stainless-steel plate is a rectangular of $25 \times 30\,\mathrm{mm}^2$ in area and 0.1 mm in thickness. Cement used in the present study is ordinary Portland cement, which is a mixture of dicalcium silicate (C2S, $2CaO–SiO_2$), tricalcium silicate (C3S, $3CaO–SiO_2$), tricalcium aluminate (C3A, $3CaO–Al_2O_3$) and tetracalcium aluminate ferite (C4AF, $4CaO–Al_2O_3–Fe_2O_3$). The content of the four main minerals is C3S:C2S:C3A:C4AF = 50:26:9:9 in weight percent. The rest is other contents such as MgO, $CaSO_4$ and so on. Sand to compose mortar is screened between 250 $\mu$m and 500 $\mu$m.

For curing of hydration reaction between cement and water, the disk samples were placed under a constant humidity condition at room temperature for two months. The disks of cement paste and mortar made up above have a porous structure. Part of the cement changes to hydrate crystals such as ettringite of $3CaO–Al_2O_3–3CaSO_4–nH_2O$ in the course of the curing time as shown in our previous study [16]. After the curing time for two months, some of the disks are coated with any of the two kinds of an epoxy polymer resin paint, which commercial name is “2-part Epoxy Waterproof”, Asahipen Co., Japan, and an acrylic-silicon polymer resin paint, which commercial name is “Water base Supercoat”, Asahipen Co., Japan. The chemical structures of (a) acrylic-silicon polymer resin and (b) epoxy polymer resin included in the two hydrophobic paints are shown in Fig. 1. The thicknesses of the paints coated on the disks of cement or mortar and the stainless-steel plates are measured from surface observation photos by a microscope. Their thickness varies from 0.10 mm to 0.14 mm. The average thickness is 0.12 mm.

### 2.2. Experiment of tritium exposure and $H_2O$ dissolution

After preparing the cement or mortar disks with or without any of the hydrophobic paints described above, HTO exposure experiment was performed inside any of the three tritium-handling facilities of Kyushu University, Tritium Process Laboratory in Japan Atomic Energy Agency (TPL-JAEA) and Hydrogen isotope Research Center (HRC) in University of Toyama according to different HTO concentrations. The above nine kinds of samples were placed in an acrylic box in a T-handling glovebox under the conditions of constant vapor pressures of HTO and $H_2O$ as schematically shown in Fig. 2. The HTO concentration in atmosphere of the acrylic box is denoted by $c_{\mathrm{HTO},0}$ hereafter. Its values are $c_{\mathrm{HTO},0} = 1.0–1.5 \times 10^{-2}\,\mathrm{Bq/cm^3}$, $7.2\,\mathrm{Bq/cm^3}$ and $740\,\mathrm{Bq/cm^3}$ according to the scale of the gloveboxes used in their respective facilities. The HTO concentration tested here ranges from around one-hundredth lower than the *MAC* in Japanese laboratory air ($c_{\mathrm{HTO,MAC}} = 0.8\,\mathrm{Bq/cm^3}$) to around 1000-holds higher than that. HTO exposure time varies from 1 to 79 days. The exposure time is so long that the HTO concentration in the acrylic box changes slightly from time to time depending on variations of room temperature, even if the laboratory room is usually air-conditioned in the daytime. This is because the HTO vapor pressure is controlled by a HTO water solution in a petri dish placed in each acrylic box. Consequently, the