

# CONTAMINATION BY TRITIUM OF WATERS AND CONCRETE IN A RESEARCH REACTOR CONTAINMENT DUE TO LEAKAGE OF HEAVY WATER

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## ABSTRACT

Approximately heavy water of ca. 28.2 L (ca. 0.34 TBq of tritium) leaked from the D<sub>2</sub>O facility of the Kyoto University Reactor (KUR) during the period of about one and half year. As a result of the incident, HTO concentrations in pool waters had increased extensively as well as in the containment building air during the period before the leak detection. The soaking effect of HTO vapor by the building concrete was also confirmed by air monitoring. Although the leak was of little radiological consequence, it served as a useful lesson of our emergency response.

## INTRODUCTION

Radiation protection both on and off the site is deeply dependent upon the availability of appropriate monitoring data both in nuclear facilities and in natural environs.

The KUR near Osaka, Japan commenced operation in 1964 at 1 MW<sub>th</sub> but the power was raised to 5 MW<sub>th</sub> in 1968. Ventilation system in the containment building works so as to cover the duration of the reactor operation cycle of 70-80 h/wk. By the routine monitoring the concentrations of radionuclides in the exhaust air from the KUR, operating at 5 MW<sub>th</sub> have been confirmed to be in the order of 10<sup>-2</sup> Bq/cm<sup>3</sup> for <sup>41</sup>Ar, 10<sup>-6</sup> Bq/cm<sup>3</sup> for HTO, 10<sup>-8</sup> Bq/cm<sup>3</sup> for <sup>133</sup>I and 10<sup>-9</sup> Bq/cm<sup>3</sup> for <sup>131</sup>I. Because of the low detection limits, as measured by a 22-L ionization chamber set for the exhaust streams, the presence of most radionuclides in the stream is usually masked by the reading of <sup>41</sup>Ar for such minor releases during normal operation of the reactor; this is especially true for HTO vapor. For this reason, monitoring of HTO vapor in the exhaust is carried out during the operation of the KUR on twice a year basis using liquid scintillation counting of water samples collected by condensation from the exhaust air.

## BACKGROUND TO THE INCIDENT

During such a routine check in fall 1987, a relatively high concentration of HTO vapor was detected in the condensate. This result, however, was attributed to probable contamination of experimental devices because the same staff of the Radiation Control Division was involved in the research of tritium metabolism and routine monitoring of HTO from the KUR stack. A water sample was not taken from the exhaust during the next monitoring period (Spring 1988) because the operation of KUR was suspended over a 3-month period beginning in April because of fixation for secondary

coolant system. Although passing over data from the exhaust air monitoring had delayed the confirmation of HTO leakage, there was another indicator of leakage. The source monitoring data obtained by the Research Reactor Division had showed gradual increases in the HTO concentration in pool water in the containment building. This was especially the case in the sub-pool (ca.  $1.0 \text{ m}^2 \times 1.7 \text{ m}$  deep), located on the top of biological shield, where the HTO concentration in water had increased 60-fold during the period of 1 y (May 1987-April 1988). Investigations into the source of this increase in the HTO concentration in water were focused mainly on the presence of irradiated materials such as lithium and boron which could be a source of HTO production by a neutron activation process. A smear test carried out on August 1988 revealed significant contamination on the front surface of a heavy water tank, and a high HTO concentration was detected in the atmosphere near the  $\text{D}_2\text{O}$  tank using a 1.5-L portable ionization chamber. These findings led to the conclusion that HTO increases in water were the result of leakage of the heavy water. Nevertheless, the mechanisms by which HTO concentrations of both the sub-pool and the primary coolant water (about  $30 \text{ m}^3$  and 9-m deep) had extensively increased during 1 year since May 1987, were unknown.

#### SURVEY OF LEAK POINT AND RECOVERY OF HTO

Under normal conditions, the aluminium  $\text{D}_2\text{O}$  tank contains approximately  $2.2 \text{ m}^3$  of heavy water with a purity of 99.7%. As is shown in Fig.1, one face of the  $\text{D}_2\text{O}$  tank, ca.1.8 m in dia., is adjacent to a graphite block layer (ca.  $1.5 \text{ m}^3$ ), 48 cm thick, with a bismuth plug. The space in front of this layer is used as an irradiation room with dimensions ca.  $2.4 \text{ m} \times 2.4 \text{ m} \times 2.4 \text{ m}$ , covered with high density concrete identical to the biological shield of the reactor core. By 5 October 1988, the concentration of HTO in condensate in air near the tank was found to be as much as 2.2 MBq/ml. Then, using an air conditioner and a 20-L water bubbling trap, a 6-wk program was started to lower the moisture in the air in the irradiation room adjacent to graphite blocks. On 16-17 November, the heavy water in the  $\text{D}_2\text{O}$  tank was withdrawn into 200 L drums and replaced by light water to carry out a leak test. An estimated loss of 28.2 L of heavy water was made by comparing the weight of the water removed with that initially loaded into the  $\text{D}_2\text{O}$  tank. Inspection with a remote TV camera failed to locate the source of the leak in any of the suspected areas such as joints or flanges in the  $\text{D}_2\text{O}$  plumbing system. Then, the high density concrete shield of the irradiation room was dismantled on 21 November 1988. The graphite layer adjacent to the  $\text{D}_2\text{O}$  tank was removed and stored in a container ( $2 \text{ m}^2 \times 2 \text{ m}$  in height) on 19 January 1989 so as to access more easily to the tank and to recover any HTO absorbed by the graphite blocks. In early February, air, heated to  $90^\circ\text{C}$  was circulated through this container with electrical condensation loop system over a 2-mo period. By this procedure ca.  $4.6 \times 10^1 \text{ GBq}$  (ca. 12.4 L of condensate) of HTO was recovered as shown in Fig.2.

On 25 January 1989, less than 100 mL of water was found in a guide tube in the  $\text{D}_2\text{O}$  tank used to hold a thermo-couple. The HTO

concentration of this water was 7.9 MBq/ml, which was close to that of the stagnant heavy water (8.1 MBq/ml) in the dead space of the D<sub>2</sub>O plumbing system, and two thirds the concentration of the heavy water in the D<sub>2</sub>O tank itself ( $1.2 \times 10^1$  MBq/ml). Therefore, the source of the HTO leak was located somewhere in the guide tube. This tube was cut on the upper part of the tank where it had been inserted, and the resulting hole was welded shut. This resulted in the isolation and sealing of the main sources of HTO in the reactor containment building by early April 1989.

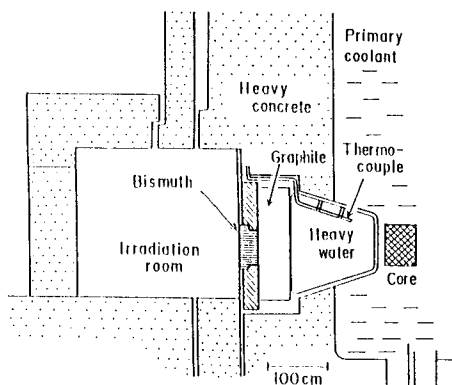


Fig.1 Cross section of KUR.

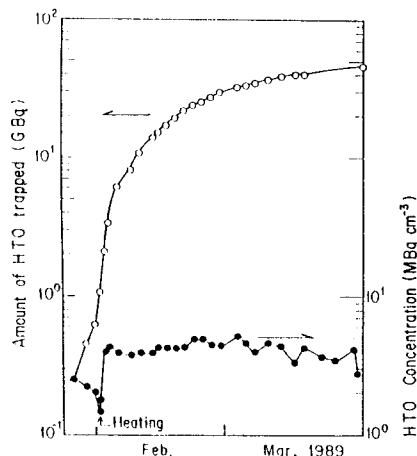


Fig.2 Amount of HTO recovered.

#### CHANGE IN HTO CONCENTRATION IN AIR

After the ventilation was shut down, the concentration of HTO vapor in air increased and reached a steady state condition within a few days. Assuming a constant production rate of HTO vapor from sources such as building concrete and neglecting physical decay process of tritium, the growth of HTO concentration in containment building air is formulated as follows:

$$d(CV)/dt = P - \mu CV \quad \dots (1)$$

where

- C : HTO vapor concentration in air (Bq/cm<sup>3</sup>),
- V : volume of the containment building (cm<sup>3</sup>),
- P : production rate (Bq/h),
- $\mu$  : leak rate (1/h), and
- t : elapsed time after turning off of ventilation (h).

Setting C=0 at t=0, the HTO concentration is given by:

$$C/C_0 = 1 - \exp(-\mu t) \quad \dots (2)$$

where

- C<sub>0</sub> : HTO concentration in air (=P/ $\mu$ V) under steady state condition (Bq/cm<sup>3</sup>).

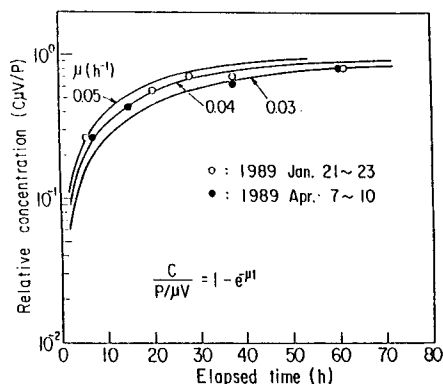


Fig.3 Growth of HTO concentration in building air.

The growth curve, as expressed by eqn (2) for HTO concentrations in air during shut down of the ventilation allows for the estimation of the leak rate, which was approximately estimated to be  $0.04 \text{ h}^{-1}$  by the curve fitting as shown in Fig.3.

The production rate, P, could be approximated from eqn (1) as the product,  $\mu C_0 V$ , for large times ( $t > 3 \text{ d}$ ). Therefore, long-term monitoring of HTO concentration in air ( $C_0$ ) under steady state conditions allows for the estimation of the attenuation rate of the HTO production rate, which depends on the HTO content in the building concrete. The concentration,  $C_0$ , in air monitored over nearly a 1-y period after venting shut down, shows attenuation rate of approximately  $5.0 \times 10^{-3} \text{ d}^{-1}$  ( $T_{1/2} = 20 \text{ wk}$ ) for the first 8-month starting on late November 1988 after transferring heavy water into drums and  $3.0 \times 10^{-3} \text{ d}^{-1}$  ( $T_{1/2} = 33 \text{ wk}$ ) since August 1989.

#### CHANGE IN HTO CONCENTRATION IN POOL WATERS

Laboratory experiments revealed that the fundamental mechanisms describing the HTO dynamics between water and air are exchange and evaporation. A model developed on the basis of this concept gave an average concentration of HTO vapor in air as soaking effect during the period for the HTO concentration in water to decrease from  $C_{w0}$  to  $C_w$  as expressed in eqn (3),

$$C_v = aH\varepsilon k_c^{-1} \{C_w - C_{w0} \exp(-at)\} / \{1 - \exp(-at)\} \quad (3)$$

where  $k_c$  : exchange rate constant between air and water (cm/h),  
 $k_e$  : evaporation rate constant (cm/h),  
 $H$  : water depth (cm),  
 $a$  :  $(k_c + k_e)/H$  (1/h),  
 $\varepsilon$  : isotopic ratio in HTO vapor and water (-),  
 $C_{w0}$  : HTO concentration in water at time 0 (unit/ml),  
 $C_w$  : HTO concentration in water at time t (unit/ml), and  
 $C_v$  : average HTO vapor concentration in condensate in air (unit/ml).

A linear regression for the attenuation of HTO concentrations observed in sub-pool water after removing sources in containment building gave a half-life of approximately 15 wk. Using a value for  $\varepsilon$  of 0.92 in eqn (3), the average concentration,  $C_v$ , in the condensate in the containment building air was calculated to be 36 Bq/ml during 50 wk starting on 20 March 1989. The average value estimated for the air corresponds to only ca.  $5 \times 10^{-5}$  times of the Administrative Level for radiation workers ( $7 \times 10^{-1} \text{ Bq/cm}^3$ ), though it was more than ca. one order larger in magnitude compared with that during normal operation.

#### CONCLUSIONS

Approximately 0.34 TBq of tritium had been leaked from the  $D_2O$  tank over a 19-mo period. More than 80% of HTO leaked was estimated to be released promptly as exhaust. Part of HTO leaked (ca. 2.5 GBq) moved into pool waters via air and increased the concentration as much as ca. 60-fold for sub-pool ( $1.7 \text{ m}^3$ ) and 5-fold for primary coolant ( $30 \text{ m}^3$ ), respectively. A significant amount of HTO vapor had been absorbed by the concrete material (4.2 GBq) and 46 GBq of HTO absorbed by graphite blocks was recovered.