Changes of Activity Concentration of Tritium in Water under the Air with Tritiated Water Vapor

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Recently, controversy continues over the cause of occurrence of tritiated water, whose activity concentration was about $7.1 \times 10^8 \text{ Bq} \cdot \text{m}^{-3}$, found in a nuclear power plant in Korea. The power plant claims that the tritiated water may have been produced by the absorption of the tritiated water vapor from the air, while some suspect that it may have leaked out from the other systems [1]. We cannot hastily determine the exact cause but we have investigated the possibility that water containing initially no tritium could be changed to tritiated water with high activity concentration of tritium when it is placed in the air containing tritiated water vapor.

It has been well known since the 1960s that tritium in the air containing tritiated vapor (HTO, T is tritium or ^3H) is easily transferred in water. This phenomenon has been used early for sampling of tritium in the air (known as air bubbler) [2–4] and still used today. It is also applied to respiratory masks for removing tritiated water vapor these days [5–7].

The relationship between the molar concentrations of tritium and hydrogen in the tritiated vapor of the air and those in (liquid phase) water at equilibrium is expressed by Equation (1), reported by Sepall and Mason [8], as follows:

$$\frac{([T]/[H])_{\text{vapor}}}{([T]/[H])_{\text{water}}} = \alpha \quad (1)$$

where T is tritium, H is hydrogen, and α is the partition constant (or equilibrium constant) and is 0.91 at 20°C.

In the above equation, molar concentrations of tritium in vapor and water can be converted to the activity concentrations of tritium in vapor and water, respectively, by the following equation:

$$[T] = \frac{m}{V} = \frac{A}{\lambda} \times \frac{1}{6.023 \times 10^{23}} \times \frac{1}{V} = \frac{S}{\lambda} \times \frac{1}{6.023 \times 10^{23}} \quad (2)$$

where m is the number of moles of tritium in vapor or water, V is the volume of vapor or water, A is the radioactivity of tritium in vapor or water, λ is the decay constant of tritium, 6.023×10^{23} is Avogadro's number, and S is the activity concentration of tritium in vapor or water.

The activity concentration of tritium in the air and that in water at equilibrium are calculated by using above two equations and the result is shown in Fig. 1.

An air with activity concentration of $3,000 \text{ Bq} \cdot \text{m}^{-3}$ (release limit of tritium in air with tritiated water vapor in Korea [9]) equilibrates at 20°C with a water with activity concentration of $1.92 \times 10^8 \text{ Bq} \cdot \text{m}^{-3}$. Also, at the same temperature, an air with activity concentration of $300,000 \text{ Bq} \cdot \text{m}^{-3}$ (derived concentration of tritium in air with tritiated water vapor in Korea [9]) equilibrates with a water with activity concentration of $1.92 \times 10^{10} \text{ Bq} \cdot \text{m}^{-3}$.

When a 1 m^3 of water with no tritium is placed in certain volume of air with 50% RH (relative humidity) at 20°C and tritiated vapor, the activity concentration of tritium in air and that in water at equilibrium after saturation by the water and tritiated water vapors are shown in Table 1.

When a 1 m^3 of water with no tritium is placed in a volume of air with 50% RH at 20°C , containing tritium, the activity

concentration of tritium in air and that in water at equilibrium are changed with the volume of air. When volumes of air with initial activity concentration of $3,000 \text{ Bq} \cdot \text{m}^{-3}$ are 10 m^3 , 100 m^3 , $1,000 \text{ m}^3$, $5,000 \text{ m}^3$, $10,000 \text{ m}^3$, $15,000 \text{ m}^3$, and $20,000 \text{ m}^3$, respectively, the activity concentrations of tritium in air at equilibrium are reduced to $0.47 \text{ Bq} \cdot \text{m}^{-3}$, $4.7 \text{ Bq} \cdot \text{m}^{-3}$, $47 \text{ Bq} \cdot \text{m}^{-3}$, $230 \text{ Bq} \cdot \text{m}^{-3}$, $440 \text{ Bq} \cdot \text{m}^{-3}$, $640 \text{ Bq} \cdot \text{m}^{-3}$, and $820 \text{ Bq} \cdot \text{m}^{-3}$, respectively, and those in water increase to $3.0 \times 10^4 \text{ Bq} \cdot \text{m}^{-3}$, $3.0 \times 10^5 \text{ Bq} \cdot \text{m}^{-3}$, $3.0 \times 10^6 \text{ Bq} \cdot \text{m}^{-3}$, $1.5 \times 10^7 \text{ Bq} \cdot \text{m}^{-3}$, $2.8 \times 10^7 \text{ Bq} \cdot \text{m}^{-3}$, $4.1 \times 10^7 \text{ Bq} \cdot \text{m}^{-3}$, and $5.3 \times 10^7 \text{ Bq} \cdot \text{m}^{-3}$, respectively. It indicates that the activity concentration of tritium in water placed in the air of $3,000 \text{ Bq} \cdot \text{m}^{-3}$ and $15,000 \text{ m}^3$ could exceed the release limit of tritiated water in Korea ($4 \times 10^7 \text{ Bq} \cdot \text{m}^{-3}$) [9]. If the initial activity concentration of tritium in air is $300,000 \text{ Bq} \cdot \text{m}^{-3}$, the equilibrium activity concentration of tritium in water placed in the air of above 150 m^3 exceeds the release limit of tritiated water.

Changes of the activity concentration of tritium in air and that in water at equilibrium with relative humidity at 20°C are shown in Table 2.

The lower the initial relative humidity of air, although the activity concentration of tritium is the same, the higher the activity concentration of tritium in air and that in water at equilibrium. The reason is that the lower the relative humidity, the more evaporation of water until it reaches the equilibrium and the volume of water decreases.

As described above, we briefly estimates the change of the activity concentration of tritium in water when the water is placed in the air containing tritiated water vapor. We have

Fig. 1. Activity concentration of tritium in air and that in water at equilibrium (20°C).

Table 1. Activity Concentration of Tritium in Air and That in Water at Equilibrium^{a)}

Initial activity concentration of tritium in air ($\text{Bq} \cdot \text{m}^{-3}$)	Phase	Activity concentration of tritium in air and that in water at equilibrium, 20°C ($\text{Bq} \cdot \text{m}^{-3}$)						
		Volume of air (m^3)						
		10	100	1,000	5,000	10,000	15,000	20,000
1,000	Air	0.16	1.6	16	76	150	210	270
	Water	1.0×10^4	1.0×10^5	1.0×10^6	4.8×10^6	9.3×10^6	1.4×10^7	1.8×10^7
3,000	Air	0.47	4.7	47	230	440	640	820
	Water	3.0×10^4	3.0×10^5	3.0×10^6	1.4×10^7	2.8×10^7	4.1×10^7	5.3×10^7
10,000	Air	1.6	16	160	760	1,500	2,100	2,700
	Water	1.0×10^5	1.0×10^6	1.0×10^7	4.8×10^7	9.3×10^7	1.4×10^8	1.8×10^8
30,000	Air	4.7	47	470	2,200	4,400	6,400	8,200
	Water	3.0×10^5	3.0×10^6	3.0×10^7	1.4×10^8	2.8×10^8	4.1×10^8	5.3×10^8
100,000	Air	16	160	1,600	7,600	15,000	21,000	27,000
	Water	1.0×10^6	1.0×10^7	1.0×10^8	4.6×10^8	9.3×10^8	1.4×10^9	1.8×10^9
300,000	Air	47	470	4,700	23,000	44,000	64,000	82,000
	Water	3.0×10^6	3.0×10^7	3.0×10^8	1.4×10^9	2.8×10^9	4.1×10^9	5.3×10^9

^{a)}Initial air of 50% relative humidity at 20°C , initial volume of water of 1 m^3 .

Table 2. Changes of Activity Concentration of Tritium in Air and That in Water at Equilibrium with RH at 20°C^{a)}

Volume of air (m ³)	Phase	Activity concentration of tritium in air and that in water at equilibrium, 20°C (Bq·m ⁻³)					
		Initial activity concentration of tritium in air					
		3,000 Bq·m ⁻³			300,000 Bq·m ⁻³		
		10% RH	50% RH	100% RH	10% RH	50% RH	100% RH
10	Air	0.47	0.47	0.47	47	47	47
	Water	3.0×10^4	3.0×10^4	3.0×10^4	3.0×10^6	3.0×10^6	3.0×10^6
100	Air	4.7	4.7	4.7	470	470	470
	Water	3.0×10^5	3.0×10^5	3.0×10^5	3.0×10^7	3.0×10^7	3.0×10^7
1,000	Air	47	47	46	4,700	4,700	4,600
	Water	3.0×10^6	3.0×10^6	3.0×10^6	3.0×10^8	3.0×10^8	3.0×10^8
5,000	Air	240	230	220	23,000	23,000	22,000
	Water	1.4×10^7	1.40×10^7	1.40×10^7	1.50×10^9	1.50×10^9	1.40×10^9
10,000	Air	470	440	410	47,000	44,000	40,000
	Water	3.0×10^7	2.8×10^7	2.6×10^7	3.0×10^9	2.8×10^9	2.6×10^9
15,000	Air	700	640	570	70,000	64,000	57,000
	Water	4.5×10^7	4.1×10^7	3.6×10^7	4.5×10^9	4.1×10^9	3.6×10^9
20,000	Air	940	820	720	94,000	82,000	72,000
	Water	6.0×10^7	5.3×10^7	4.6×10^7	6.0×10^9	5.3×10^9	4.6×10^9

RH, relative humidity.

^{a)}Initial volume of water of 1 m³.

confirmed that water with initially no tritium could be changed to tritiated water with high activity concentration. At present, we are planning to conduct experiments to verify that this phenomenon occurs in the field.

Conflict of Interest

No potential conflict of interest relevant to this article was reported.

Author Contribution

Methodology: SP Yim, CK Lee. Investigation: SP Yim, CK Lee. Writing - original draft: SP Yim. Writing - review & editing: CK Lee.

References

1. Cho HH. Tritium in air of Wolsung power plant, 1,800 times, is that normal? [Internet]. Seoul, Korea: Media Today; 2021 [cited 2021 Mar 21]. Available from: <http://www.mediatoday.co.kr/news/articleView.html?idxno=211538>.
2. Valentine AM. An investigation of a bubbler tritium sampler (Report No. LA-3916). Los Alamos, NM: Los Alamos Scientific Laboratory; 1968.
3. McConnon D. The use of water as a sampling medium for tritium oxide (Report No. BNWL-CC-547). Richland, WA: Battelle Northwest Laboratory; 1970.
4. Osborne RV. Sampling for tritiated water vapor. Proceedings of the 3rd International Congress of the International Radiation Protection Association; 1973 September 9–14; Washington, DC. p. 1428–1433.
5. Yim SP, Lim IC, Ahn GH, Seoul CW, Lee CK. Performance test of ice packed respirator for tritium removal. Proceeding of the 36th Annual Radioactive Waste Management Symposium (WM); 2010 March 7–11; Phoenix, AZ.
6. Shin JS, Lee JK. Wet-type tritium filter cartridge. KR Patent 20-0472036, 2014.
7. Radiation Safety Team of Wolsung Nuclear Power Plant #2. Introduction to performance test on tritium respirator at Wolsung Nuclear Power Plant #2 [Internet]. Gyeongju, Korea: Wolsung Nuclear Power Plant; 2013 [cited 2021 Mar 21]. Available from: https://www.kins.re.kr/nsic_info/nsic/2013/pdf/13_5.pdf.
8. Sepall O, Mason SG. Vapor/liquid partition of tritium in tritiated water. Can J Chem. 1960;38:2024–2025.
9. Nuclear Safety and Security Commission. Standards for Radiation Protection (NSSC Notice No. 2014-034) [Internet]. Sejong, Korea: National Law Information Center; 2014 [cited 2021 Mar 21]. Available from: <https://www.law.go.kr/LSW/admRulInfoP.do?admRulSeq=2100000006874&lsId=35174&chrClsCd=010202>.