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# Dynamic equations of tritium concentration during wastewater (light water) containing tritium electrolysis

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## ABSTRACT

Nuclear facilities often produce some wastewater containing tritium, which cannot be discharged directly because of the radioactivity of the tritium. It is of great significance to study the dynamic change rules of tritium concentration during wastewater containing tritium electrolysis with SPE for tritium enrichment for the estimation of the environmental tritium concentration and the recovery of tritium resources. The dynamic equation of low tritium is established on account of the electrolytic reaction and the balance equation of hydrogen isotope exchange in gas and liquid phases. The theoretically dynamic change rules of tritium concentration are studied while continuous feeding wastewater (light water) containing tritium electrolysis and batch feeding wastewater (light water) containing tritium electrolysis, so as to lay a necessary theoretical basis for the research of recycling of wastewater containing tritium.

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## Introduction

Nuclear facilities often produce some wastewater containing tritium, which cannot be discharged directly because of the radioactivity of the tritium. The tritium separation from the wastewater is feasible for recovering the heavy water, diminishing the radioactive waste and reducing the environmental burden. Especially in 2012, the Japanese nuclear power station leakage accident caused the attention of the world. The processing of radioactive wastewater emissions seriously affects the environment security and the lives of the people. Therefore, the research of water detritiation technology has very important meaning. Several processes for the tritium separation have been developed, for example, distillation, chemical exchange, catalytic exchange, and electrolysis at present. Enrichment of

tritium with electrolysis is a well-known technique for hydrogen isotope separation. The method has many advantages that it has high separation factor (SF), and can be performed in small-scale instruments. The electrolysis with the solid polymer electrolyte (SPE) has been used for tritium enrichment for the estimation of the environmental tritium concentration and the recovery of tritium resources.

Y. Ogata et al. [1] have studied electrolysis with SPE by experiments, which is used for tritium separation from heavy water. Its result shows that the tritium separation factor of the heavy water,  $SF_{D/T}$  was about 2,  $SF_{H/T}$  of about 12, for stainless steel and Ni cathodes at 20 °C. The SF values were in good agreement with the results of KaKiuchi [2] for the tritium enrichment of the environmental water, using conventional apparatuses [3–5].

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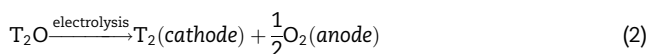
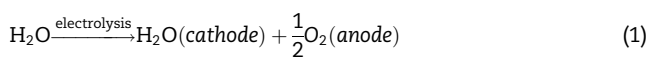
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O. M. Ivanchuk et al. [6] have studied isotope effect during heavy water electrolysis with SPE by experiments. The approximate calculation formulas of separation factor are obtained in the bi-component condition by experimental study.

All of these studies are experimental, which are bound to be affected by the operating conditions and human factors. It is of great significance to study the dynamic change rules of tritium concentration during wastewater containing tritium electrolysis with SPE. Though previously S. K. Malhotra et al. [7] studied material balance of tritium electrolysis of heavy water, and J. B. Zhou et al. [8] studied dynamic equation and the calculation methods of impurity hydrogen during high concentration heavy water electrolysis, the theoretical research on this system of light water containing tritium has not been reported. The purpose of our work is to study theoretically dynamic change rules of tritium concentration during wastewater (light water) containing tritium electrolysis, so as to lay a necessary theoretical basis for the further research.

### Electrode reactions and balance equations in electrolysis

The following reactions occur during wastewater containing tritium electrolysis [9,10]:



where  $\text{T}_2\text{O}$  is the heavy water. Oxygen is generated on the anode and released, while on the cathode, hydrogen, tritium and mixed molecules of hydrogen and tritium are produced (Fig. 1). Based on the balance reaction, Eq. (3), we can get Eq. (4), thus  $K_1$  can be calculated according to (4):



$$K_1 = \frac{[\text{HT}]^2}{[\text{H}_2][\text{T}_2]} \quad (4)$$

where HT is the mixed molecular hydrogen and tritium atom.  $K_1$  is the reactive equilibrium constant that is 2.57 at 25 °C and 1 atm pressure based on Ref. [11]; [HT],  $[\text{H}_2]$  and  $[\text{T}_2]$  are molar fractions, respectively. If the component of tritium atoms is 0.1% in cathode gases, the HT molar fraction (ratio) is calculated to be 0.19969% from (4) and the molar ratio in the form of  $\text{T}_2$  molar rate is only 0.00015547%. This shows that tritium in the gas phase presents mostly in the form of HT mixture

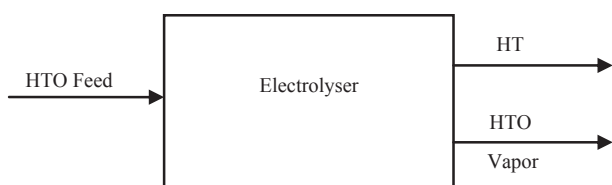


Fig. 1 – Input and output of tritium in the electrolyzer.

molecules. And therefore, when the water of trace tritium is electrolyzed, tritium produced on the cathode presents mainly in the form of mixture molecules (HT) of hydrogen and tritium.

In the same way, it accords with the balance equation,



where HTO is mixed molecular water. That is

$$K_8 = \frac{[\text{HTO}]^2}{[\text{H}_2\text{O}][\text{T}_2\text{O}]} \quad (6)$$

where  $K_8$  is the reactive equilibrium constant; [HTO],  $[\text{H}_2\text{O}]$  and  $[\text{T}_2\text{O}]$  are the molar fractions, respectively (concentration). This shows that tritium in the liquid phase presents mainly in the form of HTO molecules when the purity of light water is very high, i.e. the content of heavy water is very small [11].

### Dynamic balance equations of tritium

According to calculations from Eqs. (4) and (6) and the above discussion, we can know when solution of low tritium concentration is electrolyzed to generate tritium, and the presence of  $\text{T}_2$  and  $\text{T}_2\text{O}$  (vapor) can be ignored, tritium in gas and liquid phases presents, respectively, in the forms HT and HTO mixture molecules. We attempt here to give a complete material balance of tritium escaping from the system in the form of HT gas and as HTO vapor.

Considering an electrolyzer (Fig. 1), in which the  $\text{H}_2$  gas formed is let out,  $\text{H}_2\text{O}$  vapor escapes along with the gases evolved, and wastewater containing tritium is continuously added to maintain a constant level of electrolyte in the cell, the rate of increase in the number of HTO moles in the electrolyzer per unit time can be written as

$$\frac{d}{dt}n_{\text{HTO}}(\text{EL}) = \frac{d}{dt}n_{\text{HTO}}(\text{F}) - \frac{d}{dt}n_{\text{HTO}} - \frac{d}{dt}n_{\text{HTO}}(\text{V}) \quad (7)$$

where the term with (F) is the rate of addition of HTO in moles per second, the term with  $n_{\text{HTO}}$  is the rate of escape of HT with gas, and the term with (V) is the rate of escape of HTO as vapor.

The rate of escape of HT can be calculated from the electrolytic separation factor for tritium. Thus, if  $n_E$  is the electrolysis rate (mol/s) and  $\alpha_E$  is the electrolytic separation factor, then for low tritium concentration,  $\alpha_E$  can be defined as

$$\alpha_E = \frac{(\text{T}/\text{H})_{\text{liq}}}{(\text{T}/\text{H})_{\text{gas}}} \quad (8)$$

The number of HT moles escaping per second is

$$\frac{d}{dt}n_{\text{HT}} = \frac{n_E \cdot x}{\alpha_E} \quad (9)$$

where  $x$  is the mole fraction of HTO in the electrolyzer at time  $t$ .

The rate of escape of HTO as vapor can be calculated from vapor pressure  $P_V$  of wastewater containing tritium at the temperature of electrolysis, pressure  $P$  in the electrolyzer, flow rate  $F_g$  of the gases evolving out of the electrolyzer, and the

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