

Development Strategy for Tritium(^3H) Extraction·Removal from Liquid Radioactive Waste of Nuclear Power Plants

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1. Introduction

Among the nuclides released into the environment with nuclear power plant (NPP) operations, the tritium (half-life: 12.3, 18.6 keV, ^3H), which is a beta emitter, is the most interesting nuclide among researchers. The biological half-life of HTO ingested in the human body is about 9.7 days, and most of it is discharged out of the body. Many radiochemistry researchers have been researching to remove ^3H from the contaminated water because it has a genetic effect when it is replaced with hydrogen when absorbed into the body. However, ^3H is present in various forms such as H_2O , D_2O , HT and HTO in water and has physical and chemical properties similar to water, making it difficult to develop ^3H separation and removal technologies. [1-2].

Among the technologies developed to date, commercialized technologies include LPCE (Liquid Phase Catalytic Exchange) technology used in domestic heavy water reactors NPP (Wolsung) and CECE (Combined Electrolysis Exchange) technology used in Fukushima accident site in Japan. Both of these technologies separate ^3H by electrolysis method and cryogenic catalyst exchange method and require very expensive facility and have also a very small treatment capacity less than 100 kg/hr. For this reason, there is a limit to the processing capacity to treat ^3H from a large amount of contaminated water. The Fukushima NPP accident in Japan has generated more than 1.2 million tons of contaminated water so far, which forces the Japanese government to seriously consider ocean discharge due to a lack of storage capacity. In Korea, as ^3H is detected from urine samples of residents around Wolsong NPP, which operates heavy water reactors, concerns about ^3H and social interest in removal technology are rapidly increasing.

This research paper focuses on the development of high-capacity / high-efficiency ^3H removal technology to increase the treatment capacity, which is a limitation of the commercially available ^3H removal technology. In this works, we will introduce an approach strategy for the development of more advanced ^3H removal technology through a review of the technologies that have been developed to date, and evaluate the detailed characteristics of each technology through empirical experiments on technologies with high potential.

1.1 Objectives

The technology to be developed is a new concept of hybrid type ^3H removal technology. We plan to develop a technology of 100 L / hr that is cheaper than the current

commercialized system construction cost, can improve the removal efficiency for ^3H by 80%, and can increase the processing capacity.

2. Method and Results

2.1 Technical Characteristics for Removing ^3H

There are four technologies currently being considered. As shown in the experimental scene in Fig. 1, it is a technology to decompress and vacuum separate ^3H by using alumina or activated carbon whose surface is modified as an adsorbent. In this technology, tritiated water (HTO), which has a relatively large mass, is adsorbed to the adsorbent through the distillation under reduced pressure, where gas phase H_2O is condensed and recovered. As a result of experiments using this technique, the removal efficiency of ^3H was achieved about 45%, but further research on continuous processing and regeneration of the adsorbent is required.



Fig. 1, Experimental equipment of ^3H removal by adsorbent

The second technique uses an ion separation membrane coated with LiMn_2O_4 spinel-structure manganese oxide containing Li, which is well known as a battery material (fig. 2). The technology is an improved ^3H removal technology that promotes the removal rate and removal ability than the previously introduced technology by supporting $\lambda\text{-MnO}_2$ (HMnO_2) on the ion exchange membrane using the property that Li in the lattice is easily substituted with H^+ ions. Recently, it has been reported that about 50-60% of ^3H removal efficiency was obtained through experiments [3]. In addition, a variety of experiments are currently under way by signing an agreement with a company, TEPCO (Japan), that developed this technology.

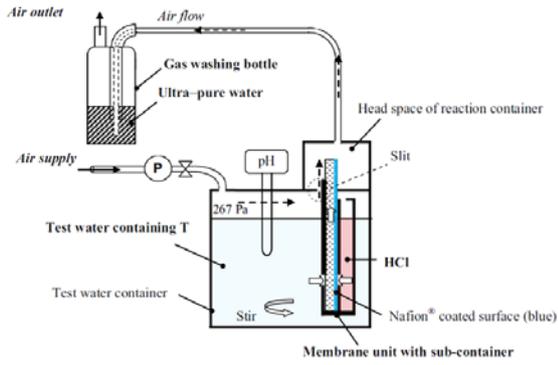


Fig. 2, Schematic diagram of ^3H removal using $\lambda\text{-MnO}_2(\text{HMnO}_2)$, reproduced for permission [3]

In the third technology, after the ^3H ions are replaced with Al ions while undergoing a two-step ion exchange process in which Al-supported resin is used, ^3H undergoes desorption and concentration processes from the resin, and finally adsorbed, enriched and stored in zeolite (fig. 3). This technology has been studied largely by DOE in the United States. Since the resin recycling process is possible, it can be configured as a continuous processing process, and the removal efficiency is known to be about 70-80%. In addition, a pre-treatment process is essential to pre-remove various interfering ions or organic substances contained in contaminated water.

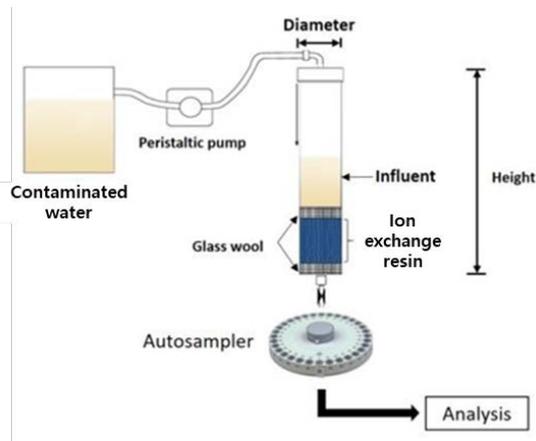


Fig. 3, ^3H removal process via technology of ion exchange resin

The fourth technique separates ^3H through an inorganic graphene oxide separation membrane and a zeolite molecular sieve membrane (fig. 4). Since this technology includes multiple layers of separation membranes, there is a space constraint that requires a huge area when making a large-capacity system. Therefore, research is currently being conducted to reduce the installation space by increasing the contact efficiency. This technology is also actively being researched around the United States' DEO, and is one of the technologies that has the advantage of being capable of large-capacity processing.

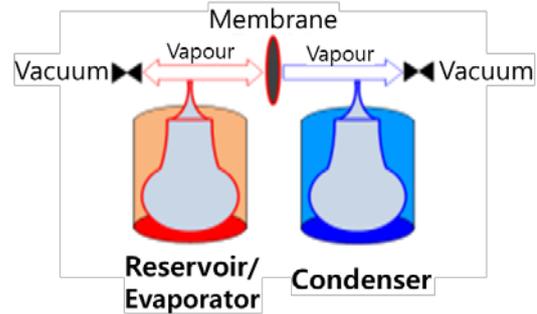


Fig. 4, ^3H removal process via membrane technology

2.2 Strategy

Currently, basic experiments and device configuration for each of the aforementioned technologies are in progress. As shown in fig. 5, additional pre-treatment process removing ions, particulates and suspended solids of the new concept will be introduced to increase the removal efficiency. We plan to develop a hybrid composite process that selects and combines the most probable technologies among the four technologies mentioned above. The treatment of separated and concentrated HTO will be reviewed with the possibility of cement solidification and recycling possible depending on the purity of separation.

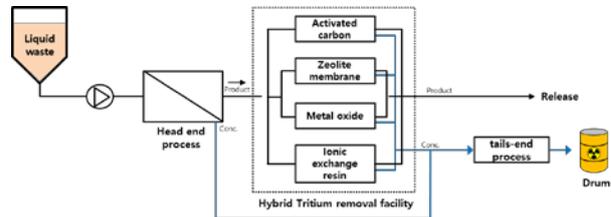


Fig. 5, Conceptual diagram of ^3H removal technology

3. Summary

Researchers from all countries with NPP have been constantly researching to develop the technologies to separate and remove the tritiated water (HTO) from contaminated water. However, the development of large-capacity treatment technology for the separation and removal of tritiated water requires a lot of time and effort, as tritiated water has similar properties to light water, both physically and chemically. Our research institute has been continuously discussing with related researchers who have various experience for developing the ^3H removal technology. To conduct research based on technologies that are likely to remove ^3H , four domestic universities in similar fields were selected and then, it plans to perform step by step from laboratory-scale research to semi-pilot level research. Through this independent study, we intend to develop a process capable of regeneration of ^3H adsorbents and continuous

processing. In addition, after developing and manufacturing a hybrid-type complex ^3H removal system that combines advantages for each technology, performance tests will be conducted. As it is still in the early stages of research, this paper briefly describes the introduction and promotion strategies of the technologies to be approached. The progress of each development stage will be introduced annually through this journal.

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