Palladium Alloy Membrane Characteristics for Purifying Hydrogen Isotopes

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1. Introduction

Tritium is a radioactive isotope of hydrogen and it has a half-life of 12.3 years; it decays to He-3 by emitting a low energy beta radiation with an average energy of 5.7 keV and a maximum energy of 18.6 keV. Transfer of environmental tritiated water to humans takes place via an inhalation, diffusion through the skin and ingestion. Radioactive waste containing tritium is continuously generated by the nuclear industry in, for example, nuclear reactor operations and a radioisotope production, and in medical research. Methods for removing tritium from a liquid waste provide an alternative to the control of tritium emissions and a personnel exposure. Combined electrolysis and catalytic exchange process is a very effective method to remove small quantities of tritium from light or heavy waste water streams. The process consists of three main steps: (a) A front end step that exchanges the tritium to a less toxic hydrogen phase. This can be performed either through a chemical exchange in the presence of a platinum supported catalyst or through the decomposition of water. (b) A back end process that purifies the tritiated hydrogen gas which evolved from the electrolysis. This can be performed through a palladium alloy membrane separator. (c) A means of storing the concentrated gas safely. Uranium or ZrCo metal is used if the storage is temporary; titanium is mainly employed for long term storage. To have a better knowledge of the tritiated hydrogen gas purification process, a test rig for the palladium alloy membrane performance has been installed. This rig is described herein, and the representative results of the hydrogen purification performance are presented [1, 2].

2. Overall Detritiation Process

In this section the overall detritiation process is shown in Figure1. The process treats tritiated water with a detritiation factor of ten. The process makes use of a hydrophobic catalyst in a liquid-phase exchange column. Tritiated water flows downward countercurrently to a rising stream of a tritiated hydrogen gas generated in the electrolytic cells. The liquid stream entering at the top of the catalyst column is made up of distilled water. The hydrogen stream from the electolyser is fed to the bottom of the column. As this hydrogen stream is enriched in heavier isotopes, a part of the stream is purified in a membrane permeator in which the water vapor and a small amount of oxygen gas are separated. The upstream hydrogen gas is vented to the atmosphere through a stack. This stream can also be fed to a fuel cell to provide a reflux to the process. Finally the purified waste hydrogen gas in the membrane permeator is safely stored in a metal getter bed or sent to the isotope separation process for a further treatment.

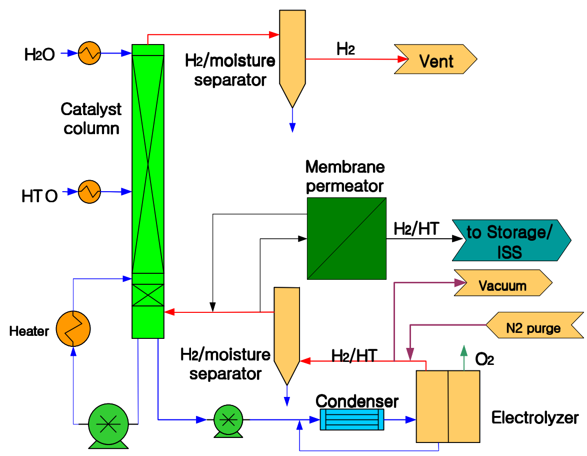

Figure1. Detritiation Process Schematic

3. Membrane Permeator Modeling

The membrane is clearly the most important part of the separation process. [3, 4] The authors selected the palladium alloy membrane for the hydrogen purification process by considering the membrane properties, such as the chemical resistance, mechanical stability, thermal stability, high permeability, and a stable operation. Hydrogen permeates through the palladium alloy membrane by adsorbing and dissociating into atoms at the metal surface, followed by a dissolution and diffusion of the hydrogen atoms through the metal under the influence of the pressure gradient applied. At the permeate side of the hydrogen atoms they recombine to the molecular form and desorb from the surface.

Sieverts and Kumbhaar observed that the solubility of hydrogen in the α-phase of the palladium has been found to be directly proportional to the square root of the hydrogen pressure [3]. This proportionality is due to the dissociation of hydrogen into atoms at the surface of the palladium. The flux of hydrogen, J, can be expressed in terms of the solution-diffusion model as

$$J = P\,[\sqrt{p'_{H2}} - \sqrt{p''_{H2}}\,] / \delta$$

where J is the flux, $cm^3/cm^2/s$, p'_{H2}, p''_{H2} are the hydrogen pressures, kgf/cm^2, δ is the thickness of the palladium, cm, P is the permeability constant, $cm^3 \cdot cm/cm^2/s/\sqrt{kgf/cm^2}$, $P = 6.52\times10^{-6}\exp(-6.38/RT)$.

4. Experimental

The authors have installed a rig to examine the applicability of the solution-diffusion model to a palladium alloy membrane Model HP-2 of Johnson Matthey Inc. Figure2 shows the design concept of the membrane permeator for the detritiation process of Figure1.

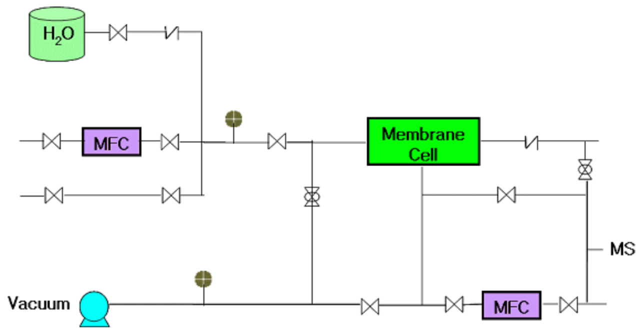

Figure2. Membrane Process Schematic

5. Results

Figure3 shows a preliminary experimental result of the membrane permeator of Figure2. As expected, a very high pure hydrogen could be obtained as permeate during the time interval of 40 to 80 min.

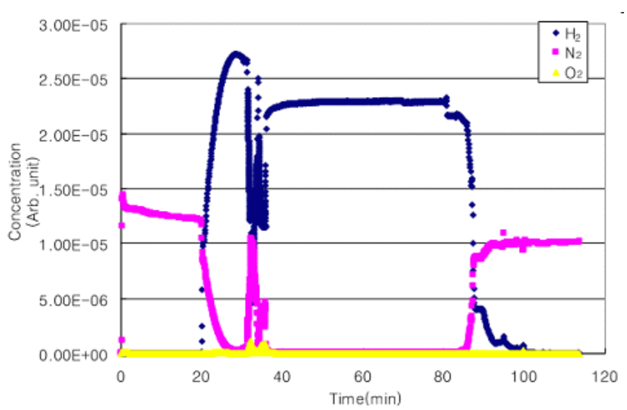

Figure3. Purity of the Permeate

Figure4 shows a preliminary experimental permeation result of the membrane permeator. The authors found that the membrane follows the solution-diffusion model very well.

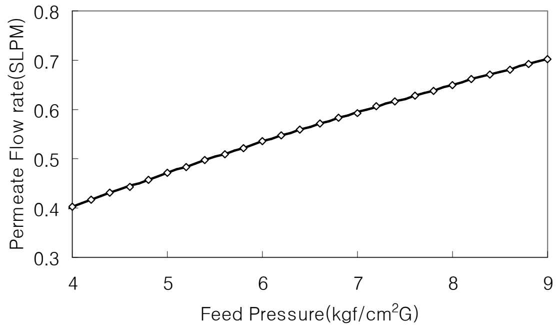

Figure4. Flow rate of the Permeate

6. Conclusion

The authors selected the palladium alloy membrane for hydrogen purification in the detritiation process. The authors have installed a rig to examine the applicability of the solution-diffusion model to a palladium alloy membrane Model HP-2 of Johnson Matthey Inc. Very high pure hydrogen could be obtained as a permeate

The solution-diffusion model can be a useful tool for designing a membrane permeator. The authors applied the solution-diffusion model to a palladium alloy membrane to obtain the design parameters for the detritiation process. The authors found that the membrane follows the solution-diffusion model very well.

Acknowledgement

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