Physicochemical conditions for effective hydrogen isotope storage and delivery

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In this study, a hydrogen isotope storage bed and a test rig were designed and fabricated to investigate the characteristics of a hydrogen isotope storage bed and obtain the effective physical-chemical conditions for hydrogen isotope storage and delivery. Depleted uranium (DU) was chosen as the storage material. Protium (H_2), the properties of which are virtually identical with those of tritium (T_2), was used to study the performance of the hydrogen isotope storage bed depending on radiochemical properties of uranium hydride. Under newly applied physical-chemical conditions, much higher and constant hydrogen recovery and delivery rates were obtained.

I. INTRODUCTION

Ternary fission is a comparatively rare nuclear fission process whereby the nuclei is split into three fission fragments. Tritium is produced this way in all nuclear reactors. The more important source of tritium is the neutron activation of deuterium in pressurized heavy water reactors (PHWRs), which use heavy water (D_2O) as the primary coolant and moderator. The large amount of deuterium oxide in the PHWRs is continuously exposed to a neutron flux. Substantial quantities of tritium are produced by the neutron activation of deuterium in the reactor core. Tritium is a radioactive isotope of hydrogen that decays into ^{3}He with a 12.3-year half-life through a beta decay. Tritium is a major source of operator exposure to radiation in a nuclear reactor, and at the same time is a key material used as a nuclear fusion fuel. To minimize this radiation exposure and utilize it for nuclear fusion, the storage and delivery of tritium in a less hazardous chemical form are important. Tritium can be stored either directly as a gaseous form or in a metal tritide form. The tritium stored in both forms can be recovered easily. The storage and delivery of tritium in a gaseous form is a simple well-established technology. However, storage of tritium in a metal tritide form is preferred because of the concern for gas leakage. For these reasons, many researches on hydrogen isotope storage materials, including zirconium cobalt (ZrCo), lanthanum pentanickel ($LaNi_5$), depleted uranium (DU), zirconium (Zr), erbium (Er), yttrium (Y), and titanium (Ti), have been widely studied (Ref. 1-4). However, researches on hydrogen isotope storage beds containing metal tritides have not been intensively conducted. In this study, we designed and fabricated a hydrogen isotope storage bed and a test rig to investigate the performance of the hydrogen isotope storage bed in different conditions and obtain better operation scenarios (Ref. 5). DU has been used as a storage material because DU does not have disproportionation at high temperatures or pressures. DU tritide can be heated to a high temperature at which it is possible to deliver the hydrogen isotopes directly. In addition, the temperature does not profoundly affect the structures of the hydrogen isotope storage bed. Protium (H_2) was used instead of tritium because the chemical characteristics of protium are virtually identical with those of tritium (T_2).

II. HYDROGEN ISOTOPE STORAGE BED & EXPERIMENTAL APPARATUS

The hydrogen isotope storage bed consists of a primary vessel and a secondary vessel. Four cylindrical and disc thermal reflectors were placed between the two vessels. The primary vessel contains approximately 1,893.75 grams of DU, which theoretically stores approximately 23.598 grams of hydrogen. It has copper foam inside for effective heat transfer, a filter tube for hydrogen inflow and outflow, and two cable heaters for temperature control. The primary vessel, secondary vessel, and measuring tank are connected to the manifold. The primary vessel is directly connected to the measuring tank with the hydrogen loading and unloading nozzle. To vacuum the hydrogen isotope storage bed and the measuring tank, a rotary pump and a turbo molecular pump are employed. To measure the vacuum, pressure, and temperature conditions, several vacuum gauges, pressure gauges, and thermocouples were placed at strategic locations (Ref. 5).

III. RESULTS AND DISCUSSION

III.A. Vacuum annealing

Vacuum annealing was conducted prior to the hydrogen recovery and delivery. The DU used was sufficiently powderized by repetitive hydriding and dehydriding in advance. The hydrogen isotope storage bed was heated at 450°C for over 12 hours with vacuum pumping to remove volatile impurities and oxides from the surface of the DU and the

copper forms. During the vacuum annealing, the primary and secondary vessels remained at a vacuum level of 10^{-2} and 10^{-4} Torr, respectively. The primary vessel was heated up to a maximum of 521°C.

III.B. Hydrogen recovery (Hydriding)

The hydrogen recovery was conducted at various initial temperatures of the DU (30, 225, and 419°C) and pressures of hydrogen (400, 700, and 900 Torr at room temperature). The secondary vessel was in a vacuum state during the hydrogen recovery because the temperature increments of the DU by the exothermic reaction were not significant enough to affect the performance of the hydrogen isotope storage bed. Although a uranium-hydrogen reaction is widely known to be the most active at between 200 and 250°C, the increment in hydrogen absorption rates at such temperature was negligible, whereas the temperature of DU remained higher than the other cases during hydrogen recovery. The effects of the initial temperatures of DU on the hydriding performance were negligible. When we increased the initial pressure of hydrogen in the measuring tank, both the hydrogen absorption rates into the DU and the temperature of the DU increased. The temperature increasing rates of the DU by the exothermic reaction were also increased. The effects of the initial pressures of hydrogen on the hydriding performance were remarkably high.

III.C. Hydrogen delivery (Dehydriding)

The hydrogen delivery was conducted with different operation scenarios. The scenarios are a typical constant heating, linear progression heating, and linear progression heating with preheating. The total elapsed times to desorb 75% of the initial absorbed amount of hydrogen into the DU were approximately 4.8, 3.6, and 2.6 hours when typical constant heating, linear progression heating, and linear progression heating with preheating were applied. The application of a simple operation scenario improved the hydriding rates significantly and led to a steady hydrogen delivery.

IV. CONCLUSIONS

The tritium storage and delivery performance in different physical-chemical conditions are investigated. To investigate the effects of physical-chemical conditions on hydrogen recovery and delivery, the initial temperatures of DU and the initial pressure of hydrogen were manipulated, and three different hydrogen delivery operation scenarios were suggested and applied. The hydrogen recovery performance of the hydrogen isotope storage bed was significantly affected by the initial pressure of the hydrogen but not by the initial temperature of the DU. With the application of higher initial pressure of hydrogen, much higher and constant hydrogen recovery rates were obtained. The hydrogen delivery performance of the hydrogen isotope storage bed was remarkably improved with simple manipulations of operation. With the combination of preheating and linear progression heating, much higher and constant hydrogen delivery rates were obtained. Our experiment results can be used for the safe storage and delivery of tritium not only for environmental protection and waste management in various nuclear fission reactors but also for future commercial use of tritium in isotope industries and nuclear fusion fuel systems.

ACKNOWLEDGMENTS

This research was supported by the National Fusion Research Institute and the National R&D Program through the National Research Foundation of Korea (NRF), which is funded by the Ministry of Science and ICT, and Ministry of Trade, Industry and Energy (2009-0070685). The views and opinions expressed herein do not necessarily reflect those of the ITER Organization. ITER is a Nuclear Facility INB-174.

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