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Assessment of amount and concentration of tritium in HTTR-IS system based on tritium behavior during high-temperature continuous operation of HTTR

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ABSTRACT

The tritium concentration in the high temperature engineering test reactor (HTTR) was measured during high-temperature (950 °C) continuous operation for 50 days, which is the first measurement of tritium concentration in the secondary helium circuit during operation using the intermediate heat exchanger, to develop an assessment base for evaluating tritium behavior in the high temperature gas-cooled reactor hydrogen production system. The tritium concentration in the primary helium gas increased after startup and peaked at 1.6×10^{-1} Bq/cm^3 (STP) at 60% reactor power. Then, it decreased slightly over the course of the normal operation phase. This decrease could be attributed to the effect of tritium chemisorption on graphite. The tritium concentration in the secondary helium gas was slightly lower than that in the primary helium gas. The tritium concentration in the secondary helium gas peaked at 4.7×10^{-2} Bq/cm^3 (STP) during the power enhancement phase. Thereafter, it decreased gradually to 2.2×10^{-2} Bq/cm^3 (STP) at the end of the normal power operation phase. As assessed herein, the concentration and total quantity of tritium in the secondary helium cooling system of the HTTR-iodine sulfur (IS) system can be maintained below the regulatory limits, which implies that the hydrogen production plant can be exempt from the safety function of the nuclear facility.

1. Introduction

Hydrogen is a clean energy medium that releases no greenhouse gases when consumed or burned. A high temperature gas-cooled reactor (HTGR) is one of the candidates for an energy supply system to produce massive amounts of hydrogen from water by thermochemical methods without releasing CO_2. Japan Atomic Energy Agency (JAEA) has conducted research and development (R&D) on HTGR technology, for example, construction and operation of the first Japanese HTGR called high temperature engineering test reactor (HTTR) (Saito et al., 1994), design of advanced commercial HTGR plants (i.e., GTHTR300 series (Kunitomi et al., 2007) and HTR50S (Ohashi et al., 2013)), and the hydrogen production technology that employs the thermochemical water-splitting iodine–sulfur (IS) process (Russell et al., 1976). Parallel to these R&D efforts, JAEA has studied the safety requirements and design considerations for the HTGR hydrogen production system to ensure reactor safety against postulated events initiated in the hydrogen plant as well as to build the coupled hydrogen plant under conventional industrial regulations (Sakaba et al., 2008). One of the requirements for classifying a hydrogen production plant as a "non-nuclear facility" is that the plant must not require the safety function for radioactive waste storage (Sato et al., 2013). In the HTGR hydrogen production system, tritium is generated by a ternary fission reaction in fuel and by the neutron absorption reactions of ^{6}Li, ^{7}Li, ^{10}B, and ^{3}He, which are present in the core graphite, control rods, and helium coolant, respectively (Ohashi and Sherman, 2007). The generated tritium is circulated in the primary coolant and permeates from the primary coolant loop to the intermediate coolant loop through the heat transfer tubes of the intermediate heat exchanger (IHX) and from the intermediate coolant loop to the hydrogen plant through the heat transfer tubes of the process heat exchangers. Therefore, in safety design, significant focus should be placed on tritium transport from the nuclear reactor to the hydrogen production plant.

The behavior of tritium in HTGRs was evaluated in several countries in the 1970s (e.g., the Dragon Reactor in the U.K. (Forsyth, 1972), Peach Bottom HTGR in the USA (Wichner and Dyer, 1979), and AVR in Germany (Steinwarz et al., 1980)). Data

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Nomenclature

Acronyms

CCT	cold charcoal trap
EFPD	equivalent full power days
HTGR	high temperature gas-cooled reactor
HTTR	high temperature engineering test reactor
IHX	intermediate heat exchanger
IS	iodine–sulfur
JAEA	Japan Atomic Energy Agency
MST	molecular sieve trap
PPWC	primary pressurized water cooler
R&D	research and development
SPWC	secondary pressurized water cooler
THYTAN	tritium and hydrogen transportation analysis code

Symbols

C_{cat}	tritium concentration in catalyst immersion water [Bq/g]
C_{HT}	HT concentration in helium gas [Bq/cm^3 (STP)]
C_{HTO}	HTO vapor concentration in helium gas [Bq/cm^3 (STP)]
C_i	tritium concentration in water of i-th water trap [Bq/g]
$C_{tritium}$	tritium (HT + HTO) concentration in helium gas sample [Bq/cm^3 (STP)]
Q_{He}	helium gas volume through helium gas sample processing train [cm^3 (STP)]
R_i	net count rate of water in vial sampled from i-th water trap and catalyst immersion water [cpm]
$W_{V,i}$	water weight in vials sampled from i-th water trap [g]
$W_{w,cat}$	water weight in catalyst immersion container [g]
$W_{w,i}$	water weight in i-th water trap [g]

from the operation of HTGRs and laboratory experiments revealed the mechanisms of tritium production, transport, and release to the environment. According to a review by Gainey (1976), calculations with some of the HTGR data showed that tritium release should be well within the U.S. Federal guidelines contemporary with the earlier HTGR operations, so further laboratory-scale work related to tritium transport in HTGRs was discontinued. However, at that time, HTGRs only produced electricity and were not targeted toward high-temperature heat applications, and the potential for tritium migration into downstream processes was not evaluated.

In the present study, tritium concentrations at several locations in the HTTR were measured during high-temperature (950 °C) continuous operation of the HTTR for 50 days over the period January 5 to March 21, 2010, to develop an assessment base for an evaluation model consistent with the transportation path of tritium and the relevant phenomena in an HTGR hydrogen production system. This is an unprecedented measurement of the tritium concentration in both the primary and secondary helium circuits of an HTGR system equipped with IHX. The technical feasibility of design consideration against tritium transport from the reactor to hydrogen production plant for ensuring exemption of hydrogen production plants from the safety function of nuclear facilities is assessed using a representative HTGR hydrogen production system, HTTR-IS, and HTTR tritium data.

The rest of this paper is organized as follows. In Section 2, the method, results, and discussion of the tritium measurement are presented. In Section 3, estimation of the concentration and the total quantity of tritium in the HTTR-IS system are shown and discussed. Final remarks and the conclusions of this study are given in Section 4.

2. Tritium measurements

2.1. Test facility

2.1.1. HTTR facility and operating conditions

The tritium measurement test was performed during high-temperature continuous operation of the HTTR for 50 days. The HTTR has two operation modes: rated operation mode and high-temperature operation mode. The measurement was conducted in the high-temperature operation mode. Major specifications of the high-temperature operation mode are listed in Table 1. The temporal trend in reactor thermal power is described in a previous report (Goto et al., 2012).

2.1.2. Location of sampling points

Primary helium gas was sampled from seven points, as shown in Fig. 1 and listed in Table 2. The reactor inlet helium gas was sampled at point 1a, which is the mixing point of the helium returning to the reactor from the IHX and the primary pressurized water cooler (PPWC). The reactor outlet helium gas was sampled at point 1b in the gap between the honeycomb baffle plate and the PPWC inlet. The PPWC outlet helium gas was sampled at the top of the PPWC at point 1c, just before it flows into the primary helium circulator. The helium gas from the helium gas circulator outlet to the IHX was sampled at point 1d in the IHX body. The helium gas inlet by the primary purification system was sampled at point 1e between the auxiliary water cooler and the primary helium purification system. The cold charcoal trap (CCT) inlet helium gas in the primary purification system was sampled at point 1f in the molecular sieve trap (MST) outlet. The CCT outlet helium gas in the primary purification system was sampled at point 1g in the CCT outlet before it connects to the CCT bypass piping.

The secondary helium gas was sampled at the four points shown in Fig. 2 and listed Table 3. Helium gas from the IHX outlet was sampled at point 2a between the secondary pressurized water cooler (SPWC) inlet and the honeycomb baffle plate. Helium gas from the SPWC outlet was sampled at point 2b in the SPWC body. Helium gas from the secondary purification system CCT inlet was sampled at point 2c in the MST outlet. Helium gas from the secondary purification system CCT outlet was sampled at point 2d in the outlet piping of the CCT before it connects to the CCT bypass piping.

2.2. Test procedure

The primary helium gas was collected from the sampling point in the HTTR, as shown in Fig. 1, into a sampling bag using the pri-

Table 1
Major specifications of high-temperature operation mode in HTTR.

Item	Condition
Reactor thermal power [MW]	30
Reactor inlet/outlet primary helium temperature [°C]	395/950
Primary helium flow rate [t/h]	36
PPWC/IHX [t/h]	24/12
Primary helium pressure [MPa]	4.0
IHX inlet/outlet secondary helium temperature [°C]	240/870
Secondary helium flow rate [t/h]	11
Secondary helium pressure [MPa]	4.1