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# Study of tritium in the primary loop of HTR-10: Experiment and theoretical calculations

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ABSTRACT

An experimental system for the measurement of tritium in the primary loop of HTR-10 has been established, sampling gas from the helium purification system. After the restart of HTR-10 in 2015, the activity of tritium in the primary loop has been obtained at three different sampling points: upstream of the copper oxide bed, downstream of the copper oxide bed, and downstream of the molecular sieve adsorber. The experimental results indicated that the activity concentration of tritium in the primary loop of HTR-10 was 1.09 (10) × $10^4$ Bq/$m^3$ $_{STP}$. The calculated value of 2.31 × $10^5$ Bq/$m^3$ $_{STP}$ was highly conservative compared to the experimental results. The discrepancy between experimental data and theoretical result may come from uncertainties in the inputs to the generation term (He-3 in the helium coolant and Li-6 in the graphite), and the ability of the graphite in the primary coolant circuit to absorb and desorb tritium. The experimental data for the activity of tritium in the primary loop of HTR-10, HTTR, and AVR are presented and compared. The chemical form of H-3 and other factors affecting the experimental measurements are discussed.

## 1. Introduction

Understanding the production and transport behavior of radioactive isotopes in the primary coolant of advanced reactors is essential to their safety analysis. Tritium is a typical radionuclide in the primary coolant of several classes of advanced reactors: High Temperature Gas Cooled Reactors (HTGRs), Fluoride-salt-cooled High-temperature Reactors (FHRs), Molten Salt Reactors (MSRs), and high-magnetic-field fusion (Xu et al., 2017; Forsberg et al., 2017; Ohashi et al., 2008; Dipu et al., 2016; Köllő et al., 2011). At the component-level, the sorption, diffusion, and exchange reactions of H-3 from helium gas into graphite, and the permeation of H-3 through heat-exchanger tubes and other metallic components have been extensively studied (Dipu et al., 2016; Tsetskhladze et al., 1988; Miles et al., 1974; Gainey, 1976; Wu and Cao, 2010). However, to date, no system-level validation of tritium transport in an advanced reactor has been performed.

The Institute of Nuclear and New Energy Technology (INET), Tsinghua University has performed several data-collection campaigns on the circulating activity in the primary circuit of HTR-10, measuring tritium, C-14, radioactive dust in the primary coolant (Xie et al., 2015, 2017; Wei et al., 2016), and radionuclide distribution in circulating graphite pebbles (Liu et al., 2017; Li et al., 2017).

HTR-10 is the first pebble bed HTGR in China and has 10 MWth. It was designed and constructed in the 1990s, brought to criticality in 2000, and reached full power operation in 2003 (Wu et al., 2002). HTR-PM is a 250 MWth pebble bed HTGR, and it is expected to be connected to the grid in 2017 (Zhang et al., 2009, 2016). HTR-10 was used for data collection pertaining to design and safety analysis of HTR-PM, as well as for the training of HTR-PM personnel.

The current paper reports on the measurement of H-3 activity in the primary coolant of HTR-10, and compares the results with values predicted by Xu et al. (2017) based on the operational history of HTR-10. The results are also compared to experimental data from AVR and HTTR (Bäumer et al., 1990; Dipu et al., 2016).

## 2. Theoretical prediction

The tritium sources in the primary coolant of HTR-10 are: (1) activation of He-3 in the primary coolant, (2) activation of Li-6 impurities in the graphite reflectors, (3) activation of Li-6 in the fuel elements, and (4) ternary fission reactions of fissionable isotopes in the fuel elements that are not particle-encapsulated (Xu et al., 2017; Cao et al., 2017;

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Ohashi et al., 2008; Gainey, 1976). Xu et al. (2017) calculated the relative contributions of each of these sources to be 53.2%, 39.6%, 7.0%, and 0.2%, respectively. The tritium sinks from the primary coolant of HTR-10 include (1) leakage of helium from the primary circuit, (2) tritium removal in the helium purification system, (3) radioactive decay, and (4) tritium permeation through the metallic heat exchanger, and (5) tritium uptake in graphite (removable graphite sphere, and fixed graphite reflectors). Xu et al. (2017) calculated the relative contributions of each of these sinks, cumulatively over a 20-year lifetime, to be 0.8%, 92.5%, 0.01%, and 6.75%, respectively; evaluation of tritium uptake in rate the in-core graphite spheres is under study (Liu et al., 2017; Li et al., 2017).

In order to predict the expected tritium activity in the primary coolant of HTR-10, we account for the sources and sinks in a zero-dimensional model of the primary coolant circuit, based on its operational history. In this calculation, we do not account for tritium uptake into graphite. This calculation approach was used by Wu and Cao (2010) and Xu et al. (2017), and was adopted in the Final Safety Analysis Report of HTR-10, which was submitted to the State Bureau of Nuclear Safety of China in 2000. The balance equation for the integral inventory of tritium in the primary loop is:

$$\frac{dN_T^{Primary}}{dt} = k\cdot\sum_X \frac{dN_T^X}{dt} + \frac{dN_T^{He-3}}{dt} + \frac{dN_T^{F-Li6}}{dt} + \eta\cdot\frac{dN_T^{R-Li6}}{dt} - (L + P_{ur} + \lambda + P_{er})N_T^{Primary} \quad (1)$$

where, $N_T^{Primary}$ is the number of H-3 atoms in the primary loop, $k$ is fraction of uranium that is not contained in the coated fuel particles (due to broken particles and free uranium contamination in the fuel elements), $\sum_X \frac{dN_T^X}{dt}$ is the generation rate of H-3 atoms from ternary fission ($s^{-1}$), summed over all fissionable isotopes, $\frac{dN_T^{He-3}}{dt}$ is the generation rate of H-3 atoms from the activation of He-3 in the primary loop in the core ($s^{-1}$), $\frac{dN_T^{F-Li6}}{dt}$ is the generation rate of H-3 from the activation of Li-6 in the fuel elements ($s^{-1}$), $\frac{dN_T^{R-Li6}}{dt}$ is the generation rate of H-3 from the activation of Li-6 in the graphite reflectors ($s^{-1}$), $\eta$ is the H-3 release fraction from the graphite reflectors in the high temperature range of the core, $L$ is the release rate of the helium coolant in the primary loop ($s^{-1}$), $P_{ur}$ is the fractional purification rate of helium purification system ($s^{-1}$), $\lambda$ is the decay constant of H-3 ($s^{-1}$), and $P_{er}$ is the fractional permeation rate of H-3 to the secondary loop ($s^{-1}$). Time is defined as the time that has passed since the initial start of the reactor in December 2000.

We assume that the helium leakage rate from the primary loop is constant, and therefore the loss of H-3 by this route is proportional to its concentration in the coolant. The permeation rate of H-3 to the secondary loop, through the metallic heat exchangers, is calculated as proportional to the partial pressure of H-3 on the primary side. The heat exchangers operate at 104–440 °C tube temperature at 10MWth, and have steam on the secondary side. In the primary loop of HTR-10. The design set-point for $H_2$ content in the helium is less than 30 ppm by volume, and the primary coolant pressure is 3 MPa; so the design $H_2$ partial pressure is 90 Pa. At this partial pressure, the permeation rate is proportional to the differences of the square root of the partial pressures on either side of the metallic heat exchangers (Forcey et al., 1988). For simplification of the analysis, in this study we approximate the permeation rate as linearly dependent on the tritium partial pressure. Since the partial pressure variation is not large, and the uncertainties in all other tritium removal rates are significant, the authors consider this approximation to be acceptable. For simplification, zero tritium concentration is assumed on the secondary side of the heat exchangers; this is an approximation, since there is a non-zero content of tritium in the water, but the distortion due to this assumption is minimal. We also assume that the pressure in the primary loop is constant, and therefore the tritium concentration and tritium partial pressure in the primary coolant are proportional to the total inventory of H-3 (Ohashi and Sherman, 2007). Helium purification is performed on a small bypass steam, which is approximately 5% of the total helium circulation rate. For the purpose of this calculation, we assume 100% removal of tritium from the side-stream that circulates through the purification system (Gainey, 1976).

The ternary fission rate is given by:

$$\frac{dN_T^X}{dt} = \frac{P_{th}}{g}\cdot y_X\cdot f_X \quad (2)$$

where, $N_T^X$ is the number of the H-3 generated by the ternary fission reaction of the nuclide $X$, $P_{th}$ is the thermal power of the reactor (MW), $g$ is the energy released per fission (MW·s), $y_X$ is the average yield of H-3 per fission of the nuclide $X$, and $f_X$ is the fission fraction from nuclide $X$. The source of unencapsulated fissionable isotopes in the fuel spheres is from broken TRISO fuel particles and uranium contamination in the matrix graphite. In HTR-10, the dominant ternary fission contributions were from U-235, Pu-239, and Pu-241.

The He-3 production rate was calculated as detailed by Xu et al. (2017):

$$\frac{dN_T^{He-3}}{dt} = \sigma_{He-3}\phi N_{He-3}^0\cdot\left(\frac{\sigma_{He-3}\phi}{L+\sigma_{He-3}\phi}\cdot e^{-(L+\sigma_{He-3}\phi)t} + \frac{L}{L+\sigma_{He-3}\phi}\right) \quad (3)$$

where, $N_{He-3}^0$ is the number of the He-3 atoms in the core of HTR-10 at the initial start, $\sigma_{He\text{-}3}$ is the cross section of the activation reaction of He-3 (n, p) H-3 (barn), and $\phi$ is the core-averaged thermal neutron fluence rate in the core ($cm^{-2}\cdot s^{-1}$), which is considered constant with time.

The Li-6 activation reaction rate in the fuel spheres is defined by:

$$\frac{dN_T^{F,Li-6}}{dt} = N_F\cdot\frac{\int_{t_{FE}=0}^{t_{FE}=T}\sigma_{Li-6}\phi n_{F,Li-6}^0\cdot e^{-\sigma_{Li-6}\phi t_{FE}}dt_{FE}}{T} \quad (4)$$

where, $N_F$ is the number of fuel elements in the equilibrium reactor core, $\sigma_{Li\text{-}6}$ is the cross section of the activation reaction of Li-6 (n, α) H-3 (barn), $n_{F,Li-6}^0$ is the number of Li-6 atoms in a fresh fuel element, $t_{FE}$ is the dwell time for a fuel element in the reactor core, and $T$ is the average time of discharge of a fuel element. The tritium produced in the graphite matrix of the fuel elements is assumed to release fully in the primary coolant. This is an over-prediction, possibly by up to a factor of two, of the tritium source term to the primary circuit, given that the graphite matrix has a non-negligible solubility for hydrogen isotopes, and a possibly low-enough diffusion coefficient for the diffusion timescale to be on the order of hours or days.

The Li-6 activation reaction rate in the graphite reflectors is defined by:

$$\frac{dN_T^{R,Li-6}}{dt} = \sigma_{Li-6}\phi_R N_{R,Li-6}^0\cdot e^{-\sigma_{Li-6}\phi_R t} \quad (5)$$

where, $\phi_R$ is the volumetrically-average thermal neutron fluence rate in graphite reflectors ($cm^{-2}\cdot s^{-1}$), and $N_{R,Li-6}^0$ is the number of Li-6 atoms in graphite reflectors at the initial start of the reactor. Only a fraction, $\eta$, of the tritium that is produced in the graphite reflector blocks is assumed to release in the primary coolant. It is possible that at the beginning of life of HTR-10, the graphite blocks behaved as tritium sinks, absorbing tritium from the primary coolant. Therefore, for the beginning of life of HTR-10, the source term to the primary coolant may be over-predicted.

Equations (1)–(5) are used to calculate the activity concentration of tritium in the primary loop of HTR-10, for comparison to the experimental data reported here. The input parameters are presented in more detail by Xu et al. (2017) and are summarized in Table 1 and Table 2. The parameters correspond to the operational conditions at the time when the measurements were made, to the degree that the information was available and public.