Nuclear Engineering and Design 323 (2017) 39–45

Contents lists available at ScienceDirect

Nuclear Engineering and Design

journal homepage: www.elsevier.com/locate/nucengdes

Experimental study on the content and distribution of key nuclides in an irradiated graphite sphere of HTR-10

Hong Li, Xuegang Liu, Feng Xie*, Fuming Jia

Institute of Nuclear and New Energy Technology, Collaborative Innovation Center of Advanced Nuclear Energy Technology, Key Laboratory of Advanced Reactor Engineering and Safety of Ministry of Education, Tsinghua University, Beijing 100084, China

ARTICLE INFO

Keywords:
HTR-10
An irradiated graphite sphere
Radial distribution
H-3
C-14

ABSTRACT

An irradiated graphite sphere of the 10 MW high temperature gas-cooled reactor (HTR-10) has been studied experimentally with a mechanical disintegration way and a suite of radiochemical analysis methods, including the total α/β counting measurement and the absolute activity measurement with a liquid scintillation counter and a high-purity germanium detector connected to a multichannel analyzer. The key nuclides in this irradiated graphite sphere were determined, which were H-3, C-14, Co-60, and Cs-137. The radial distributions of the specific activity for each nuclide were obtained experimentally and compared with theoretical calculations. The production mechanisms of H-3, C-14, Co-60, and Cs-137 in the irradiated graphite sphere of HTR-10 were discussed individually, and the generation sources for each nuclide were analyzed explicitly. Current results can supply important experimental data for source term analysis of HTR-10, and provide useful research reference for the decommissioning work and waste minimization of high temperature gas-cooled reactors (HTGRs).

1. Introduction

The Very High Temperature Gas Cooled Reactor System (VHTR), as a development of high temperature gas-cooled reactors (HTGRs), has been selected for a candidate of the Generation IV systems by the Generation IV International Forum (GIF) for the production of process heat, electricity, and hydrogen (NERAC and GIF, 2002). A high level of passive safety performance, high generating efficiency, and high temperature heat output promise a broad commercial utilization for HTGRs (Zhang and Yu, 2002). For the pebble bed high temperature gas cooled reactor, the performance of the fuel spheres in the core plays a crucial role with regard to nuclear safety. The nuclides produced in the core are the original source of radioactive substances into primary coolant and auxiliary systems in a nuclear power plant. Thus, the determination of the source term in the reactor core can supply important information to understand the behavior of fission and activation products and provide reliable foundation to evaluate the radiation level of the nuclear facility. Apart from the routine supervision of the coolant activity, another way of quantification of radioactivity release from the fuel elements is the investigation of spherical elements discharged from a pebble bed reactor or irradiated in a Material Test Reactor (MTR). Wenzel et al. (1979) measured the content and radial distribution of Carbon-14 (C-14) in spent fuel spheres experimentally and indicated that 96% of the C-14 was produced in the matrix graphite of the fuel spheres in Arbeitsgemeinschaft Versuchsreaktor (AVR). Schenk et al. (1988) went even further and examined in numerous experiments the transient fission products release behavior and concentration profiles of fission products in fuel spheres heated under simulated accident conditions (IAEA, 1997; IAEA, 2012).

The 10 MW high temperature gas-cooled test reactor (HTR-10) is the first gas-cooled pebble-bed reactor in China, which has been designed, built and operated by the Institute of Nuclear and New Energy Technology (INET), Tsinghua University, China (Wu and Zhang, 2004). It is a helium-cooled, graphite-moderated, and thermal neutron spectrum reactor, adopting the concept of 60 mm diameter spherical fuel elements which contain tristructural-isotropic (TRISO) coated fuel particles (Tang et al., 2002). The primary helium pressure, the core inlet temperature, the core outlet temperature, and the primary coolant flow rate of HTR-10 were 3.0 MPa, 250 °C, 700 °C, and 4.32 kg/s, respectively (Wu et al., 2002). In December 2000, HTR-10 reached first criticality, and since 2003, it was successfully demonstrating the expected safety features of a pebble bed HTGR in various experiments (Zhang et al., 2009). From July 2007 to November 2014, HTR-10 was shut down. During the year of 2015, HTR-10 was operated at 2.9 MW for about 90 days for the training of personnel that will operate the HTR-PM, the first high temperature gas-cooled reactor pebble-bed module demonstration at commercial scale in China. Before 2014, the HTR-10 was operated about 225 equivalent full power days. As a test

* Corresponding author.
E-mail address: fxie@tsinghua.edu.cn (F. Xie).

http://dx.doi.org/10.1016/j.nucengdes.2017.07.033
Received 15 March 2017; Received in revised form 12 June 2017; Accepted 28 July 2017

reactor, the HTR-10 was used to conduct experimental measurement on the activity concentration of radioactive dust, H-3 and C-14 in the primary loop which supplied valuable data for the source term analysis in HTR-10 and HTR-PM (Wei et al., 2016; Xie et al., 2015).

In the equilibrium core of HTR-10, there are 27,000 fuel elements totally. The initial load of fuel elements into the reactor contains a fraction of 47% of graphite spheres without coated particles inside serving as moderator. During the transition to the equilibrium core, graphite spheres will be gradually discharged from the reactor and replaced with fuel elements. Since the spent fuel element from the reactor core will possess strong radiation, it is difficult to measure the nuclides in the fuel element directly. However, graphite spheres which have experienced the same neutron radiation and are composed of the same matrix graphite material, will be the ideal substitute to study the source term from the matrix material in the fuel element. The graphite matrix is made up of natural graphite, electro graphite and resin in a certain percentage (Zhao et al., 2006). In this paper, we report the complete measurement results for an irradiated graphite sphere from HTR-10 reactor core. The experimental data include the total α/β counting measurement and the absolute activity measurement with a liquid scintillation counter and a high-purity germanium detector connected to a multichannel analyzer. The distribution of the specific activity of key nuclides in the sphere are determined experimentally. Theoretical calculations for the specific activity of each nuclide are provided with the parameters from the design data and/or experimental values. By comparing the calculational and experimental results, the generation source and reduction method of fission and activation products in the matrix graphite are proposed, which can provide useful research reference for the decommissioning work and waste minimization of HTGRs.

2. Experiment

During the shutdown stage of HTR-10, several graphite spheres from the reactor core were discharged in 2014 for experimental research. To determine the distribution of the nuclides in the matrix graphite, the graphite samples at different radial positions of the sphere need to be prepared (Wang et al., 2014). A mechanical method to disintegrate the graphite sphere has been proposed. A homemade drilling machine (SIEG SUPER X3, from Shanghai SIEG Machinery Co., Ltd) with a special annular drill of which the inner diameter is 9 mm was adopted and the cylindrical graphite stick sample through the center of the irradiated graphite sphere was obtained, as indicated in Fig. 1. Then a series of graphite powder samples at unique positions of the graphite sphere were formed with a grinder machine.

The graphite powder samples were first measured with a total α/β analyzer (BH1216III, from CNNC Beijing Nuclear Instrument Factory) and a high-purity germanium detector connected to a multichannel analyzer (GC3018 detector, from Canberra Company), which are non-destructive examination methods. The total β counting rate for each graphite sample provides the qualitative radial distribution information about nuclides in the sphere. The γ spectra supply energy peaks and positions of γ emitter nuclides in the sphere. However, pure β emitting nuclides, such as H-3 and C-14, must be investigated in a further process where the graphite samples are put into a combustion vessel vented with oxygen (1180B, from Parr Instrument Company). After the combustion, H-3 and C-14 in the graphite are converted into $^{14}CO_2$ and water vapor, and adsorbed with NaOH solution in the combustion vessel completely. Then the aqueous samples were analyzed with an automatic potentiometric titrator (809Titrando, from Metrohm Company) to determine the total carbon content and a liquid scintillation counter (Quantulus 1220, from Perkin Elmer Company) to measure the activity of H-3 and C-14 in the liquid sample. The specific activity of H-3 and C-14 in the solid graphite sample can be deduced quantitatively. The conversion efficiencies during the combustion and adsorption process have been considered strictly. The detail information about the whole experimental method for the source term analysis of the graphite sphere of HTR-10 will be published in a separate paper (Liu et al., 2017).

Fig. 1. Drilling machine with a special annular drill and cylindrical graphite stick sample of the irradiated graphite sphere of HTR-10.

3. Results and discussions

3.1. Experimental results

The spectrum of the total β counting rate per gram along with the position of the sample in the graphite stick from the irradiated sphere of HTR-10 is shown in Fig. 2. It indicates that the counting rate at the surface is higher and in the interior of the irradiated sphere is nearly uniform. This phenomena can be explained by a homogenous distribution of the β-radioactive nuclides inside the irradiated graphite sphere except at the surface which is an interface between the graphite spheres and primary helium or other materials. The total α/β counting measurement can supply a qualitative distribution of the aggregate activity in the graphite sphere, whereas the information about specific nuclides needs to be investigated with a high-purity germanium detector and a liquid scintillation counter which can distinguish different types of nuclides.

Fig. 3 exhibits the β spectra of the graphite sample at 36.60 mm position of the graphite sphere from the measurement with a liquid scintillation counter. The separate peaks indicated in the spectra belong to H-3 and C-14. Fig. 4 shows the radial distribution of the specific activity of H-3 and C-14 in the irradiated graphite sphere. In the interior

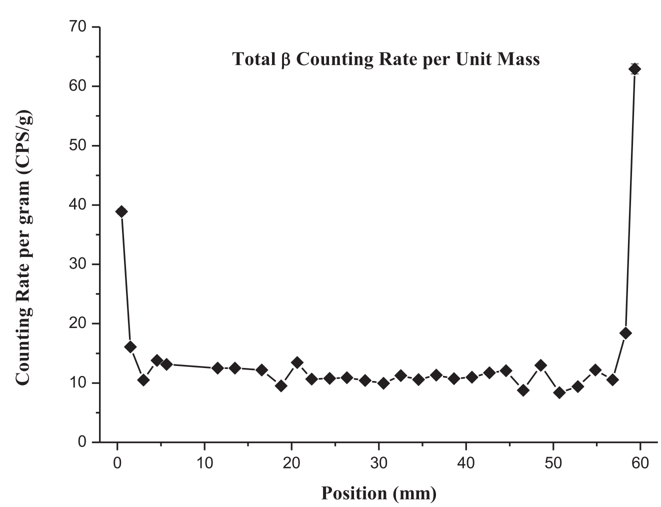

Fig. 2. Radial distribution of total β counting rate per unit mass of the irradiated graphite sphere of HTR-10.