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A comprehensive study on source terms in irradiated graphite spheres of HTR-10

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ABSTRACT

With previously developed experimental methods which include the preparation and measurement process for the graphite sample, two new irradiated graphite spheres with surface γ dose rates of 51.00 μSv/h and 0.14 μSv/h from the reactor core of the 10 MW high temperature gas-cooled reactor (HTR-10) have been investigated experimentally. The total β counting rate, the β spectra and the γ spectra for each graphite sample of irradiated graphite spheres were recorded with a total α/β counting measuring apparatus, a liquid scintillation counter and a high-purity germanium detector connected to a multichannel analyzer, respectively. Combined with previous experimental data of two irradiated graphite spheres with surface γ dose rates of 25.10 μSv/h and 1.17 μSv/h, the types of key nuclides in the irradiated graphite sphere of HTR-10 were determined, which were H-3, C-14, Co-60, Cs-137, Eu-152 and Eu-154. The distributions for each nuclide in four irradiated graphite spheres were compared. The generation mechanisms of H-3, C-14, Co-60, Cs-137, Eu-152 and Eu-154 in the irradiated graphite sphere of HTR-10 were discussed and analyzed. Based on all the experimental data regarding impurities and uranium contamination in the matrix graphite of HTR-10 available, a sensitivity analysis was performed to explain the effect of impurities and uranium contamination on the specific activity of key nuclides in the graphite sphere. The influence of the neutron flux and the dwell time in the core on the specific activity of key nuclides was also considered. The differences of experimental specific activities among these irradiated graphite spheres were compared and explained. Current comprehensive studies on irradiated graphite spheres of HTR-10 can provide valuable information for the source term analysis, waste minimization and radiation protection of high temperature gas-cooled reactors (HTGRs).

1. Introduction

In the Generation IV Program, the very high temperature reactor (VHTR) has been identified as one of the six technologies for development as a next generation nuclear energy system (NERAC and GIF, 2002). Compared to the current high temperature gas-cooled reactor (HTGR), the VHTR is expected to supply electrical production with a high efficiency and provide versatility in process-heat generation or co-generation at a high temperature level. However, with respect to the safety assessment of VHTR, current reliable data about radionuclides including fission products and activation products in the primary circuit, especially in the reactor core is rather rare (Morris et al., 2008; Kissane, 2009). The relevant information can be derived from the operating experience and experiments on the high temperature gas-cooled reactors (HTRs). For a pebble bed core, the Arbeitsgemeinschaft Versuchsreaktor (AVR) in Germany had accomplished a series of experimental programs to study the radioactive source term and made up a knowledge base for understanding the behavior of fission products and activation products in HTGRs (Bäumer and Barnert, 1990; IAEA, 1997; IAEA, 2012).

After the decommissioning of AVR and the thorium high temperature reactor (THTR-300) in Germany, the 10 MW high temperature gas-cooled test reactor (HTR-10) is the only pebble bed reactor working in the world, which is the first gas-cooled pebble bed test reactor in China. It uses helium as the primary coolant and graphite as a moderator and reflector (Wu et al., 2002). The spherical fuel elements with 60 mm in diameter embedded tristructural-isotropic (TRISO) coated particles are adopted, which are composed of two parts: a fuel zone with 50 mm in diameter and a fuel-free shell of 5 mm in thickness (Tang et al., 2002). The

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HTR-10 attained first criticality in December 2000, realized full power operation at the beginning of 2003, and demonstrated several expected safety features of a pebble bed HTGR by July 2007 (Zhang et al., 2009). Then it was shut down and totally HTR-10 was operated about 225 equivalent full power days (EFPDs). In 2015, HTR-10 was restarted and operated at 2.9 MW for about 90 days (Wei et al., 2016). In order to obtain more valuable information about radioactive source terms in the primary circuit and reactor core, a series of experiments has been conducted in HTR-10, including: (1) measurement of the activity concentration of H-3 and C-14 in the primary coolant, (2) measurement of the concentration and particle distribution of radioactive dust in the primary circuit, and (3) measurement of the content and distribution of key nuclides in the irradiated graphite sphere from the reactor core (Xie et al., 2018, 2015, 2017; Liu et al., 2017; Li et al., 2017).

All fission products and activation products in a power plant come from the reactor core. To determine source terms in the core will be essential to study the transport behavior of fission products and activation products in the primary circuit and auxiliary systems. However, it is generally known to be difficult to determine the core temperature precisely with respect to the pebble bed reactor which might affect the performance of fuel elements. The direct investigation of irradiated fuel elements can lead to an extensive demand for experimental tools and radiation protection. For the initial core of a pebble bed reactor, a certain percentage of graphite spheres without fuel particles will be loaded into the core to balance the excess reactivity of the basically fresh fuels. From the transition core to the equilibrium core, graphite spheres will be gradually taken out. These graphite spheres have the same size and material of matrix graphite as fuel elements, and experienced nearly the same neutron flux and energy spectrum in the core. Since there are no fuel particles inside, the radiation level of the irradiated graphite sphere is rather low (usually about 1 μSv/h with the maximum less than 60 μSv/h), which makes it ideally suitable to obtain the radiological information about the pebble bed reactor core on one hand. On the other hand, to study the irradiated graphite sphere can provide unambiguous data about source terms in the nuclear graphite which has been widely used in HTGRs.

Liu et al. (2017) has established the analytical methodology to study the source term in the irradiated graphite sphere and determined the analytical procedure and parameters. Later, this analytical methodology was successfully applied to investigate the content and distribution of key nuclides in an irradiated graphite sphere of HTR-10 experimentally (Li et al., 2017). However, the key nuclides and corresponding specific activities exhibit distinct characters with different irradiated graphite spheres. In this paper, we will combine all the experimental data available from four graphite spheres irradiated in the reactor core of HTR-10 to compare the types, specific activities and distributions of key nuclides therein. Meanwhile we will gather all the information about impurities in the matrix graphite available to do a sensitivity analysis with the variation of neutron flux and dwell time in the core. The study whose results are presented here deals with the detailed investigation of four irradiated graphite spheres discharged from the reactor core during the shutdown stage of HTR-10 in 2014. As all graphite spheres loaded into the core were part of the initial core of HTR-10, their irradiation time of 225 EFPDs is precisely known. The fact that these four graphite spheres do not contain any coated fuel particles has the advantage that any measured radioactivity must originate from either natural contamination of the graphite material with uranium and impurities, respectively, or from activity that was transported with the cooling gas and deposited on the sphere surfaces, while an origin from the coated particles can be excluded. Therefore the measurements of surface contamination (such as Cs-137) provide valuable information on the overall fuel performance of the HTR-10 core, while other nuclides identified (such as Co-60) show the level of impurities in the graphitic material.

2. Experiment

The detailed experimental method and process can be found in Liu et al. (2017). Briefly to say, a mechanical method was adopted to obtain a cylindrical graphite stick sample through the center of the irradiated graphite sphere with a homemade drilling machine (SIEG SUPER X3, from Shanghai SIEG Machinery Co., Ltd). The graphite stick of 60 mm in length and 7 mm in diameter was used to prepare graphite powder samples at different radial positions of the graphite sphere. Fig. 1 shows the cylindrical graphite stick sample drilled from the irradiated graphite sphere.

As indicated earlier, the graphite powder samples were measured first with a total α/β analyzer (BH1216III, from CNNC Beijing Nuclear Instrument Factory). Then a high-purity germanium detector connected to a multichannel analyzer (GC3018 detector, from Canberra Company) was used to record the γ spectra of the solid sample. After that, the graphite powder samples were combusted sufficiently in a vessel (1180B, from Parr Instrument Company) and the exhaust gas was absorbed in NaOH solution. Finally, an automatic potentiometric titrator (809Titrando, from Metrohm Company) was applied to determine the carbon content in the liquid sample and the liquid scintillation counter (Quantulus 1220, from Perkin Elmer Company) was used to measure the β spectra of the liquid sample.

By now, four irradiated graphite spheres discharged from the reactor core of HTR-10 have been investigated experimentally. The experimental results of the first two graphite spheres have been presented in previous literatures (Liu et al., 2017; Li et al., 2017). In this article, all the experimental data available were considered for comparison and analysis. Table 1 lists the mass, the surface γ dose rate and the average total β counting rate per gram for the four irradiated graphite spheres, which were denoted as A, B, C and D for convenience.

Fig. 1. Cylindrical graphite stick sample drilled from the irradiated graphite sphere of HTR-10.