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REVIEW

Nuclear graphite for high temperature gas-cooled reactors

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Abstract: Since its first successful use in the CP-1 nuclear reactor in 1942, nuclear graphite has played an important role in nuclear reactors especially the high temperature gas-cooled type (HTGRs) owing to its outstanding comprehensive nuclear properties. As the most promising candidate for generation IV reactors, HTGRs have two main designs, the pebble bed reactor and the prismatic reactor. In both designs, the graphite acts as the moderator, fuel matrix, and a major core structural component. However, the mechanical and thermal properties of graphite are generally reduced by the high fluences of neutron irradiation of during reactor operation, making graphite more susceptible to failure after a significant neutron dose. Since the starting raw materials such as the cokes and the subsequent forming method play a critical role in determining the structure and corresponding properties and performance of graphite under irradiation, the judicious selection of high-purity raw materials, forming method, graphitization temperature and any halogen purification are required to obtain the desired properties such as the purity and isotropy. The microstructural and corresponding dimensional changes under irradiation are the underlying mechanism for the changes of most thermal and mechanical properties of graphite, and irradiation temperature and neutron fluence play key roles in determining the microstructural and property changes of the graphite. In this paper, the basic requirements of nuclear graphite as a moderator for HTGRs and its manufacturing process are presented. In addition, changes in the mechanical and thermal properties of graphite at different temperatures and under different neutron fluences are elaborated. Furthermore, the current status of nuclear graphite development in China and abroad is discussed, and long-term problems regarding nuclear graphite such as the sustainable and stable supply of cokes as well as the recycling of used material are discussed. This paper is intended to act as a reference for graphite providers who are interested in developing nuclear graphite for potential applications in future commercial Chinese HTGRs.

Key Words: Nuclear graphite; High temperature gas-cooled reactors; Irradiation, microstructure; Physical, mechanical and thermal properties

1 Introduction

The phrase nuclear graphite began to be used at the end of 1942 when the first nuclear fission occurred in the graphite moderated nuclear reactor CP-1 [1]. From the early 1960s, the United Kingdom, the United States and Germany began to develop high temperature gas-cooled reactors (HTGRs). Japan began the construction of a 30 MW_{th} high temperature test reactor (HTTR) in 1991, which reached its first criticality in 1998. In China, a 10 MW experimental high temperature gas-cooled reactor (HTR-10) [2, 3], whose design started in 1992 and construction commenced in 1995, reached it criticality in the end of 2000, and its full power in the beginning of 2003. Since the Fukushima accident in March, 2011, the public has paid more and more attention to the safety of nuclear power. As a candidate reactor for the Generation-IV reactors, the construction of a 2×250 MW high temperature gas-cooled reactor pebble-bed module (HTR-PM) with inherent safety is underway in Shidao Bay, Rongcheng of Shandong province, China and is expected to complete in 2017 [4]. In both of the research and commercial HTGRs, the reactor reflectors and cores have been constructed by structural graphite components. Past designs represent two primary core concepts commercially favored for HTGRs: the prismatic block reactor (PMR) and the pebble-bed reactor (PBR) [2]. In both of the HTGR concepts the polycrystalline graphite not only is a major structural component which offers thermal and neutron shielding and provides channels for fuel and coolant gas, channels for control and safety shut off devices, but also acts as a moderator and matrix material for the fuel elements and control rods and a heat sink or conduction path during reactor trips and transients.

The polycrystalline graphite exhibits significant importance in HTGRs because of its outstanding nuclear physical properties such as high moderating and reflecting efficiency, a relatively low atomic mass and a low absorption cross-section for neutrons, in addition to high mechanical strength, good chemical stability and thermal shock resistance, high machinability and light weight [5]. The following example illustrates the importance of nuclear graphite in more details. For the thorium high temperature reactor (THTR) in Germany

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with a power of 300 MWe, nearly 400 000 kg of nuclear graphite has been used [2]. In China, approximately 60 tons of graphite was used in HTR-10 [3], and more than 1 000 tons of nuclear graphite will be used in HTR-PM as the structural material and matrix graphite of pebble fuel elements [4]. The raw materials of matrix graphite of fuel elements for HTR-10 and HTR-PM such as natural flake graphite and artificial graphite powder are supplied by Chinese domestic providers [6,7]. The behavior of the individual fuel particles and the matrix graphite material in which the particles are encased are not considered here. However, it should be noted that although the graphite technology associated with the matrix graphite is related to that of the main structural graphite such as the moderator there are differences as non-graphitized materials and natural flake graphite are used in the matrix graphite. Because so far no qualified domestic nuclear graphite is available, all the structural nuclear graphite materials for HTR-10 and HTR-PM are imported from Toyo Tanso of Japan. In April 2015, China Nuclear Engineering Corporation Ltd (CNEC) announced that its proposal for two commercial 600 MWe HTGRs (HTR-600) at Ruijin city in Jiangxi Province had passed an initial feasibility review. The HTR-600 is planned to start construction in 2017 and for grid connection in 2021 [8]. In order to achieve the economy and security of supply, the structural nuclear graphite must be provided by domestic providers in China in the future. Fortunately, with the rocketing development of photovoltaic industry in China, several Chinese companies have emerged which can produce the fine-grained isotropic, isostatic molded, high strength graphite in large scale. Some of the manufacturers with state-of-the-art graphite manufacture capabilities should be chosen as the potential candidate providers of the structural nuclear graphite for HTGRs based on qualification programs.

However, during the operation of a reactor, many of the graphite physical properties are significantly changed due to the high fast neutron doses. The physical, mechanical and chemical properties of graphite can be influenced negatively by irradiation induced damage， which would lead to the failure of graphite components. In pebble-bed HTGRs such as HTR-PM in China, the core support graphite structure is particularly considered permanent, although it is expected that certain high neutron dose components (inner graphite reflector) will be replaced during the whole lifetime of the reactor. During the life time of the reactor, the reflector graphite would be subjected to a very high integrated fluence of fast neutrons of around 3×10^{22} n/cm^2 (E > 0.1 MeV) [9, 10]. Therefore, the pre-irradiation and post-irradiation comprehensive properties of nuclear graphite candidates must be thoroughly examined and evaluated. Those properties of nuclear graphite are strongly dependent on the extent of anisotropy, grain size, microstructural orientation and defects, purity, and fabrication method.

In this paper, basic nuclear requirements of nuclear graphite are presented and the specifications such as the manufacture, material properties with three primary areas (physical, thermal and mechanical) and irradiation responses of nuclear graphite suitable for HTGRs are elaborated， which could be a reference for the potential providers who are anxious to develop the nuclear graphite for future commercial HTGRs of China. The long-term considerations such as those involving the cokes and recycle for nuclear graphite are also discussed.

2 Nuclear requirements of graphite for HTGRs

2.1 Fission reactions with neutrons

The tremendous energy produced in HTGRs is from the fission of isotopes such as ${}_{92}U^{233}$, ${}_{92}U^{235}$, and ${}_{94}Pu^{239}$. Fission of a heavy element, with release of energy and further neutrons, is usually initiated by an impinging neutron. The fission of ${}_{92}U^{235}$ can be described as:

$$ {}_{92}U^{235}+{}_{0}n^{1}\rightarrow A+B+v*n+\sim200\text{ MeV} \qquad (1)$$

The average yield per fission of ${}_{92}U^{235}$ is about 2.5 fast neutrons. The energy of neutrons released from the fission reactions can be described by a Maxwellian distribution, with an average value of approximately 2 MeV. The probabilities of the fission reactions initiated by neutrons (the cross section) are inversely proportional to the velocity of the neutrons. It is essential to slow down the “fast” neutrons yielded by fission to “thermal” neutrons with lower energies (~0.025 eV at room temperature), which correspond to a neutron velocity of 2.2×103 m/s. The slowing down process results principally from energy transfer during elastic collisions between the neutrons and medium which is commonly called “moderation” and the non-absorbing medium where the moderation takes place outside the fuel is termed “moderator”. As is known, the nuclear fuel for HTGRs is commonly a mixture of low-enriched ${}_{92}U^{235}$ and ${}_{92}U^{238}$. Once the moderated thermal neutrons return to the fuel, they are most likely to cause fission in the ${}_{92}U^{235}$, instead of being captured by ${}_{92}U^{238}$.

2.2 Nuclear requirements for a good moderator

There are two very fundamental nuclear requirements for any moderator in HTGRs. First， it must have a small cross section for neutron absorption. Second， fast neutrons must be effectively slowed down to thermal neutrons over short distances and within few collisions in the moderator. Thus， the probability of neutron absorbed by the moderator impurities and, ${}_{92}U^{238}$ or absorbing structural materials in a reactor is reduced. Therefore, a good moderator material should exhibit a high slowing down power and low adsorption ability [11, 12]. Furthermore, the moderator material should be economically acceptable and compatible with the other materials used in the core of reactor, and maintain physical and chemical stability against the bombarding neutrons.

According to the fundamental consideration of Newton’s law, the more energy loss of neutron per collision takes place when the target nuclei have the lower atomic mass. The