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GRAPHITE FOR HIGH TEMPERATURE GAS-COOLED NUCLEAR REACTORS

ASME STANDARDS TECHNOLOGY, LLC

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FOREWORD

This report is intended as an introduction to the burgeoning field of graphite high temperature gas-cooled nuclear reactors with particular emphasis on nuclear graphite.

There is a brief review of the use of bulk graphite as a moderator in fission reactors around the world from the beginnings in 1942 to Generation IV prismatic and pebble bed High Temperature Reactor (HTR) potential. The characteristics, manufacture, properties and irradiation behavior of bulk graphites are outlined. A bibliography is provided for further study.

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ABSTRACT

This technical report presents the basic information relative bulk graphite production, structure, chemical properties, physical properties and neutron irradiation behavior. Bulk graphite characteristics, its manufacture, properties and irradiation behavior as well as a new generation of nuclear grades are briefly reviewed. An overview of graphite moderated gas-cooled reactor designs is also presented. The report serves as a summary of the training seminar on Nuclear Graphite conducted during the ASME Boiler and Pressure Vessel Code week, October 30–November 3, 2006, in Louisville, KY.

There is no universally accepted code for the design of graphite moderator structures. The history of graphite moderated reactors is traced from the beginnings in 1942 to the most recent utility start-up in 1989. Developments have continued over the intervening years especially in the area of helium cooled High Temperature Reactors. Prismatic 30MWth, and pebble-bed 10MWth, test reactors were brought into operation in Japan and China, respectively.

1 INTRODUCTION

Graphite has extraordinary abilities to both slow down fast neutrons and to sustain the collateral damage. In particular, structural integrity is retained over a wide range of neutron fluence and reactor temperatures such that commercial graphite moderator systems have run for over forty years.

Graphite is a member of a class of materials known as moderators which are effective in slowing high energy fission neutrons to thermal energies so as to maximize fission collisions. (Other moderators include beryllium, beryllium oxide, heavy water and water as well as certain metal hydrides and certain hydrocarbons.) The slowing down of neutrons is the result of multiple collisions with the nuclei of the moderator.

Graphite produced synthetically in large blocks is particularly attractive as a moderator because it is produced in quantity around the world by relatively simple industrial processes in a wide range of sizes and grades at reasonable cost. (In this report, blocks of synthetic graphite will be termed bulk graphite as distinct from other forms such as single crystal, powder and fibers.) The blocks are readily machined into quite intricate shapes. Furthermore, graphite is refractory in the sense that, in the absence of both air and neutron irradiation, at temperatures up to about 2000˚C, structural integrity is retained. The strength is actually enhanced: for instance, the tensile strength of graphite at 2000˚C is about 50% higher than its room temperature value.

Thus, it should come as no surprise that the first fission reactors in the USA (1942), the USSR (1947), the UK (1947), Belgium (1956) and France (1956) were all graphite moderated. The early civil power reactors in France, the UK and the USSR were large-scale derivatives and currently form the backbone of nuclear electricity generation in the UK.

The refractory capability of graphite moderated reactors is evident in the increase in coolant gas outlet temperatures from 140˚C in the early air-cooled versions to 950˚C in the helium cooled pilot units in Germany and Japan. There is great interest around the world in the commercial exploitation of the high helium gas temperatures for the production of hydrogen, sea water desalination and direct cycle power generation.

Currently, there is no universally accepted code for the design of graphite moderator structures. In 1990, ASME (Section III, Div. 2) issued a draft code proposal for Graphite Reactor Core Supports. More recently, a diverse group of Generation-IV Very High Temperature Reactor (VHTR) stakeholders have expressed the need for a unified approach to design code. The stakeholders include the DOE and the NRC in the USA, the nuclear regulator in the Republic of South Africa and the designers AREVA, General Atomics and the PBMR Company. Therefore, an ASME Project Team on Graphite Core Components was established in 2002, initially under Section II. The Team was re-assigned to Section III in 2006. The work of the Project Team had matured and justified the preparation of a training seminar on Nuclear Graphite for presentation during the Louisville ASME Code week, Oct. 30–Nov. 3, 2006. The seminar focused on bulk graphite production, structure, chemical and physical properties and neutron irradiation behavior.

The aim of this technical report is to capture the essentials covered in the Louisville seminar and to provide a brief introduction to nuclear graphite.

2 DEFINITIONS

(See also ASTM C 709 on Terminology Relating to Manufactured Carbon and Graphite)

Anisotropy – defined in terms of properties:

> ***With grain*** – parallel to extrusion direction, perpendicular to molding axis.
>
> ***Against grain*** – perpendicular to extrusion axis, parallel to molding axis.

Crystallite – fundamental region of three dimensional order as revealed by X-ray diffraction.

Grain – filler particle (calcined coke, recycled graphite).

Isotropy Ratio – usually defined in terms of the ratio of the thermal expansion coefficient (measured over a defined temperature range) (against-grain)/(with-grain).

Isotropic Graphite – a graphite in which the ratio of the against-grain to with-grain value of the coefficient of thermal expansion measured from 25˚C to 500˚C is between 1.0 and 1.1.

Near-Isotropic Graphite – a graphite in which the ratio of the against-grain to with-grain value of the coefficient of thermal expansion measured from 25˚C to 500˚C is between 1.1 and 1.15.

Nuclear Graphite – any bulk graphite with the desired properties, suitable and known irradiation behavior, with unwanted impurities removed. (See also ASTM D7219 Standard Specification for Isotropic and Near-Isotropic Nuclear Graphites.)

RMBK – reaktor bolshoy moshchnosti kanalniy (Russian)

3 GRAPHITE MODERATED GAS-COOLED REACTORS

3.1 Developments 1942–1989

The early graphite moderated reactors in the USA, the USSR, the UK and France were dedicated to the production of plutonium, the fundamental investigations of fission and graphite behavior and the preparation of radioisotopes. The operating temperatures were initially relatively low (typically <150˚C) and cooling was achieved indirectly with air initially and more commonly with water. The Hanford reactor cores were contained in a mixed gas blanket of helium and carbon dioxide. However, the potential for harnessing the heat released to raise steam for turbine generation of electricity was recognized.

The first generation of graphite moderated civilian power reactors in France and the UK were cooled directly with carbon dioxide. In the reactors of the former Soviet Union, heat was removed from the RBMK 1000MWe cores indirectly by cooling water. The RBMK graphite core was contained in a mixed gas blanket of helium and nitrogen, typically a 90%/10% mixture.

The first electric utility systems in the UK (so-called Magnox because of the alloy used to contain the fuel) were relatively small (49MWe per reactor, Calder Hall 1957), growing to 490MWe per reactor in the final build (Wylfa 1972, scheduled to close in 2010). A complication with these commercial Magnox reactors was the concomitant radiolytic oxidation. Some mitigation was achieved with small amounts of hydrogen and methane in the coolant circuit. The same grade of medium grain, high purity, extruded needle coke graphite, grade PGA, was used in every Magnox reactor.

A second generation of graphite moderated electric utility reactors were built in the UK between 1976 and 1989 with a combined generating capacity of almost 9GWe. These so-called Advanced Gas-cooled Reactors (AGR) were all relatively large at 555MWe to 625MWe per reactor. Each reactor required about 3000 tonnes of graphite. The graphite grade, GCMB (also known as Gilsocarbon), was a medium grain molded isotropic type using coke produced from Gilsonite, a natural asphalt occurring in Colorado and Utah. A typical arrangement of the graphite moderator blocks in an AGR core is shown in Figure 1. The last AGR station is scheduled to be closed in 2022, representing an average generating life of about 35 years.