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USDOE ART PROGRAM: Graphite – Selection and Acquisition Strategy

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OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE US DEPARTMENT OF ENERGY

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Materials Science and Technology Division

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ABREVIATIONS AND ACRONYMS

AGC	advanced graphite creep
ANSTO	Australian Nuclear Science and Technology Organisation
ART	Advanced Reactor Technology
ASME	American Society of Mechanical Engineers
ASTM	American Society for Testing and Materials
CEA	Alternative Energies and Atomic Energy Commission
DOE	US Department of Energy
GCR	gas-cooled reactor
GIF	Generation IV International Forum
HDG	high-dose graphite
HTGR	high-temperature graphite reactor
HTR	high-temperature reactor
HTR-DM	High-Temperature Reactor – Demonstration
HTTR	High-Temperature Test Reactor
INEET	Institute of Nuclear and New Energy Technology
JRC	Joint Research Centre
MMR	Multi Megawatt Reactor
MSR	molten salt reactor
MTR	material test reactor
NBG	nuclear block graphite
NGNP	Next Generation Nuclear Plant
PMB	Project Management Board
PRC	People's Republic of China
RSA	Republic of South Africa
USNC	Ultra-Safe Nuclear Corporation
VHTGR	Very High Temperature Gas-Cooled Reactor
VHTR	very-high-temperature reactor
XE	X-Energy

ABSTRACT

The previous selection and acquisition strategy document for this project dates from 2007.[1]. Since then, significant changes have occurred with reactor vendors and graphite manufacturers, as well as changes in the membership of the Gen IV International Forum. These factors were considered when formulating a revised selection and the strategy for the selection. In the new graphite selection and acquisition plan, it has been determined that no graphite billets will be purchased until there is new graphite production (nuclear block graphite [NBG]-18 and PCEA, a specific grade designation for medium-grained extruded grade of graphite produced by GrafTech). Existing supplies of NBG-17, NBG-18, PCEA, IG-110, and 2114 are considered enough for future work.

1. INTRODUCTION

The previous selection and acquisition strategy document for this project was published in 2007[1]. Since then, significant changes have occurred with reactor vendors and graphite manufacturers. For example, General Atomics is no longer working on a prismatic block graphite high-temperature reactor (HTR) concept. Moreover, the PBMR Company (South Africa) is no longer a reactor vendor.

A multitude of molten salt–cooled reactor vendors have entered the market, and it is not clear how their presence will alter the acquisition of graphite, if at all. It is probable that graphite selection criteria applied for a molten salt–cooled reactor will differ from that used by gas-cooled reactor designers. To date, molten salt–cooled reactor vendors have not declared a candidate graphite, but the superfine and ultrafine grained graphite included in the plans described herein should be of interest to the molten salt reactor (MSR) community.

Manufacturers of graphite have undergone many changes. SGL is no longer vibrationally molding graphite at their Meitingen, Germany, facility, but they have vibrationally molded a fresh batch of NBG-18 at a different European plant. GrafTech or GTI has been organizationally restructured, and any new production of PCEA will be performed by Amsted Graphite Materials[2] at their Clarksburg, West Virginia, plant.

As previously reported[3] we to the Materials Project Management Board (PMB) of the Generation IV International Forum (GIF):

> *The USA graphite selection and qualification strategy has been established and only minor changes to the graphite selection will be considered in future research tasks. These minor changes include the selection of super-fine and ultra-fine graphite grades that may be useful to Molten Salt Reactor designs. No further explicit contribution is planned for this task.*

Consequently, the United States is obliged to update their plan at this time. However, there have been significant changes to the GIF since the US strategy for graphite selection and qualification was established in 2007. The CEA (France) is no longer involved in graphite-moderated reactors and thus is no longer a member of the very-high-temperature reactor (VHTR) Project Plan. Moreover, in preparation for Brexit, or the withdrawal of the United Kingdom from the European Union (EU), the United Kingdom has become a member a of the GIF VHTR project and hence a member of the PMB. Previously, the United Kingdom had been involved in the GIF through their membership of the European Union, represented by the Joint Research Centre (JRC). The European Union has analyzed at the behavior of NBG-17, a material that has been de-emphasized by the United States.

Similarly, the People’s Republic of China (PRC) has joined GIF and is represented by the Institute of Nuclear and New Energy Technology (INEET), Tsinghua University. The greatest portion of their work will be on Toyo Tanso grade IG-110, which will be the graphite for their Demonstration High Temperature Reactor (HTR-DM), a pebble-bed graphite-moderated reactor currently under construction.

Other changes to the GIF include the addition of Australia as represented by the Australian Nuclear Science and Technology Organisation (ANSTO), and the Republic of South Africa (RSA) withdrawal from the GIF.

These changes, along with changes in reactor vendors, the type and design of reactor being offered, the resurgence of interest in the MSR, along with changes in the worldwide graphite market, have necessitated a review of the current US strategy.

This document sets forth the revised US plan for the selection and acquisition of graphite.

2. MANUFACTURE OF GRAPHITE

Graphite is a composite material manufactured from filler coke and pitch binder. Nuclear graphites are usually manufactured from isotropic cokes derived from petroleum or coal tar and are formed in a manner to make them isotropic or near isotropic. Figure 1 shows the major processing steps in the manufacture of nuclear graphite. After baking (carbonization), the artifact is typically impregnated with a petroleum pitch and re-baked to densify the part. Impregnation and re-bake steps may be repeated several times to ensure the required density. Graphitization typically occurs at temperatures >2,500°C. Additional halogen purification may be required. Typical manufacturing times for a production lot of graphite are 6–9 months. The forming and densification processes impart property variations within the billet: the properties will be different in the forming direction compared with the perpendicular to forming direction. Moreover, a density gradient will exist from the billet's edge to the center. These variations must be quantified for the selected grades of graphite. In addition, variations in properties will occur from billet to billet within a batch, and variations will also be found between production lots.

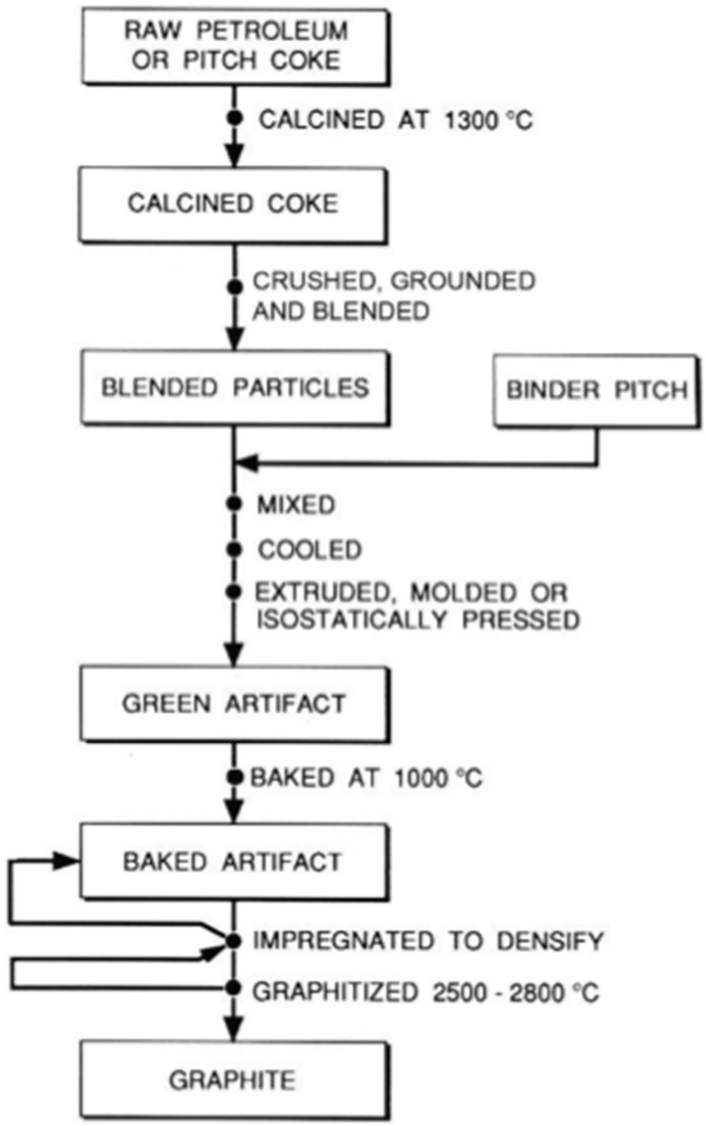

Figure 1. The process steps in the manufacture of nuclear graphite.[4]

In extruded graphite, the in-billet variations will be significant and can exceed the lot-to-lot variations. In isostatically molded graphite, the in-billet variations will be smaller than those in the extruded graphite and will be on the order of the lot-to-lot variations. Finished graphite billets are machined to the complex geometries required for the reactor components, including fuel elements, reflector blocks, core support posts. Machined parts are assembled to form the core assembly.

The properties of graphite are a direct consequence of the raw materials used in their manufacture and formation method. The type and source of the coke used in manufacture is crucial because the coke's properties largely dictate the properties and behavior of conventionally manufactured graphite. In conventional nuclear graphite, the selection of the coke is paramount. Reactor designers desire isotropic irradiation behavior to minimize differential irradiation-induced dimensional changes and subsequent stress buildup. Modern nuclear graphites achieve this behavior through a combination of an isotropic coke and a specialized forming method. Secondary coke graphites use a nonconventional manufacturing process to achieve the desired isotropic irradiation response and do not depend on starting with an isotropic coke. Essentially, anisotropic cokes are fabricated into graphite and are then ground up to become the starting filler in a conventional process. Because of the long graphite manufacturing process, the time taken to develop new graphite grades or to introduce a new source of coke is significant, taking several years.

The American Society for Testing and Materials (ASTM) provides two specifications for nuclear graphite[5,6] which provide minimum requirements for production, traceability, and properties. Any new graphite that is purchased by the US program should, as a minimum, comply with the relevant ASTM specification.

3. FACTORS INFLUENCING GRAPHITE SELECTION

Several factors may affect the grade of graphite selected, as listed below:

1. Function of component
2. Irradiation lifetime (if known)
3. Existence and extent of irradiation database
4. Extent of grade characterization
5. Extent of collaboration (Gen IV)
6. Product size and availability of grade
7. Longevity of supply, graphite and filler coke
8. Cost

These factors are discussed below.

Function of component. The function of a graphite component within the reactor will be predicated on the type of high-temperature graphite reactor (HTGR) being considered. In a pebble-bed reactor such as X-Energy's XE-100 or the HTR-DM under construction in the PRC, the graphite components form the core in which the pebble form fuel flows (Figure 2).

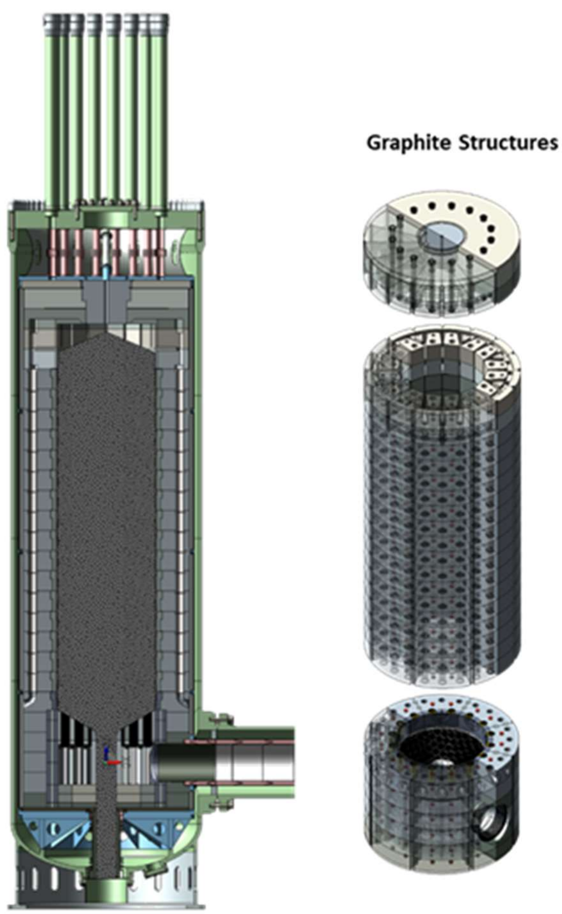

Figure 2. Cross section of the XE-100 reactor showing graphite core structures.

Irradiation lifetime. The graphite components are often considered core lifetime components. To limit the neutron dose to front-facing core components, the graphite core is sometimes segmented, and different grades may be used. Moreover, some fuel-facing sections of the core may be replaceable. In a prismatic core HTGR such as the Ultra-Safe Nuclear Corporation (USNC) Multi-Megawatt Reactor (MMR) or the High-Temperature Test Reactor (HTTR)[7] in Japan, the fuel is in stick form and is inserted directly into the graphite to form a fuel element. Additional nonfueled graphite blocks may be required to provide moderation. The graphite would be discharged from the reactor with the spent fuel. In designs such as the HTTR[7] (Figure 3), the fuel is contained in a graphite fuel pin which is then housed in the graphite fuel element. This allows the total neutron dose to the fuel element to be reduced if needed, allowing for the re-use of the refueled element.

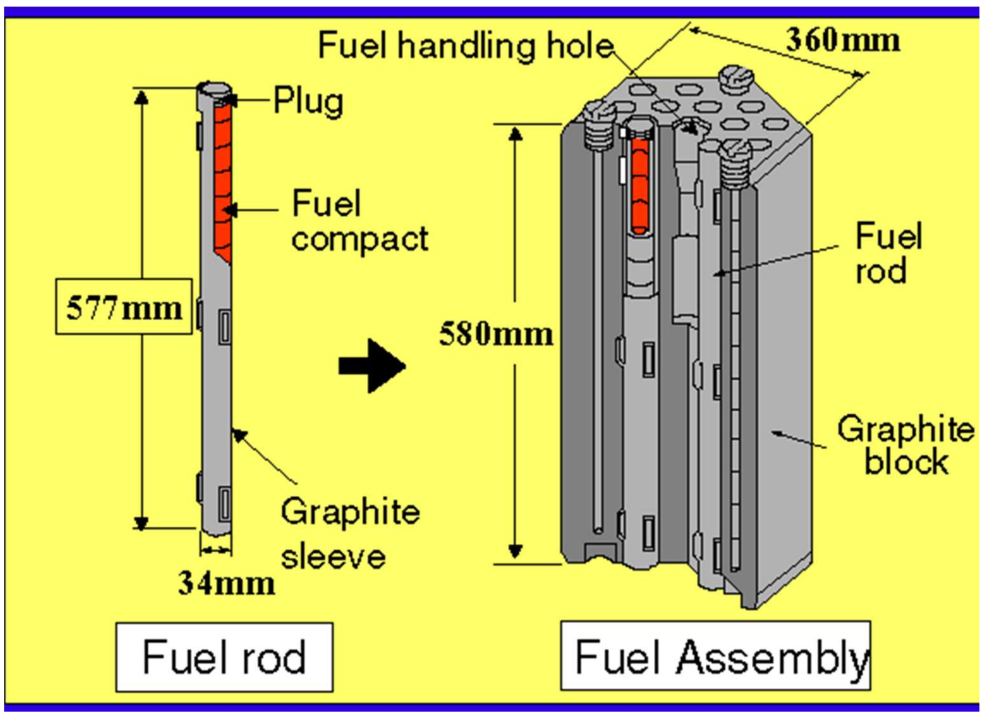

Figure 3. Configuration of the HTTR fuel element.[7]

Extent of grade characterization. To date, vendors of MSRs have not identified specific grades of graphite. Some fine-grained graphite grades with smaller pores are included in the high-dose graphite (HDG) capsules, which will be of interest to the MSR community. If the MSR is to be of the pebble-bed design in which the fuel pebbles are buoyant and thus the core is inverted, then there will be no sensitivity to pore size because there will be no fuel salt intrusion, and all the grades in the HDG capsules are relevant. The irradiation lifetime of a grade can determine whether it is selected as a core graphite. Graphite components are often safety-relevant parts, so they must maintain their geometries or adequate strength margins throughout their lifetimes. A graphite's lifetime is frequently taken as the dose at which a graphite returns to its original volume (after shrinkage), although the American Society of Mechanical Engineers (ASME) HTR graphite code takes a more lenient cohesive life limit. The core designer will want the longest possible life for the core graphite, especially if the graphite is the material used in a lifetime component.

Existence and extent of irradiation database. The existence and extent of an available irradiation database for the graphite grade is influential in its selection. The reactor designer and the licensor require behavioral data over the reactor core's projected operating envelope. Such data, including creep behavior data, are expensive and time consuming to obtain because there are few MTRs available worldwide. Most manufacturers have relied upon national programs to supply such data, but a few have contracted privately to perform high-dose MTR programs.

Extent of collaboration (Gen IV). When considering the extent of any data of a particular graphite, collaboration with our Gen IV partners must be considered to avoid duplication of effort. For example, the United States deemphasizes NBG-17 because it is included in the EU program. Similarly, a grade that has not been fully characterized in order to establish its baseline properties and variability should not be used for graphite core components. Grades such as IG-110 have been in continuous production and thus have production history to support their use.

Product size and availability of grade. The reactor designer must also consider the product (billet) size and availability of a grade of graphite before selecting it for a core component.

Longevity of supply, graphite and filler coke. The longevity of supply of a grade and its key ingredients, especially the filler coke, should also be considered. Mid-production changes in processing and feedstocks have the potential to alter the graphite's performance.

Cost. The price per pound of nuclear graphite varies from grade to grade according to the processing route. Therefore, a final consideration will be the cost of the graphite. This is tied to the component's lifetime and its potential re-use, but generally, the reactor designer will select the graphite with the lowest cost commensurate with the desired performance.

4. AVAILABLE GRAPHITE GRADES AND IRRADIATION CAPSULES

The available (as of Feb 2020) grades of nuclear graphite are summarized in Table 1:

Table 1. Available nuclear graphite grades (as of February 2020)

Graphite	Manufacturer	Coke type	AGC [a] and HDG [b] experiments	Prismatic candidate	Pebble-bed candidate	Of interest to MSR community
PCEA	AGM [c]	Pet	All	✓	✓	
NBG-18	SGL	Pitch	All		✓	
NBG-17	SGL	Pitch	All	✓ (non-US)		
IG-110	Toyo Tanso	Pet	All	✓	✓	✓
IG-430	Toyo Tanso	Pitch	AGC-1, 2	✓		✓
2114	Mersen	Non-Pet	3 onwards	✓	✓	✓

[a] AGC = advanced graphite creep
[b] HDG = high-dose graphite
[c] AGM = Amsted Graphite Materials

Further data on these graphite grades (Table 1) are given in Table 2

Table 2. Manufacturing details for the graphite grades presented in Table 1

Graphite grade	Forming method	Density (g/cm^3)	Filler size (µm)
PCEA	Extruded	1.83	
NBG-18	Vibrationally molded		1,800 maximum
		1.82	
NBG-17	Vibrationally molded		900 maximum
IG-110	Isostatically pressed	1.77	10 mean
IG-430	Isostatically pressed	1.82	10 mean
2114	Isostatically pressed	1.81	13

Grade 2114 was added to the irradiation program after AGC-2 because (1) it was considered that another superfine grain isotropic graphite would be of interest to MSR vendors, and (2) there was a desire to include a graphite from Carbone USA, subsequently Mersen. As a result, the number of grade 2114 specimens in capsule HDG-1 has been increased to ~50. Details of the irradiation capsules are presented in Table 3.

Table 3 Details of the graphite irradiation program

Capsule	Irradiation temperature	Accumulated dose	Major grades in capsule
AGC-1	470-716	2.73–6.92	IG-110, IG-430, PCEA, NBG-17, NBG-18, H-451
AGC-2	541-681	2–4.7[a]	IG-110, IG-430, PCEA, NBG-17, NBG-18, H-451
AGC-3	748-918	0.9–3.7[b]	IG-110, IG-430, PCEA, NBG-17, NBG-18, 2114
AGC-4	900 (des)	3.5 (des max)	IG-110, IG-430, PCEA, NBG-17, NBG-18, 2114
HTV	1500, 1200, 900 (Des)	1.49–3.34	NBG-17, H-451, IG-110, PCEA, NBG-18, 2114
HDG-1	600 (des)	10 (des max)	Re-irradiated AGC-2 plus new specimens
HDG-2	800 (des)	10 (des max)	Re-irradiated specimens from AGC-3 and -4

[a] capsule center section (i.e., creep section only)
[b] Entire capsule

It was originally planned that the AGC series of capsules would comprise six capsules at design irradiation temperatures of 600, 900, and 1200°C. However, the demise of the Next Generation Nuclear Plant (NGNP) project and the Very High Temperature Gas-Cooled Reactor (VHTGR) negated the need for such high-temperature graphite data. Moreover, it was further recognized that the program needed to increase the maximum dose attained by the capsules. Consequently, capsules HDG-1 and -2 were instigated; their design temperatures —700 and 800°C, respectively—were chosen with the gas-cooled reactor (GCR) in mind, although 700°C is very relevant to the MSR.

The AGC and HDG capsules will yield irradiation creep data, in addition to irradiation effects data.

5. REVISED SELECTION AND ACQUISITION PLAN

5.1 THE SELECTION AND ACQUISITION PLAN

The revised selection and acquisition plan has three major goals:

1. to support the current and planned irradiations (AGC 1 to 4 and HDG 1 and 2),
2. to support the ongoing characterization program, and
3. to support new production of existing grades.

5.1.1 SUPPORT CURRENT IRRADIATIONS

The first goal is to support the grades of graphite in our current irradiation capsules; that is, AGC-4 and the HDG series. The specific grades are NBG-17 (now deemphasized), NBG18, PCEA, IG-110, IG-430, and 2114. It should be noted that HDG-1 and HDG-2 include superfine and ultrafine grain size isotropic, high-strength grades such as IG-110, IG-430 and 2114, which are of interest to the MSR community. Moreover, there are fewer specimens of NBG-17 since there are no US HTR vendors who have selected this grade.

5.1.2 SUPPORT GRADES IN CHARACTERIZATION PROGAM

Grades that have only been partially characterization should be completed, except for NBG-17 which is not being considered by any US HTR vendors.

5.1.3 SUPPORT NEW PRODUCTION OF EXISTING GRADES IN PROGRAM

New NBG-18 should be irradiated in the next available HDG capsule. Similarly, if PCEA goes back into production, then new PCEA should be irradiated in the next available HDG capsule.

In summary, the new graphite selection and acquisition plan stipulates that no new billets will be purchased until there is new graphite production (NBG-18, PCEA), and existing supplies of NBG-17, NBG-18, PCEA, IG-110, IG-430, and 2114 are considered enough for future work.

6. QUALITY ASSURANCE

"The described technical work scope and related activities were conducted in accordance with the applicable requirements of the ASME NQA-1-2008 standard (including the 1a 2009 Addendum) entitled *Quality Assurance Requirements for Nuclear Facility Applications.* Project and activity-specific information concerning ORNL's application of the standard's requirements is provided in Document #QAP-ORNL-NR&D-01 entitled *Quality Assurance Plan for Nuclear Research and Development Conducted at the Oak Ridge National Laboratory."*

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