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THE NEXT GENERATION NUCLEAR PLANT GRAPHITE PROGRAM

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ABSTRACT

Developing new nuclear grades of graphite used in the core of a High Temperature Gas-cooled Reactor (HTGR) is one of the critical development activities being pursued within the Next Generation Nuclear Plant (NGNP) program. Graphite's thermal stability (in an inert gas environment), high compressive strength, fabricability, and cost effective price make it an ideal core structural material for the HTGR reactor design. While the general characteristics necessary for producing nuclear grade graphite are understood, historical "nuclear" grades no longer exist. New grades must be fabricated, characterized, and irradiated to demonstrate that current grades of graphite exhibit acceptable non-irradiated and irradiated properties upon which the thermo-mechanical design of the structural graphite in NGNP is based.

The NGNP graphite R&D program has selected a handful of commercially available types for research and development activities necessary to qualify this nuclear grade graphite for use within the NGNP reactor. These activities fall within five primary areas; 1) material property characterization, 2) irradiated material property characterization, 3) modeling, and 4) ASTM test development, and 5) ASME code development efforts. Individual research and development activities within each area are being pursued with the ultimate goal of obtaining a commercial operating license for the nuclear graphite from the US NRC.

INTRODUCTION

The Next Generation Nuclear Plant (NGNP) will be a helium-cooled High Temperature Gas Reactor (HTGR) with a large graphite core. Graphite physically contains the fuel, acts as the neutron moderator for this thermal reactor, provides an enormous heat sink for passive safety measures during off-normal (accident) conditions, and comprises the majority of the core volume. The basic technology for inert gas-cooled HTGR design is well established from the early graphite piles of the 1940s to the fully commercial reactor designs operating in the 1980s. These past designs represent the two primary core configurations commercially favored for gas reactors; the solid-block prismatic or the pebble-bed graphite core. While the USA has focused on a prismatic design, active interest in the Pebble Bed design is increasing primarily from the activities of PBMR (Pty.) Ltd. in South Africa (*1,2*). Graphite research for both designs are similar with the pebble-bed core requiring higher cumulative doses and the prismatic core experiencing higher temperatures. Estimated thermal and irradiation conditions for both prismatic and pebble bed core designs are listed in Table 1.

Table 1. Expected reactor operating conditions (*3*)

Parameter	Prismatic	Pebble-bed
Temperature (Normal operations)		
Inner reflector blocks	1050°C	600-1040°C
Fuel centerline	1200-1250°C	<1100°C
Peak fast fluence (FPY @ > 0.1 MeV)		
Inner reflector	$1.7 – 12.2x10^{20}$ n/cm^2	$1.6 – 12.2x10^{20}$ n/cm^2
Dose ($0.78x10^{21}$ n/cm^2 = 1 dpa)	0.19 – 0.85 dpa/FPY	0.18 – 0.85 dpa/FPY

All HTGR designs utilize interlinking graphite blocks to form an annular core configuration, Figure 1. The inner graphite reflector ring is expected to receive the highest operating temperature and dose rate in the reactor core other than the actual graphite fuel elements. The primary material issue for these inner and outer reflector blocks is one of long-term irradiation stability since the expected temperature ranges and applied stress levels are well within the limits for graphite. The reflector blocks are the primary structural component of the active core and for the pebble bed design are expected to stay inside the reactor as long as possible (15-20 years). At those exposure times the physical dimensions of the graphite blocks are expected to change dramatically as a result of irradiation induced dimensional changes (new basal plane formation between existing crystal planes arising from free interstials created during irradiation). Irradiation induced swelling well after turnaround is expected with increasing temperature accelerating these dimensional changes.

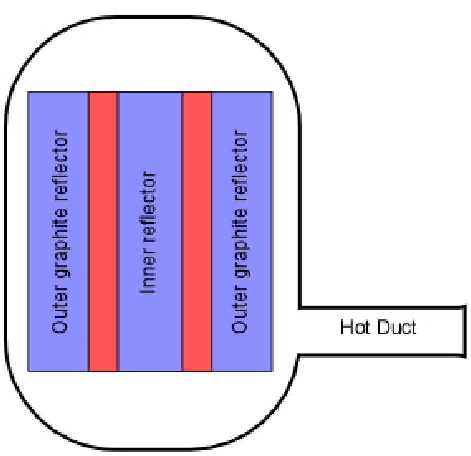

Figure 1. Schematic of annular graphite core for HTGR.

Unfortunately, neither graphite moderated HGTRs nor new nuclear grade graphite types have been built or fabricated since the 1980s. This requires not only the development of new nuclear graphite types but also re-establishing expertise in utilizing nuclear graphite in support of the NGNP program. This paper will discuss the individual research and development activities ongoing with the NGNP graphite program. These include the thermal, physical, and mechanical testing programs for the selected graphite types, an irradiated material property (irradiation creep) testing program, microstructural as well as whole-core modeling efforts, and the development of new ASTM test standards deemed necessary for graphite components in nuclear applications. Finally, our thoughts on gaining approval of the data contained within data packages to be vetted by the ASME and hopefully approved by the US NRC will be presented.

DEVELOPMENT OF NUCLEAR GRADE GRAPHITE

The nuclear graphite type H-451 previously used in the United States for HTGR graphite components is no longer available. New graphite types have been developed and are considered suitable candidates for the new NGNP reactor design. However, to support the design and licensing of NGNP core components within a commercial reactor design a complete properties database for these new, available, candidate grades of graphite must be developed. Data are required for the physical, mechanical (including radiation-induced creep), and oxidation properties of the graphite types (*4*). Moreover, the data must be statistically sound and take account of intra-billet, inter-billet, and lot-to-lot

Table 2. Candidate nuclear grade graphite types for NGNP (*4*)

Grade	Manufacturer	Coke type	Comments
IG-430	Toyo Tanso	Pitch coke	Isostatically molded, candidate for high-dose regions of NGNP concepts
NBG-17	SGL	Pitch coke	Vibrationally molded, candidate for high-dose regions of NGNP prismatic core concepts (not currently commercially available)
NBG-18	SGL	Pitch coke	Vibrationally molded, candidate for high-dose regions of NGNP pebble bed concepts; PBMR reflector graphite
PCEA	GrafTech International	Petroleum coke	Extruded, candidate for high-dose regions of NGNP prismatic core concepts
PGX	GrafTech International	Petroleum coke	Large blocks for permanent structure in a prismatic core (used in HTTR)
2020	Carbone of America	Petroleum coke	Isostatically molded, candidate for permanent structures in a prismatic core

variations of properties within the graphite. These data are needed to support the ongoing development of the risk-derived (probabilistic) American Society of Mechanical Engineers (ASME) graphite design code which is a departure from the deterministic approach traditionally used for codification. This is a consensus code being prepared under the jurisdiction of the ASME by gas cooled reactor vendors and NGNP researchers and stakeholders.

It's easily seen that the reactor type and operating conditions have major influences on the selection of graphite, Table 1. For a prismatic core design graphite with small grain size is necessary to accommodate the many fuel and coolant channels drilled throughout the fuel block. Pebble-bed reflector block designs do not have this requirement for the graphite since the fueled region does not have solid fuel blocks (*5*). Larger grain sized material can therefore be used in this design. In addition, due to anticipated operating conditions the reflector blocks in the pebble-bed core will experience much longer service times and thus much higher doses than anticipated for a prismatic graphite type. With such contrasting requirements the testing of multiple graphite types is needed.

Six graphite types (from four manufacturers) have been selected as commercially available, or near commercially available, candidates for the NGNP graphite R&D program. Experimental graphite grades are not being investigated as the NGNP is considered a near-term project, Table 2. Of these selected candidates the NBG-18 and PCEA grades are considered major grades most likely to meet the initial pebble-bed and prismatic design requirements, respectively. To determine the long term material property changes to these candidates an extensive characterization program has been implemented. The primary activities are outlined below.

Material property characterization

As noted earlier, a material property database recording the as-received material property values for each graphite type and the changes in those properties resulting from in-reactor exposure must be developed. Physical, mechanical, and thermal properties of the graphite types as well as the statistical variations of those properties within the graphite billets are being characterized in support of the as-received material database.

Baseline, “as-received” material properties for each graphite type are needed to establish accurate thermal and mechanical response of the graphite core. Since material properties are expected to vary throughout the rather large billets or bricks of graphite, mapping the magnitude and spacial positions of variability is important to determining an individual component’s material properties. To enable credible core designs and to support the ongoing development of a probabilistic graphite design methodology, the maximum variability within graphite components must be well characterized. A typical cutting plan illustrating the size, number and positions of various test coupons from a graphite billet is shown in Figure 2.

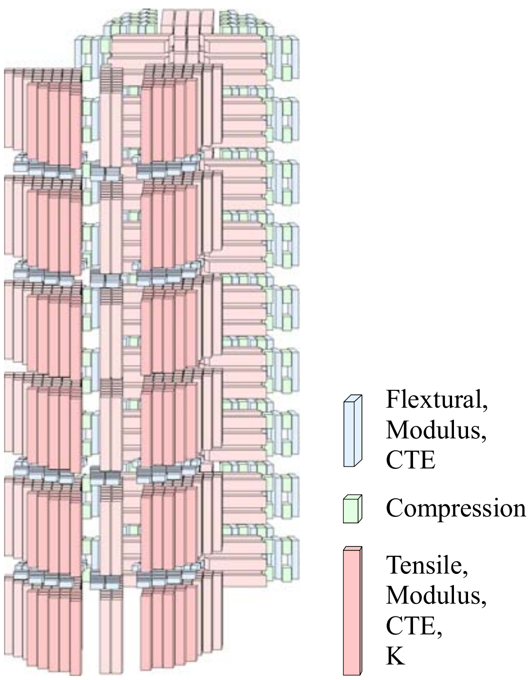

Figure 2. Schematic illustrating the different types of test blanks machined from a billet and the anticipated tests associated with those test coupons.

As depicted in Figure 2, a number of different tests can be conducted from the same test coupons allowing material property relationships to be determined (i.e. the effects of bulk density on thermal conductivity values). Both “with-grain” and “against-grain” material properties are tested to map differences resulting from fabrication processes. The NGNP program has developed an extensive sample cutting and sectioning plan for the graphite billets to guarantee not only statistically valid sample numbers but also spacial validity so that microstructural changes within the bulk material affecting material property changes are well characterized.

Once the baseline material properties for each graphite type have been established the irradiation induced or enhanced changes must be determined. The most important of these properties is the irradiation induced creep rate as a function of temperature and dose. Thermal creep of graphite is not expected at the temperatures experienced in the reactor core (< 1100°C). However, irradiation induced creep in graphite is expected at these temperatures and will play a significant role in the irradiated behavior of the graphite during reactor service. A series of irradiation experiments will be required to determine the graphite response under irradiation as compared to the baseline material properties from “as-received” graphite billets.

Irradiated material property characterization

The ATR Graphite Creep (AGC) experiments are designed to provide irradiation creep rates for moderate doses and higher temperatures of leading graphite types that will be used in the NGNP reactor design. The experiments are designed to provide not only static irradiation material property changes but also to determine irradiation creep parameters for actively stressed (i.e., compressively loaded) specimens during exposure to a neutron flux. The temperature and dose regimes covered by the AGC experiment are illustrated in Figure 3.

As shown, the dose and temperature is bounding for the prismatic reactor design (dpa ~ 5 – 6 at 1100°C) for both fuel and front facing reflector blocks. The dose limit of the experiment is intentionally below the expected point of turnaround for the current NGNP graphite types for normal operating temperatures. Only AGC-6 experiment (6 – 7 dpa at 1200°C) may approach expected turnaround limits for the selected NGNP graphite types. As such, to determine when (and if) turnaround will occur for the selected NGNP graphite during exposure in the AGC-6 capsule, an additional experiment – the high-temperature vessel (HTV) has been postulated. The HTV 1 and 2 capsules are simple “drop-in” capsules with the exposure parameters illustrated in Figure 3. As shown, these experiments are operated at much higher temperatures (inducing faster turnaround) but at lower doses. As this is a simple dimensional change experiment to determine when turnaround may occur, the graphite is not loaded during irradiation.

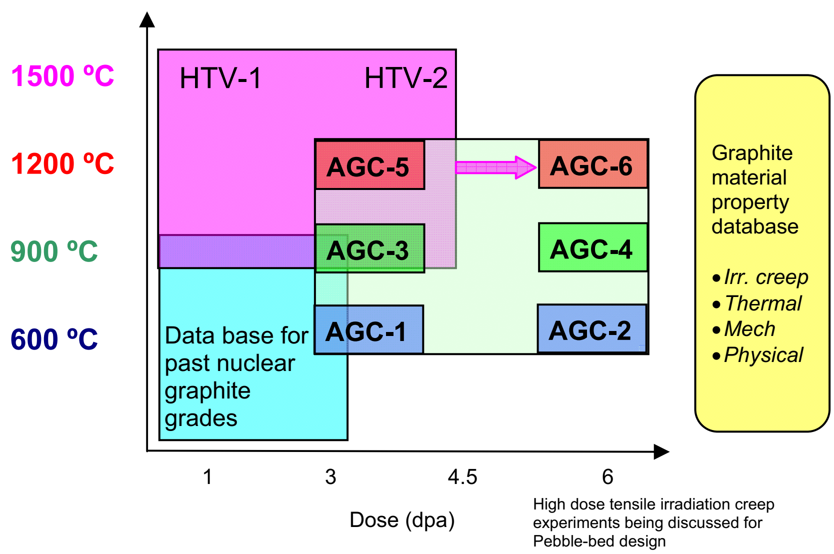

Figure 3. Schematic diagram illustrating dose and temperature ranges for AGC and HTV experiments.

The prismatic NGNP design assumes that fuel and reflector blocks can be replaced well before turnaround should occur at normal operating temperature (typically < 5 – 6 dpa). The AGC experiment should fully bound the graphite operating conditions within a prismatic design. However, the pebble-bed NGNP design assumes that the front facing reflector blocks will stay in reactor well beyond turnaround (typically 12-15 dpa) allowing minimal reactor shutdown time. While the AGC experiment will not fully bound pebble-bed graphite requirements it will certainly provide preliminary data for the first 30-40% of the expected dpa levels for these graphite components. Once the preliminary data has been established for the pebble-bed graphite high dose experiments can be considered at a later time. Since the graphite in these experiments is expected to experience turnaround the applied loads will need to be tensile loads.

Post irradiation examination and testing of the irradiated samples will follow. The dimensional changes of the loaded specimens will establish the creep rate while changes in the measured material properties of the unloaded specimens can be determined for the candidate graphite types.

Modeling

Sophisticated models will be required to allow designers to assess the condition of graphite components and core structure design margins at any point in the lifetime of the reactor. These models should describe any anticipated interactions between graphite components, specifically, the behavior of the stack of graphite blocks making up the core moderator and reflector. Specific models should be able to calculate external loads imposed upon the graphite components, internal stresses resulting from radiation and temperature induced dimensional changes, movement of components (i.e., dimensional clearances for control rod insertion), and estimates of residual strength both with and without environmental attack (i.e., air-ingress during off-normal event).

Modeling the behavior of a graphite core is complex and will require some fundamental understanding of the graphite physical, thermal, and mechanical behavior as a function of irradiation temperature and neutron fluence. However, the primary objective of these models is to provide the ability to calculate in-service stresses and strains in graphite components and estimate the structural integrity of the core as a whole. Thus, understanding of fundamental mechanistic material behavior during operation will be limited to those aspects required to understand the response of the entire core both during normal operation and during off-normal events (e.g. predict seismic behavior of the core). While a physics-based understanding of microstructural damage and its effects on material structure and properties will provide an initial start to estimating the amount of changes to a graphite component the actual degree of change is unique to the specific nuclear graphite grade and these fundamental principles must be supplemented with actual experimental material property data to provide a complete analysis of the core behavior.

Finally, microstructural and even experimental material tests obtain data from small volumes of material, not large components. Behavioral models can provide point-to-point flux, temperature, and stress state estimates for all components (or parts of components) throughout the entire core. A whole core model will therefore use a combination of experimentally derived material properties underpinned by an understanding of the fundamental physics to account for all variations possible within the graphite components of the core. Consequently, a major goal is the development and validation of multi-scale models for determining the behavior of graphite core components, and whole graphite cores for use in licensing and continued operational safety assessments. This activity is considered critical to obtaining a commercial operating license for the nuclear graphite from the US NRC.

STANDARDS AND CODIFICATION ACTIVITIES

The NGNP graphite program will use the ASME codification process to verify the viability and validity of the nuclear grade graphite used in the reactor core. The ASME simply provides an independent and rigorous method of vetting the graphite material property data and models. This decision was made with the assumption that the licensing review process should be accelerated since the regulators are already assured that the data and models were developed through an approved ASME methodology.

Recently, there has been significant activity within the ASME to develop appropriate codes for using graphite in nuclear reactor applications. This is a consensus code being prepared by an international working group of graphite experts to insure universal acceptance by all countries worldwide. One of the significant issues being addressed by this working group is the inclusion of a probabilistic failure methodology rather than the strict use of a traditional deterministic approach which is more conservative. It is our assumption that developing a code utilizing more probabilistic methods will provide a more realistic failure determination for the brittle, ceramic-like graphite than a strict deterministic methodology that has been used previously for metallic (ductile) systems.

Graphite fabrication results in a distribution of flaws (i.e. pores and microcracks) throughout the graphite microstructure, Figure 4. Due to the brittle nature of graphite failure and this distribution of flaws within the microstructure the graphite therefore has a distribution of ultimate strengths within the billet and from billet to billet. It is our view that a deterministic design approach that relies on a single failure load is not completely adequate for this type of material. Statistical or probabilistic design approaches can more accurately reflect the stochastic material strengths. Probabilistic design methodology combines the statistical nature of the strength-controlling flaws with the mechanics of the crack growth to allow for the multiaxial stress states, concurrent flaw populations, and component size/scaling effects.

This focus on a design approach which utilizes more probabilistic mythology requires an adequate failure theory of graphite under multiaxial loads and a program that takes the statistical behavior of graphite strengths and integrates over the population of flaws to obtain survival probability.

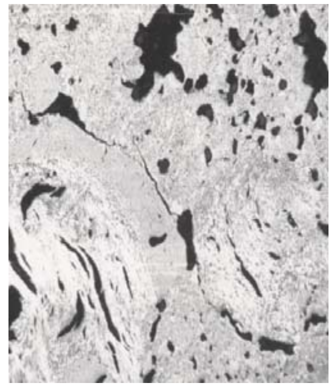

Figure 4. An optical photomicrograph of the microstructure of grade H-451 graphite revealing the presence of pores, coke filler particles, and cracks.

To this end, the ASME code stresses material property data of graphite over a large range of billets and lots to provide not only the statistical variability needed but also the multiaxial material properties necessary to support such a probabilistic failure methodology.

Finally, it is assumed that the ASME will only approve of a nuclear graphite material property database if it is populated with data obtained from ASTM approved test standards. Most of the non-irradiated standards already exist with some small exceptions such as oxidation testing and fracture toughness testing. Irradiated material property standards are problematic due to the limited size of the irradiation volumes inside material test reactors. At issue is the small test specimen size necessary for irradiation tests and the statistical validity of the resulting material properties measured from these diminutive samples. A large portion of the ASTM approved tests are affected by this issue. Consequently, there are significant activities in this area. However, it is believed that the existing ASTM standards are adequate to fully characterize these new, available, candidate grades of graphite.

SUMMARY AND FURTHER COMMENTS

NGNP Graphite program has selected six candidate graphite types viewed as “commercially available” with two grades (PCEA & NBG-18) designated as major grades which will most likely meet the initial pebble-bed and prismatic design requirements. However, in order to support a statistically based probabilistic design methodology an extensive non-irradiation material characterization program must be implemented for all major types of graphite.

In addition, an expensive irradiated characterization program is underway which will gather data on the important parameter of irradiation creep but also the material property changes as a function of creep and dose. These irradiation induced changes to the graphite material properties are needed for multi-scale behavior models to provide point-to-point flux, temperature, and stress state estimates for all components (or parts of components) throughout the entire core. This activity is considered critical to obtaining a commercial operating license for the nuclear graphite from the US NRC.

Finally, two long-term activities must be considered in such a research program; establishing alternative coke sources and development of viable graphite recycle and reuse options. Currently, no coke source is capable of sustaining long term (60+ years) consistent graphite that can be used over the entire lifetime of a reactor. This does not even consider the possibility of additional graphite moderated reactors after this initial program. A production method should be developed that would be completely different from the current by-product coke oven route and thus not subject to the same environmental and economic pressures.

Further, discharged graphite from reactor could be taken to a dedicated facility, ground into powder, and annealed (~3000°C) at high temperature with the activity being captured. The graphite could then be reconstituted into new billets, or used as an additive in the production of new, recycled billets. Technology to achieve these two activities is currently available and could eventually aid in the viability of these unique reactor types.

ACKNOWLEDGMENTS

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REFERENCES

1. E. Wallace, R. Matzie, R. Heiderd, J. Maddalena, “From field to factory – Taking advantage of shop manufacturing for the pebble bed modular reactor”, Nuc. Eng. and Design, 236 (2006) 445-453.
2. A. Koster, H. Matzner, and D. Nicholsi, „PBMR design for the future“, Nuc. Eng. and Design, 222 (2003) 231-245.
3. W. Windes, T. Burchell, and R. Bratton, “Graphite Technology Development Plan” INL/EXT-07-13165, Idaho National Laboratory, September 2007
4. T. Burchell, R. Bratton, and W. Windes, “NGNP Graphite Selection and Acquisition Strategy”, ORNL/TM-2007/153, September 2007
5. T. R. Allen, et.al. “Materials Challenges for Generation IV Nuclear Energy Systems”, Nuclear Technology, 162 (2008) 342-357