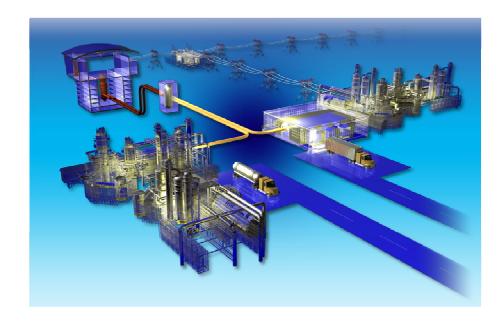
# **NGNP High Temperature Materials White Paper**

June 2010



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June 2010

Idaho National Laboratory
Next Generation Nuclear Plant Project
Idaho Falls, Idaho 83415

Prepared for the
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Office of Nuclear Energy
Under DOE Idaho Operations Office
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# **Next Generation Nuclear Plant Project**

# **NGNP** High Temperature Materials White Paper

# INL/EXT-09-17187

# June 2010

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#### **ABSTRACT**

The Next Generation Nuclear Plant (NGNP) will be a licensed commercial high temperature gas-cooled reactor (HTGR) plant capable of producing electricity and high temperature process heat for industrial markets supporting a range of end-user applications. The NGNP Project has adopted the 10 CFR 52 Combined License (COL) application process, as recommended in the *Report to Congress*, dated August 2008, as the foundation for the NGNP licensing strategy. Nuclear Regulatory Commission (NRC) licensing of the NGNP plant utilizing this process will demonstrate the efficacy of licensing future HTGRs for commercial industrial applications. This white paper is one in a series of submittals that will address key generic issues of the COL priority licensing topics as part of the process for establishing HTGR regulatory requirements.

The result of reviews of existing policies, regulations, and guidance associated with acceptable materials for HTGR applications is documented. It includes development of a process for high-temperature component material selection and evaluation, leading to recommendations for qualification and acceptance of HTGR components. Metallic and nonmetallic materials proposed for high-temperature service within the NGNP are identified and assessed in terms of supporting codes and standards and the existing bases for design and qualification. As part of this assessment, the processes for establishing the expected material performance requirements under operating and accident conditions are also described.

The information in this paper is intended to serve as the basis for interactions with the NRC staff. The NGNP Project wishes to obtain comments on the adequacy of the planned approach and feedback on a number of issues that have the potential to significantly impact the effort and schedule to prepare a COL application for the HTGR-based NGNP.

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### **ACRONYMS**

AGC Advanced Graphite Capsule

AGR Advanced Gas-cooled Reactor

ASME American Society of Mechanical Engineers

ASTM ASTM International

ATR Advanced Test Reactor

AVR Arbeitsgemeinschaft Versuchsreaktor (Germany)

B&PV boiler and pressure vessel

CFR Code of Federal Regulations

CFRC carbon fiber reinforced carbon

COL Combined License

CSC core structure ceramic

CTE coefficient of thermal expansion

DBA Design Basis Accident

DEB Design Basis Event

DOE Department of Energy

DPP Demonstration Power Plant

EDN equivalent dido nickel

EU European Union

FSV Fort St. Vrain

GDC General Design Criteria (10 CFR 50 Appendix A)

GE General Electric

GTMHR gas turbine modular helium reactor

HTGR high temperature gas-cooled reactor

HTR high temperature reactor

HTR-10 High Temperature Test Reactor (China)
HTTR High Temperature Test Reactor (Japan)

INL Idaho National Laboratory

ISI in-service inspection

IHX intermediate heat exchanger

JAEA Japan Atomic Energy Agency

LBE Licensing Basis Event

LWR light water reactor

MHTGR modular high temperature gas-cooled reactor

MQP Materials Qualification Plan

MTR Materials Test Reactor

NGNP Next Generation Nuclear Plant
NRC Nuclear Regulatory Commission

NUREG Nuclear Regulatory Commission Report

ORNL Oak Ridge National Laboratory
PBMR Pebble Bed Modular Reactor

PGA Pile Grade A

PIRT Phenomena Identification and Ranking Table

PRA Probabilistic Risk Assessment

RDMCI Requirements for the Design and Manufacture of the Ceramic Internals

RG Regulatory Guide

RIM Reliability and Integrity Management

RPV reactor pressure vessel

SECY NRC Commissioner's Document (acronym)

SRC structural reliability class
SRP Standard Review Plan

SSC structures, systems, and component

SST stainless steel

THTR Thorium High Temperature Reactor

VHTR very high temperature reactor

# **NGNP High Temperature Materials White Paper**

# 1. INTRODUCTION

# 1.1 Purpose

This paper is one in a series of white papers that address key generic licensing issues in preparation for the submittal of a Combined License (COL) application for the Next Generation Nuclear Plant (NGNP). The NGNP will use advanced, high temperature gas-cooled reactor (HTGR) technologies to demonstrate the integration of a nuclear heat source, providing electricity and/or process steam, with one or more industrial applications. The purpose of these white paper submittals is to reduce the time required for COL application review by identifying and addressing key regulatory issues and obtaining agreements for achieving their resolution with the Nuclear Regulatory Commission (NRC).

The scope of this paper is to review the existing policies, regulations, and guidance associated with acceptance of materials for nuclear reactor applications and to assess the bases for their implementation in the system components of the HTGR. Following review of the existing regulatory framework for materials, a process is developed for high temperature component material selection and evaluation, leading to recommendations for qualification and acceptance. The principal materials proposed for application in the NGNP primary system are then identified, along with the proposed approaches for establishing regulatory compliance. For those cases in which the established regulatory infrastructure for qualification and acceptance is determined to be sufficient, regulatory issues are identified along with proposed bases for their resolution.

The design of the HTGR is in the initial conceptual design phase, so final component specification and material selection has yet to be performed. Still, typical component performance requirements and candidate materials for specific applications are evaluated to identify potential qualification and acceptance gaps.

# 1.2 Objectives of the White Paper

Development of this white paper considers applicable information from sources such as (1) past papers on materials, as provided in attached reference, (2) NRC regulatory guidance, (3) insights gained from NRC public meetings, (4) available industry standards, (5) Modular High Temperature Gas-cooled Reactor (MHTGR) licensing documents, (6) Pebble Bed Modular Reactor (PBMR) licensing documents, (7) American Society of Mechanical Engineers (ASME) Codes and Code Cases, and (8) other gas-cooled reactor documents.

The primary objectives of this white paper are to:

- Summarize the existing regulatory policies and guidance that may apply to materials expected to be used in HTGRs.
- Describe an approach for selecting materials, identifying properties, qualification, and accepting materials for key gas-cooled reactor components.
- Discuss the influence that material selection and code requirements may have on licensing basis events (LBEs), including design basis accidents (DBAs).
- Discuss any needed codes and standards work, including the status and schedule for code and standards activities already in progress.

• Identify policy and technical issues that should be discussed and resolved with the NRC.

The desired outcome of this white paper is to obtain NRC agreement with the recommended approach for qualification and regulatory acceptance of materials for the high temperature service conditions of the HTGR. Specific topics for which NRC feedback is requested are identified in Section 5, Outcome Objectives.

# 1.3 Related Licensing Topics

Two related licensing topics have been identified that, while beyond the scope of this paper, have the potential to influence the selection and qualification of high temperature materials for service in HTGRs. These topics are summarized as follows:

- NRC acceptance of HTGR LBE selection and categorization. LBEs are event scenarios considered in the licensing process and used to derive regulatory requirements for design certification. LBEs include normal plant operation, events anticipated to occur over the life of the plant, and off-normal events as required by 10 CFR Part 52, including infrequent Design Basis Events (DBEs) and rare events beyond the design basis.
- NRC acceptance of HTGR structures, systems, and components (SSC) classification. The classification of SSCs with respect to safety functions provides an essential input to the establishment of design and performance requirements.

The NGNP reactor design and its unique passive and inherent safety characteristics rely on key material properties to define performance during normal operation, anticipated operational occurrences, and accident conditions within the design basis. In addition to the qualification of materials properties that provide for acceptable performance during normal operation at elevated temperatures in a helium environment under neutron irradiation, the qualification of materials for use in the NGNP reactor must include certain material properties relied upon during accident scenarios. Further, the materials qualification basis must provide assurance that such properties stay within their design range for the life of the component.

The licensing and technical issues and the recommended resolutions associated with LBE selection and SSC classification will be discussed in separate white papers; however, the basic assumption made in this white paper is that the resolutions of these issues are such that the fundamentals of the HTGR safety case are preserved.

# 2. CURRENT REGULATORY BASIS

#### 2.1 Materials

Light water reactors (LWRs), which are the basis for current NRC commercial reactor regulations, typically use metals for their primary loop components because of the relatively low temperatures encountered in these reactors. HTGR technology requires expanding the use of primary loop component materials to include nuclear grade graphite, composite materials, and other ceramics where temperatures are higher than those allowed for metals.

All commercial nuclear power plants currently operating in the United States use water as both the heat transport medium and the neutron moderator. The HTGR concept discussed in this paper; however, uses helium as the heat transport medium and nuclear grade graphite as the neutron moderator. This represents a significant difference in reactor technology. With the exception of Fort St. Vrain (FSV) and Peach Bottom 1, no graphite-moderated gas-cooled reactors have been licensed commercially to operate within the United States. The NRC conducted preliminary safety reviews for the large gas-cooled reactors in the 1970s and for modular high temperature reactors in the 1980s and 1990s. More recently, interactions took place between the NRC and General Atomics on the MHTGR and gas turbine modular helium reactor (GTMHR) design and with Exelon and Pebble Bed Modular Reactor (Pty) Ltd. (PBMR) on the PBMR design.

Metallic components used in the primary system of an HTGR include the reactor vessel, cross vessel, piping, steam generator vessel, and/or intermediate heat exchanger (IHX), as well as components within these vessels, including the core barrel, core support structures, and steam generator tubes. In addition, nuclear grade graphite is used for HTGR fuel blocks in prismatic reactors, and for reflector blocks and core support components in all HTGRs. Baked carbon is used in conjunction with the reflector blocks in some designs to provide a higher degree of thermal insulation between the core and metallic components. Other ceramic, composite or metallic materials may be used for the cross-vessel liners, reflector supports and/or core reactivity control elements.

HTGR primary loop components operate in a different environment (helium with controlled levels of impurities) and, in many cases, at higher temperatures during both normal operation and LBEs than those applicable to LWRs. Materials needed to manufacture such HTGR components are, in general, commercially available. Some have been used in HTGRs both within and outside of the United States. This paper proposes a path for regulatory acceptance, qualification, and/or approval of these materials for use in an HTGR environment.

The sections that follow provide an assessment of NRC regulations, regulatory guidance, policy statements, standards, and past precedents that are considered relevant to materials used in nuclear reactor components. The objective is to identify regulations that may be applicable to or provide insights regarding the regulatory basis for qualification of materials for HTGRs.

# 2.2 NRC Regulations

This section identifies NRC regulations that may have potential relevance to materials used in HTGR primary system components. Because current regulations have been established primarily for application to LWR technologies, it is natural that most existing regulations are for metallic materials. This review did not identify regulations that refer specifically to graphite components; however, some insights may be obtained from regulatory requirements applicable to metallic materials.

The NRC regulations applicable to LWR primary system components are provided in the following *Code of Federal Regulations* (CFR) sections:

- 10 CFR 50.55a, "Codes and Standards"
- 10 CFR 50.34, "Contents of Applications; Technical Information," particularly Section (a)(3) addressing "Principal Design Criteria"
- 10 CFR 50.61, "Fracture Toughness Requirements for Protection Against Pressurized Thermal Shock Events"
- 10 CFR Part 50, Appendix A, "General Design Criteria (GDC)," 4, 10, 14, 15, 30, and 31
- 10 CFR Part 50, Appendix B, "Quality Assurance Criteria for Nuclear Power Plants and Fuel Reprocessing Plants"
- 10 CFR Part 50, Appendix G, "Fracture Toughness Requirements"
- 10 CFR Part 50, Appendix H, "Reactor Vessel Material Surveillance Program Requirements."

The quality assurance criteria and requirements provided in 10 CFR Part 50 Appendix A-GDC 1 and Appendix B generally apply to nuclear reactor components, irrespective of the component operating temperature. Therefore, these requirements are not discussed in this paper. Key elements of the other identified regulations are summarized below.

- 10 CFR 50.55a. Section 50.55a requires that SSCs must be designed, fabricated, erected, constructed, tested, and inspected to quality standards commensurate with the importance of the safety function to be performed. In addition, 10 CFR 50.55a requires that systems and components of boiling and pressurized water nuclear power reactors meet applicable requirements of the ASME Boiler and Pressure Vessel (B&PV) Code.
  - Section 50.55a also includes a provision for the applicants to propose alternative solutions provided (a) the proposed alternatives would provide an acceptable level of quality and safety, or (b) compliance with the specified requirements of this section would result in hardship or unusual difficulty without a compensating increase in the level of quality and safety.
- 10 CFR 50.34. Under the provisions of 10 CFR 50.34, 52.47, 52.79, 52.137, and 52.157, an application for construction permit, design certification, combined license, standard design approval, or manufacturing license, respectively, must include the principal design criteria for a proposed facility. The principal design criteria establish the necessary design, fabrication, construction, testing, and performance requirements for SSCs important to safety. It must provide reasonable assurance that the facility can be designed, constructed, and operated without undue risk to public health and safety.
- 10 CFR 50.61 and Appendices G and H to 10 CFR Part 50. This information addresses fracture toughness and associated surveillance requirements for ferritic materials used in the pressureretaining components of the reactor coolant pressure boundary. These requirements, which pertain specifically to LWRs, are designed to provide adequate margins of safety during normal operation, including anticipated operational occurrences and system hydrostatic tests. These requirements are included in both Section III and Section XI of the ASME Code.
- 10 CFR Part 50, Appendix A, "General Design Criteria (GDC)." The GDC in 10 CFR Part 50, Appendix A establish minimum requirements for the principal design criteria for LWRs similar in design and location to plants for which construction permits have already been issued. Some GDC are generally applicable to other types of nuclear power units, except for those that are LWR technology specific. GDC that are technology specific to LWRs may, however, provide guidance in establishing the principal design criteria for non-LWR reactor technologies.

The following GDC may be relevant to both metallic and nonmetallic materials unless specifically indicated:

- GDC 4, "Environmental and Dynamic Effects Design Bases," requires that SSCs important to safety shall be designed to accommodate the effects of and to be compatible with the environmental conditions associated with normal operation, maintenance, testing, and postulated accidents, including loss-of-coolant accidents. These SSCs shall be appropriately protected against dynamic effects, including the effects of missiles, pipe whipping, and discharging fluids that may result from equipment failures and events and conditions outside the nuclear power unit. However, dynamic effects associated with postulated pipe ruptures in nuclear power units may be excluded from the design basis when analyses reviewed and approved by the NRC demonstrate that the probability of fluid system piping rupture is extremely low under conditions consistent with the piping design basis.
- GDC 10, "Reactor Design," requires that the reactor core and associated coolant, control, and protection systems be designed with appropriate margin to ensure that specified acceptable fuel design limits are not exceeded during any condition of normal operation, including the effects of anticipated operational occurrences.
- GDC 14, (metals only) "Reactor Coolant Pressure Boundary," requires that the reactor coolant pressure boundary be designed, fabricated, erected, and tested so as to have an extremely low probability of abnormal leakage, of rapidly propagating failure, and of gross rupture.
- GDC 15, "Reactor Coolant System Design," requires that the reactor coolant system and associated auxiliary, control and protection systems be designed with sufficient margin to assure that the design conditions of the reactor coolant pressure boundary are not exceeded during any condition of normal operation, including anticipated operational occurrences.
- GDC 30, (metals only) "Quality of Reactor Coolant Pressure Boundary," requires components that are part of the reactor coolant pressure boundary to be designed, fabricated, erected, and tested to the highest quality standards practical. Means shall be provided for detecting and, to the extent practical, identifying the location of the source of reactor coolant leakage.
- GDC 31, (metals only) "Fracture Prevention of Reactor Coolant Pressure Boundary," requires that the reactor coolant pressure boundary be designed with sufficient margin to ensure that, when stressed under operating, maintenance, testing, and postulated accident conditions, (1) the boundary behaves in a nonbrittle manner, and (2) the probability of rapidly propagating fracture is minimized. The design shall reflect consideration of service temperatures and other conditions of the boundary material under operating, maintenance, testing, and postulated accident conditions and the uncertainties associated therewith in determining (1) material properties; (2) the effects of irradiation on material properties; (3) residual, steady state, and transient stresses; and (4) size of flaws.

The NRC regulations identified in this section are potentially applicable to high temperature components of the NGNP reactor. The interpretation and application of these current NRC regulations must consider the differences between the principal safety functions of the HTGR and the LWR technologies in addition to the inherent reactor characteristics and passive core decay heat removal capabilities of HTGRs. In addition to determining which current NRC regulations may not apply to HTGRs due to their unique characteristics, it is important to determine whether those unique characteristics create the need for additional regulatory guidance and agreements to complete the NGNP design and license application.

# 2.3 NRC Policy Statements

The NRC has published no policy statements that explicitly address the performance of materials for HTGR components. However, the Commission has made policy statements and other official comments on advanced reactor issues which address some of the issues associated with the use and acceptance of materials used at HTGR conditions. These include:

- The *Policy Statement on the Regulation of Advanced Reactors* (October 2008)
- Commission vote on the NRC Commissioner's Document SECY-03-0047, "Policy Issues Related to Licensing Non-Light Water Reactor Designs"
- SECY-08-0019, "Licensing and Regulatory Research Related to Advanced Nuclear Reactors"
- SECY-10-0034, "Potential Policy, Licensing, and Key Technical Issues for Small Modular Nuclear Reactor Designs."

The *Policy Statement on the Regulation of Advanced Reactors* has been revised several times since its original release in 1986. The most recent revision, published in the *Federal Register* on October 14, 2008, does not differ substantially from previous versions in its discussion of the Commission's expectations with regard to advanced reactor features; rather, the Commission added a discussion concerning emergency preparedness and security in light of the events of September 11, 2001, and subsequent new regulatory requirements for enhanced security and protection from aircraft attacks.

While the *Policy Statement* delineates those attributes of advanced plant designs that the Commission finds highly desirable, it does not specifically refer to the materials used to fabricate plant structures or components. However, several points can be read as being broadly applicable to the plant materials, including those used in the HTGR. These points include:

- Designs that minimize the potential for severe accidents and their consequences by providing sufficient inherent safety, reliability, redundancy, diversity, and independence in safety systems, with an emphasis on minimizing the potential for accidents over minimizing the consequences of such accidents.
- Designs that provide easily maintainable equipment and components.
- Design features that can be proven by citation of existing technology or that can be satisfactorily established by commitment to a suitable technology development program.
- Designs that incorporate the defense-in-depth philosophy by maintaining multiple barriers against radiation release and by reducing the potential for, and consequences of, severe accidents.

These attributes will be considered when selecting materials to be used in HTGR designs.

In SECY-03-0047 and the corresponding Staff Requirements Memorandum, the NRC addressed a number of key policy issues that it had been identified in early discussions with the designers of and prospective license applicants for non-LWRs. One such issue is related to the potential use of international codes and standards where the U.S. codes and standards incorporated in the NRC's regulations did not adequately address non-LWR designs. The NRC's direction to the staff was to "Review international codes and standards only as part of an application or preapplication review. The staff should gain experience through review of international codes and standards during the preapplication and application reviews of non-LWRs then apply the lessons-learned from these reviews to their activities involving our domestic codes and standards committees."

# 2.4 NRC Regulatory Guidance

The regulatory guidance for the design, fabrication, and inspection of nuclear reactor components includes Regulatory Guides (RGs) 1.84, 1.87, 1.147, 1.174 and 1.178. These guides provide additional insight into acceptable methods and criteria for nuclear primary system components in support of regulatory requirements discussed in Section 2.2.

#### 2.4.1 RG 1.84

RG 1.84, Rev. 34, "Design, Fabrication, and Materials Code Case Acceptability," ASME Section III provides guidance on the acceptable uses of ASME Section III code cases applicable to materials and component design, fabrication, examination, and testing. The ASME code cases referenced in this regulatory guide for Class 1 components are currently applicable only to LWR metallic materials.

#### 2.4.2 RG 1.87

RG 1.87, Rev. 1, "Guidance for Construction of Class 1 Components in Elevated-Temperature Reactors" (Supplement to ASME Section III Code Cases 1592, 1593, 1594, 1595 and 1596) describes five code cases that provide guidance for the construction of components subject to elevated temperature service, including HTGR components. RG 1.87 states that the service temperatures and load conditions for HTGRs are such that time-dependent phenomena such as creep and relaxation are important. It further states that Subsection NB of Section III of the ASME Code does not provide adequate guidance for construction of components subject to elevated-temperature service, thus leading to the development of the five ASME code cases as an interim step. The referenced code cases cover design, fabrication, installation, examination, testing, and protection against overpressure for such components. They reflect both time-independent and time-dependent materials properties and structural behavior (elastic and inelastic) by considering the following modes of failure:

- Ductile rupture from short-term loadings
- Creep rupture from long-term loadings
- Creep-fatigue failure
- Gross distortion caused by incremental collapse and ratcheting
- Loss of function caused by excessive deformation
- Buckling caused by short-term loadings
- Creep buckling caused by long-term loadings.

RG 1.87 also states that component designs should accommodate the required in-service inspection (ISI) and surveillance programs for material or component integrity. Finally, it states that the materials evaluations should address representative environmental factors such as compatibility with the coolant (helium) and potential contaminants in the coolant, irradiation effects that might induce ductility loss, and aging resulting from prolonged exposure to elevated temperature.

The code cases referenced within RG 1.87 were superseded by ASME Code Cases N-47 through N-51 (with numerous revisions) and, subsequently, by Section III, Subsection NH. To date, Subsection NH has attained acceptance as a basis for regulatory compliance for only one specific application. The current version of 10 CFR 50.55a(b)(1)(vi) states:

(vi) *Subsection NH*. The provisions in Subsection NH, "Class 1 Components in Elevated Temperature Service," 1995 Addenda through the latest edition and addenda incorporated by reference in paragraph (b)(1) of this section, may only

be used for the design and construction of Type 316 stainless steel (SST) pressurizer heater sleeves where service conditions do not cause the component to reach temperatures exceeding 900°F.

#### 2.4.3 RG 1.147

RG 1.147, Rev. 15, "In-service Inspection Code Case Acceptability, ASME Section XI, Division 1," provides guidance on ASME Section XI code cases oriented to ISI programs that are generally acceptable to the NRC staff. The code cases identified in this Regulatory Guide are incorporated by reference within 10 CFR 50.55a for application to LWRs. While their use for HTGRs is not specifically addressed, many of the permitted examination and repair activities addressed by the Section XI Code cases could potentially be applied to HTGR components.

#### 2.4.4 RG 1.174 and RG 1.178

NRC recently provided guidance for application of risk-informed methodologies in meeting current regulations. RG 1.174, Rev. 1, "An Approach for Using PRA in Risk-Informed Decisions on Plant-Specific Changes to the Licensing Basis," and RG 1.178, Rev. 1, "An Approach for Plant-Specific Risk-Informed Decision Making for In-service Inspection of Piping," provide an acceptable path to establish risk-informed technical specification modifications and ISI programs for LWR components and piping systems. Although these guides cannot be directly applied to HTGRs because they are linked to LWR risk metrics of core damage frequency and large release frequency, they do provide a potential path for the development of risk informed decisions that link requirements to plant-specific risk metrics.

#### 2.4.5 Other Guidance

NRC report NUREG-0800, the Standard Review Plan (SRP), provides detailed guidance to the NRC staff for regulatory reviews (e.g., construction permits, operating licenses, design certifications) of LWRs. However, it is also useful to designers, applicants, and licensees, insofar as it describes the acceptance criteria that the staff applies in its reviews. SRPs may also refer to Regulatory Guides in describing acceptable methodologies. Of interest to this discussion are the SRP Sections 5.2.1.1, "Compliance with the Codes and Standards Rule"; 10 CFR 50.55a, 5.2.1.2, "Applicable Code Cases"; and 5.2.3, "Reactor Coolant Pressure Boundary Materials." Although these SRP sections are not directly applicable to primary systems in HTGRs, they do indicate that material selection considerations should include the evaluation of issues such as susceptibility of the material in the reactor coolant pressure boundary to cracking and corrosion, fracture toughness, compatibility of the materials with the reactor coolant (including contaminants in the coolant), and compatibility of the materials in the reactor coolant pressure boundary with the materials in the insulation. These material selection considerations are applicable to the material section process for any reactor design, including HTGRs.

Furthermore, in 2003, the NRC published NUREG/Contractor Report CR-6824, "Materials Behavior in HTGR Environments," which addresses the performance of metallic components in high temperature helium-cooled reactors. This document includes information on HTGR materials properties and environmental effects on the behavior of metallic components in gas-turbine HTGR technology with a core outlet temperature range of 850 to 900°C (1562 to 1652°F). As noted in Section 2 of that report, the selected materials should have adequate performance over long service life at temperatures in the range of 900 to 950°C (1652 to 1742°F).

# 2.5 Regulatory Precedents

The NRC regulatory precedents for graphite-moderated HTGRs and, more specifically, modular HTGRs were developed in two distinct time periods. Early safety reviews include those performed for Peach Bottom 1, FSV, and the large HTGR designs by General Atomics. More recent licensing interactions include preapplication reviews of the MHTGR, early preapplication reviews of the GTMHR, and two separate series (PBMR/Exelon in 2002 and PBMR (Pty) Ltd. in 2007) of early licensing interactions in support of the PBMR design.

NRC regulatory experience with these reactor concepts began with the U.S. Department of Energy (DOE)-sponsored MHTGR program. The results of the NRC's review of that concept are published in NUREG-1338, "Draft Preapplication Safety Evaluation Report for the Modular High-Temperature Gas-Cooled Reactor," which was initially issued by the NRC in 1989 and updated in 1995. NUREG-1338 notes that lower-level criteria for the design and review of the MHTGR primary system, such as those contained in the SRP, do not exist in a form approaching that for LWR primary systems. NUREG-1338 goes on to note that certain LWR criteria for primary systems are helpful and important in guiding the MHTGR conceptual design, but that significant gaps remain, particularly those related to safety issues.

During its 2001 preapplication review of PBMR, the NRC staff provided feedback on various technical, safety, and policy issues raised by Exelon for that reactor concept. With regard to a path for the NRC review and approval of materials used in the construction of HTGR components, the staff provided the following direction:

...A list should be provided of all materials used for the reactor pressure vessel and its appurtenances, core support structures, primary system boundary, connecting piping, and other components important to safety and the applicable material specifications, design stress and time at temperature and other environmental conditions. The identification of the grade or type and conditions of the materials to be placed in service would also be required. If the code approved material specifications for the intended applications are not available, relevant material specifications should be developed following the format of [ASTM International] specifications. The subject specifications should be supported by the data and information as identified in ASME Code, Section III, Appendix IV, for approval of the new materials. Additional information unique to the application in the PBMR environment and condition shall also be provided...

# 2.6 PIRT Workshops

The NRC staff conducted a series of workshops in 2007 that applied the PIRT methodology as a means of identifying and prioritizing HTGR-related issues specific to the NGNP.<sup>3</sup> The purpose of these workshops was to assist NRC in prioritizing research and allocating available resources. High-temperature materials and graphite are among the several subjects addressed within these workshops.

A follow-on workshop was conducted in March 2009 to further assess the status of worldwide research on nuclear graphite and identify the technical gaps between the planned DOE research and the outcome of the Graphite PIRT conducted earlier.

These workshops informed the NGNP Research and Development Program Plan revisions in 2008 and 2010 and also informed the selection of technology studies (e.g., reconciliation studies of design data needs against PIRT findings and reactor pressure vessel [RPV] material alternatives studies) that were awarded to the HTGR design suppliers in 2008 and 2009.

# 2.7 ASME Code Development

ASME has organized a "Working Group on Nuclear High Temperature Gas—Cooled Reactors" within the framework of the B&PV Committee on Construction of Nuclear Facility Components (Section III). The charter of the Working Group is given as follows:

The Working Group shall develop rules for the construction of Nuclear High Temperature Gas-Cooled Reactors (HTGR) within Section III Division 5 Part 1. The rules of Part 1 shall constitute the requirements for the construction of the nuclear HTGR facility components such as pressure vessels, piping, pressure retaining portions of rotating equipment including pumps, blowers, turbines and compressors, valves, heat exchangers and for core support structures, both metallic and nonmetallic, and for containment or confinement structures. The rules shall contain requirements for materials, design, fabrication, testing, examination, inspection, certification, and the preparation of reports. The Working Group shall identify research and development efforts required to support the technical development of these code rules. Coordination with BPV XI on in-service inspection (ISI) issues shall be maintained.

An important document in guiding their mission is the *Roadmap for the Development of ASME Code Rules for High Temperature Gas Reactors* that was developed by an ASME Project Team for HTGR Code Development. The effort to develop this roadmap was originally sponsored by the NRC. Draft versions of this roadmap have been used to determine Working Group efforts and tasks sponsored by the DOE in FY 2009 and FY 2010. The roadmap is expected to be issued in 2010 as an ASME publication by ASME Standards Technology, LLC.

Additional groups have been organized within the ASME Board on Nuclear Codes and Standards infrastructure that support the development needs of HTGRs in areas relevant to materials. The NRC participates in these B&PV committee groups to provide regulatory perspective. These are:

- Subgroup on High Temperature Reactors. Within the B&PV committee on Construction of Nuclear Facility Components, the Subgroup on High Temperature Reactors continues activities related to Section III with specific focus on HTGRs and liquid metal reactors. The Section III Working Groups for HTGRs and liquid metal reactors report to this subgroup.
- Subgroup on Elevated Temperature Design. Within the B&PV Committee on Construction of Nuclear Facility Components, the Subgroup on Elevated Temperature Design continues activities related to Section III, Subsection NH, plus Code Cases N-499, N-253 and N-201. The applicability of these standards includes but is not unique to HTGRs.
- Subgroup on Graphite Core Components. The ASME Subgroup on Graphite Core Components was organized within the B&PV Committee on Construction of Nuclear Facility Components to establish rules for materials selection, design, construction, examination, inspection, and certification of graphite core components and core assemblies. The draft graphite code document has passed ballot and is currently being incorporated into ASME B&PV Section III, Division 5. For further details, see Section 3.3.5.
- Special Working Group, High Temperature Gas Cooled Reactors. The B&PV Committee on Nuclear In-service Inspection (Section XI) has established a Special Working Group, High Temperature Gas Cooled Reactors. The immediate objective of this group is to develop a plan for a rewrite of Section XI, Division 2 to address in-service inspection, evaluation, and repair/replacement activities for next generation HTGRs. A draft of the revised Section XI, Division 2 is currently under review within the ASME committee structure.

DOE has entered into a multiyear Cooperative Agreement with ASME Standards Technology, LLC for the Generation IV Reactor Materials project. The scope includes development of technical basis documents necessary to update and expand codes and standards for application in future Generation IV nuclear reactor systems that operate at elevated temperatures. The following tasks have been undertaken to date. In those cases, where reports documenting the results have been completed and approved, they are identified in the list that follows as "(STP-NU-xxx)." The following additional tasks have been proposed:

- Task 1 Allowable Stresses in Section III, Subsection NH on Alloy 800 H and Grade 91 Steel (STP-NU-020)
- Task 2 Regulatory Safety Issues in Structural Design Criteria (STP-NU-010)
- Task 3 Improvement of Subsection NH Rules for Grade 91 Steel (STP-NU-013)
- Task 4 Updating Nuclear Code Case N-201
- Task 5 Creep-Fatigue Data and Evaluation Procedures for Grade 91 Steel and Hastelloy XR (STP-NU-018)
- Task 6 Operating Condition Allowable Stress Values
- Task 7 ASME Code Considerations for the IHX
- Task 8 Creep and Creep-Fatigue Crack Growth
- Task 9 Update NH Simplified Elastic and Inelastic Methods
- Task 10 Update NH Alternative Simplified Creep-Fatigue Design Methods
- Task 11 New Materials for NH
- Task 12 Nondestructive examination and ISI Technology for high temperature reactors (HTRs; Funded by NRC)
- Task 13 Recommend Allowable Stress Values (planned for 2010)
- Task 14 Corrections to Stainless Steel Allowable Stress (planned for 2010).

# 2.8 Summary

The components of the HTGR operate at higher temperatures in different environmental conditions (helium with controlled levels of impurities) and with different performance requirements than those experienced in LWRs during normal and accident conditions. The candidate materials being considered for primary HTGR components are generally commercially available and are in use in high temperature applications in other industries and, in some cases, have been used in HTGR applications in the U.S. and other countries. However, the regulatory bases for these materials in the United States are limited for metallic materials and nearly nonexistent for nonmetallic materials. This section of the white paper reviewed the current body of NRC regulations, regulatory guidance, policy statements, standards, and past precedents and identified those that are judged to be applicable to candidate HTGR materials.

It is concluded that current regulations are adequate for the licensing of the NGNP, however, gaps exist where NRC approval will be required for the specific approaches proposed as the bases for qualification and regulatory approval. This may include NRC approval for the use of specific materials and/or operating conditions. These gaps and proposed approaches will be identified as the design of the NGNP reactor continues. Further development of the regulatory infrastructure will be desirable in support of follow-on commercial plants

#### 3. MATERIAL SELECTION AND QUALIFICATION

The materials for the HTGR will be selected using a rigorous approach that will consider component functional and performance requirements, safety classification, and code and regulatory compliance. Candidate metallic and nonmetallic materials are discussed below in relationship with specific applications. This discussion will include potential applications for the candidate material, key considerations, related experience, and the current qualification status.

# 3.1 Overall Material Selection Approach

The general material selection approach for the HTGRs is summarized in Figure 1.

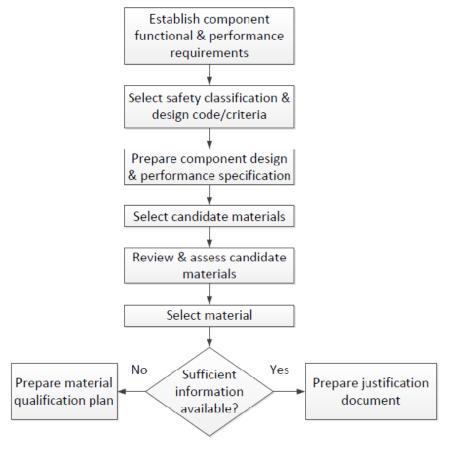


Figure 1. Material selection approach for HTGR components.

The material selection process begins by establishing component functional and performance requirements under normal and accident conditions. For example, the main functions of the core barrel under normal operating conditions are providing lateral and axial support for the graphite core structures and acting as a flow barrier between the hot helium gas in the core and the relatively cool gas in the outer annulus. Important material properties supporting these functions include high temperature strength and resistance to fracture, creep, fatigue, corrosion, neutron irradiation, and thermal aging. In addition to the above mentioned normal operation functions, the core barrel must also resist seismic loads and effectively transfer heat away from the core during a conduction cooldown event. The latter function defines the need for critical material thermal properties, such as emissivity and thermal conductivity.

Each function has a set of requirements, including performance, that must be met for the range of normal and off-normal events. For those off-normal events within the design basis, SSCs relied on to meet the HTGR-specific safety functions for public safety are classified as safety-related.

The above information would then be used to develop the component design and performance specification, which will state the component material and mechanical requirements under normal and accident conditions. For example, some key material property requirements for graphite reflector blocks, as well as for the fuel blocks in prismatic designs, may include emissivity, heat capacity, and thermal conductivity. The mechanical property requirements for these graphite components will include compressive strength at design temperatures.

Material candidates will then be selected that meet, or potentially meet, all the requirements of this specification, while considering the level of existing applicable code and regulatory compliance. Priority is given to candidates that are accepted by industry consensus codes (ASME B&PV) and the NRC at the proposed design and accident conditions. If such materials are not available, materials will be considered that meet a consensus code or are in the process of being added to a code but have not been accepted by the NRC. If there is not a codified material available that can meet design conditions, material candidates will be selected based on applicable available information.

Once material candidates are selected for a component, the materials will be assessed based on a series of key attributes, which may include:

- Code acceptability, limitations, and requirements. Each candidate material is assessed based on the acceptability, limitations, and requirements of the ASME Code for nuclear facility components. For example, Section III, Subsections NB and NH are used for Class 1 components. Both subsections contain rules for materials, design, fabrication, examination, testing, and overpressure relief of Class 1 components. The rules of Subsection NB guard only against time-independent failure modes. Subsection NH extends specific rules of Subsection NB to elevated temperature service, provided the designer can demonstrate that the combined effects of temperature, stress level, and duration of loading do not introduce significant creep effects. Note that codification by the ASME is not required for qualification of a material for a specific application; however, the qualification process is generally more straightforward when a material is covered by the ASME Code.
- Existing design, fabrication, and operating experience. Existing design, fabrication and operating
  experience are considered for each candidate material. Design experience is the depth and breadth of
  analysis that has been performed regarding the material's performance under design conditions for the
  proposed application. Fabrication experience will mainly focus on lessons learned from forming,
  machining and welding of the candidate material. Operating experience will focus on HTGR
  applications, but other industrial experience will also be considered.
- Ability to procure. The ability to procure the material in the necessary form(s) and to the requisite specification and quality must be evaluated given the availability and capability of global resources. The ability to procure a material is impacted by acceptance of the material by a standards body. In the United States this is typically an ASTM International Standard. This can be a significant issue with composite materials, for example, which are not currently covered by a procurement standard. Standardization is also a prerequisite for acceptance into the ASME Code.
- Technical maturity. The technical maturity of each material under consideration will be evaluated. For the purpose of this paper, technical maturity is defined as the amount and quality of existing data and operating experience for a given application. In general, the greater a material's maturity, the less research will be required to qualify the material. A technically mature material is ideal, but materials that require significant research may be necessary to meet the requirements of certain applications.

- Fabrication assessment. Each candidate material will undergo a fabrication assessment to determine whether the material can be economically formed, machined, welded, etc., to meet the requirements of the component. The ability to properly heat treat very large components for a very high temperature reactor (VHTR) is also an important criteria.
- *Performance assessment*. A performance assessment of each potential material for a given application will be conducted to determine which candidate best meets the application requirements.
- Compatibility with environment. The compatibility of each candidate material with its operating environment will be considered. The corrosion resistance of each material will be assessed based on the temperature and reactive species present during normal and accident conditions. Components in close proximity to the core will also be evaluated for irradiation effects. The potential for degradation in properties associated with aging will also be considered.
- Regulatory acceptance review. A regulatory acceptance review will be performed for the group of candidate materials to project the likelihood of ASME code and regulatory approval and the amount of effort required to obtain such approval.
- *Cost*. The relative cost of each candidate material will be assessed to determine the most economic material choice.

The material that best meets these assessment criteria will be selected for the component under consideration.

At this point, a determination will be made as to whether sufficient information is available to qualify the selected material for this application. If the required information is currently available, then a justification document will be prepared for qualification. If not, a document will be prepared to identify the current gaps in data and provide a detailed plan to obtain this data.

### 3.2 Metallic Materials

The following sections briefly discuss some candidate materials for the main components of the primary loop of the HTGR, such as the RPV, cross vessel, steam generator and reactor internals. The effects of welding on metallic materials are not discussed here, but will be considered during the evaluation process for qualification. Note that all stated operating and accident temperatures are estimates and may change during the design process. It is important to note that material characterization and potential qualification have been under active consideration for some candidate materials for several years. As described in Section 2.7 of this report, ASME Standards and Technology, LLC has sponsored a number of studies relating to the Code status of specific materials including Alloy 800H, Grade 91 steel and Alloy XR. The Generation IV International Forum Materials Program Management Board coordinates materials research and development relevant to VHTR systems from eight signatories. This work includes activities on Grade 91 steel and Alloys 800H and 617. Other candidate materials such as Alloy X were actively investigated for earlier programs, but are not currently under investigation for VHTR applications.

#### 3.2.1 SA-508/SA-533

# 3.2.1.1 Relevant Applications of SA-508/533

Manganese-nickel-molybdenum low-alloy steel is being considered as the main material of construction for the vessel system, which consists of the RPV, steam generator vessel and the cross vessel. ASME SA-508 Grade 3 Class 1 is used for forgings and SA-533 Type B Class 1 is used for plate. These materials are advantageous because they represent an excellent compromise between relatively

high mechanical properties, thus limiting the pressure vessel wall thickness, good aging resistance, and toughness.

# 3.2.1.2 Important Considerations for SA-508/533

Important considerations for material selection and qualification for the vessel system are dependent on the function of the specific component. When selecting a material for the reactor vessel, the following characteristics must be considered: high temperature mechanical properties, toughness, thermal properties, corrosion resistance, and possible effects of irradiation and thermal aging. The main factors affecting the steam generator vessel and cross vessel are their mechanical properties and corrosion resistance, since these components will not be significantly affected by conduction cooldown events or irradiation.

The focus for high temperature mechanical properties will be on accident conditions, since there is extensive experience with this material at the projected operating temperature of 325°C (617°F). ASME Code Case N-499-2 sets temperature and time limits for accident conditions, which consider strength, fatigue, creep, and stress to rupture. The HTGR design will conform to the code case limits so that further analysis of these properties will not be required.

Thermal properties such as emissivity and thermal diffusivity are integral to assessing the material's ability to meet the design requirements during a conduction cooldown event. Emissivity is a measure of a material's ability to radiate heat energy, and thermal diffusivity is the ratio of thermal conductivity to volumetric heat capacity. Measurements of emissivity will be required after oxidation in air and helium to determine the most appropriate values. Nominal values of thermal diffusivity are available in ASME Section II, Part D at temperatures up to 815°C (1499°F).

The effects of oxidation in the helium environment will need to be evaluated, including during hot transient conditions. Furthermore, oxidation effects caused by potential air ingress or steam/water ingress events will need to be considered. However, given the present extensive database and the large material thicknesses involved, oxidation effects are not expected to be significant, making the need for new data unlikely.

Any hardening and embrittlement of the HTGR RPV material caused by neutron exposure should be much less than in LWRs because the end of life vessel fluence of the HTGR will be at least an order of magnitude less than a typical LWR vessel. However, the radiation spectrum differs between these two designs and neutron embrittlement of the vessel will still need to be considered.

#### 3.2.1.3 Related Experience with SA-508/533

For over 40 years, Mn-Ni-Mo low-alloy steel has been used for LWR pressure vessel components in the United States and abroad. During this time, the material has been improved by limiting some trace elements. Weldability and toughness were improved by restricting the allowable carbon content. Toughness was further improved by reducing the allowable amount of sulfur and phosphorus. Reducing the amount of phosphorus also decreased the material's sensitivity to thermal aging. This experience is applicable, since the HTGR RPV normal operating temperatures will be similar to those of existing LWRs.

### 3.2.1.4 Current Qualification Status of SA-508/533

One of the objectives of any design option that will permit use of SA-508/SA-533 steel is to keep the vessel system wall temperatures within the acceptable range permitted by the ASME Section III Code. SA-508/SA-533 steels are ASME Code approved for Class 1 nuclear components and Subsection NB rules are applicable up to 371°C (700°F) for normal operation. By operating within the requirements of

the ASME Code, it will be possible to take advantage of the years of operating experience with LWR reactor vessels. ASME also allows limited high temperature excursions under Code Case N-499-2, but this code case has not yet been accepted by the NRC.

Code Case N-499-2 permits the use of SA-508/SA-533 for nuclear applications at temperatures up to 538°C for abnormal situations. The code case states that the component design will be based on a maximum cumulative time of 3,000 hours at metal temperatures in the range of 371 to 427°C (700 to 800°F) and 1,000 hours at metal temperatures exceeding 427°C (800°F) and up to 538°C (1000°F). The code case also requires that the number of anticipated events where metal temperatures exceed 427°C (800°F) be limited to a total of three. In the case of the HTGR, an abnormal situation requiring the use of Code Case N-499-2 might result from an accident involving a depressurized conduction cooldown event; however, such an event is not anticipated to occur more than once during the lifetime of the facility.

The ASME Code is also sufficient to permit the use of SA-508/SA-533 steel in the cross-vessel and the steam generator vessel because these components are not significantly affected by a conduction cooldown event or exposure to neutron radiation. However, the code does not address key requirements of the RPV design, such as emissivity and thermal aging. The qualification of this material for the RPV will thus require consideration of these factors.

# 3.2.2 Alloy 800H

# 3.2.2.1 Relevant Applications of Alloy 800H

Metallic components in contact with hot helium gas could potentially be fabricated from Alloy 800H. For an HTGR, these components include the core support structure (including the core barrel), outer control rod cladding, control rod guide tubes, upper plenum shroud, lower plenum sidewall thermal barrier, hot duct liner, and hot end-steam generator tubing. Alloy 800H is an iron-nickel-chromium material that is designed for high temperature service where resistance to creep and rupture is required. The operating temperature for the areas where Alloy 800H is being considered will depend on the outlet temperature of the reactor core. The current plan for the HTGR is to use an outlet temperature of 750 to 800°C (1382 to 1562°F).

#### 3.2.2.2 Important Considerations for Alloy 800H

Important considerations for material selection and qualification for the above mentioned applications are dependent on the specific component attributes, which include operating and design temperature, environment, proximity to the core, and function.

For the core support structure, outer control rods, control rod guide tubes, upper plenum shroud, and lower plenum sidewall thermal barrier, material selection and qualification is based on high temperature strength, time dependent stresses, irradiation effects, thermal aging effects, and corrosion resistance. The core barrel material will also need to possess appropriate levels of emissivity and thermal diffusivity. Material selection and qualification for the hot end steam generator tubing is based mainly on high temperature strength, time dependent stress effects, thermal conductivity, and corrosion resistance. Material selection and qualification for the hot duct liner is based mainly on high temperature strength, corrosion resistance, and time dependent stress effects such as creep and stress rupture.

High temperature strength and time dependent stress effects such as creep and stress rupture for Alloy 800H are covered by ASME Code Case N-201-5 for core support structures and Subsection NH for Class 1 components. Both Code Case N-201-5 and Section III, Subsection NH permit Alloy 800H to operate up to 760°C (1400°F). Furthermore, N-201-5 and Subsection NH state that there is no significant time dependent effect on the stress allowables for Alloy 800H up to about 427°C (800°F). This temperature limit was based on a component life of 300,000 hours. Time dependent effects related to the 60-year

design life will be evaluated during qualification. Other components will see temperatures above 427°C (800°F) and some nonstructural components (e.g., hot-gas-duct liner) will possibly operate as high as 800°C (1472°F) for the life of the plant. Consequently, allowable Alloy 800H stresses will need to be extended to encompass the 60-year design life for the HTGR to a minimum of at least 800°C. Studies sponsored by ASME Standards and Technology, LLC have determined that there is currently sufficient information available to extend Code qualification to 850°C for a maximum use temperature for 500,000 hours design life for lower temperatures.

The core support structure, control rod guide tubes, and upper plenum shroud operate below 400°C during normal operation. Therefore, high temperature strength and time dependent stress effects such as creep and stress rupture are not considered a significant concern for these components under normal operating conditions. Further evaluation is required to determine if the core support structure will exceed the 760°C (1400°F) limit during a conduction cooldown event and for how long. If the core support structure is predicted to exceed 760°C (1400°F) during a conduction cooldown event, an extension of Section III, Subsection NH to higher temperatures may be required for the use of Alloy 800H. In STP-NU-20,<sup>4</sup> it is concluded that existing data are sufficient to extend the stress allowables to 600,000 hours at 900°C (1652°F). The control rod guide tubes and upper plenum shroud will likely exceed the current code temperature limit during a pressurized conduction cooldown event and, therefore, Alloy 800H will require further evaluation for fitness of use at these higher temperatures.

In the prismatic reactor, the lower plenum barrier will operate around 660°C (1220°F) during normal conditions, which is within the limit set by Code Case N-201-5. However, the code case does show an impact on the stress allowables at this temperature because of extended operation. An evaluation will be required to determine if the lower plenum barrier can meet the allowables over the life of the plant. If it cannot meet the code, an evaluation of existing data or the selection of alternate materials and possible further testing will be required for qualification. A conduction cooldown event would not likely cause the temperature of this component to exceed 760°C (1400°F), but this must be confirmed during design.

During normal operation, the outer control rods will operate around 440°C (824°F), which is within the limit set by Code Case N-201-5. However, N-201-5 does show a small impact on the stress allowables at this temperature caused by extended operation. An evaluation will be required to determine if this component can meet the allowables over the 60-year life of the plant. Because of the proximity to the core, this component will also likely exceed the current code temperature limit for Alloy 800H during a conduction cooldown event and, therefore, will require further evaluation at these high temperatures for qualification. Note that the inner control rods in prismatic fuel reactors may see operating and accident temperatures of about 800°C (1472°F) and 1400°C (2552°F), respectively. Thus, Alloy 800H is not a viable option unless the inner control rods are not inserted during an accident. Note that both the outer control rods and the inner control rods can be replaced if necessary during the facility lifetime. This option will be evaluated. Irradiation effects must also be considered for the control rods, but sufficient data currently exists to undertake such an evaluation.

The hot end steam generator tubing will operate at about 600°C (1112°F) during normal conditions, which is within the limit set by Subsection NH. However, Subsection NH does show an impact on the stress allowables at 600°C (1112°F) caused by extended operation. An evaluation will be required to determine if the hot end steam generator tubing can meet these allowables over the 60-yr life of the plant. If the present allowables are not sufficient, then an evaluation of existing data may be required for qualification. A conduction cooldown event would not likely cause the temperature of this component to exceed 760°C (1400°F), but this will be confirmed during design.

Some HTGR steam generator designs may involve a bimetallic weld between Alloy 800H tubing and ferritic steel tubing, likely 2.25Cr-1Mo. It is not certain how this bimetal would be incorporated into the ASME Code. A conservative approach might be to consider the bimetal to possess high-temperature

properties equivalent to those of the 2.25Cr-1Mo steel. However, special consideration will need to be given to corrosion questions, especially where alternating wet-dry conditions might exist

The hot duct liner will operate at the approximate reactor outlet temperature of 750 to 800°C (1382°F to 1472°F), depending on the final design under normal conditions and the higher of these temperatures is above the limit set by ASME Section III Subsection NH. However, the hot duct liner will be under minimal stress so that the high temperature strength and time dependent stress effects that drive the 760°C (1400°F) limit may not directly apply. Further, as already noted earlier, data already available would support operation at even higher temperatures. During a pressurized conduction cooldown event, the hot duct liner may exceed the current code temperature and, if so, would require further evaluation at these high temperatures for qualification.

Since the outer control rods, control rod guide tubes, upper plenum shroud and core support structure are in close proximity to the core, irradiation effects also need to be evaluated for qualification. However, sufficient data already exist for this purpose. The lower plenum thermal barrier and hot duct liner will not likely accumulate enough fluence over the life of the plant to experience detrimental effects from irradiation. This will be confirmed during the design process.

Increasing temperature tends to accelerate the corrosion of all materials, including Alloy 800H, in a helium gas environment. At temperatures below 475°C (887°F), extended operation studies have shown that corrosion of Alloy 800H in impure helium gas is minimal. At temperatures below 900°C (1652°F), extended operation studies in impure helium gas have shown that Alloy 800H develops chromia scales along with significant internal oxidation of aluminum and subsurface depletion of Cr. However, it must be noted that the corrosion behavior is quite sensitive to levels and ratios of active impurity species and this must be considered in the evaluation process.

Ensuring the performance of Alloy 800H components under accident conditions will require consideration of thermal properties such as emissivity and thermal diffusivity, as these are integral to the material's ability to meet applicable design requirements during a conduction cooldown event. Measurements of emissivity will be required after oxidation in air and helium to determine the most appropriate values. Nominal values of thermal diffusivity are available in ASME Section II, Part D at temperatures up to 815°C (1499°F).

#### 3.2.2.3 Related Experience with Alloy 800H

Alloy 800H has been used in high temperature components of HTGRs for over 20 years of plant operation. FSV, the Arbeitsgemeinschaft Versuchsreaktor (AVR), and the Thorium High-Temperature Reactor (THTR) used Alloy 800H for steam generator tubing and heat exchanger components with success. These plants operated for a combined total of 34 years, with AVR operating for 20 years. The steam generator inlet temperature was around 950°C (1742°F) for AVR, 775°C (1427°F) for FSV, and 750°C (1382°F) for THTR; however, actual metal temperatures were significantly lower.

Extensive studies evaluating the effects of operating temperature on the performance of Alloy 800H have been performed. The minimum creep rate versus stress at 593 to 760°C (1100 to 1400°F) was determined using regression analysis. Fatigue behavior of Alloy 800H has been evaluated from room temperature to 760°C (1400°F) and low-cycle and high-cycle fatigue data were taken at 760°C (1400°F). These studies form part of the foundation for ASME's current provisions for the use of Alloy 800H up to 760°C (1400°F). In addition, there is extensive data for these properties at temperatures through 1000°C (1832°F).

# 3.2.2.4 Current Qualification Status of Alloy 800H

Code Case N-201-5 is an ASME approved addition to Section III, Subsection NG that allows the use of Alloy 800H for core support structures at temperatures up to 760°C (1400°F). High temperature Class 1 components are covered by Subsection NH, which also sets a limit for Alloy 800H of 760°C (1400°F). Since some components may exceed these temperature limits during normal operation or a conduction cooldown event, one of the following will be required: (1) modifying the Code to allow the use of Alloy 800H at higher temperatures; (2) qualifying Alloy 800H for the specific component based on existing data, and potentially further testing, without code qualification; (3) modifying the design to stay below the temperature limit; or (4) fabricating these components from a different material. Since some components operate in the time-dependent stress regime, Alloy 800H stress allowables will need to be extended to encompass the 60-year design life of the HTGR. The draft version of German Standard KTA 3221 covers the use of Alloy 800H up to 1000°C (1832°F). An ASME and DOE joint effort is currently underway to obtain the basis of the KTA 3221 draft standard, including information on the quality assurance program under which the data were collected. The acquisition of these data may support an increase of the ASME allowable operating temperature for Alloy 800H.

The ASME Code covers Alloy 800H in terms of high temperature strength and time dependent stress effects, such as creep and stress rupture. However, the code does not address other key requirements of the design of these components, such as the emissivity, corrosion resistance, thermal aging, and irradiation effects.

## 3.2.3 Alloy X/XR

## 3.2.3.1 Relevant Applications of Alloy X/XR

Alloy X and Alloy XR are being considered for reactor core and core support structures that will experience temperatures greater than about 750°C (1382°F) during normal operation or under accident conditions. For the current HTGR design with an outlet temperature of about 750 to 800°C (1382 to 1472°F), these components include the control rod guide tubes, control rods, upper plenum shroud thermal barrier, and hot-gas-duct liner. Alloy X is a nickel-chromium-iron-molybdenum alloy that possesses a combination of corrosion resistance, ease of fabrication, and high temperature strength. The main differences between Alloy X and Alloy XR are in ASME codification and cobalt limits. Alloy X has been around longer, and is contained in the ASME Code, but it contains sufficient cobalt (0.5 to 2.5%) to warrant evaluation of dose impact at high neutron fluence conditions. Alloy XR is a proprietary version of Alloy X developed by the Japan Atomic Energy Agency (JAEA) for the High Temperature Test Reactor (HTTR), which has lower limits on cobalt and other restrictions on minor elements that are said to optimize high-temperature strength properties. It is not currently covered by an ASTM International standard and is not in the ASME Code. There do not appear to be any commercial vendors for this material.

#### 3.2.3.2 Important Considerations for Alloy X/XR

Important considerations for material selection and qualification for the above mentioned applications are dependent on the specific component attributes, which include operating and design temperature, environment, proximity to the core, and function.

For the control rod guide tubes, outer control rods and upper plenum shroud, material selection and qualification are based on high temperature strength, time dependent stress effects, thermal aging effects, irradiation effects, and corrosion resistance. For the hot duct liner, material selection and qualification are based on high temperature strength, aging effects, corrosion resistance, and time dependent stress effects such as creep and stress rupture.

Alloy X is permitted to operate at up to 427°C (800°F) according to ASME Section III and up to 899°C (1650°F) according to Section VIII. Note that the 899°C (1650°F) limit in Section VIII does not apply to nuclear applications, but it is considered useful guidance. Some components will see temperatures above 427°C (800°F) during normal operation. Consequently, Alloy X stress allowables will need to be extended to encompass the 60-year design life for the HTGR.

The projected operating temperature for the control rod guide tubes and the upper plenum shroud are both below 400°C, which is within the limit set by Section III of the ASME Code. Therefore, high temperature strength and time dependent stress effects such as creep and stress rupture are not considered a concern for these components under normal operating conditions. Further evaluation is required to determine if these components will exceed the 427°C (800°F) limit during a conduction cooldown event and for how long. Remaining below the Section VIII limit of 899°C (1650°F) suggests that this material may be acceptable, but since Section VIII does not apply to nuclear components, further evaluation will be required for qualification.

During normal operation, the projected temperature for the outer control rods is about 440°C (824°F), which is only slightly above the 427°C (800°F) limit set by Section III, Subsection NB. Therefore, high temperature strength and time dependent stress effects such as creep and stress rupture are unlikely to be of concern for this component under normal operating conditions, but this will be confirmed by evaluation. During an accident, this component will likely exceed 899°C (1650°F), thus requiring additional evaluation should this material be selected. An alternative under consideration is using a high temperature composite for the control rods.

The projected operating temperature for the hot duct liner is 750 to 800°C (1382 to 1472°F), which is below the 899°C (1650°F) limit in Section VIII. Again, remaining below the Section VIII limit does not negate the need for further evaluation or further codification. However, the hot duct liner will be under minimal stress, so the high temperature strength and time dependent stress effects that drive the 899°C (1650°F) limit may not directly apply. During a pressurized conduction cooldown event, the hot duct liner may exceed the current ASME Section III Code temperature and therefore require further evaluation at these high temperatures for qualification.

The outer control rods, control rod guide tubes, and upper plenum shroud are in a significant neutron radiation field, so irradiation effects will also have to be evaluated for qualification. The hot duct liner will not likely accumulate enough fluence over the life of the plant to experience detrimental effects from irradiation, but this will be confirmed during design.

Studies have been performed to measure the corrosion resistance of Alloy X and Alloy XR in simulations of the HTGR environment. In 870°C (1598°F) impure helium gas, extended operation studies have shown that Alloy X exhibits better resistance to oxidation than Alloy 800H and, over time, the oxidation rate decreased three times faster in Alloy X compared to Alloy 800H (the relative behavior depends on the impurities). A direct comparison of Alloy X and Alloy XR in 1000°C (1832°F) impure helium gas showed that Alloy XR had improved resistance to intergranular attack and internal oxidation. Further corrosion testing may be required once the accident temperature and environment are better quantified. However, all of the components being considered are composed of relatively thick sections, so that the overall effects of corrosion are likely to be minimal. Thermal aging may be a significant issue for this alloy, depending on the temperature range of application. At 750°C, aging has been shown to result in room temperature ductility of less than 15% after exposure of several thousand hours. The potential for interaction between aging and environmental effects has not been extensively studied.

Studies have shown that carburization of Alloy X in helium is heavily dependent on temperature and oxygen potential. Carburization is mitigated at higher oxygen potential levels because of the formation of a protective oxide film on the alloy. In a helium gas environment with low oxidation potential,

carburization of Alloy X is minimal at temperatures of 650°C (1202°F). Also, carbon pickup does not appear to increase in the period from 3,000 to 10,000 hours. However, carburization does increase with time at temperatures of ≥800°C (1472°F) at low oxygen potentials. Most components currently considering the use of this material will operate well below 650°C (1202°F) during normal operation and the duration of an accident would not be long enough to develop significant carburization. However, the hot duct liner may operate close to 800°C (1472°F) during normal operation. The modified version of this alloy, Alloy XR, has been used in the HTGR environment at 850°C (1562°F) for over 10 years in the HTTR, a 30 MWth prototype HTGR in Japan, without any reported degradation because of carburization. However, conservatism suggests that the potential for carburization of the hot duct liner should be further evaluated.

#### 3.2.3.3 Related Experience with Alloy X/XR

Alloy X has found a wide range of uses in gas turbine engines for combustion zone components such as transition ducts, combustor cans, spray bars, and flame holders as well as in afterburners, tailpipes, and cabin heaters. Components of industrial furnaces have also used this alloy because of its high resistance to oxidizing, reducing, and neutral atmospheres. Furnace rolls of this alloy were still in good condition after 8,700 hours at 1177°C (2150°F). Alloy X is also used in the chemical process industry for retorts, muffles, catalyst support grids, furnace baffles, tubing for pyrolysis operations, and flash drier components. There has been no recent activity through either ASME Standards and Technology or the Generation IV International Forum to determine the current status of this alloy for VHTR application.

In the nuclear industry, the Japanese HTTR has been operating for over 10 years with a Alloy XR hot duct liner and intermediate heat exchanger piping at a normal operating temperature of 850°C (1562°F) and at limited operation up to 950°C (1742°F). Given the success that the nuclear industry and other industries have had using Alloy X and Alloy XR in high temperature components, these alloys have been brought under consideration for components of the HTGR.

Studies have been conducted in the United States and Japan to evaluate the tensile, creep rupture, low-cycle fatigue, creep fatigue properties, and fatigue crack growth of Alloy X, specifically for application in the helium environment of an HTGR. Testing of rupture time variation with applied stress showed that Alloy X would not rupture at 7 MPa (1000 psi) and 871°C (1600°F) during the 60-year life of the plant. Test data also indicated that the creep rate at 871°C (1600°F) would be insignificant. These studies may later form part of the foundation for ASME allowing use of Alloy X at up to 871°C (1600°F) during normal operation.

## 3.2.3.4 Current Qualification Status of Alloy X/XR

Based on the ASME Code, the maximum allowable temperature for Alloy X per Section III is 427°C (800°F), and per Section VIII, Division 1 it is 899°C (1650°F). The 899°C (1650°F) limit specified in Section VIII is not specific to nuclear applications, but it is considered useful guidance. As some components will see temperatures above 427°C (800°F) during normal operation, Alloy X stress allowables will need to be extended to encompass the 60-year design life for HTGRs. Industry experience reports that Alloy X can be used at temperatures up to 871°C (1600°F) during normal operation and could be allowed to operate at up to 938°C (1720°F) for up to 3,000 hours. This, however, will need to be supported by quality assured references. These projected temperature allowables would likely meet the design and conduction cooldown event conditions for the applicable metallic components of the HTGR design. Ideally, the results of these studies can be used to expand Code Case N-201-5 and Section III, Subsection NH to include Alloy X. The use of Alloy X in the reactor internals adjacent to the core will also need to be evaluated, because of the potential for high dose issues related to cobalt content (nominal range 0.5 to 2.5 wt%). Alloy XR does not have significant levels of cobalt, but it is currently not in the ASME Code.

At stresses below 7 MPa (1,000 psi), industry experience reports that Alloy XR can be used at temperatures up to 927°C (1700°F) during normal operating conditions and up to 938°C (1720°F) for under 3,000 hours, but more data would have to be gathered on the use of Alloy XR before the qualification process could proceed.

The ASME Code does not currently address key requirements of the design of Alloy X/XR components such as corrosion resistance, thermal aging effects, irradiation effects, high temperature strength, and time dependent stress effects such as creep and stress rupture, so the qualification of these materials will require further evaluation.

## 3.2.4 Modified 9Cr-1Mo

# 3.2.4.1 Relevant Applications of Modified 9Cr-1Mo Steel

Modified 9Cr-1Mo (9Cr-1Mo-V) (Grade 91) steel is being considered for the core support structure, including the core barrel. Modified 9Cr-1Mo steel experiences only a gradual reduction in strength at temperatures up to 450°C (842°F), but above that, allowable stresses for all low alloy steels drop off considerably. However, modified 9Cr-1Mo has an advantage over other steels because it retains its strength much better at these elevated temperatures.

## 3.2.4.2 Important Considerations for Modified 9Cr-1Mo

Material selection and qualification for the core support structure is based on high temperature strength, time dependent stress effects, thermal aging, emissivity, thermal diffusivity, irradiation effects, and corrosion resistance.

According to Section III, Subsection NB of the ASME Code, modified 9Cr-1Mo (Grade 91) is permitted to operate at up to 371°C (700°F), and according to Section III, Subsection NH up to 649°C (1200°F). However, subsection NH is not directly applicable to the core support structure because it deals only with Class 1 components. Still, it is considered useful guidance. Based on Subsection NH, there is no significant effect of temperature on stress allowables at temperatures below 371°C (700°F). This temperature limit was based on a component life of 300,000 hours. Whether this remains true for the HTGR 60-year design life will be evaluated during qualification.

During normal operation, the core support structure will operate at about 350°C (662°F), which is below the 371°C (700°F) limit set by Section III, Subsection NB. The high temperature strength and time dependent stress effects such as creep and stress rupture are therefore not considered a concern under normal operating conditions. Further evaluation is required to determine if these components will exceed the 649°C (1200°F) limit during a conduction cooldown event and for how long. Remaining below the 649°C (1200°F) limit will not guarantee acceptability, but this limit is considered helpful guidance for qualification.

The effect of thermal aging on the properties of Modified 9Cr-1Mo (Grade 91) is negligible over the temperature range of 300 to 600°C (572 to 1112°F) for times to 75,000 hours. Yield strength, ultimate tensile strength, and ductility are not significantly affected. Therefore, thermal aging of this material is not expected to be an issue.

Thermal properties such as emissivity and thermal diffusivity are important to passive heat removal capability. Measurements of emissivity will be required after oxidation in air and helium to determine the most appropriate values. Nominal values of thermal diffusivity are available in ASME Section II, Part D at temperatures up to 815°C (1500°F).

Ferritic steels are susceptible to neutron embrittlement over extended periods of operation in high fluence locations. The effects of irradiation on strength and ductility are dependent on irradiation temperature and dose (dpa). Considerable hardening occurs for irradiations below 400°C (752°F) but hardening decreases rapidly as irradiation temperature is increased. Essentially no hardening is experienced by the time the irradiation temperature reaches 500°C (932°F). Data are available for irradiations from 50 to 600°C (122 to 1112°F) for doses to 60 dpa, well above what is expected for the application of Modified 9Cr-1Mo. The projected end-of-life fluence of the Modified 9Cr-1Mo core support structure will be used to evaluate the viability of this material for the intended applications.

No particular corrosion concerns are expected for Modified 9Cr-1Mo at the service temperatures of the HTGR. The effects of oxidation in the helium environment during hot transient conditions will need to be evaluated; however, given the thickness of the components of interest, no significant issues are anticipated.

#### 3.2.4.3 Related Experience with Modified 9Cr-1Mo

9Cr-1Mo low alloy steel was originally developed for the fast breeder reactor starting in the 1970s. It was found to have lower thermal expansion, higher thermal conductivity, and improved oxidation resistance compared to traditional power plant steels, such as 2.25Cr-1Mo low alloy steel. The addition of niobium, vanadium, and nitrogen created Modified 9Cr1-Mo (9Cr-1Mo-V), which exhibits a substantial increase in creep-rupture strength. Modified 9Cr-1Mo was certified by ASME in the 1980s and is now widely specified for electric utility power plants and is moving into the oil and gas industry. For example, Modified 9Cr-1Mo has been used for tubing in the super-heaters of power boilers for over 20 years. It has been used for piping applications up to 593°C (1100°F) and in tubing up to 565°C (1050°F). Modified 9Cr-1Mo has had great success in the fossil industry; however, some failures have occurred and these were traced to a lack of quality assurance. Special care must be taken during processing, fabrication, and installation to create and maintain the proper microstructure to obtain the desired material properties. It is not currently possible to insure that the steel is properly heat treated through-thickness by means of nondestructive examination. The necessity for preweld and post-weld heat treatment makes onsite fabrication of components from this steel problematic.

Extensive studies have been conducted on Modified 9Cr-1Mo to evaluate tensile strength, creep rupture, and low-cycle fatigue properties in a high temperature environment. Long-term aging effects on mechanical properties have also been determined. After aging at 482°C (900°F) for 75,000 hours, little effect was noticed on the ultimate tensile strength at temperatures up to 500°C (932°F). For the aged material, creep rates at 575°C (1067°F) at 14.5 ksi showed no acceleration and only about 1% strain after 20,000 hours. For material aged at 650°C (1202°F) for 10,000 hours, the rupture life at 14.5 ksi and 600°C (1112°F) was about 30,000 hours. Extrapolation to a test temperature of 500°C (932°F) gives a rupture life at 14.5 ksi that far exceeds the proposed 60-year life of the plant. Low-cycle fatigue data indicated a higher cyclic strength for the hot-rolled material compared with the hot-forged material. Fatigue crack growth testing at 538°C (1000°F) concluded that the crack propagation rate was similar with both product forms. Fracture toughness is good and relatively constant with a  $K_{JQ}$  value of ~275 MPa[m]<sup>1/2</sup> from room temperature through 200°C (392°F); irradiation to 3 dpa reduces  $K_{JQ}$  to ~100 MPa[m]<sup>1/2</sup>, but this is still a substantial value.

#### 3.2.4.4 Current Qualification Status of Modified 9Cr-1Mo

Based on the ASME Code, the maximum allowable temperature for Modified 9Cr-1Mo per Section III Subsection NB is 371°C (700°F), and per Section III, Subsection NH is 649°C (1200°F). Code Case N-201-5 is currently being expanded to include Modified 9Cr-1Mo because N-201-5 is applicable to core support structures, while Subsection NH is for Class 1 components.

The ASME Code covers Modified 9Cr-1Mo in terms of high temperature strength and time-dependent stress effects such as creep and stress rupture. However, the code does not address key requirements for the design of these components such as the emissivity, corrosion, thermal aging, and irradiation effects. Therefore, the qualification of this material will require further effort.

#### 3.2.5 2.25Cr-1Mo

## 3.2.5.1 Relevant Applications of 2.25Cr-1Mo

Grade 22 of 2.25Cr-1Mo (as specified in Subsection NH) is being considered for cold-end steam generator tubing that will be exposed to helium and water during normal operation. Further, 2.25r-1Mo might also apply to other components, such as the core barrel and the steam generator vessel. The allowable stress for 2.25Cr-1Mo is similar to Modified 9Cr-1Mo up to about 430°C (806°F), but above this temperature its strength drops off significantly relative to Modified 9Cr-1Mo. However, the cold-end steam generator tubing will experience a maximum temperature of only about 400°C (752°F) during normal operation, so 2.25Cr-1Mo may be a suitable option.

## 3.2.5.2 Important Considerations for 2.25Cr-1Mo

For the cold-end steam generator tubing, material selection and qualification is based on high temperature strength, thermal aging effects, time dependent stress effects, thermal conductivity, and corrosion resistance.

According to Section III, Subsection NB of the ASME Code, 2.25Cr-1Mo Grade 22 is permitted to operate at up to 371°C (700°F). Accordance to Section III, Subsection NH, 2.25Cr-1Mo in the annealed form may be operated up to 649°C (1200°F). The latter is well above the projected operating temperature for these components (about 400°C [752°F]). Based on Subsection NH, there is no significant effect on stress allowables caused by operation to 300,000 hours at temperatures below about 371°C (700°F). The 60-year design life proposed for the HTGR must be evaluated during qualification to determine if time-dependent effects, such as creep and stress rupture, must be taken into account. Further evaluation is required to determine if these components will exceed the 649°C (1200°F) limit during a loss-of-forced-convection event and for how long. Remaining below the 649°C (1200°F) limit will not guarantee NRC acceptance, but this limit is considered helpful guidance for qualification.

Thermal conductivity is integral to assessing the ability of tubing to transfer heat efficiently from the primary helium gas to the secondary side water. Measurements will be required after oxidation in air and helium to determine whether degradation of heat transfer properties will need to be taken into account.

Previous HTGR steam generators (e.g., FSV) have used 2.25Cr-1Mo tubes in high temperature aerated water with success. The corrosion behavior of this material will still need to be evaluated for projected operating temperature and water chemistry conditions to identify whether or not additional testing is required.

Field experience has shown the 2.25Cr-1Mo steels are virtually immune to wet steam erosion-corrosion in LWR nuclear applications. The main difference between traditional reactor steam generator tubing and tubing in the HTGR steam generator is that the temperature is significantly higher. This higher temperature is advantageous because maximum erosion-corrosion takes place at about 180°C (356°F) and then falls off. In the HTGR cold-end steam generator tubing, the water temperature will be about 400°C (752°F), which should cause the formation of a protective layer of Fe<sub>3</sub>O<sub>4</sub>.

No significant corrosion effects related to the helium environment are expected for 2.25Cr-1Mo at the service temperatures projected for the cold-end steam generator tubing of the HTGR.

# 3.2.5.3 Related Experience with 2.25Cr-1Mo

The well-established Grade 22 of 2.25Cr-1Mo has been used in both fossil and nuclear power plants. This material has been used in boiler and pressure vessels in fossil plants at operating temperatures around 400°C (752°F). The Japanese HTTR has operated for over 10 years with a 2.25Cr-1Mo reactor vessel and heat exchanger vessel with an operating temperature of about 395°C (742°F). This material was also used in the FSV and THTR for steam generator tubing.

A substantial database is available on the tensile, creep, fatigue, and creep-fatigue properties of 2.25Cr-1Mo ferritic steel. This database also includes the effect of long-term aging on microstructural changes and mechanical properties at temperatures consistent with the cold helium side of the HTGR.

#### 3.2.5.4 Current Qualification Status of 2.25Cr-1Mo

The maximum allowable temperature for 2.25Cr-1Mo Grade 22 per Section III, Subsection NB is 371°C (700°F) per Section III, Subsection NH is 593°C (1100°F). Even though the use of this material is allowed under ASME Section VIII at up to 649°C (1200°F), significant decreases in strength occur above about 427°C (800°F) and will need to be considered during design. Further evaluation will be required to determine the temperature of the steam generator tubes during a conduction cooldown event. Higher strength versions of this steel and the vanadium modified version are not in Section III of the Code.

The ASME Code covers 2.25Cr-1Mo Grade 22 in terms of high temperature strength and time dependent stress effects such as creep and stress rupture. However, the Code does not address key requirements of the design of these components, such as corrosion resistance and thermal aging effects. Therefore the qualification of this material will require some small amount of further evaluation.

# 3.2.6 Type 316 Stainless Steel

#### 3.2.6.1 Relevant Applications of Type 316H Stainless Steel

Type 316H austenitic stainless steel is being considered as a material option for the HTGR core barrel assembly and other reactor metallic internal components that would experience temperatures above 593°C (1100°F) during service, either in normal or transient operation. Type 316, most likely in the lower carbon 316L version, is also a potential material for the steam generator tubing, which experiences maximum tube metal temperatures of about 620°C (1150°F). If the steam side corrosion potential can be controlled (to prevent stress corrosion cracking), a substantial cost saving can be achieved in replacing the Alloy 800H tubing with Type 316H or 316L tubing.

# 3.2.6.2 Important Considerations for Type 316H Stainless Steel

Material selection criteria for the core barrel assembly are dominated by high temperature strength, thermal conductivity, and resistance to oxidation and neutron irradiation.

The high strength and creep resistance of 316H, as specified in the ASME code, is ensured by controlling the carbon content and the microstructural grain size of the finished product. The carbon content is controlled between 0.04 and 0.06% and the grain size is specified to be in the range of ASTM 3–6. These controls are considered essential for operating temperatures between 427 and 593°C (800 and 1100°F). The effects of these controls are not stated for higher temperatures, but they are still considered to be beneficial.

The emissivity of the core barrel surface is an important parameter in heat removal from the core via the core barrel sides, especially during conduction cooldown events. The emissivity of austenitic stainless steel is highly dependent on the surface conditions, and typical total emissivity values reported are 0.11

for machined surfaces and up to 0.38 for sandblasted surfaces. The higher the emissivity, the more efficient the heat removal from the core. Increased heat removal from the core correspondingly results in lower maximum fuel temperatures, as well as lower metal temperatures for the core barrel and RPV. Furthermore, emissivity is temperature-dependent, and over the measured temperature range of 200 to 700°C (392°F to 1292°F), oxidized surfaces exhibit an average emissivity of about 0.85. This is also achievable through the application of thin, ceramic-like coatings, e.g., Tyrano Coat TYR-1181.

Because of the presence of minor concentrations of impurities in the helium coolant of HTRs, the austenitic stainless steels will tend to form protective, stable oxide scales. Because of the rather low design temperatures of up to  $600^{\circ}$ C (1112°F), the expected scale growth rates will be slow and a typical scale thickness at the end of the 60-year design life should be  $<10\mu m$ .

The fast neutron (E>0.1MeV) fluence levels for the core support structures and other metallic internals are typically below  $1\times10^{19}$  n/cm<sup>2</sup>. This is below the level ( $\sim1\times10^{21}$  n/cm<sup>2</sup>) at which austenitic stainless steels begin to show significant irradiation-induced increases in tensile strength, along with associated reductions in ductility.

#### 3.2.6.3 Related Experience with Type 316H Stainless Steel

Austenitic steels of Type 304 and Type 316 are commonly used for LWR internals, such as fuel support structures, core barrels, and flow baffle plates. These are, however, all low temperature applications in aqueous conditions, and the materials used are the low-carbon versions (Type 304L or Type 316L).

#### 3.2.6.4 Current Qualification Status of Type 316H Stainless Steel

The maximum allowable temperature for Type 316H SST is 427°C (800°F) for ASME Section III, Subsection NB (Class 1 Components) and Subsection NG (subsection applicable to core support structures). It is, however, allowed for use at temperatures up to 816°C (1600°F) in Subsection NH and Code Case N-201-5, which comprise extensions to Subsections NB and NG, respectively. These parts of the ASME code cover Type 316H in terms of high temperature strength, creep and creep-fatigue effects up to a design life of 300,000 hours. However, the Code does not address other key requirements associated with the design of these components, such as thermal aging effects and neutron embrittlement. Therefore, the qualification of this material will require some further evaluation to address these effects.

## 3.2.7 Alloy 617

#### 3.2.7.1 Relevant Applications of Alloy 617

Alloy 617 has superior strength and creep resistance compared to Alloy 800H or Alloy X above 800°C (1472°F) and would be the preferred choice for applications at the highest temperatures where mechanical property considerations dominate. Applications might include the hot-duct liner or a high temperature IHX. Alloy 617 contains a significant amount of cobalt (10 to 15 wt%) that is an important contributor to high temperature strength and resistance to carburization, but it would preclude application in control rod sleeves or other applications where a significant amount of irradiation can occur.

#### 3.2.7.2 Important Considerations for Alloy 617

Alloy 617, also designated as Inconel 617, was initially developed for high temperature applications above 800°C (1472°F). It is often considered for use in aircraft and land-based gas turbines, chemical manufacturing components, metallurgical processing facilities, and power generation structures. The alloy was also considered and investigated for the HTGR programs in the United States and Germany in the late 1970s and early 1980s.

Alloy 617 has substantial creep strength at temperatures above 870°C (1598°F), good cyclic oxidation and carburization resistance, and good weldability. It also has lower thermal expansion than most austenitic stainless steels and high thermal conductivity relative to the other candidates. It retains toughness after long-time exposure at elevated temperatures and does not form intermetallic phases that can cause embrittlement.

Alloy 617 is less resistant to oxidation than Alloy X, although earlier VHTR programs concluded that the behavior was adequate. This alloy is more prone to grain boundary oxidation than Alloy X because of the formation of aluminum rich grain boundary oxides. This type of oxidation would be particularly deleterious for use in thin sections associated with some compact heat exchanger designs. Preliminary testing indicates Alloy 617 has better resistance to carburization than either Alloy 800H or Alloy X.

Aging effects on Alloy 617 are quite complex and are not well understood. Observations and predictions of which precipitates form in Alloy 617 at given temperature ranges have not been consistent. A comprehensive review of the precipitates in Alloy 617 was performed recently and it was clear from the review that the kinetics of the precipitation and coarsening processes were important in determining the effects of aging on properties. It appears precipitates may form at initial exposure and the alloy may become stronger. But most of the precipitates will be dissolved after long-term exposure in the temperature range of interest to the NGNP IHX, and the alloy will depend on solid solution strengthening at long times. Aging at 700 to 750°C (1292 to 1382°F) results in reduction in tensile and impact properties, however, these effects are less pronounced at higher temperatures.

The grain size also plays an important role in the strength of the alloy. For general applications, a grain size of ASTM No. 6 ( $\sim$  45 $\mu$ m) or coarser is typically preferred, but it has been shown that creep strength increases with increasing grain size so microstructures of 100–200  $\mu$ m grain size are often produced. A tradeoff exists, however, when fatigue is an issue, since finer grain sizes are preferred for fatigue resistance. In addition, for compact IHX, the thin sheet form restricts large grain size. Whether the grains will significantly coarsen after the dissolution of certain grain boundary precipitates at long-term exposure is not clear.

#### 3.2.7.3 Relevant Experience with Alloy 617

During early development, Alloy 617 was systematically studied by Huntington Alloys, Inc., and when Alloy 617 was considered for the HTGR, it was extensively investigated by Huntington, Oak Ridge National Laboratory (ORNL), and General Electric (GE). The Huntington data were used to develop ASME B&PV Code applications covered by Section I and Section VIII Division 1 and in a Draft Code Case for Section III.

Both the ORNL-HTGR and GE-HTGR studies generated data from Alloy 617 that had been aged and/or tested in simulated HTGR helium. The helium impurities used in those studies were the same as those considered for the NGNP system but the concentrations were different. Germany also extensively investigated Alloy 617 for its HTGR and other programs.

Over the past 5 years, interest in the behavior of Alloy 617 for VHTR applications has renewed. Activities carried out by the Korean Atomic Energy Research Institute, AREVA, French Atomic Energy Commission, and NGNP Research and Development Program are coordinated through the Gen IV International Forum Materials Program Management Board. There have been fundamental studies of Alloy 617 corrosion in VHTR atmospheres and development of predictive models for environmental effects. The creep and creep-fatigue behavior have been investigated in VHTR helium and investigators made a comparison of behavior under these conditions to newly generated data in air for the same material heats.

#### 3.2.7.4 Code Status

Alloy 617 is not currently qualified for use in ASME Code Section III, although it is allowed in Section I and Section VIII, Division 1 (nonnuclear service). A draft Code Case for incorporating Alloy 617 in Section III was developed and submitted in the early 1990s. Efforts to gain approval from the ASME Code committees were stopped because of termination of the associated VHTR programs. Comments obtained from ASME on the draft Code Case are being used to guide current research and development activities which are intended to update and resubmit the Code Case for approval in Section III, Subsection NB and NH.

Additionally, the German HTR program generated sufficient data for this material to be included in the draft version of German standard KTA 3221, for temperatures up to 1000°C (1832°F) and design periods up to 100,000 hrs.

## 3.2.8 Other Metallics

The metallic materials considered in this paper highlight potential options for select components in the primary loop. The need for additional or alternative metallic materials may become evident as the design progresses.

# 3.3 Graphite Materials

Material selection for the graphite components will be based on the same general principles discussed in Section 3.1. The grades of graphite that were used for previous HTGRs are no longer available. New grades of graphite have been developed based on the strengths and weaknesses of those previous grades. In order to qualify these new grades, testing is currently underway to obtain physical, thermal, mechanical (including radiation-induced creep), and oxidation properties. In some cases, past historical data and experience are being used (and discussed herein) to address the graphite selection and qualification approach.

A distinguishing feature of HTGR concepts is the extensive use of graphite in reactor internal components, including the core fuel blocks in the case of prismatic concepts. These graphite components are relied upon to establish core geometry, serve as the moderator in support of the nuclear heat generation process, and direct the flow of helium coolant. They also serve as a path for passive removal of heat in the case of certain licensing basis events, passive heat removal capability being fundamental to the HTGR safety concept.

This white paper addresses the use of graphite in core structural components. While HTGR fuel typically employs graphite, graphite fuel matrix (or fuel compact) materials are to be addressed separately in white papers specific to fuel qualification. In the case of prismatic-fuel reactors, the fuel elements (also referred to as fuel blocks), excluding the contained fuel compacts, are considered to be structural graphite. In the case of pebble-bed reactors, the fuel pebbles are considered part of the fuel, rather than core structures. The reflectors are classified as structural graphite components in both prismatic and pebble reactor concepts.

## 3.3.1 Graphite Applications

Graphite is used for the main core components in both prismatic and pebble-bed HTGR concepts, including in reflectors (typically top, bottom, and side reflectors), core support blocks, core support posts, and outlet plenum blocks. Graphite is also used for the fuel blocks (elements) that contain the fuel compacts in prismatic designs.

Both design concepts employ permanent and replaceable reflector components, the permanent sections usually being the peripheral reflector regions exposed to a much lower fluence and irradiation temperature. In prismatic designs, refueling outages occur at approximately 18-month intervals, during which one-half of the fuel elements are replaced. The replaceable inner reflector elements are typically changed out at approximately 6-year intervals during one of the refueling outages, leaving only the permanent reflectors to last the lifetime of the plant.

Pebble-bed concepts have historically had no design provisions for replacement of the reflector components, implying that these were expected to last the lifetime of the reactor. However, the PBMR 400 MWth design included a replaceable reflector concept, since structural analyses of graphite reflector components used with the highest utilization (in the most extreme fluence-temperature regions) indicated that these would not last the 36 equivalent full-power year reactor design life. Reflector components in the PBMR 250 MWth reactor, which is strongly based on the earlier German High Temperature Reactor (HTR) Module design, see a substantially lower fluence-temperature regime, resulting in a significantly longer life of just over 40 years, based on the available data and analysis methods. The life of this material could approach the 60-year target without the need for replacement through refinements in analytical methods, improvements in design data inputs, and refinement of the reflector component designs, as well as surveillance, testing, inspection, and maintenance. Hence, the expected lifetime of the graphite components in pebble bed designs and the measures to be taken to extend the safe operating lifetime of these components to 60 years, if needed, is an issue that requires further consideration.

The fluence-temperature exposure conditions of the graphite components differ between the prismatic and pebble-bed concepts, mainly because of differences in the fuel design and core configuration. For the same reactor inlet temperature, reactor outlet temperature system design pressure, and coolant flow rate, the nominal operating fuel temperature in the pebble design is expected to be ~865°C (1589°F), with peak fuel temperatures <1200°C (2192°F). Consequently, the graphite reflector components would be exposed to temperatures ranging from 250 to 800°C (482 to 1472°F) under normal conditions and peak temperatures approaching 1100°C (2012°F) during accident conditions.

In the prismatic design, the fuel blocks will operate between about 350 and 1200°C (662 and 2192°F) and the replaceable reflectors will operate between about 350 and 800°C (662 and 1472°F) during normal conditions, with the precise range in temperature determined by the specific core design employed. The permanent reflectors, core support, and outlet plenum will have significantly less fluence compared to the inner core graphite, even though they will operate for 60 years, because of their distance from the fuel region. During normal operation, these permanent components will operate between about 350 and 800°C (662 and 1472°F). Some portions of graphite core components may reach 1400°C (2552°F) during accident conditions.

## 3.3.2 Graphite Selection and Qualification Approach

Material selection for the graphite components will be based on the same general principles discussed in Section 3.1. One significant difference is that, up until now, only minimal guidance has been available from established regulatory requirements or the ASME Code regarding the use of these materials. This situation is expected to evolve, since a consensus ASME code on graphite component design for HTGRs has been prepared by the ASME Subgroup on Graphite Core Components and is expected to be published in 2011 (See Section 3.3.5 below). As explained in Section 3.3.5, the addition of nuclear grade graphite to the ASME Code would be ideal, but it is not required for qualification. Until the graphite code is published and accepted by the NRC, graphite material selection will focus on existing design and operating experience with both past and currently available grades of reactor graphite. Fabrication experience and technical maturity are additional selection criteria that must be considered. Notwithstanding these criteria, the performance and compatibility of candidate materials with the operating environment will be assessed. Testing is currently underway to qualify the new grades of

graphite, including tests to obtain data on physical, mechanical, and oxidation relevant properties (including the effects of irradiation and irradiation-induced creep). The extent of qualification testing undertaken thus far on new candidate HTGR grades will be considered as part of the selection process.

# 3.3.3 Candidate Graphite Grades

Nuclear graphite grades employed in past HTGR plants or developed for previous concept designs are no longer available; however, a selection of candidate grades are currently available from the major graphite suppliers. These candidate grades build on past experience, more recent developments and state-of-the-art nuclear graphite developments, and they satisfy requirements for both HTGR concepts. Table 1 provides a list of present candidate graphite grades for the HTGR and summarizes their areas of application relating to prismatic- or pebble-type designs.

Table 1. Candidate nuclear graphite grades for HTGR application.

Grade	Supplier	Key Characteristics	Existing Precedent	Area of Application
NBG-17	SGL Carbon Company	Medium grain, pitch coke filler, pitch binder, vibration molded	No	Reflector & fuel elements, prismatic type
NBG-18		Medium grain, pitch coke filler, pitch binder, vibration molded	No, based on past Grade ATR-2R	Reflector blocks, pebble type, permanent reflector blocks, prismatic type
PCEA	Graftech International	Medium grain, petroleum coke filler, pitch binder, extruded	No	Reflector & fuel elements, prismatic type
IG-110	Toyo Tanso	Fine grain, petroleum coke filler, pitch binder, isostatic-molded	Yes, used in HTTR & HTR-10	Reflector & fuel elements, pebble & prismatic type
IG-430		Fine grain, pitch coke filler, pitch binder, isostatic-molded	No	Reflector & fuel elements, prismatic type
S2020	Carbone Lorraine	Fine grain, petroleum coke filler, pitch binder, isostatic-molded	Yes, FSV	Core support posts, prismatic type

Grade selection is determined according to specific requirements of the HTGR type, including techno-economic factors. For example, raw material characteristics and availability relating to the filler coke would be a determining factor in the grade selected. NBG-17, NBG-18, and IG-430 are made from coal derived pitch coke, while grades PCEA, S2020, and IG-110 are made from crude oil derived petroleum coke. Pitch cokes are made from coal tar, which is produced as a by-product in coke ovens. Previous German developments focused on pitch coke for their graphite development program, following the oil crisis of 1978. Because of economic and environmental factors, Japan is currently the only source of pitch coke. Conversely, petroleum coke accounts for by far the largest tonnage of coke made worldwide, and is available domestically. Oil refineries are run to optimize the production of fuels, so petroleum cokes made from the heavy end of the distillation process will have variable quality and properties dependent on the crude source and refinery operation. However, on the west coast of the United States, certain smaller refineries have developed a niche business supplying specialty isotropic cokes made from sweet light crude.

Graphite material selection criteria further stem from the functional requirements of the graphite core components for the specific reactor type (pebble or prismatic). A key requirement in prismatic designs is the need for fine grained graphite, with its correspondingly higher strength, for the fuel elements to ensure an adequate number of grains across the thickness of the graphite webs between the fuel compacts and the coolant holes. Relatively high thermal stresses are generated in the thin graphite ligaments between the

coolant and fuel channels in these elements. Ideally, a ligament should be no thinner than 10-times the maximum grain size.

There is also a substantial database on irradiated properties for grades (now historical) that are similar to NBG-18 (ATR-2E, ATR-2R, and VQMB), which provide valuable insights regarding the NBG-18 irradiation behavior that is to be expected, as well as being useful for preliminary design purposes. Importantly, this historical data indicates that both ATR-2E and ATR-2R (bearing close resemblance to NBG-18) exhibited good dimensional change behavior under irradiation. That is, they underwent low maximum shrinkage with small differences in dimensional changes in the with-grain and against-grain orientations, these factors are very important in terms of reducing internal stress in core components.

As another example, Grade IG-110 has already established significant design and operating experience, being employed for both fuel elements and reflector blocks in the HTTR (Japan) and reflector components in the HTR-10 (China). The HTTR is a 30 MWth test reactor of the prismatic design, in operation since 1998. The HTR-10 is a 10 MWth pebble-bed design that went critical in 2000. Grade IG-110 is further earmarked as the candidate grade for the reflector blocks of the scaled-up GTHTR-300 prismatic design concept (Japan) and HTR-PM pebble-bed design concept (China). There is a significant irradiated properties database for IG-110 over a range of HTGR applicable temperatures, but only to limited fluence. This is in line with the low peak fluence requirements of prismatic designs, however, the HTR-PM pebble-bed concept would necessitate that higher fluence irradiation data for IG-110 at representative operating conditions be acquired prior to construction or start-up. The little available high fluence data for IG-110 at 600°C (1112°F) indicates similar dimensional change behavior similar to that of historic coarser grained materials, such as ATR-2E. Irradiated properties data for IG-110 tends to be presented without directional orientation on the basis that the material is isotropic; however, this is an aspect that needs some verification, since unirradiated properties measured for IG-110 in different orthogonal directions can show some variation.

Other considerations such as supplier capability and grade manufacturability may also feature strongly amongst the selection criteria. All major manufacturers typically have the capability to produce extruded or isostatically-molded products, with vibration-molded materials being less common. The various suppliers typically have a preference based on historical developments and expertise. For example, SGL carbon has a history of producing extruded and vibration-molded product for the former German program and, therefore, favor extruded or vibration molded products based on isotropic filler coke. This is strongly related to the need for large graphite billets for pebble bed designs, where medium grain graphite is better suited to large block manufacture. This provides some advantage over the extrusion process in terms of the maximum size of blocks and properties achievable; hence, the development of grades NBG-17 and NBG-18, which benefit from the experience of the German program.

Toyo Tanso has focused its efforts on isostatic pressing, based on its own developments and its affiliation to the HTTR test reactor development. IG-110 represents the flagship HTGR graphite grade of the company, where the use of very fine-grain petroleum coke-based raw material provides an isotropic, high strength material well suited to a prismatic HTGR application. The isostatic-pressing technique has been further extended to develop Grade IG-430, an isostatic-pressed, pitch coke derivative that fills the gap for a higher strength, high-conductivity, isotropic graphite for VHTR applications. However, the process to achieve this small grain size is said to limit the maximum size of graphite blocks that can be produced, which could pose difficulties when fabricating the larger structural components. However, it is interesting to note in this regard that the scaled up HTR-PM pebble bed design, which requires large graphite sections, currently plans to utilize IG-110 as reflector graphite. The requirement for fine-grained graphite is less significant for the outer permanent reflector elements of the prismatic design, where coarser grained graphite can be employed, as is the case with the application of PGX graphite in the HTTR.

## 3.3.4 Graphite Properties

Typical properties of the candidate grades are given in Table 2. All of the candidate graphite grades have low ash concentration, reflecting a qualitatively low level of catalytic impurities, which can potentially enhance graphite oxidation reactions with primary coolant impurities.

Table 2. Typical graphite vendor properties of candidate graphite grades.

		PROPERTY*						
			TI 1	Dynamic	T 1	6	A 1	
Grade	Density (g.cm <sup>-3</sup> )	CTE (10 <sup>-6</sup> °C <sup>-1</sup> )	Thermal Cond. (W.m <sup>-1</sup> .K <sup>-1</sup> )	Young's Modulus (GPa)	Tensile Strength (MPa)	Comp. Strength (MPa)	Ash Content (ppm)	Isotropy Ratio
NBG-17	1.84	4.5, 4.6	130	11.0	19		<300	1.02
NBG-18	1.86	4.6	136	11.9, 11.6	20.8, 20.4	77.4, 78.5	< 300	1.02
PCEA	1.83	3.5, 3.7	162, 159	11.3, 9.9	21.9, 16.9	60.8, 67.6	< 300	1.05
IG-110	1.77	4.5	120	9.8	25.3	76.8	<100	<1.10
IG-430	1.82	4.8	140	10.8	37.2	90.2	<100	<1.10
S2020	1.77	4.3	85	10.7	29	93	500	1.14

<sup>\*</sup>Both with-grain and against grain values given where available or applicable

These grades all qualify as isotropic or near-isotropic in accordance with ASTM D7219-08. The degree of isotropy is defined by the ratio of the larger to the smaller coefficient of thermal expansion (CTE) in the with-grain and against-grain directions. ASTM D7219-08 recommends that material exposed to high neutron flux regions of the HTGR be isotropic. It is well established on the basis of past research and operating experience that highly anisotropic grades develop large differences in their irradiation-induced dimensional changes in the with-grain and against-grain orientations, resulting in excessively large internal stresses within the graphite components. This stress can be substantially reduced in reflector components by utilizing more isotropic grades.

Importantly, ASTM D7219-08 specifies a range of physical and mechanical properties for isotropic grades that allows for a broad range of nuclear graphite grades as far as raw materials, forming method, purity level, and actual properties are concerned. More anisotropic grades not precisely meeting the D7219-08 degree of isotropy requirement may still be applied in HTGR applications, provided due diligence is paid to material behavior under irradiation, operating conditions (fluence-temperature), and design considerations. These grades can be applied to lower fluence regions as recommended by ASTM D7301-08, which would typically apply to the outer reflector regions.

It is important to recognize that the degree of isotropy only serves as an initial indicator of the graphite behavior under irradiation. End-product isotropy is influenced by raw material, grain size, forming method, and heat treatment. Graphite billets can be fabricated by extrusion, isostatic molding, or vibration molding. Extrusion tends to yield graphites that are less isotropic and less dimensionally stable under irradiation than molded graphites, although isotropy can be improved remarkably through careful control of raw material and processing. Isostatic-molded graphite is commonly available in smaller sizes than extruded grades, while vibration molding are available for larger block sizes.

The selection of graphite materials for HTGR core components is based on physical properties (density, CTE, thermal conductivity, specific heat capacity), mechanical properties (strength, Young's modulus, fracture toughness), neutronic properties (neutron absorption cross-section), response to irradiation, and resistance to chemical attack (e.g., oxidation).

The mechanical properties of graphite provide the basis for its use as a structural material to establish and maintain the geometric characteristics of the HTGR to assure the ability to insert reactivity control materials and to provide the required channels for the flow of helium coolant. The neutronic properties of graphite support its neutron moderator and reflector functions, while minimizing the development of activated byproducts. Physical properties such as thermal conductivity, specific heat capacity, and emissivity are critical to the HTGR safety concept, providing for thermal energy storage and transport, thus limiting fuel temperatures during certain LBEs and associated DBAs involving conduction cooldown.

The sections that follow further elaborate upon the properties of graphite and the significance of those properties in HTGR design and operation.

# 3.3.4.1 Properties of Ideal Nuclear Graphite

The properties most relevant for ideal nuclear reflector graphite are summarized in Table 3. These requirements are based on previous experience gained in the manufacture and application of nuclear grade graphite for reactor core internals, as well as more recent developments in nuclear graphite technology.

It can be seen from Table 3 that the properties of graphite can be classified within four general categories. <sup>5,6</sup> The first category relates to structural functions. Important structural properties include density, strength, anisotropy, and CTE. Dimensional changes under irradiation also play a key role in the useful structural life of the graphite reflector components.

The efficiency of graphite as a neutron moderator/reflector is characterized by two properties, the density and neutron absorption cross-section, which is a function of graphite impurities. Thermal conductivity provides a measure of the heat transport capabilities of graphite, which are important to the HTGR safety concept.

#### 3.3.4.2 Manufacturing Considerations

Graphite products are manufactured for a wide variety of conventional applications, ranging from electric motor brushes to arc-furnace electrodes. Nuclear applications of graphite date from the Chicago Pile in 1942 and, since that time, over 100 graphite moderated reactors have been constructed, including six HTGRs, of which two (the HTTR in Japan and HTR-10 in China) are presently in operation.

The ideal requirements for nuclear grade graphite are summarized in Section 3.3.4.1 above. In considering these requirements, there are a number of raw material and process variables that can be combined to produce graphites with the desired properties.

#### **Raw Materials**

The raw materials for nuclear graphite include coke, binder, and impregnation materials. Coke is a solid carbonaceous material that is most frequently derived as a byproduct of crude oil processing or the destructive distillation of coal. Pitch, which is used as the binder, is a solid (at ambient temperature) or is a highly viscous carbonaceous liquid that is also most frequently derived from petroleum crude oil or coal. Pitch is mixed with the coke to provide a material that may be molded or extruded into the desired component shape. Impregnation materials such as pitch are used for further processing to provide for increased density and strength.

Table 3. Ideal requirements for reflector graphite.

Property	Required Range	Reason	Performance Attributes
Density	1.7–1.9 g/cm <sup>3</sup>	High density is indicative of lower porosity, provides for more effective neutron moderation/reflection per unit volume, and in general, also indicates higher strength.	Neutron efficiency Structural integrity
Neutron absorption cross-section	<5 mbarn	Required for neutron efficiency of the core. The limiting neutron absorbency is that of pure carbon (~3.5 mbarn).	Neutron efficiency
Thermal conductivity at room temperature	>100 W/m/K	Indicative of a high degree of graphitization and typically the level required for effective heat transfer in HTGR applications.	Heat transport
Purity (total ash content)	<300 ppm	Required to minimize activation and reduce susceptibility to catalytic oxidation. It is possible to manufacture graphite with much higher purity levels using a dedicated purification step. The selected and specified purity may vary depending on the function of the components. This decision will be based on a cost-benefit analysis.	Component activity levels during replacement and/or disposal. Graphite Oxidation under normal and accident conditions.
Tensile strength	>15 MPa (tensile)	Adequate strength is required for structural component integrity. The strength reserves offered by the material must exceed the allowable operating component stresses.  Higher strengths are achievable with isostatic-molded, fine grain graphite, but these typically possess lower fracture toughness. This is a trade-off that must be taken into account in the design.	Structural integrity
, ,	$3.5 \text{ to } 5.5 \times 10^{-6} \text{ K}^{-1}$	A higher value is indicative of the coke isotropy and hence isotropy of the graphite. This implies that the graphite will have better dimensional stability when subjected to fast neutron irradiation. However, lower CTE can be beneficial in terms of thermal stress.	Structural integrity
CTE isotropy ratio	<1.10	Indicative of the bulk graphite isotropy.	Structural integrity
Dynamic elastic modulus	8–15 GPa	Higher modulus is typically associated with a higher strength material, but increased sensitivity to thermal stresses. Thus, values at the lower end tend to be more beneficial.	Structural integrity
Dimensional changes with irradiation	Minimal shrinkage over the applicable fluence range and minimal differences in the with-grain and against-grain directions	This is mainly a function of temperature and fluence, but is strongly dependent on the graphite grade. Dimensional changes strongly influence the level of internal stresses generated in core components when subjected to fast neutron irradiation and are critical in determining their useful life.	Structural integrity

The properties of graphite, and particularly the irradiated properties, are highly dependent upon both the raw materials (particularly the coke) and the processing steps that are described below. For this reason, a change in the source and/or characteristics of the raw materials may require requalification of the graphite for nuclear service. Precursor materials will change over time. The current NGNP Project is performing research to understand the effects on graphite properties with changing precursor materials.

#### **Process Variables**

Process variables include the particle size distribution, production steps, and degree of purification.

The particle size distribution is generally classified by maximum grain size into coarse-, medium-, and fine-grain material. Typically, medium-grain graphites have been used for nuclear applications in the United Kingdom, Germany, and the United States. Recently, fine-grain materials have been used for HTTR in Japan and HTR-10 in China.

In terms of fabrication techniques, graphite is manufactured in several stages:

- 1. Procurement of raw materials including coke, pitch, etc.
- 2. Grinding coke particles to the required size by milling.
- 3. Mixing of ground coke particles with pitch to form a visco-elastic mass.
- 4. Green fabrication of the billet by molding or extrusion.
- 5. Baking of the billet at 850 to 1200°C (1562 to 2192°F) for 30 to 70 days to remove the volatile carbon artifacts.
- 6. Density increase by impregnation with pitch, generally the bake-impregnate-rebake process is done three times for a nuclear grade graphite.
- 7. Graphitization of the billet at 2500 to 3200°C (4532 to 5792°F) for up to 15 days. Higher temperatures are better; however, temperatures beyond 2800°C (5072°F) are difficult to achieve in large size billet production because of furnace limitations.
- 8. Purification (may be combined with graphitization).
- 9. Machining to final size and shape.

The impregnation and baking stages are often repeated to improve mechanical strength and to produce a higher density product.

## 3.3.5 ASME Code for Graphite Structures

As renewed interest in HTGR applications emerged, an ASME Section III Subgroup on Graphite Core Components was established in 2004. The charter of the subgroup includes the establishment of rules for materials selection and qualification, design, fabrication, testing, installation, examination, inspection, certification, and preparation of reports for manufacture and installation of nonmetallic internal components for fission reactors, including graphite but excluding nuclear fuel.

#### 3.3.5.1 Changing the Approach of the Prior ASME draft Code Case

The international committee first reviewed the prior draft of the ASME graphite core component code case and decided that approach was inadequate for today's design and regulatory licensing. The prior draft was a deterministic approach that paralleled that of metallic core components. The prior code only addressed nonirradiated graphite use and did not endorse design of irradiated graphite core components. Graphite was treated as a linear Hookean material with no strength in the plastic regime. Graphite core components that suffered cracking were considered failed and would be removed at the earliest convince.

# 3.3.5.2 Current Philosophy of Current ASME Graphite Core Component Design Rules

The current rules apply to graphite core components utilized in a high-temperature, graphite-moderated, gas-cooled fission reactors. Graphite core components include fuel blocks, reflector blocks, shielding blocks, and any keys or dowels used to interconnect them. The rules also apply to the

arrangement of graphite core components that form the graphite core assembly. The rules do not apply to fuel compacts and pebbles, bushings, bearings, seals, blanket materials, instrumentation, nor core restraints.

The committee concluded that the designer should account for the effects of irradiation on the thermal and mechanical properties of the graphite in the design of the graphite core. The design must also consider the statistical variation of strength within the billet, as well as the variations resulting from manufacture in different production runs. The committee endorsed the use of ASTM D7219-08 that provides guidance on statistical sampling and provided the minimum mechanical parameter values for near-isotropic nuclear grade graphite.

The code provides for a simplified deterministic design methodology for irradiated and nonirradiated graphite using ultimate strengths determined from Weibull statistics of the graphite billet strengths. This is a conservative method which, if not met, does not mean the component is rejected. The code also provides a full analysis method that uses a probabilistic design approach incorporating Weibull statistics to determine the probability of failure. The margin to failure is determined by comparing the probability of failure to the three structural reliability classes (SRC) defined in the code. Each SRC is assigned a maximum probability of failure, based on the safety function and expected operational transients. The SRCs and their respective maximum probability of failure are shown in Table 4.

Table 4. Maximum probability of failure for each safety class.

Structural Reliability Class	Maximum Probability of Failure
SRC-1	1.0E-4
SRC-2	1.0E-2
SRC-3	1.0E-1

A third design method is provided on the basis of the testing of full-scale graphite components. The test shall be designed to ensure that the loads determined from the tests conservatively represent the load-carrying capacity of the actual graphite core component for the specified loadings. The test objective is to demonstrate that the probability of failure is within the SRCs in Table 4. The test results shall provide values with a 95% certainty, as represented by a single-sided confidence level and envelope all appropriate design and service loadings.

The code's use of probabilistic design departs from standard ASME design code methodologies. The ASME code has not previously addressed irradiation induced changes to material properties. This will be the first time in ASME code history that the ASME Board of Governors has approved of this approach. The code includes instructions on how to collect the material properties changed by irradiation as a function of temperature. Irradiation induced creep is included as one of the parameters that must be measured.

Another deviation from past ASME metallic codes is allowance of cracks in the graphite components. The rules require the core designer to demonstrate through analyses or testing that cracked graphite core components can maintain their assigned safety function and that the graphite component is remotely retrievable when cracks of a specified size and orientation are present. This puts a high reliance on developing ISI techniques that provide visual and in-situ measurement of graphite properties.

The code also deviates from the ASME standard practice of defining primary, secondary, and membrane stresses. Mechanical loads on the graphite core components are small compared to stresses induced by irradiation damage. These irradiation-induced stresses follow thermal and neutron fast fluence gradients within the block and are the critical stresses in the graphite blocks while the mechanical stresses (primary, secondary, and membrane) do not challenge the graphite core components strength margins.

#### 3.3.5.3 Status of the ASME Graphite Core Component Code

As of March 2010, the ASME graphite core component code was approved by ASME and will undergo editing by the ASME editors for publication in the 2011 update of the ASME Code. Additional editorial comments approved by the graphite core component committee are being addressed in parallel with the ASME staff's editorial process. It is anticipated the graphite design rules will be published in the ASME Code under ASME Section III Division 5 Subgroup on High Temperature Reactors. Currently, Division 5 is still under development and has not been officially recognized by the ASME organization.

# 3.3.6 Graphite Materials Qualification

This section addresses the proposed bases for graphite materials qualification. The following subsections summarize the key functions and requirements associated with the graphite reflector components and the associated material qualification requirements for HTGR application. The key elements of the graphite Material Qualification Plan are discussed through the use of examples in Section 3.3.6.4. This is followed by an overview of the main test parameters and material characterization that will be needed to provide the as-manufactured and irradiated properties of candidate graphite grades for the HTGR application.

### 3.3.6.1 Key Functions and Requirements to be Validated

The graphite components of HTGRs fulfill the following safety-related functions:

- Maintain core geometry
- Contain fuel compacts within the fuel elements (prismatic reactors)
- Provide undisturbed access for the insertion of reactivity control material
- Passively remove core heat, primarily by radial conduction from the fuel to the core barrel, during off-normal events when forced cooling is not available
- Control chemical attack by limiting oxidation for off-normal events involving ingress of water or air gas mixtures.

Key requirements associated with these functions are summarized below.

#### **Design Life**

The design life of the HTGR plant is 60 years. The graphite core components are generally subdivided into two groups, replaceable components and the fixed or permanent reflectors, as required by the particular HTGR concept design. Components that cannot be designed for the full plant life (such as reflector graphite components in high flux regions) need to be designed to be replaceable and, thus, allocated a design life. Both permanent and replaceable graphite core components must adequately perform their safety-related functions for the duration of the allocated design life and this must be validated for their respective operational periods.

Prismatic HTGR designs are refueled at approximately 18 month intervals during which time typically one-half of the fuel elements are replaced. Replaceable reflector elements adjacent to the active core may also be changed out during such refueling outages, typically at 6-year intervals, leaving only the permanent reflectors to last the lifetime of the plant.

Earlier pebble-bed concepts with shorter design lifetimes (e.g., 20–30 years) did not require provisions for replacement of reflector components. Based on presently available data and analysis methods, the most highly irradiated reflector components are based on the earlier German HTR Module design and would therefore have a projected life of just over 40 years. There is a possibility that this life

could be extended further, approaching the 60-year HTGR target, without the need for replacement. However, this would require refinements in analytical methods, improvements in design data inputs, and the optimization of reflector component designs as well as an effective surveillance, testing, inspection, and maintenance program. At most, one reflector replacement outage may be necessary for a 250 MWth pebble bed design with a 750°C (1382°F) outlet temperature for a 60-year life. The actual design life of the replaceable reflector components will need to be established based upon the results of materials irradiation tests, supplemented by data obtained through actual operating experience.

#### **Service Conditions**

The fluence levels and irradiation temperatures seen by the various graphite reflector components are dependent upon both the reactor type and their locations within the reactor, i.e., service conditions seen by graphite components within pebble bed reactors are typically more limiting, because of their longer design lives and the correspondingly higher fluence levels that will be accumulated by some of these components. Normal operating temperatures for the pebble bed graphite reflector range between 250 and 800°C (482 and 1472°F). The maximum temperatures that would be seen by the reflector graphite during certain LBEs are less than 1200°C (2192°F). Only components in close proximity to the pebble fuel are subjected to high fluence levels that may limit their lifetime to less than that of the design life of the plant as a whole, this maximum fluence being slightly above  $1.1 \times 10^{22}$  n.cm<sup>-2</sup> EDN (equivalent dido nickel) (15 dpa). The affected parts of these components do not serve a structural function.

### 3.3.6.2 Characterization of Unirradiated Graphite

As-manufactured graphite material must be characterized to ensure its suitability for use in the HTGR and to generate design data for graphite core components in low fluence areas that can effectively be treated as unirradiated. All design-relevant properties must be characterized (density, CTE, Young's modulus, thermal conductivity, tensile and compressive strength, specific heat capacity, fatigue strength, oxidation resistance, and neutronic properties) along with their temperature dependence.

It is important that the characterization program include enough material batches to adequately assess the material variability. Graphite properties generally display significant variability within-billet, within-batch and from batch-to-batch. This variability has to be assessed for the candidate graphite grade. The degree of variability is influenced to some extent by the forming method and processing history, but is largely inherent to polygranular graphite materials. For a probabilistic graphite component lifetime prediction approach, the Weibull material parameters must also be established with sufficient confidence, which requires that a statistically sufficient number of samples be tested.

## 3.3.6.3 Characterization of Irradiated Graphite

During reactor operation, graphite undergoes structural changes because of fast neutron irradiation that, in turn, lead to changes in most of its physical and mechanical properties (see Appendix B). The irradiation conditions seen by various reflector graphite components are strongly dependent on their location within the reactor. The property changes with irradiation that the design must account for are:

- Dimensions
- Coefficient of thermal expansion
- Thermal conductivity
- Tensile strength
- Young's modulus.

The need to account for these fluence-related property changes is specified by the current draft ASME Code for Graphite Component Design for HTGRs in Section GB-2200, "Material Properties for Design," and Subsection GB-2220, "Irradiated Material Properties." Important design considerations with respect to irradiation-induced changes in graphite properties are discussed below.

Dimensional changes physically influence the core structures and must be accommodated for in the reactor core design to ensure the accomplishment of safety functions (reliable insertion of control rods and passive heat removal capability during LBEs involving conduction cooldown). Therefore, the dimensional change behavior of the particular candidate grade requires proper evaluation to ensure that its influence on the component lifetime in-reactor is adequately assessed.

Thermal conductivity of the graphite is very important during normal operation in ensuring adequate heat transfer to the coolant and regulation of fuel temperature within acceptable limits. Additionally, it plays a key role in heat removal during low frequency LBEs involving conduction cooldown. In general, graphite thermal conductivity decreases dramatically with low fluence, reaches a saturation value that persists for a portion of the design fluence range, and is followed by a secondary reduction because of more advanced material degradation. Again, the design inputs that must be considered for prismatic and pebble types will differ, the latter operating the graphite into higher fluence regimes.

Other important design properties such as CTE, Young's modulus, and strength also need to be evaluated with irradiation temperature and fluence for graphites used in both reactor types. Both CTE and Young's modulus increase with fluence and reach peak values, after which they decrease below their unirradiated values. Both are important to the evaluation of thermal stress and irradiation creep in the graphite parts. Strength also increases and follows a similar progression as Young's modulus with fluence and temperature. From a design perspective, the available strength reserve must be compared against the stress within the graphite component based on the fluence-temperature history of the component.

Irradiation-induced creep in graphite is also a key design parameter whose vital role is to relieve internal stresses generated by irradiation-induced dimensional changes in graphite core components. In the absence of this stress-relieving mechanism, reactor components would fail at a much earlier stage of irradiation. While extensive fundamental research and data gathering have been conducted on this phenomenon over the past 50 years, or so, there is still need for improvement in the available creep models and their range of applicability in terms of fluence and temperature. Evolution of creep strain with fluence in irradiated graphite (differential strain between stressed and unstressed) specimens is characterized by primary, secondary, and tertiary regimes, much like those observed in metals. There is evidence to suggest that the normalized creep strain (normalized to initial elastic strain) is similar for different grades of graphite, lending support to the theory that this creep behavior is not material grade specific. Additional creep data would be useful in supporting this position and extending its application for a broader fluence-temperature range and for a wider variety of nuclear graphite grades. This may help in rationalizing and minimizing the need for costly irradiation creep experiments for current reactor graphite grades or grades that may be developed in the future.

Another important aspect related to the characterization of irradiated materials properties is the use of recognized materials test standards, typically ASTM standards, for generating the material data. This is important towards ensuring that the data capture and analysis follows recognized test procedures and that data acquired by different test facilities are directly comparable. One of the major challenges in this area is the need by Materials Test Reactor (MTR) facilities to utilize subsize test specimens (nonconformant to that specified or recommended by the applicable standard) for the graphite irradiations. This need stems from the requirement to irradiate a statistically representative population of test specimens for each property to be measured, and the limited irradiation capsule volume for accommodating test specimens. Both of these factors add to the cost of the irradiation test programs. Effort is underway within the ASTM D02 Technical Committee on Manufactured Carbon and Graphite (Nuclear Applications) to develop test

standards for the irradiation of graphite specimens that do not conform to conventional ASTM standard test specimen sizes. As part of this effort, round robin testing would be conducted to establish the degree of reproducibility associated with using the mini-test specimens that would be recommended for irradiation and to demonstrate that properties measured on subsize specimens are valid when compared to those obtained on standard specimens.

## 3.3.6.4 Graphite Development Programs

Graphite development programs are presently underway in support of HTGR initiatives in the United States, Europe, and China. A brief overview of these programs is provided in the following sections.

#### **PBMR Graphite Development Program**

In support of the proposed PBMR Demonstration Power Plant (DPP) in South Africa, a detailed Materials Qualification Plan (MQP) was developed for NBG-18 graphite that is consistent with the requirements of the Requirements for the Design and Manufacture of the Ceramic Internals (RDMCI) and local regulatory requirements stipulated by the South African National Nuclear Regulator. The objectives of this MQP are two-fold:

- To characterize the as-manufactured and irradiated properties of NBG-18 graphite as a basis for confirming its suitability for use in the PBMR and its compliance with the requirements established for the PBMR reflector components.
- To validate the PBMR analytical models for predicting the behavior of irradiated graphite.

As of the time of this paper, several batches of pre-production NBG-18 graphite have been acquired and characterized. The characterization of as-manufactured properties is effectively complete.

Graphite irradiation tests programs are also planned in support of the PBMR, but are presently on hold. The present "PBMR-Specific Materials test reactor Program" (PSMP) is specific to NBG-18 graphite and covers the operating fluence-temperature envelope projected for the DPP-400 reactor design. Some modifications of the PSMP will be required to address the lower-temperature range associated with the reactor inlet conditions of the present reference 750°C steam cycle design for the HTGR.

The approach taken by PBMR is to utilize the historical database characterizing the irradiated properties of the earlier German ATR-2E and VQMB graphites since these grades are believed to be similar to NBG-18 (i.e., all are medium grain, pitch coke graphite and are molded). The PSMP will be used to validate the use of this existing database in developing analytical models for the initial design and structural analysis of the graphite core structures, and to supplement this database in areas where data is sparse or unavailable.

The overall strategy is for the irradiated properties database developed through the PSMP to be sufficiently complete by initial plant startup to confirm or improve the accuracy of analytical models for graphite design and to justify operation of the plant over a substantial portion of its life. Thereafter, the remainder of the PSMP will substantially lead the actual operation of the initial plants.

### **NGNP Graphite Development Program**

With the advent of the NGNP project, an advanced graphite development effort was initiated in the United States and structured within a program led by Idaho National Laboratory (INL). The activities associated with this program fall into the following areas:

- Unirradiated and irradiated material properties characterization
- Modeling

- ASTM test development
- ASME Code development.

The graphites used in earlier HTGRs built in the United States (e.g., H-451) are no longer available. The NGNP graphite development effort addresses a range of potential materials, as well as historical grades, utilized in contemporary HTGR applications (HTTR and HTR-10), which are included for comparison. The grades presently being considered within the NGNP Advanced Graphite Development Program are summarized in Table 5.

Six graphite grades (from four manufacturers) were initially selected as the principal candidates for the HTGR application: NBG-17, NBG-18, PCEA, PGX, 2020, and IG-430.7 An additional graphite grade, 2114, has since been added to the recommended list. The latter is a pitch coke, isostatically molded graphite that is a potential replacement for grade 2020.

Respectively, NBG-18 (coarse grain size, pitch coke, vibration molded) and PCEA (medium grain size, petroleum coke, extruded) graphites are considered to be grades most likely to meet the initial pebble-bed and prismatic design requirements. The other recommended grades are included for the following reasons and are all fine-to-medium grain graphites that could potentially be used to support a prismatic core design:

- NBG-17: Pitch coke, vibration molded candidate for high dose regions of the core (not currently available commercially)
- PGX: Petroleum coke, extruded, large blocks for permanent structures (used in HTTR)
- 2020: Petroleum coke, isostatically molded for permanent structures in the core
- IG-430-Pitch coke, isostatically molded, candidate for high dose regions of the core.

Table 5. Graphite grades in the NGNP development program.

Grade	Supplier	Forming Method	Coke Type	Application		
Recommended Grades						
NBG-17	SGL Carbon	Vibration molded	Pitch coke	Prismatic fuel & reflector		
NBG-18*		Vibration molded	Pitch coke	Pebble bed reflector		
PCEA*	Graftech	Extruded	Petroleum coke	Prismatic fuel & reflector		
PGX		Extruded	Petroleum coke	Prismatic permanent reflector		
2020	Carbone	Isostatically-molded	Petroleum coke	Prismatic fuel & core supports		
2114	Lorraine	Isostatically-molded	Pitch coke	Core support; 2020 replacement		
IG-430	Toyo Tanso	Isostatically-molded	Pitch coke	Prismatic fuel & core supports		
Other Grades Co	nsidered					
H-451 (Historical grade)	Great Lakes	Extruded	Petroleum coke	Provides a basis for comparison		
IG-110	Toyo Tanso	Isostatically-molded	Petroleum coke	HTTR, HTR-10		
NBG-10	SGL Carbon	Extruded	Pitch coke	Replaced by NBG-18		
NBG-25		Isostatically-molded	Petroleum coke	Core support		
HLM		Extruded	Petroleum coke	FSV permanent reflector		
PPEA	Graftech	Extruded	Pitch coke	Pebble bed reflector		
PCIB		Isostatically-molded	Pitch coke	Core support		
* Major grades; primary reference for pebble bed (NBG-18) and prismatic (PCEA) reactors.						

Additional graphite types are also being considered for various reasons. H-451, which was used in the FSV reactor, is being used as a primary historical reference for irradiations. IG-110 is used in the HTTR and HTR-10 reactors. Other grades are being considered based on reactor designer interest.

Complete properties data need to be developed for the graphite(s) eventually selected for the NGNP. Once the baseline material properties for the selected graphite grade(s) have been established, irradiation induced property changes must then be determined, including the characterization of irradiation induced creep. Determining these properties are important data needed for the design to satisfy the safety-related functions indentified in Section 3.3.6.1. These data will be developed through a series of irradiation experiments in the Advanced Test Reactor (ATR) at INL and HTV-1,2 at the High Flux Isotope Reactor at ORNL.

As noted above and in Appendix B, irradiation creep plays an important role with respect to the operating life of structural graphite components. This property is also difficult to measure and requires the development of an Advanced Graphite Capsule (AGC) designed to characterize the irradiation induced creep rate as a function of temperature and fluence. A cross-section through the AGC irradiation capsule is given in Figure 2. The approach used in the ATR experiments is to irradiate matched pairs of stressed and unstressed graphite samples. This is achieved by taking advantage of the axial flux symmetry in the ATR to irradiate compressively stressed specimens above the core centerline and unstressed specimens below the core centerline in each of six vertical channels in the capsule around the capsule periphery. This arrangement maximizes the number of specimens that can be tested at the target temperature range along the 4-foot usable vertical section of the ATR core. Target temperature ranges are maintained by gamma heating, selective neutron shielding, selected inert sweep gas ratios of helium and argon in the gas jacket of the capsule, and varying gas channel widths in the vertical orientation. The center channel is used for nonstressed drop-in samples. The creep strain is determined from the difference in irradiation-induced dimensional changes between the stressed and unstressed samples.

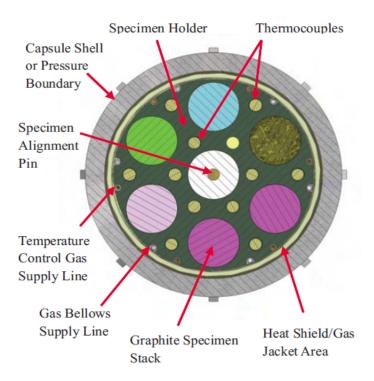


Figure 2. Cross section through typical AGC.

The distribution of AGC experiment irradiations showing planned neutron dose and temperatures is given in Figure 3. The AGC experiments will be conducted at 600°C (1112°F), 900°C (1652°F), and 1200°C (2192°F). At each temperature, two different capsules will be irradiated to different fluence levels; the first from 0.5 to 4 dpa and the second from 5 to 7 dpa. AGC-1,2 capsules cover the 600°C (1112°F) irradiations while AGC-3,4 and AGC-5,6 cover 900°C (1652°F) and 1200°C (2192°F) respectively.

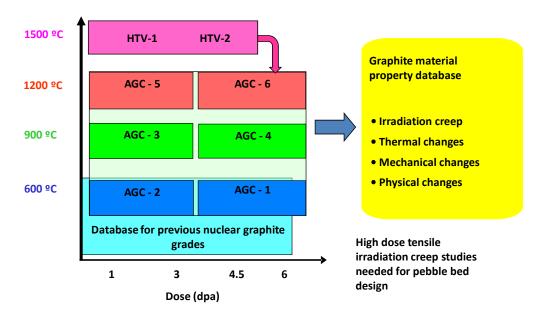


Figure 3. Planned dose and temperature distributions for the AGC experiments.

The HTV irradiations shown in Figure 3 are drop-in experiments only, and may be required prior to irradiation of the AGC-6 capsule, since the AGC-6 capsule is the only one in the series that may approach expected turnaround limits for the selected graphite types. The HTV experiments will be operated at much higher temperatures (inducing faster turnaround) but at lower doses so that turnaround for these materials can be established prior to the irradiation of AGC-6. The prismatic HTGR design assumes that fuel and reflector blocks will be replaced well before turnaround. The pebble bed HTGR design assumes that the front facing reflector blocks adjacent to the pebble fuel will stay in the reactor beyond turnaround to maximize service life. The peak 7 dpa dose in the AGC experiments supports both prismatic and pebble-bed graphite designs; however, relevant volume contraction data will be provided at the AGC peak doses. Higher dose volume expansion experiments can be considered to support the pebble bed concept at a later time.

Post-irradiation examination and testing of the irradiated samples will be conducted at INL and ORNL facilities.

AGC-1 is the first of the six experiments designed for the ATR and will focus on the prismatic fluence range. The AGC-1 experiment was inserted in the ATR during Cycle 145A in September 2009 and is scheduled to be discharged in Cycle 148b in October 2010. As of January 2010, AGC-1 had attained approximately one-third of its target fluence. Preirradiation characterization of graphite samples for AGC-2 is presently underway.

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#### **Other Graphite Development Programs**

Additional graphite development and irradiation test programs are underway in the European Union (EU) and China. The EU FP5 and FP6 Materials Test Reactor Programs include irradiation of all the candidate grades mentioned above, as well as additional piggyback grades and historical (archive) grades. The major graphite grades being tested in the EU program are shown in Table 6.

Table 6. Major graphite grades in the EU FP graphite irradiation program.

Grade	Supplier	Forming Method	Coke Type
NBG-10		Extruded	Pitch coke
NBG-17		Vibration molded	Pitch coke
NBG-18	SGL Carbon	Vibration molded	Pitch coke
NBG-20		Extruded	Petroleum coke
NBG-25		Isostatically-molded	Petroleum coke
PPEA		Extruded	Pitch coke
PCEA	Graftech	Extruded	Petroleum coke
IG-110	T T	Isostatically-molded	Petroleum coke
IG-430	Toyo Tanso	Isostatically-molded	Pitch coke

Test specimens from these grades were irradiated at 750°C (1382°F) up to high fluence (in excess of 20 dpa), beyond turnaround. A second phase of irradiation at 950°C (1742°F) up to high fluence (>20 dpa) is also underway. These irradiations at HFR Petten (Netherlands) aim to provide irradiated properties data that can be used to compare irradiation behavior and post-irradiation properties of the different reactor grades available today, both between these grades and against their historical counterparts.

Graphite irradiation tests programs are also planned by China in support of the HTR-PM, a steam cycle pebble bed concept designed as a commercial follow-on to the HTR-10. As with the PBMR development effort, the Chinese program is graphite-specific and covers the operating fluence-temperature envelope expected for the HTR-PM. Since both the PBMR and HTR-PM designs are strongly related to the German HTR Model, the fluence-temperature envelope would be expected to be similar for both. However, unlike its German predecessor, which employed coarse grain, pitch coke nuclear graphite as reflector material (e.g. ATR-2E, ASR-1RS, PXA2N), the HTR-PM will employ fine-grained, isostatically-molded, petroleum-coke based IG-110 as the reflector graphite. This follows from the use of IG-11 for the HTR-10 graphite reflector.

# 3.3.7 Graphite Operational Considerations

## 3.3.7.1 Approach for Identifying and Validating the Operational Period

During reactor operation, graphite undergoes structural changes that lead to changes in most of its physical and mechanical properties. The changes in properties and their consequent influence on the graphite structures must be accommodated in reactor core design. The most significant design related property changes include changes in dimensions, CTE, strength, modulus, and thermal conductivity, plus the effects of irradiation creep. Oxidation of graphite components must also be considered, however, its influence on component strength and, hence, structural integrity is not expected to be significant for events within the design basis.

Given the expected changes in materials properties during operation, the requirement is to ensure the safe performance of the graphite components under all applicable operating conditions over their allocated design life. For the pebble-bed designs, reflector replacement may be required once during the reactor lifetime. The following approach will be utilized for pebble-bed designs and is also applicable for extending the life of replaceable reflectors in prismatic designs:

1. The design life of graphite core structures will be initially established on a conservative basis, using probabilistic analysis methods.

It is reiterated here that the initial design life is based upon the criterion that the probability of a failure in the most highly stressed reflector components will not exceed the levels specified in Table 4. The most highly stressed components are those adjacent to the pebble fuel, which receive the highest fluence. These components will be designed to be replaced, where appropriate. Nonreplaceable graphite reflector components are exposed to only modest fluence levels and are expected to have lifetimes that substantially exceed the design life of the plant.

In the absence of irradiated properties data for the specific graphite in question, conservative estimates must account for uncertainties in the materials models used for design. This additional conservatism results in lifetime estimates for the most highly irradiated components that are less than those expected when materials-specific data are available. The design life estimates will be updated as materials test reactor data become available.

2. The operational life of the most highly irradiated graphite components will be further evaluated in service through Reliability and Integrity Management (RIM) Program.

Given the conservative basis upon which the initial design life is established and the inherent fault tolerance of the reflector design, the actual operational lifetime is expected to be substantially greater than that predicted. This will be evaluated during service through a combination of improved modeling and a RIM program. The latter is conceptually addressed in the following subsection.

## 3.3.7.2 RIM Program

The reliability of a nuclear plant and its systems and components is determined by the design, fabrication, inspection, surveillance, operation, and maintenance procedures used to build and operate the plant and its systems and components. Each of these aspects contributes in varying degrees to the reliability of the plant, its systems and components. In order for a nuclear plant to have a level of reliability that will satisfy both safety and economic goals, an appropriate combination of these contributors to reliability must be identified and implemented. The objective of the RIM program is to define, evaluate, and implement strategies to ensure that reliability targets for passive components are defined, achieved, and maintained throughout the plant lifetime. RIM programs relevant to graphite components will differ for prismatic and pebble concepts and, for the HTGR, are yet to be developed for either. However, some general observations can be made.

#### **Surveillance**

Given the passive nature of graphite structural components, operational surveillance can provide a significant amount of information regarding the ability of the graphite components to fulfill their functions, including those associated with safety. For example, the maintenance of core geometry may be inferred by normal and stable operation of the reactor and the associated heat transport system. The ability to reject heat via the passive pathway to the reactor cavity cooling system may be confirmed in part by observing heat transport to the reactor cavity cooling system during normal operation. In the case of the pebble bed reactor, the normal circulation of fuel provides additional insights.

The practice of including small samples of surveillance material in positions that see a higher flux than the actual component to be evaluated is common in the operation of LWRs. Such samples are

typically extracted during maintenance outages. With prismatic reactors, however, the effects of fluence can be confirmed by evaluating the fuel blocks and replaceable reflector elements that are periodically removed from the reactor. Through such means, there is a potential that the operating lifetime of these components can be extended.

## **Testing**

The principal active function supported by the graphite core structures is reactivity control. This is accomplished by providing unimpeded access for the insertion and withdrawal of the control rods (reactivity control system) and for insertion and removal of the small absorber spheres (reserve shutdown system). Functional testing of these systems will take place at appropriate times during normal production operation and during maintenance shutdown periods.

#### **In-service Inspection**

ISI requirements for graphite structures will be driven by accessibility during planned refueling or maintenance outages. This is considered to be acceptable given the conservative design bases and inherent fault tolerance of HTGR reflector designs, plus the above mentioned opportunities for surveillance and testing.

This is judged to be true of both pebble bed and prismatic block concepts prior to approaching the service life limit of the most highly irradiated components. For the prismatic block concept, accessibility to the graphite structures will be available during periodic refueling outages. For the pebble bed, as the initially predicted design life of the reflector is approached, the most highly irradiated reflector components adjacent to the pebble core would be inspected during planned maintenance outages. Should it be decided not to exchange the replaceable reflector components at that time, further regular inspections would be completed (typically during plant maintenance shutdown periods). If the reflector components are ultimately replaced, the life of the components could again be guaranteed and inspection would no longer be necessary until the service life limits of the replaced components is reached.

The inspection conducted to determine the need for replacement would need to address the following issues:

- The continuing ability of the reflector to maintain core geometry and to allow unimpeded insertion of the control and shutdown elements
- Changes in the graphite material properties relevant to the continued safe operation of the reactor during normal operation and LBEs.

The technical approach for undertaking a graphite ISI program under actual HTGR operating conditions remains to be assessed in a comprehensive way. Some possibilities are:

- Visual Inspection The possibility exists for visual inspection of exposed graphite reflector surfaces and/or reactivity control system penetrations during refueling or maintenance shutdown periods. For pebble bed design, a comprehensive inspection of the reflector surfaces adjacent to the core would require unloading of the fuel. Parameters assessed as part of such an inspection (subject to the technical limitations of the inspection techniques) may include identification of any cracks, abnormalities, or swelling in the graphite components, as well as determining changes in dimensions and/or surface conditions.
- Volumetric Inspection Volumetric inspection techniques have not yet been developed and qualified for graphite. Research on volumetric inspection techniques for graphite is ongoing at INL and other locations.

- In Situ Measurements It would be ideal to estimate the material properties from in situ measurements that could be completed during a maintenance shutdown, without defueling the core. Several alternatives for this type of measurement have been considered for use in reactors. Among alternatives are the following:
  - Micro hardness testing to determine the residual stress state of components. This approach is under development by JAEA for the HTTR graphite reflector blocks.
  - Detecting cracks by means of eddy current measurement. This technique has been applied
    extensively in the British advanced gas-cooled reactor for crack detection and monitoring with
    good success. The equipment and methods are available but would need to be adapted to pebblebed or prismatic type cores.
  - Trepanning material samples from the reflector adjacent to the core. Extensive trepanning has been undertaken in the United Kingdom on both Magnox and advanced gas-cooled reactor cores. The trepanning sampling strategy, extraction method, and analysis techniques for trepanned samples have been extensively developed in the United Kingdom. The extracted samples provide valuable real-time data on the core graphite condition in critical areas. Trepanning has been extensively used to monitor radiolytic oxidation in CO<sub>2</sub>-cooled cores.

#### Maintenance

Maintenance of graphite structures will be accomplished via planned replacement of the various components.

Given periodic replacement of the replaceable reflector components of the core during refueling outages, no additional planned maintenance activities are anticipated for the graphite structures of prismatic reactor concepts. Still, the designs of prismatic reactor concepts typically provide for the replacement of permanent graphite structures should unanticipated degradation occur.

In the case of pebble bed reactors, provisions will be made to replace the most highly irradiated graphite components when they are determined to have reached the limit of their service life. Based on present conservative estimates, this would occur once, at most, during the 60-year life of the HTGR plant. As is the case with prismatic designs, the possibility exists to replace other permanent graphite structures, should unanticipated degradation occur.

It is envisaged that the reflector components removed from the core during the replacement operation would be examined. The aim of this examination would be to evaluate the accuracy of analytical predictions, identify any failures that may have occurred, and, where possible, identify the cause of such failures or degradation.

#### 3.4 Ceramic Insulation Materials

Graphite, the principal material used in HTGR core structures, has a relatively high thermal conductivity. The high conductivity of graphite is advantageous in terms of transport of heat from the fuel to the helium coolant during normal operation and via the Reactor Cavity Cooling System during certain LBEs. In some applications, however, it is desirable or necessary to control the flow of heat from the graphite core structures to adjacent metallic components (e.g., core support structure) to avoid excessive temperatures. Ceramic insulation may be used in conjunction with the graphite core structures to achieve this objective. Two classes of ceramic insulation have been used in HTGRs to date, baked carbon and fused or sintered quartz. While quartz-based materials provide a greater degree of insulation, baked carbon is often utilized, where practical, based on economic considerations and the similarity of its properties (e.g., neutronic properties, coefficient of thermal expansion) to those of the adjacent graphite core structures.

Given the above, a discussion of baked carbon for use as core insulation is given in this section, which includes additional details regarding the manufacturing processes, properties, and prior uses of baked carbon. The approach for the design and structural evaluation is summarized, as well as the bases for qualification of insulation components.

## 3.4.1 Relevant Applications

A typical use of ceramic insulation in HTGR's is found in the reference PBMR design, in which baked carbon is being considered for use in the lowermost layers of the bottom reflector, below the core outlet plenum. The purpose of this insulation is to ensure that the service temperatures of the metallic Core Barrel Support Structure are maintained within allowable limits. The bottom reflector insulation layers extend across the full cross-section of the core structure. As noted above, baked carbon is preferred for use as an insulator in this area because it offers an economic alternative with attractive physical and mechanical properties that are similar to those of the reflector graphite structures. Baked carbon also presents advantages in terms of manufacturing. Much larger billets can be produced compared with other types of ceramic insulation, such as fused silica. Baked carbon is also more easily machined to the desired final configuration. It is also worth noting that baked carbon may be obtained from the same supplier as the reflector graphite, which offers specific advantages in terms of both procurement and fabricating experience.

For example, the current PBMR design utilizes SGL Carbon NBC-07 as the reference baked carbon material in conjunction with the NBG-18 graphite reflector components. Other baked carbon/core structure graphite combinations could be used, depending on specific reactor design considerations. However, since the insulation requirements of other HTGR reactor designs have not yet been identified, NBC-07 baked carbon will be used in this section as a basis for discussing the primary considerations in the use of ceramic insulation material.

# 3.4.2 Important Considerations

#### 3.4.2.1 Manufacturing

NBC-07 baked carbon is essentially identical to NBG-18 graphite in terms of raw materials and initial processing steps. It is manufactured from the same raw materials as NBG-18, utilizing an isotropic pitch coke filler and coal tar pitch binder and it is formed by vibration molding. The grain size of NBC-07 is identical to that used for NBG-18, thus qualifying NBC-07 as a medium-grain carbon. It is pitch impregnated and re-baked once, following a similar processing route as NBG-18 except for a higher baking temperature of 1100°C. The principal difference in processing is that the NBC-07 baked carbon is not graphitized.

## 3.4.2.2 Properties

Key properties of NBC-07 baked carbon insulation are summarized in Table 7 and compared with NBG-18 graphite and the target values for nuclear graphite. Also included in the table are the properties of the ASR-0RB baked carbon insulation used in the Japanese HTTR reactor (see Section 3.4.3 that follows). As can be seen from the table, the most significant difference between NBC-07 and NBG-18 is the higher thermal conductivity of the latter. NBC-07 also exhibits a somewhat higher compressive strength and elastic modulus, plus a CTE that is highly compatible with NBG-18.

Table 7. Properties of NBC-07 and ASR-0RB carbon insulation compared to the target value for nuclear

graphite.

Property	Unit	ASR-0RB Carbon	NBC-07 Carbon	NBG-18 Graphite	Target Value for Nuclear Graphite
Bulk Density	g/cm <sup>3</sup>	1.6	1.7	1.87	1.7–1.9
Coefficient of Thermal Expansion	$\times 10^{-6}$ /°K	4.4	4.6-4.8	4.5-4.6	3.5-5.5
Thermal Conductivity	W.m <sup>-1</sup> .K <sup>-1</sup>	10	4.9-5.0	140–145	>100
Tensile Strength	MPa	17.8	15	20	>15
Compressive Strength	MPa	50.4	138.5	77–78	>50
Elastic Modulus	GPa	8.7	15.7	12	8-15
Ash Content	ppm	5000 max.	4100 max.	< 300 avg.	< 300 avg.

# 3.4.3 Related Experience

Baked carbon insulation has been previously used in HTGRs, including the HTTR and HTR-10.8,9

In the HTTR, blocks of nuclear grade ASR-0RB carbon are used to insulate the metallic core support structures from excessive heat that would otherwise flow from the bottom of the graphite core structures. These insulating blocks were required to keep these metallic structures below 500°C.

The HTR-10 reactor, which bases its design strongly on the German HTR-Module concept, also uses carbon thermal insulation in the core to protect metallic components. Carbon block insulation is used at the top, bottom and around the core reflector graphite blocks. The bottom carbon insulation blocks are composed of three layers, which support the bottom and side reflector graphite blocks. Shaped carbon blocks are used at various locations in the core structures to insulate selected components from hot gas flow. The carbon blocks around the reflector, at the top of the reflector and the upper insulation layer below the reflector have 5 wt% boron carbide added to the carbon to reduce neutron irradiation to the adjacent metallic components in these areas. The carbon insulation blocks were produced by the Lanzhou Carbon Works to ensure low thermal conductivity and good dimensional stability at high temperature.

It is also worth noting that baked carbon was employed in past German pebble-bed designs such as the AVR and THTR-300, and was intended as bottom reflector ceramic insulation for the HTR-Module and HTR-500 concept designs. For the HTR-Module concept design, Grade AK4 baked carbon manufactured by the SIGRI Company (predecessor to SGL Carbon) was the identified candidate material. This baked carbon had a nominal, indicated bulk density of just over 1.5 g.cm-3, CTE of 3.5 × 10-6°C-1 and thermal conductivity of about 3.8 W.m-1.K-1. In addition, HTR-Module design documents indicate that a 5 wt% boron carbide surrogate of the AK4 baked carbon was under consideration for shielding purposes in lower temperature areas. Boron carbide pin inserts in graphite core components were identified as the most likely design option in higher temperature core locations that were identified as needing neutronic shielding.

## 3.4.4 Approach to Design and Qualification

As is the case with the graphite and carbon fiber reinforced carbon (CFRC) components discussed in other sections, there are at present no established industrial standards for the design and analysis of ceramic insulation components in nuclear applications. For example, PBMR selected the requirements for the design and manufacture of the ceramic internals (see Appendix A) as the most suitable basis for

ceramic insulation design and for assessments of structural reliability. The applicable methods are similar to those used to characterize graphite. In this regard, the following are noted:

- A Structural Reliability Class (in this case SRC-3) is assigned to the ceramic insulation components. As with graphite, the SRC is related to the component functions.
- Based upon the assigned SRC, the allowable failure probabilities are  $1 \times 10^{-2}$  for Load Category A and  $5 \times 10^{-2}$  for Load Category B.
- The Weibull distribution functions of the component-specific strength values are experimentally determined.

Structural loads imposed on the ceramic insulation are determined by analysis. During normal operation and for anticipated operational occurrences, such loads primarily relate to compressive stresses associated with the weight of core structure ceramic (CSC) components above. For DBEs, Operating Basis Earthquake loads must also be addressed. Thermally-induced stresses associated with temperature gradients and transient thermal loadings will be modest in the ceramic insulation, based on both the material properties and the specific application.

The temperatures seen by baked carbon insulation during normal operation are limited to 600°C. The peak temperature that could be seen during transients is approximately 800°C. At these temperatures, physical properties changes (notably thermal conductivity) are expected to be negligible.

The use of ceramic insulation in the CSC is restricted to areas where it is not exposed to significant fast neutron irradiation (e.g., <1018 n.cm-2 EDN) and, consequently, irradiation-induced changes in properties will be negligible. The baked carbon insulation will be designed for the full HTGR plant life of 60 years.

# 3.4.5 Material Qualification Plan

An MQP has been developed for NBC-07 baked carbon. The objectives of the MQP are to characterize the properties of the insulation, to confirm its suitability for use in the HTGR and to demonstrate compliance with requirements established for the insulation components. The properties to be developed in accordance with the MQP for NBG-07 baked carbon insulation are summarized in Table 8.

Table 8. Characterization of baked carbon insulation.

Property	Units	Test Method	Conditions
Bulk Density	kg.m <sup>-3</sup>	ASTM C559	RT
Mean Coefficient of Thermal Expansion	$\times 10^{-6}  \mathrm{K}^{-1}$	DIN 51 909	20-800°C
Isotropy Ratio	_	DIN 51 937	_
Electrical Resistivity	μOhm.m	DIN 51 911	RT
	xx -1 xz-1	DIN 51 908	RT
Thermal Conductivity	W.m <sup>-1</sup> .K <sup>-1</sup>	DIN 51 936	20-1000°C
Specific Heat Capacity	J.kg <sup>-1</sup> .K <sup>-1</sup>	_	20-1000°C
Emissivity (Total, Normal)	_	ASTM E307	20-800°C
Water Absorption	_	ASTM C20	RT
Towards Character	MD-	DIN 51 914	RT
Tensile Strength	MPa	_	Elevated Temperature
Tensile Weibull Modulus	_	As per RDMCI	RT

Property	Units	Test Method	Conditions
Tensile Weibull Characteristic Strength	MPa	As per RDMCI	RT
Communication Street	MD-	ASTM C695	RT
Compressive Strength	MPa	_	Elevated Temperature
Elavural Strangth (4 naint)	MDo	DIN 51 944	RT
Flexural Strength (4-point)	MPa	_	Elevated Temperature
Dynamia Elastia Madulus	GPa	ASTM C747	RT
Dynamic Elastic Modulus	Gra	_	Elevated Temperature
Static Elastic Modulus	GPa	ASTM C749	RT
Poisson's Potio (v)		_	All Temperatures
Poisson's Ratio (v)		_	Elevated Temperature
Friction Coefficient		ASTM C808,	RT-900°C (Helium)
(Carbon-Graphite)	_	ASTM G115	
Chemical Analysis	ppm	DIN 51 096	As per test method
Ash Content	ppm	DIN 51 903	As per test method
Equivalent Boron Content (EBC)	ppm	PBMR Procedure	N/A
Open Porosity	%	DIN 51 918	RT
Pore Size Distribution	_	DIN 66 133	_
Air Permeability	Darcy	DIN 51 935	_
Air Reactivity	ug.g <sup>-1</sup> .h <sup>-1</sup>	SGL Procedure	400°C, 24h
BET Surface Area	$m^2.g^{-1}$	DIN ISO 9277	Elevated Temperature

# 3.5 Composite Materials

Ceramic composites represent a promising class of candidate materials for HTGRs, because of their unique properties in a high temperature helium environment. The most important of these properties are their high strength and toughness, resistance to very high temperature, low coefficient of thermal expansion and excellent resistance to thermal creep. For the near-term, CFRC is considered a potentially attractive, but not essential, alternative to metallic materials in low-fluence regions. Among the available classes of ceramic composites, CFRCs are the most technically mature, can be deployed more readily in the near-term and at lower cost. Other ceramic composites (C-SiC and SiC-SiC) have been suggested for advanced HTGRs applications, particularly for components involving higher temperatures and/or fluences (e.g., reactivity control components).

This section begins with an overview of potential CFRC applications in Section 3.5.1. This is followed in Section 3.5.2 by a discussion of important considerations related to the selection, manufacture and operational conditions associated with CFRC components in these applications. The historical basis and related experience for the use of CFRC in HTGR applications are described in Section 3.5.3. An overview of the approach to CFRC component design and qualification is provided in Section 3.5.4.

Additionally, all but the simplest structures of composite materials will have to be qualified through test of actual production articles (and segments extracted from actual production articles). The basic structure of the composite materials use in the components is non-homogeneous and grossly anisotropic.

# 3.5.1 Relevant Applications

In principle, CFRC components can fulfill a broad range of requirements in HTGR environments with the proviso that their application is limited to specific conditions. Typically, these would be applications where the maximum fluence is limited to a few dpa and the components are not likely to suffer significant oxidation damage during normal operation or LBEs, particularly if the components of interest are safety related. Further discrimination can be made with respect to the maximum fluence as it determines whether the material can be regarded as effectively unirradiated. Therefore, for a maximum limiting fluence, there would be a need for further irradiated properties data.

The following provides a typical examples of HTGR applications for which CFRC materials are being considered:

- Top reflector supports (pebble bed)
- Upper plenum insulation supports (prismatic)
- Upper core restraint devices (prismatic)
- Core lateral restraints
- RSS channel interface tubes
- Core outlet connection nozzle (between core outlet plenum and internal hot gas duct)
- Control rod components (advanced application involving high fluence).

To date, there has been limited development of CFRC components for the above applications. Early development work addressed the upper core restraints and control rod components for prismatic designs, including the manufacture of prototype control rod components at ORNL. More recently, PBMR has undertaken development of top reflector supports (Tie Rods) and core lateral restraint components (Racetrack Straps) for the DPP.

## 3.5.2 Important Considerations

This section identifies the characteristic properties of CFRC materials that could lead to their selection in HTGR components. This is followed by a discussion of materials processing and operational considerations

#### 3.5.2.1 Characteristic Properties of CFRCs

Characteristic properties of CFRCs that should be considered when evaluating their use in HTGR components include the following:

- Heat resistance in an inert atmosphere to temperatures in excess of 2000°C
- High specific strength and rigidity
- Low density and low thermal expansion
- Extremely high resistance to thermal shock
- Good to excellent electrical conductivity and thermal conductivity
- Anisotropy: in materials with aligned carbon fibers, the flexural and tensile strength and electrical and thermal conductivity have different values for orientations parallel and perpendicular to the fiber orientation
- Excellent fatigue resistance, even at high temperatures

- Excellent resistance to thermal creep at temperatures up to 1600°C
- Pseudoplastic fracture behavior
- Relative chemical inertness
- Moderate resistance to fast neutron irradiation damage
- Production of high purity grades is possible.

# 3.5.2.2 Manufacturing and Processing Considerations

CFRCs are comprised of two components- highly ordered carbon fibers and a carbon matrix. They are most commonly made by gradually building up a carbon matrix on a fiber preform through a series of impregnation and pyrolysis steps. Although more expensive than graphites, CFRCs are considerably stronger and tougher than graphites and retain many of the desirable attributes of graphite, including excellent machinability, high thermal conductivity and low thermal expansion. CFRC materials are typically described as being unidirectional (1-D), two-directional (2-D), or three-directional (3-D). This indicates the number of fiber bundle directions that the composite possesses. In 2-D CFRCs, the fibers, in the form of multi-filament tows, are woven into a cloth or, alternatively, carbon filaments may be sprayed from a spinneret to form a felt or mat. The woven cloth is then layered to form the desired thickness. Tubular forms are also considered to be 2-D components in most cases. In a 3-D CFRC, the fiber bundles are usually orthogonal.

The manufacture of carbon based composites begins with the production of the carbon fibers, which at the crystal structure level is comprised of an array of graphite crystallites with their layered crystal structure preferentially aligned with the fiber axis. The fibers are, therefore, highly anisotropic and, for example, may have an elastic modulus in the fiber direction (a-axis) that is greater than 100 times the modulus perpendicular to the fiber (c-axis). For commercial high performance CFRC, the fiber precursor material is generally either polyacrylonitrile (PAN) or mesophase pitch. PAN-based carbon fibers are far more resistant to compressive failure than their pitch-based counterparts. They are used predominantly in high strength, high temperature applications and represent ~90% of the total carbon fiber production. However, the PAN-based carbon fibers do not achieve tensile modulus and thermal conductivity values

comparable to those of fibers produced from mesophase pitch, and the latter are used where those properties are important.  $^{10}$ 

The processing of carbon fibers, whether from PAN- or pitch-based precursors, is quite similar. Production of both of the fiber types involves spinning, oxidative stabilization in air at 200 to 400°C, and high temperature carbonization and graphitization. Important steps in fiber processing are thermal stabilization at 200-400°C under tension to preserve the fiber molecular structure generated during drawing and carbonization at 1000-1500°C in an inert atmosphere to convert the polymer chain structure of the fibers into bundles of linked graphite crystallites. Additional heat treatment reduces strength but dramatically raises tensile modulus, which is important for applications demanding high rigidity. Interestingly, PAN based carbon fibers develop a fibrillar microstructure (Figure 4), which contains regions of undulating ribbons. This structure is much more resistant to premature tensile failure resulting from microscopic flaws than microstructures with extended

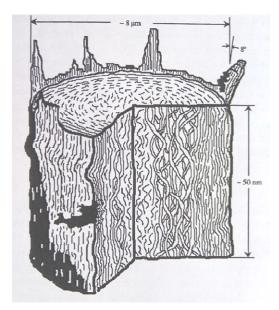


Figure 4. Illustration of the fibrillar texture of a carbonized PAN fiber.

graphitic regions transverse to the fiber axis, such as those seen in mesophase pitch-based carbon fibers. For this reason, PAN-based fibers tend to develop exceptional tensile strengths. The leading PAN-based fiber, T300, is widely used in the aerospace industry; primary suppliers are Toray and Cytec. A potential issue with these fibers is lot-to-lot variation in properties (as much as 15%).

Pitch-based carbon fibers are unique in that they can develop extended graphitic crystallinity during carbonization/graphitization. The mesophase pitches used for the production of high-modulus fibers are most commonly formed by the thermal polymerization of petroleum or coal tar-based pitches. Petroleum-based pitches now dominate because of environmental concerns relative to the coal-tar pitches. The petroleum pitch is commonly formed as a by-product during cracking of the heavy oil fraction of crude oil. A mesophase pitch is produced by heating the pitch in an inert atmosphere for an extended period of time at 400 to 550°C. The procedures used in the manufacture of carbon fibers from mesophase pitch are quite similar to those discussed in the paragraph above for PAN-based fibers. The four basic steps are melt-spinning, oxidative stabilization, carbonization and graphitization. After carbonization, the fibers are heated to the graphitization temperature range (>1700°C) that is required to develop their high strength, high modulus properties. During graphitization, dislocations are removed from the carbon structure of the fibers and this eventually results in the formation of a three-dimensional graphite lattice.

The second phase in CFRC production is the creation of a carbon matrix around the fibers. The two most common methods of accomplishing this are chemical vapor deposition and vacuum or pressure impregnation with resin or pitch. The usual commercial method for the production of CFRC items is resin or pitch-impregnation of preforms that are built up from carbon fiber textiles (woven or non-woven) or yarns. Resin can be injected at temperatures as low as 65°C; pitch is typically injected at 300°C. A commonly used petroleum pitch is A-240; phenolic resins such as 91LD and SC1008 are aerospace qualified and readily available, but offer no advantage over pitches except in a few special applications involving processing of complicated shapes.

In the chemical vapor deposition method, the preform (usually formed from several layers of woven carbon fabric in the desired final shape) is heated in a furnace pressurized with an organic gas such as methane or acetylene. The gas decomposes under the high temperature and pressure conditions existing in the furnace and deposits layers of carbon onto the preform. The gas must diffuse through the entire preform to make a uniform matrix; therefore, the process is very slow. It usually requires several weeks and numerous processing steps to make a single part and is, thus, expensive.

A typical production sequence involving impregnation begins with an initial impregnation under vacuum and is followed by carbonization of the impregnated part at 650 to 1000°C to convert the organic matrix to an amorphous carbon material. Carbon yield is ~50% of the volume of pitch or PAN resin injected. Subsequent impregnations and carbonizations may be done at high pressure to improve the carbon yield in each cycle to ~85%. Each impregnation and carbonization cycle is followed by a graphitization treatment at 2000 to 2800°C. The number of cycles required to produce high density material is typically three to five. As a general rule, the thermal and mechanical properties of the material are found to improve with density; however, there is a trade-off between the properties obtained and the increased cost of multiple impregnation cycles. The final graphitization may be done at temperatures in the lower end of the graphitization temperature range to minimize cracking in the matrix and at the fibermatrix interface. A typical CFRC will contain a volume fraction of fibers of from 40 to 50%. Additionally, the material will contain between 35 and 40% impregnant derived matrix graphite. The remainder of the composite volume is porosity distributed in the matrix and at the fiber/matrix interface.

The CFRC can be purified by means of additional halogen treatment at high temperature, typically above 2000°C. This ensures that the oxidation catalyzing impurities are minimized and that the oxidation resistance of the CFRCs is suitable for the HTGR application.

# 3.5.2.3 Properties

The material properties of CFRC are strongly influenced by the fiber fraction, fiber type employed, fiber dimensions, fiber lay-up orientation and/or textile weave type (architecture), matrix material type, the individual properties of the fiber and matrix, details of the manufacturing process and the graphitization heat treatment temperature.

Fiber properties depend on the precursor material (PAN or Pitch), production processes, including the tensioning step, and the degree of graphitization. The matrix precursor material and its manufacturing method also influence the properties of the finished composite. Although this may at first seem to present an overwhelming and confusing number of possibilities, it also allows the opportunity to select and tailor materials and processes to achieve a CFRC with physical and/or mechanical properties optimized for the intended application.

Some typical physical and mechanical values for CFRC materials are shown in Table 9 in relation to properties of "isotropic" graphite.

Table 9. Typical properties of graphite and CFRC materials at room temperature.

Table 9. Typical properties of graphic and CTRC materials at room temperature.				
Property	1-D CFRC (parallel to fibers)	3-D CFRC	Fine Grained Isotropic Graphite	
Density [g/cm <sup>3</sup> ]	1.7–1.8	1.7–1.8	1.75–1.85	
Thermal Conductivity [W/m·K]	400–600	100–200	90–200	
Coefficient of Thermal Expansion [10 <sup>-6</sup> /K]	0.1–2.0	0.1–0.2	2–5	
Young's Modulus [GPa]	150–250	75–125	10–15	
Bending Strength [MPa]	50–150	Values not available	40–70	
Tensile Strength [MPa]	300–900	150-400	40–60	
Compressive Strength [MPa]	200–500	100–200	100–200	
Fracture Toughness [MPa·m <sup>1/2</sup> ]	2–3	4–6	<1	

The densities here are typical of CFRC materials with multiple (3 to 5) densification cycles; some commercial products with single densification cycles have densities as low as 1.35 g/cm<sup>3</sup>. Note that the thermal conductivity values given for the isotropic graphite and the 3–D CFRC are essentially independent of orientation (the measurement direction). Values shown for the 1-D CFRC are for the direction parallel to the fibers; thermal conductivity in directions perpendicular to the fiber axis can be up to 25 times smaller.

Similar considerations/trends apply to the other properties listed for 1-D materials, except for the coefficient of thermal expansion, as noted below. However, the reduction factors for the perpendicular versus the parallel direction are typically much smaller than for thermal conductivity. With regard to the exception, the coefficient of thermal expansion in directions perpendicular to the fiber axis is 4 to 10 times larger than that parallel to the fiber axis.

In the context of the above, the following observations are important to the application of advanced, high-temperature CFRC materials in HTGR applications:

- Tensile and compressive strength and modulus values of CFRC are superior to those of graphite and increase over the entire range of temperatures of relevance for gas-cooled reactors.
- Both fracture toughness and impact resistance of CFRC are better than for graphite.

- Thermal conductivity is high but very sensitive to CFRC architecture, heat treatment and fiber properties.
- Thermal expansion is essentially zero over a wide range of temperature.
- Thermal shock poses little, if any, problem because of the good thermal conductivity and low thermal expansion.

Thermal creep does not occur at even the highest temperatures of interest for gas-cooled reactors; however, irradiation creep may occur in high fluence environments (not applicable for present HTGR CFRC components).

# 3.5.2.4 Operational Considerations

For the near-term application at 700 to 750°C ROT, the use of CFRC would be limited to areas where it is not exposed to significant fast neutron irradiation and, consequently, irradiation-induced changes in properties will be negligible. Irradiation-induced mechanical property changes at fluences below  $2 \times 10^{20}$  n/cm<sup>2</sup> EDN for an irradiation temperature of between 400 and 600°C are expected to be enveloped by the values given in Table 10.

Table 10. Irradiation induced property changes for CFRC.

	1 1 7	2	
Property	Unit	Value	
Dimensional Change		<0.1%	
Tensile Strength	MPa	+3 to >10%	
Young's Modulus	GPa	+5 to >10%	
CTE	× 10 <sup>-6</sup> °C <sup>-1</sup>	+3 to >10%	
K <sub>IC</sub>	MPa m 1/2	No detrimental change	
Thermal Conductivity	W m <sup>-1</sup> K <sup>-1</sup>	-25%	

For higher fluence exposure, irradiation testing would be required to ascertain the magnitude of irradiation-induced changes in these parameters from both a design and operational perspective.

The potential for oxidation of CFRC components must be considered for the operating conditions and LBEs within the design basis. Relevant parameters are the oxidation resistance of the material, the impurity levels in the helium coolant and the temperatures seen by the components in service. Regarding the first, the CFRC grades used in HTGRs would typically be subjected to a halogen purification step at high temperature. This would ensure that the level of impurities available to catalyze an oxidation reaction is limited and imparts better oxidation resistance to the CFRCs for application within the reactor. Given the typical specifications for the helium coolant in HTGRs and the nature and locations of the components identified in Section 3.5.1, oxidation effects within the design basis are not expected to be significant.

## 3.5.3 Related Experience

CFRC materials are widely used in consumer and industrial products. Of particular note is their use in aerospace applications, including critical structural components, such as aircraft braking systems, pressurized cabin structures, wings and other airfoils. Despite their wide commercial application, there has been no experience with CFRC materials in an operational nuclear power plant, including HTGRs.

Although operating experience with CFRC materials in HTGR environment is non-existent, it is worth noting that CFRC materials were under investigation in the German HTR development program for

specific applications. The extent of research and testing achieved at the time was limited but points to the potential of CFRC materials under consideration.

# 3.5.4 Approach to Design and Qualification

## 3.5.4.1 Design

As is the case with graphite, there are at present no established industrial standards for the design and analysis of CFRC components in nuclear applications. For example, the RDMCI document was developed as the basis for CFRC design and for assessments of structural reliability. In this regard, the following are noted:

- An SRC is assigned to each of the respective components. As with graphite, the SRC is related to the component functions
- Based upon the assigned SRC, the allowable failure probability is determined
- The Weibull distribution functions of the component-specific strength values are experimentally determined and the corresponding allowable stress values are calculated
- Based on the above, load limits corresponding to the allowable probability of failure are developed for the component in question

The RDMCI was developed for graphite and carbon parts and is based on the draft German design code for HTR ceramic core internals (KTA-3232). It does not specifically address the design requirements for core internals made from ceramic matrix composite materials such as CFRC. The design basis for CFRC materials is in the form of a set of extended rules to the RDMCI. This extended set of rules recognizes the unique properties and, hence, design requirements pertaining to composite materials for HTGRs. The extended rules relative to the RDMCI include consideration of oxidation and fluence effects; however, oxidation and fluence effects would be minimal for the conditions seen by CFRCs within the design basis of the NGNP.

#### 3.5.4.2 Qualification

As an example, two different CFRC grades were selected for the Tie Rods (top reflector support) and Racetrack Straps (core restraints). The main difference in these CFRC materials is the fiber architecture, which is tailored to the specific application. These CFRC materials were selected in lieu of metallic alternatives on the basis of their high temperature strength and creep resistance, as well as their low thermal expansion properties.

For both the tie rods and racetrack straps, extensive component level tests were performed. Due to the specific geometry and loading configuration of these parts, it was necessary to determine the component failure loads under representative loading conditions. This could not be satisfied through material specimen tests. Both subscale and full-scale Tie Rod and Racetrack Strap components were tested on a preliminary basis to establish component failure data. However, further work is needed to obtain statistically significant data.

In addition to component failure load tests, both Tie Rod and Racetrack Strap cyclic loading (fatigue) tests were conducted on a preliminary basis to establish safe-cycling load limits for these components in service. Sub-scale components were cycled at varying load ranges exceeding 80% of the mean failure load for these components and the number of cycles to failure recorded. Given their low-cycle fatigue application in the DPP, the design working load was set at the load at which no failure occurred after 2000 cycles. Additional work is required to improve on the statistics of the available fatigue data and full-scale cyclic loading tests were also planned.

Also, where material testing cannot satisfy component design requirements or provide the requisite design data, component tests must be conducted.

# 3.5.4.3 Status of Codes for Composite Materials

### ASME

At present, there are no established design codes or standards addressing composite materials for HTGR applications that are analogous to the ASME Code for metallic components. However, there is a plan within ASME to address this shortcoming through the ASME Subgroup on Graphite Core Components (SGCC). This subgroup has been officially sanctioned by the Board on Nuclear Codes and Standards as part of the B&PV Code Section III infrastructure. The SGCC has concentrated its efforts to date on nuclear graphite (see Section 3.3.4); however, high temperature composites are also a part of the subgroup charter. The ASME Code section on nuclear graphite will be issued in 2010 and the expectation is that the composites code will be addressed thereafter.

- Establishing an ASME Code framework for the use of CFRC and/or SiC/SiC composites in a HTGR will require at a minimum the development of:
- Design codes, which list "rules" and guidelines for employing composite materials and incorporating them into component designs
- Design codes which regulate the certification procedures for processing materials, fabricating components, and assembling final designs
- Rules for testing material and components, as appropriate, in a manner that will produce valid and statistically significant data to support the design.

### **ASTM**

- ASTM test standards for ceramic matrix composites are developed through ASTM Subcommittee C28.07 on Ceramic Matrix Composites. Presently available standards are:
- C1275-00 (2005) e1 Standard Test Method for Monotonic Tensile Behavior of Continuous Fiber-Reinforced Advanced Ceramics with Solid Rectangular Cross-Section Test Specimens at Ambient Temperature
- C1292-00(2005) Standard Test Method for Shear Strength of Continuous Fiber-Reinforced Advanced Ceramics at Ambient Temperatures
- C1337-96(2005) Standard Test Method for Creep and Creep Rupture of Continuous Fiber-Reinforced Ceramic Composites under Tensile Loading at Elevated Temperatures
- C1341-00(2005) Standard Test Method for Flexural Properties of Continuous Fiber-Reinforced Advanced Ceramic Composites
- C1358-05 Standard Test Method for Monotonic Compressive Strength Testing of Continuous Fiber-Reinforced Advanced Ceramics with Solid Rectangular Cross-Section Test Specimens at Ambient Temperatures
- C1359-05 Standard Test Method for Monotonic Tensile Strength Testing of Continuous Fiber-Reinforced Advanced Ceramics with Solid Rectangular Cross-Section Test Specimens at Elevated Temperatures
- C1360-01 Standard Practice for Constant-Amplitude, Axial, Tension-Tension Cyclic Fatigue of Continuous Fiber-Reinforced Advanced Ceramics at Ambient Temperatures

- C1425-05 Standard Test Method for Inter-laminar Shear Strength of 1-D and 2-D Continuous Fiber-Reinforced Advanced Ceramics at Elevated Temperatures
- C1468-00 Standard Test Method for Trans-thickness Tensile Strength of Continuous Fiber-Reinforced Advanced Ceramics at Ambient Temperature
- C1469-00(2005) Standard Test Method for Shear Strength of Joints of Advanced Ceramics at Ambient Temperature
- C1557-03 e1 Standard Test Method for Tensile Strength and Young's Modulus of Fibers.

### **Other Standards**

Limited standardization of composite materials has been addressed in the following full-consensus standards organizations:

- Comité Européen de Normalisation (CEN) Subcommittee TC184/SC1 on Ceramic Composites
- International Organization for Standardization Technical Committee TC206 on Fine (Advanced, Technical) Ceramics.

In addition, other noteworthy standards for CFRCs have been developed by the Department of Defense and NASA High Speed Research/Enabling Propulsion Program in the United States and the Petroleum Energy Center (PEC) in Japan.

The Department of Defense Handbook on Composite Materials consists of five volumes. Volumes 1 through 3 cover polymer matrix composites, Volume 4 covers metal matrix composites and Volume 5 (which includes composite materials discussed in this report) covers ceramic matrix composites. It appears that the information in Volume 5 will directly support ASME codification activities. Volume 5 is organized into four parts:

- Part A Introduction and Guidelines
- Part B Design and Supportability
- Part C Testing
- Part D Data Requirements and Data Sets.

### 4. MATERIALS ISSUES FOR CONSIDERATION

Given the existing regulatory foundation summarized in Section 2, and the technical status of prospective materials for the NGNP described in Section 3, this section summarizes materials-related issues that need to be addressed in the course of licensing an NGNP. Issues pertaining to metallic materials are summarized in Section 4.1, while issues pertaining to nonmetallic materials are summarized in Section 4.2. In some cases the issues discussed in this section are presently being addressed within the NGNP design or development programs or in related activities, such as the ASME/DOE cooperative agreement for code development in support of HTGRs, and this section summarizes the present status. In other cases, a specific response is requested from the NRC. For the latter, the specific questions presented for NRC response are found in Section 5 of this report.

## 4.1 Metallic Materials

# 4.1.1 Negligible Creep Limits for Extended Lifetime

The design life proposed for the HTGR plant is 60 years, which is significantly longer than the initial design life of commercial LWR plants now in operation. The current negligible creep temperature limit for SA508/SA533 low alloy steel is specified to be 371°C (700°F) in the ASME Code for Section III components. The 371°C (700°F) limit was established over 30 years ago, using existing data for 300,000 hours that were employed for the design of LWRs, which operate closer to 315°C (600°F). Since HTGR vessels are expected to operate at higher temperatures, and for longer times, the creep database were revaluated to determine if the codified limit was sufficiently conservative. These preliminary assessments using extrapolation of data to 600,000 hours indicate that the negligible creep limit for SA-508/SA-533 is closer to 350°C (662°F) for a 500,000 to 600,000 hour life. <sup>11</sup>

If the negligible creep limit is reduced to 350°C (662°F) repercussions might include modifying the maximum temperature limit for this material in Section III, Subsection NB of the ASME Code and the corresponding requirements of ASME Code Case N-499-2. Both of these actions could affect the HTGR design, so evaluation of this temperature limit will be taken into consideration during the HTGR design.

# 4.1.2 Application of Code Case N-499-2 in RPV Design

In present NGNP concepts, which are expected to have reactor outlet temperatures in the range of 750 to 800°C (1382 to 1472°F), the primary helium pressure boundary vessels, including the Reactor Vessel, will employ conventional metallic materials currently approved for nuclear service within Section III of the ASME B&PV Code. In the case of the reactor vessel, the material of choice is expected to be SA-508/SA-533 low-alloy steel. With the exception of the reactor vessel, the components would operate within the negligible creep range at all times and for all events within the plant lifetime. The reactor vessel would also be designed to operate in the region of negligible creep during normal operation and for anticipated operational occurrences that are expected to occur within the lifetime of a given plant. However, there is a potential that the temperature limits of Section III, Subsection NB would be exceeded for short periods of time during low frequency design basis events involving conduction cooldown. For such events, it is proposed to apply ASME Code Case N-499-2, which provides for short-term operation at temperatures up to 538°C (1000°F).

### 4.1.3 Application of Metallic Materials at Elevated Temperatures

Metallic materials seen as potential candidates for elevated temperature components (vessels, support structures, hot duct liners, etc.) were identified and discussed in Section 3.2. At present, the material candidates include SA508/SA533 low-alloy steel, Alloy 800H, Alloy X, Alloy 617, Modified 9Cr-1Mo

steel, 2.25Cr-1Mo steel, and Type 316 SST. Most of these materials are candidates for more than one high-temperature component and no final material/component matches have been made. All of the alloys above are mature in terms of database and industrial experience.

Table 11 shows the status of the development of ASME B&PV codes for the materials of interest. It can be seen that all of the potential metallic materials, with the exception of Alloy 617, have some degree of ASME Section III approval for nuclear service (Subsections NB, NC and/or ND). Additionally, all of the materials except Alloy X have provisions for nuclear service at temperatures where allowable stresses can be time-dependent (Subsection NB with Code Case N-499-2 for SA508/SA533, Subsection NG with Code Case N-201-5, and Subsection NH for the others). However, only the use of SA508/SA533 in accordance with Subsection NB, NC, and ND applications is currently fully accepted by the NRC.

Table 11. Current ASME code limits for potential HTGR materials.

Alloy	Applicable ASME Code Section	Prescribed Limits
SA-508/SA-533	Section III, Subsection NB, NC, ND	371°C (700°F)
SA-508/SA-533	Section III, Subsection NB Code Case N-499-2	371°C (700°F) to 427°C (800°F) for 3,000 hours (Level B)
		427°C (800°F) to 538°C (1000°F) for 1,000 hours (Level C or D)
		Maximum of 3 events over 427°C
SA-508/SA-533	Section VIII, Division 1	427°C (800°F)
316 SS	Section III, Subsection NH	816°C (1500°F)
316 SS	Section III, Subsection NB, NC, ND	427°C (800°F)
316 SS	Section III, Subsection NG with Code Case N-201-5	816°C (1500°F)
Alloy 800H	Section III, Subsection NB, NC, ND	427°C (800°F)
Alloy 800H	Section III, Subsection NG with Code Case N-201-5	760°C (1400°F)
Alloy 800H	Section III, Subsection NH	760°C (1400°F)
Alloy 800H	Section VIII, Division 1	899°C (1650°F)
Alloy X	Section III, Subsection NB, NC, ND	427°C (800°F)
Alloy X	Section VIII, Division 1	899°C (1650°F)
Alloy 617	Section VIII, Division 1	899°C (1650°F)
Modified 9Cr-1Mo	Section III, Subsection NB, NC, ND	371°C (700°F)
Modified 9Cr-1Mo	Section III, Subsection NH	649°C (1200°F)
Modified 9Cr-1Mo	Section VIII, Division 1	649°C (1200°F)
2 <sup>1</sup> / <sub>4</sub> Cr–1 Mo, Grade 22	Section III, Subsection NB, NC, ND	371°C (700°F)
2 <sup>1</sup> / <sub>4</sub> Cr–1 Mo, Grade 22	Section III, Subsection NG with Code Case N-201-5	593°C (1100°F)
2 <sup>1</sup> / <sub>4</sub> Cr–1 Mo, Grade 22	Section III, Subsection NH	593°C (1100°F)
2 <sup>1</sup> / <sub>4</sub> Cr–1 Mo, Grade 22	Section VIII, Division 1	649°C (1200°F)

The prescribed limits shown in the table do not in all cases and for all alloys sufficiently cover the temperatures and times associated with the elevated temperature components for which they are proposed. In an attempt to remedy this deficiency, the following ASME-based activities are in progress or being proposed:

- Reevaluation of the ASME Section III negligible creep temperature limits (currently 371°C (1000°F) based on 300,000-hour service) for SA508/SA533 low-alloy steel. This re-evaluation is needed because of the possibility that service for 500,000 hours may result in the need to impose a lower negligible creep temperature limit (see Section 4.1.1 above).
- Similarly, the 427°C (800°F) negligible creep temperature limits for Alloy 800H, Type 304 and 316 SST, and Alloy X will be revisited in light of the projected 60-year service life of the HTGR. The negligible creep temperature limit of 371°C (1000°F) for Modified 9Cr-1Mo and 2.25Cr-1Mo will also be reevaluated based on the longer expected service life.
- Alloy 800H is qualified under Section III, Subsections NH and NG and Code Case N-201-5 for operation at temperatures to 760°C (1400°F). All of these need to be extended to temperatures >800°C (1472°F), preferably up to 950°C (1742°F), to cover operating, off normal, and accident conditions for various high-temperature components. German Standard KTA 3221 allows use of Alloy 800H up to 1000°C (1832°F).
- Efforts are needed to include Alloy X and Alloy 617 in ASME Section III, Subsections NH and NG and Code Case N-201-5 to at least 900°C (1652°F).
- Efforts are underway to include Modified 9Cr-1Mo in ASME Section III, Subsection NG and Code Case N-201-5 at temperatures up to 650°C (1202°F).
- Finally, consideration is now being given on how to incorporate the effects of chemical environment, thermal aging and irradiation on properties into the various ASME codes, particularly those that permit operation at high temperatures.

In the present designs, the use of metallic materials at elevated temperatures beyond the time-independent limits of ASME Section III is limited to components within, but excluding, the primary helium pressure boundary. For those components, which exceed the time-independent limits, NRC acceptance of the following will be required:

- Code Case N-201-5 This is an ASME-approved addition/modification to Section III, Subsection NG that provides for the design and construction of core support structures for temperatures above 371°C (1000°F) and up to 816°C (1500°F), depending on material, during both normal operation and duty cycle events. The scope of N-201-5 currently includes Type 316 Stainless Steel, 2.25Cr-1Mo steel, and Alloy 800H.
- 2. Section III, Subsection NH This is an ASME-approved code for elevated temperature service of metals (Alloy 800H, Types 304 and 316 SST, Modified 9Cr-1Mo, and 2.25Cr-1Mo) for Class 1 components.

### 4.1.4 Extended Role of Metallic Materials in LBEs

Assessment of plant response during normal, off-normal, and postulated accident conditions provides the basis on which plant safety analyses are developed. The HTGR includes passive safety features in which material properties are relied upon to accomplish safety functions. Therefore, complete plant response to accident prevention and/or mitigation functions depends on qualified material properties. This reliance on material performance during normal and accident environment conditions is fundamental to the materials selection and qualification process.

Strength, corrosion properties, thermal aging behavior, and the influence of radiation are among the additional factors to code status that must be considered in the selection and qualification process. For components that must perform a passive safety function during licensing basis events, additional material properties such as heat capacity, emissivity, thermal conductivity, thermal coefficient of expansion, or other physical properties may be relied on. Also important are the environmental conditions and time during normal operating or accident conditions that could influence material properties. Therefore, the material certification processes must consider the passive safety functions of the components to identify and include additionally unique material properties for qualification.

The plant design and investment protection characteristics also play an important role in determining required material properties. For example, a design goal of HTGR technology that influences material selection and qualification is the capability to return to commercial operation following any LBE, including the DBEs from which deterministic DBAs are derived. This goal potentially results in additional material performance and qualification requirements beyond the normal range of operating parameters. Material performance requirements during LBE conditions must therefore be addressed as part of the material qualification and codification process, from both licensing and investment protection viewpoints.

In order for a metallic material and, more generally, any structural material to be considered for use in the HTGR, it must be qualified for the appropriate service conditions and environment. In this usage, qualification implies that the material has been evaluated, based on a set of experimental data sufficient to reliably describe its behavior, and found to be able to meet the requirements placed upon it by the design for conditions of operation.

### 4.1.5 Alternate Methods for Materials Qualification

Metallic materials that are candidates for application in the HTGR have been discussed in Section 3.2. In terms of the basis for qualification, these materials fall into several categories:

- Materials contained and operated within the limits of a code or standard that the NRC has accepted
- Materials contained and operated within the limits of a code or standard that has been accepted by a standards body but which the NRC has not yet accepted
- Materials that are not contained and/or operated within the limits of a code or standard available at the present time and for which design will be based upon first principles, with appropriate supporting qualification programs.

In the case of the latter, the behavior and performance characteristics of the materials will need to be qualified for use based on analysis and/or testing specific to the HTGR application. It is anticipated that the qualification program would be based upon attributes similar to those supporting code case development, but with more focus on the specific needs of the HTGR. These attributes may include:

- Development of a bounding set of performance criteria based on the design requirements for all anticipated modes of operation, including licensing basis events
- Compilation and analysis of appropriate existing industry data
- Performance of tests and experiments necessary to supplement existing data
- Development of analytical and/or empirical models sufficient to describe the material behavior under the anticipated HTGR conditions
- Development of a qualification package sufficient to support use of the material in the HTGR.

The plans and required schedules for these qualification efforts will be developed on a case specific basis as the design of the HTGR progresses and specific material needs are developed.

### 4.1.6 PIRT-Identified Phenomena

In 2007, the NRC (with support from DOE) conducted a series of Phenomena Identification and Ranking Table (PIRT) workshops for a range of HTGR technology-specific topics. The PIRT workshops were based on very high temperature HTGRs, with reactor outlet temperatures to 1000°C (1832°F). While the reactor outlet temperatures of present HTGR designs are significantly lower, the high-temperature materials and graphite PIRT topics are of interest to this white paper.

Metallic materials phenomena considered in the workshops included conventional material properties such as strength, creep, fatigue, and the associated aging in a potential 60-year lifetime for some of the plant components. The service conditions considered covered a range that included both chemical attack and thermal cycling; they also encompassed irradiated material properties for metallic and nonmetallic components in or near the core and the primary system. The maintenance of adequate safety margins over time was a major concern for these PIRT reviews. The PIRT results indicated that the most significant phenomena associated with the materials in HTGRs include those related to:

- High temperature stability and components' ability to withstand service conditions
- Long-term thermal aging and environmental degradation
- Issues associated with fabrication and heavy-section properties of the RPV.

A number of other lower ranked issues were also identified by the PIRT panel for other components. These included material performance issues associated with candidate materials in the reactivity control elements, power conversion unit, helium circulator, RPV internals (core barrel, supports, restraints, and insulation), and primary system valves.

A range of candidate designs envisioned for the HTGR were reviewed against the PIRT results. This review concluded that no additional phenomena, other than the negligible creep issue discussed in Section 4.1.1 above, have been introduced or revealed by the proposed configurations.

### 4.2 Nonmetallic Materials

# 4.2.1 Graphite

### 4.2.1.1 Materials Selection and Qualification

Nuclear graphite has been successfully employed in the construction and operation of gas-cooled reactors, including HTGRs, for over 50 years. However, no industrial code or regulatory basis presently exists to support NRC regulatory approval of graphite structures in HTGRs. An ASME code for graphite structures is presently being developed as Section III, Subsection NG; however, it has not yet been approved by ASME or formally reviewed by the NRC.

The grades of nuclear graphite previously employed in HTGRs have varied considerably, and graphite source materials used in the manufacture of many earlier grades can no longer be obtained. However, a selection of candidate grades is currently available from the major graphite suppliers. These candidate grades build on past experience and more recent developments in the state-of-the-art of nuclear graphite. They are evaluated here to satisfy the requirements of both the pebble and prismatic HTGR concepts. Different reference grades have been selected for these respective reactor concepts, based on specific differences in requirements; for example, the large block sizes utilized in the pebble-bed reflectors and the small grain size requirements associated with the thickness of webs between fuel

compacts and helium coolant passages in prismatic fuel blocks. The accumulation of high fluence at the end of service life is a defining characteristic of certain pebble bed graphite components adjacent to the pebble core.

The properties and behavior of nuclear graphite under irradiation vary significantly as a function of source materials, fabrication processes, and heat treatment. However, experience indicates that materials produced using similar source materials and processing will possess similar as-manufactured properties and will exhibit similar trends in behavior under irradiation. The present graphite MTR programs are based on the premise of using a limited number of test specimens for calibrating and validating analytical models of graphite behavior that were developed from large legacy databases.

As an example, Figure 5 illustrates the relationship between the legacy German database and the PBMR Specific Materials Test Reactor Program irradiation conditions that were selected earlier for the proposed PBMR Demonstration Power Plant in South Africa. The solid blue line in the figure represents the projected temperature-fluence envelope at the end of service life for components that serve a structural function (SRC-1, as defined in Table 4), whereas the dotted red line denotes a similar envelope for the most highly-irradiated nonstructural components adjacent to the pebble fuel (SRC-2). As shown in Figure 5, the primary and secondary MTR data are designed to both confirm the applicability of the historical data and to supplement that data where required.

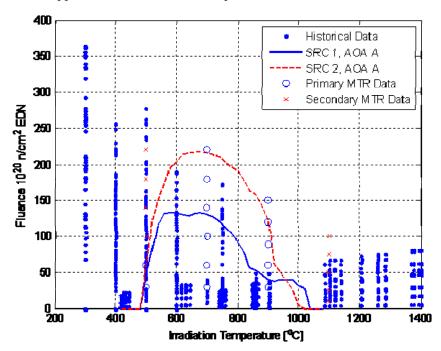


Figure 5. Comparison of the MTR data and PBMR Demonstration Power Plant service conditions.

Finally, the proposed service life of the graphite components in the PBMR implies the need for a relatively lengthy MTR program. On this basis, the PBMR approach is to acquire MTR data for a significant portion of the service life prior to the start of the lead reactor. The balance of the MTR data would be acquired in such a manner that it substantially leads the actual operation of the reactor.

The most highly irradiated components of the prismatic reactor design are the fuel blocks and replaceable reflectors. These components are replaced before they acquire fluences that would be associated with significant degradation. Accordingly, the requirements of the corresponding MTR program are limited.

### 4.2.1.2 Basis for Establishing Component Lifetimes

Graphite, located in the high-fluence regions of the core, undergoes structural changes during reactor operation that lead to changes in dimensions and in most of its physical and mechanical properties. The changes in properties and their consequent influence on the graphite structures must be accommodated in reactor core design. Oxidation must also be considered, but its influence on component strength and, hence, structural integrity is not expected to be significant for events within the design basis.

Given the expected changes in materials properties during operation, the requirement is to ensure the safe performance of the graphite components under all applicable operating conditions over their allocated design life. Meeting this requirement is considered more challenging for pebble bed designs, in which reflector replacement is projected to occur only once during the reactor's lifetime, if at all. By contrast, the replacement of fuel elements and reflector blocks in prismatic designs occurs routinely and at lower fluence

The design life of graphite core structures is based upon the criterion that the probability of a failure in the most highly stressed reflector components will not exceed a specified level. It will be established on a conservative basis using the probabilistic analysis methods noted in Section 3.3.5.2. Nonreplaceable graphite reflector components are exposed to modest fluence levels and are expected to have lifetimes that substantially exceed the design life of the plant. In the absence of irradiated properties data for the specific graphite in question, conservative estimates must account for uncertainties in the materials models used for design. The design life estimates will be updated as materials test reactor data become available.

The operational life of the most highly irradiated graphite components in pebble bed reactors will be further evaluated in service through programs involving visual examination, detection, and evaluation of cracks by eddy current techniques, trepanning of small material samples from the reflector adjacent to the core, or evaluation of replaced components post-service.

## 4.2.1.3 PIRT-Identified Phenomena

The scope of the PIRT-identified phenomena related to nuclear graphite component include graphite properties such as strength, creep, stress, fatigue, and any associated aging in a potential 60-year lifetime for some of the core components. The scope also includes oxidation and the aspects of helium gas impurities as well as the effects of gamma and neutron irradiation. The PIRT results indicated that the most significant phenomena in the graphite area include:

- Irradiation effects on material properties (expansion/contraction, thermal conductivity)
- Consistency of graphite quality and performance over the service life.

The PIRT panel also concluded that theories that can explain graphite behavior have been postulated and, in many cases, shown to represent experimental data well. Thus, much of the data needed is confirmatory in nature. However, these theoretical models still need to be tested against data for the new generation of nuclear graphites and extended to neutron doses and temperatures typical of proposed HTGR designs. It is anticipated that current and planned future graphite irradiation experiments will provide the data needed to validate many of the currently accepted models and confirm and validate designs.

# 4.2.2 Baked Carbon Insulation

Baked carbon insulation has also seen prior service in HTGR reactors. As with graphite, there is no existing industrial code or regulatory basis to support licensing approval; however, unlike graphite, none is presently being developed. Based on its similarity to graphites, the approach to materials selection and

qualification would be similar. Unlike graphite, baked carbon insulation is not expected to see significant fluence during service. The basis for design and assessing the structural adequacy of baked carbon insulation components is proposed to be identical to that of effectively unirradiated graphite.

# 4.2.3 Composites

To date, there has been no application of Carbon Fiber Reinforced Carbon (CFRC) composites in nuclear reactors, including HTGRs. However, some development work on CFRC components was done in Germany. As is the case with baked carbon insulation, there is no existing industrial code or regulatory basis to support licensing approval. It is believed, however, that composites will be addressed in the future in an ASME Section III initiative by the same group that is presently developing the ASME code for graphite core components. In present applications, including those proposed for the HTGR, the components of interest would see extremely low doses. Materials qualification would largely be based on testing.

# 5. OUTCOME OBJECTIVES

The overall objective of this materials white paper is to provide the basis for early interactions with the NRC regarding the employment of materials in the primary system of the HTGR in advance of a construction and operating license application. Through this process, a further objective is to minimize the time required for design certification through early identification and, where possible, resolution of issues. At the present time, feedback is being sought from the NRC on the specific issues identified within this section for metallic and nonmetallic materials, respectively.

## 5.1 Metallic Materials

Responses are requested to the following questions related to metallic materials.

- 1. Based on the information and justifications presented in Section 3.2, along with future plans, does application of the ASME Code for metallic materials provided in this report form a reasonable basis for the design and qualification of components, including the following ASME Code Case issues?
  - a) Assuming resolution of the negligible creep limit and appropriate adjustments, if any, to Section III and Code Case N-499-2, does NRC agree that ASME Section III, Subsection NB, supplemented by Code Case N-499-2 for low frequency events, provides a reasonable basis for developing the design certification application for the RPV?
  - b) Code Case N-201-5 is an ASME-approved addition/modification to Section III, Subsection NG that provides for the design and construction of core support structures for temperatures above 371°C (700°F) and up to 816°C (1500°F), depending on material, during both normal operation and duty cycle events. The scope of N-201-5 currently includes Type 316 SST, 2.25Cr-1Mo steel and Alloy 800H. The issue to be resolved is whether NRC agrees with the application of ASME Code Case N-201-5 as a basis for regulatory compliance for core support components.
  - c) Section III, Subsection NH is an ASME-approved code for the design of elevated temperature service of metals (Alloy 800H, Types 304 and 316 SST, Modified 9Cr-1Mo and 2.25Cr-1Mo) for Class 1 components. The issue to be resolved is whether NRC agrees with the application of ASME Subsection NH as a basis for regulatory compliance for Class 1 components (e.g., reactor pressure vessel, control rods) in elevated temperature service for identified metallics including Alloy X and Alloy 617.

### 5.2 Nonmetallic Materials

Based on the information and justifications presented in Section 3.3, along with future plans for graphite material:

- 1. Does application of the emerging ASME Code for graphite provide a reasonable basis for the design and qualification of components?
- 2. Does an experimental program characterizing the fluence/temperature response of graphite that leads the actual operation of the plant by at least 10 years provide a reasonable basis for licensing the initial operation of a HTGR lead plant?
- 3. Does the RIM Program provide a reasonable basis for assessing the condition of graphite components in service?

Based on the information and justifications presented in Sections 3.4 and 3.5, along with future plans, for ceramic and composite materials:

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- 1. Does application of the requirements for the Design and Manufacture of the Ceramic Internals provide a reasonable basis for the design and qualification of the CSC components (in the absence of established industrial codes and standards)?
- 2. Is the selection of CFRC reasonable for certain structural components, potentially including safety-related components, given the functions and requirements associated with their application (e.g., low or no fluence) and their properties relative to metallic alternatives?

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# Appendix A Historical Experience with Nuclear Graphite

# Appendix A Historical Experience with Nuclear Graphite

# **Experience with Nuclear Graphite**

Graphite has been used for core structural components in nuclear reactors for over 60 years. There is a substantial base of experience with the use of graphite in commercial high temperature gas-cooled reactors (HTGRs), both CO<sub>2</sub> and helium-cooled. The summary of experience discussed here is from the United Kingdom, Germany, United States, Japan, and China; however, there has also been extensive experience with graphite moderated reactors in both France and Russia.

### **United Kingdom**

The first commercial reactors in the United Kingdom were of the natural uranium-fueled Magnox type. Four Magnox reactors were initially built at Calder Hall and achieved full power operation between 1956 and 1960. Each reactor had around 1,700 fuel channels moderated by graphite with a design heat output of 180 MWth. A further nine Magnox stations were built between 1962 and 1971, the largest and last being a twin reactor station at Wylfa, each reactor having over 6,100 channels and a design heat output of more than 1800 MWth. The first plant closure came in 1989 and the final closure is scheduled soon after 2010. Magnox plants of UK-design were also built in Japan (Tokai) and Italy (Latina). French-designed Magnox plants were built in France (reactors) and Spain (Vandellos). All these plant are shut down and in the process of decommissioning.

The graphite used in the UK-based Magnox reactors was Pile Grade A (PGA) graphite made from 'needle' coke which had a needle-like appearance after crushing. The anisotropic graphite crystal structure tends to be preferentially aligned along the length of these needle-like grains. Since the PGA reactor components were formed by extrusion, which preferentially aligns the needle-coke grains in the product, preferential alignment of the graphite crystallites occurs along the extrusion direction resulting in a highly anisotropic bulk material. Production graphite had a density of about  $1.7 \text{ g/cm}^3$ , was quite porous and had moderate strength with highly anisotropic properties (isotropy ratio of 2.2). Therefore, the dimensional change behavior of PGA graphite under irradiation was very anisotropic. In the direction parallel to the direction of extrusion, the graphite shrank progressively with increasing dose at all irradiation temperatures of interest. In the perpendicular direction, however, the graphite shrank at irradiation temperatures greater than around  $300^{\circ}\text{C}$  ( $572^{\circ}\text{F}$ ), but below this temperature it exhibited growth, the lower the temperature, the greater the rate of growth. Nominal graphite operating temperatures ranged between 200 and  $400^{\circ}\text{C}$  (392 and  $752^{\circ}\text{F}$ ) with peak fluence at end of life expected to be about  $6 \times 10^{21}$  n.cm<sup>-2</sup> EDN (equivalent dido nickel).

The Magnox reactors were followed by Advanced Gas Cooled Reactors (AGRs), the first two of which were commissioned in the United Kingdom at Hinkley Point and Hunterston in 1976. Overall, seven stations were built using four different designs over the years. Experience confirmed that the dimensional change rates exhibited by PGA graphite were unacceptable for the much higher irradiation fluence and temperature combination that would be attained in commercial AGRs. Nominal graphite operating temperatures in the AGR ranged between 250 to 650°C (482 to 1202°F), with a peak design fluence in excess of 5 × 10<sup>21</sup> n.cm<sup>-1</sup> EDN. Therefore, development and testing of graphites with greater dimensional stability and higher strength were undertaken. Another problem in both the Magnox and AGR reactors was radiolytic oxidation caused by radiolysis of the CO<sub>2</sub> coolant and subsequent graphite oxidation. Several materials were produced, but the graphite eventually chosen for the first AGR at Dungeness, and all subsequent AGRs, was made using Gilsonite pitch coke. The Gilsonite pitch coke grains have an approximately spherical shape and, by using a molding process to make the graphite

blocks, near isotropic graphite was produced. The resulting graphite was referred to as Gilsocarbon graphite. However, available sources of Gilsonite pitch were eventually exhausted and are no longer available.

#### **United States**

Fort St. Vrain was a U.S. designed and built, helium-cooled, graphite moderated HTGR that utilized a <sup>235</sup>U– thorium fuel cycle, a prismatic fuel element design and a prestressed concrete reactor vessel. Gross reactor power output was 842 MWth and 330 MWe. The reactor core consisted of vertical columns of hexagonal graphite fuel-moderator elements and graphite reflector blocks grouped into a cylindrical array and supported by a graphite core support structure. The active core had the approximate shape of a right circular cylinder with an equivalent diameter of approximately 5.9 m (meter) and a vertical height of approximately 4.8 m. The side reflector had a mean thickness of about 1.2 m, giving an overall mean core and reflector diameter of approximately 8.3 m. The top reflector had an effective thickness of about 1 m and the bottom reflector had an effective thickness of about 1.2 m, giving an overall assembly height of about 6.9 m. The core was contained within a steel core barrel that provided lateral constraint and support for the fuel and reflector columns. The active core was composed of 1,482 hexagonal graphite fuel elements stacked in 247 vertical columns. The individual graphite fuel elements were approximately 35.6 cm across the flats and 78.7 cm high. The replaceable reflector elements assemblies were composed of 2,188 hexagonal graphite blocks. Some of these blocks incorporated boronated graphite material for shielding purposes. The permanent reflector block and spacer assemblies were composed of 312 graphite blocks and included boronated graphite material in the top or the peripheral spacers.

The graphite used for the Fort St. Vrain (FSV) core support blocks was PGX graphite, a medium-grain graphite with A low coefficient of thermal expansion (CTE) and elastic modulus, marginal strength (mean tensile ~8 MPa) and high ash content (7,000 ppm max). The core support posts were made from ATJ graphite, a fine-grain, isostatically-molded grade of high strength (27 MPa WG, 23 MPa AG) and very low CTE (2.2–2.6 × 10<sup>-6</sup>°C<sup>-1</sup>), relatively low modulus (9.7–9.5 GPa) and high thermal conductivity (125–110 W.m<sup>-1</sup>.K<sup>-1</sup>), giving this grade good thermal shock resistance and an isotropy ratio of 1.18. The permanent side reflectors were made from HLM graphite, a medium-grain, extruded-grade of marginal strength (tensile~12 MPa), high anisotropy ratio of 1.5, high thermal conductivity (> 150 W.m-1.K-1) and high ash content (1,000–3,000 ppm). The primary fuel element and reflector structural material in the initial FSV core and the first and second reloads was H-327 graphite, an anisotropic grade made using needle coke with an isotropy ratio in excess of 2.0. After some time, the high anisotropy of H-327 was recognized as being problematic. Beginning with the third reload, H-451 graphite, an extruded near isotropic grade made from petroleum coke, was substituted for the fuel and reflector elements. This remained the primary graphite material used beginning with Reload 3. Analysis confirmed that H-451 graphite improved the mechanical, thermal, and fluid flow characteristics of the reactor.

The design life of the core fuel elements and the replaceable reflector elements adjacent to the core in the FSV reactor was 1,800 effective full-power days. The remaining permanent reflector elements, blocks, and spacers had a design lifetime of 30 years. The cumulative clearances between fuel and control rod columns across any core diameter was specified to limit the maximum possible bowing deflection of any individual column to ensure that insertion of the control rods and reserve shut-down neutron absorber material were not restricted under any conditions for normal or abnormal operation.

### **Germany**

The first German pebble bed reactor was the 15 MWe Arbeitsgemeinschaft Versuchsreaktor (AVR), or research reactor, which began operation in 1967. The performance of this pebble bed reactor was generally good throughout its 21 years of operation. The reflector graphite employed in the AVR was Grade ASR/AMT, a highly anisotropic, petroleum coke grade. Despite the large difference in dimensional

change of the ARS grade with-grain and against-grain, no operational problems were attributed to graphite material performance, primarily because of the limited irradiation exposure of the top and side reflector components. Whereas the top reflector only accumulated a fast neutron fluence of <2 dpa ( $<1.5 \times 10^{21}$  EDN) at  $1000^{\circ}$ C, the side reflector received four times this fluence in the peak flux regions, but at lower temperatures, so that the anisotropic dimensional changes which did occur were not of operational consequence.

The AVR was followed by the 300 MWe thorium high temperature reactor (THTR-300), which began operation in 1985. The graphite used in the THTR-300 was Grade PXA2N, an isotropic, medium grain nuclear graphite made from Gilsonite pitch coke. By this time, the German graphite development program had reached an advanced stage, and it was well recognized and understood that control of graphite isotropy and processing parameters could dramatically improve material properties and behavior under irradiation.

THTR-500, a higher capacity follow-on to THTR-300, was designed, but never built. However, the graphite materials and reflector design associated with THTR-500 have been used, in part, as the basis for the current pebble bed modular reactor design. The HTR-Module was another pebble-bed reactor concept that was designed and reviewed by the German licensing authorities for a site-independent licensing permit. Though never built, it was a key milestone in the evolution of the inherent and passive safety concepts that are prevalent in current modular HTGR concepts. As with present modular HTGR designs, fuel temperatures were to be limited during design-basis accidents by inherent and passive features. Design documentation and safety analysis report information pertaining to the HTR-Module indicate that the concept graphite core structures design was based on ASR-1RS graphite, a grade that was in development at the time. Grade ASR-1RS represented the apex of the German graphite development effort in following respects:

- 1. It was a medium-grain (1.0 mm), isotropic pitch coke, vibration molded grade
- 2. The coke was subjected to a specialized secondary coking technique to reduce the dependence of the final graphite properties on the coke source and to achieve the highest level of isotropy attainable at that time
- 3. It was tested under irradiation to compare against other development grades at the time and excellent isotropy in irradiation behavior, including very low dimensional shrinkage was confirmed
- 4. Several batches were produced over a period of a decade or so to evaluate the influence of raw materials, refine processing parameters, and optimize material properties.

Perhaps the biggest disadvantage with ASR-1RS was the high cost of the secondary coking process, which stemmed from the significant additional processing steps required.

## Japan

Renewed development of HTGRs began with construction of the High Temperature Engineering Test Reactor (HTTR) in Japan. The HTTR is a 30 MWth helium-cooled high-temperature prismatic fuel reactor that reached initial criticality in 1998 and has operated at reactor outlet temperatures of up to 950°C. A wide variety of commercially available graphites were examined as candidates for the graphite core components. Most core components (reflector and fuel elements) are fabricated from IG-110 nuclear graphite and the larger core support blocks are fabricated from PGX nuclear graphite. Since inception, there has been ongoing development and testing of both graphite material and core components with extensive publications regarding the performance of IG-110 under a variety of test conditions This has included data on physical and mechanical properties at ambient and high temperature, fatigue, irradiation creep, and oxidation. Importantly, there has been a sustained effort in the development of surveillance and

in-service inspection techniques for the HTTR to assess the condition of the core components, both in situ and out of core.

The HTTR graphite structural design was based on conventional stress analysis approaches, similar to that used for metallic components, and an expanded version of the original draft Code for graphite core components issued to the ASME for review and comment in 1990. Significant extensions made by the Japan Atomic Energy Research Institute, now the Japan Atomic Energy Agency to this previous draft code related to the allowable stress limits for irradiated components, treatment of fatigue and buckling limits (for core supports), oxidation, and irradiation creep.

### China

The most recent addition to the HTGR family is the Chinese HTR-10, a 10 MWth pebble bed test reactor that reached initial criticality in 2002. This is a scaled-down design that also draws significantly from the HTR-Module concept. However, the graphite selected for the HTR-10 was IG-11, an isostatic-molded, fine-grain material, in contrast to the medium-grain reflector grades that were the focus of the German program. Given the short-term experience of the HTR-10, there is little published information regarding the graphite core component performance to date. However, the Institute of Nuclear and New Energy Technology, the Chinese developer, has revealed that the purified version of IG-11, i.e., IG-110, will be employed for the graphite core in the scaled up, twin 250 MWth HTR-PM plant, with construction beginning by 2013.

# **Summary**

It can be said that the quality of reactor graphite grades has improved over the years with significant improvements in the density, strength, isotropy, and purity of current grades. It is also clear that these modern grades have been developed based on the strengths and weaknesses of past grades as assessed from their performance in test reactors and in HTGR components. The physical, thermal and mechanical properties of these modern graphite grades are currently being evaluated by INL and ORNL for use in HTGR applications, and by other parties involved in HTGR research and development in Europe, China, Japan, and Korea. These test programs cover both unirradiated and irradiated conditions.

# Status of Graphite Qualification for the HTGR

Recent HTGR graphite experience in the United States has been mainly with Grades H-327 and H-451 at FSV. However, these graphites are no longer available because the coke source used to manufacture the graphite is not available. New graphite grades, such as those discussed in Section 3.3.3, have been designed based on the strengths and weaknesses of H-451 and other previous grades' performance in lab tests and in HTGR components. A complete properties database for these newly available candidate grades of graphite must be developed to support the design and licensing of HTGR core components. Data are required for thermal, mechanical (including radiation-induced creep), and oxidation properties of these graphite grades. Moreover, the data must be statistically sound and take account of in-billet, between billet, and lot-to-lot variations in properties.

Testing is currently underway at Idaho National Laboratory (INL) to gather data on these new graphite grades. The INL graphite development program provides for a number of capsules that will be used to characterize the effects of irradiation, temperature, and compression simultaneously over a range of temperatures and fluences (see Section 3.3.6). Existing data and related international programs undertaking the characterization of irradiation effects on these grades of graphite may also be used in the qualification process.

# Appendix B

**Graphite Structure and the Effects of Irradiation** 

# Appendix B Graphite Structure and the Effects of Irradiation

In its perfect form, the crystal structure of graphite consists of tightly-bonded (covalent) sheets of carbon atoms in a hexagonal lattice network as shown in Figure B-1. The sheets are weakly bound with van der Waals type bonds in an ABAB stacking sequence

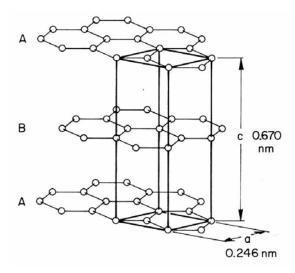


Figure B-1. Crystal structure of graphite.

Changes in graphite properties resulting from nuclear irradiation are the result of displacements of atoms in the crystal structure. The primary atomic displacement (primary knock-on carbon atoms) produced by energetic particle collisions produce further carbon atom displacements in a cascade effect. The cascade carbon atoms are referred to as secondary knock-on atoms. The displaced secondary knockon atoms tend to be clustered in small groups of 5 to 10 atoms, and for most purposes it is satisfactory to treat the displacements as if they occur randomly. The total number of displaced carbon atoms will depend upon the energy of the primary knock-on carbon atoms, which is itself a function of the neutron energy spectrum, and the neutron flux. Once displaced, the carbon atoms recoil through the graphite lattice, displacing other carbon atoms and leaving vacant lattice sites. However, not all of the carbon atoms remain displaced. The displaced carbon atoms diffuse between the graphite layer planes in two dimensions and a high proportion will recombine with lattice vacancies. Others will coalesce to form C2, C3, or C4 linear molecules. These in turn may form the nucleus of a dislocation loop—essentially a new graphite plane. Interstitial clusters may, on further irradiation, be destroyed by a fast neutron or carbon knock-on atom (irradiation annealing). Adjacent lattice vacancies in the same graphitic layer are believed to collapse parallel to the layers, thereby forming sinks for other vacancies which are increasingly mobile above 600°C and, hence, can no longer recombine and annihilate interstitials.

These changes in the graphite structure under irradiation produce corresponding changes in the properties of graphite, which are summarized in the sections that follow.

# **Dimensional Changes with Neutron Irradiation**

A principal result of carbon atom displacements is crystallite dimensional change. Interstitial defects will cause crystallite growth perpendicular to the layer planes (c-axis direction), and relaxation in the layer plane because of coalescence of vacancies will cause a shrinkage parallel to the layer plane (a-axis direction). The damage mechanism and associated dimensional changes are illustrated in Figure B-2.

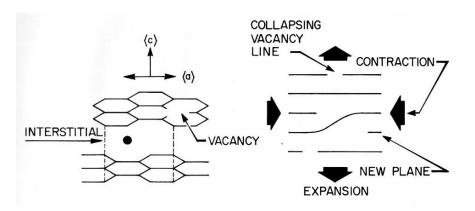


Figure B-2. Neutron irradiation damage mechanism illustrating induced crystal dimensional changes.

Irradiation-induced dimensional changes can be very large, in well-ordered graphite materials, such as pyrolytic graphite, which has frequently been used to study neutron-irradiation induced dimensional changes. Polygranular graphites, which are more typical of materials used in high temperature gas-cooled reactors (HTGRs), possess a polycrystalline structure, usually with significant texture resulting from the manufacturing method. Consequently, structural and dimensional changes in polygranular graphites are a function of both the crystallite dimensional changes and the graphite's texture. In polygranular graphite, thermal shrinkage cracks that occur during manufacture and that are preferentially aligned in the crystallographic a-direction will initially accommodate the c-direction expansion, so mainly a-direction contraction will be observed. The graphite thus undergoes net volume shrinkage. With increasing neutron dose (displacements), the incompatibility of crystallite dimensional changes leads to the generation of new porosity oriented parallel to the basal planes, and the volume shrinkage rate falls, eventually reaching zero. The graphite now begins to swell at an increasing rate with increasing neutron dose. The fluence at which the volume change with irradiation switches from the initial contraction phase to the volume expansion mode is termed the turnaround point. At extremely high fluence levels, the accumulation of pores and microcracks effectively leads to a loss of the material integrity or cohesion. This is referred to as the cohesive life limit. These trends are illustrated in Figure B-3 and Figure B-4 for a relatively isotropic graphite.

The rate of shrinkage, the maximum shrinkage observed, the turnaround fluence, and expansion rate are strongly influenced by the actual irradiation temperature. Historically, the fluence at which the graphite dimensions returned to their original values, defined as the return-to-original-volume was considered as a measure of the useful life of the graphite. Reactor grades that returned to original volume at higher fluence at a given temperature were regarded as having a longer life. For the German program, volumetric swelling amounting to 10% beyond the initial value was used as the end-of-life criterion.

Analyses, however, indicate that the stresses resulting from dimensional changes in irradiated graphite components are typically more limiting in terms of component life than volumetric swelling criteria. Stresses arising from dimensional changes must be assessed in combination with other stresses, in evaluating the likelihood of component failure. End-of-life is said to be reached when the likelihood of failure exceeds established limits. It must also be pointed out that the external stresses imposed on core components while under irradiation will alter their dimensional change behavior. A stressed graphite component under irradiation will undergo irradiation creep. The irradiation creep strain is defined as the difference in dimensional change between stressed and unstressed material irradiated under the same conditions of fluence and temperature. Therefore, the creep strain has also to be considered in the total stress analysis of the irradiated component.

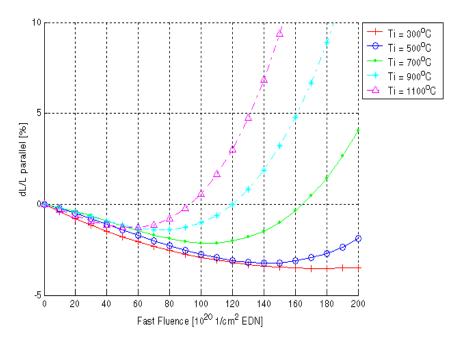


Figure B-3. Typical irradiation-induced dimensional changes in reactor graphite, parallel direction.

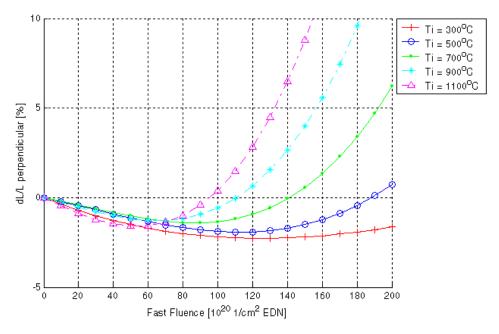


Figure B-4. Typical irradiation-induced dimensional changes in reactor graphite, perpendicular direction.

# **Thermal Conductivity Changes with Neutron Irradiation**

Thermal conductivity is critical to HTGR design, as it plays a key role in determining the ability to transfer decay heat from the core during conduction cooldown events, thus limiting maximum fuel temperatures. This property is controlled by raw materials, processing (e.g., forming method) and heat treatment temperature and the graphite irradiation fluence-temperature history. High graphitization temperature (>2700°C) is required during the final stage of billet manufacture to ensure sufficient thermal conductivity for HTGR applications.

Fast neutron fluence typically produces a rapid decrease in the thermal conductivity of graphite from the value at low fluences to an intermediate saturation level. This saturation value persists over a significant portion of the remaining fluence range until further irradiation-induced structural changes in the graphite (notably pore generation at and beyond the dimensional change turnaround fluence) cause a secondary reduction in thermal conductivity (decline). As the irradiation temperature increases, thermal annealing of irradiation damage causes the reduction in thermal conductivity to be reduced. For example, at 300°C (572°F), graphite exposed to a fast fluence of about  $5 \times 10^{21}$  n/cm² (EDN) will have a thermal conductivity of about 10% that of unirradiated graphite. Graphite exposed to a fast fluence of  $5 \times 10^{21}$  n/cm² (EDN) at 600°C (1112°F) will only drop to about 40% of its unirradiated value. Typical irradiation-induced thermal conductivity changes in reactor graphite are illustrated in Figure B-5. The stages in these changes are summarized in Table B-1.

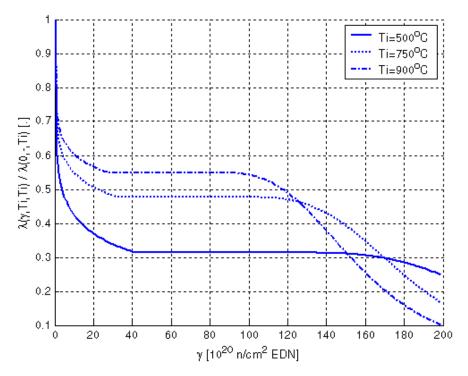


Figure B-5. Typical irradiation-induced thermal conductivity changes in reactor graphite.

Table B-1. Stages of change in thermal conductivity because of irradiation.

Phase	Description
Nonirradiated (virgin material)	The material is in its virgin state.
Initial breakdown in thermal conductivity (low dose)	Degradation by neutron-induced point defects in the crystal lattice. The conductivity drops steeply with dose in this stage.
Saturation (intermediate dose)	Thermal annealing counteracts the neutron-induced defect formation; the thermal conductivity degradation reaches a steady state value where the generation and annealing of single (point) defects occur at an equal rate. The level at which the thermal conductivity saturates is a function of the temperature at which the irradiation takes place.
Secondary breakdown in thermal conductivity (high dose)	Large volumetric expansion caused by pore generation within the material causes a secondary breakdown in the thermal conductivity.

It has been established that the saturation thermal conductivity level is practically independent of graphite grade when the relative thermal conductivity change (% based) is considered. This is important from a design perspective. Additionally, the thermal conductivity at the irradiation temperature is always higher for the operational temperatures of interest than the value measured at room temperature, a factor which must be accounted for in design. It may be further considered that there is some recovery in thermal conductivity when irradiated graphite is heated past its irradiation temperature (normal operation temperature in this instance). This may present some margin for heat transfer under accident conditions. However, this latter aspect would require verification based on the specific operating conditions of the HTGR concept.

# **Specific Heat Capacity**

Specific heat capacity, which is the energy required to increase the temperature of a unit mass of a material by unit temperature, is an important property of graphite in HTGR applications. The relatively high specific heat capacity of graphite tends to moderate transients and enhances its capability to store thermal energy during the initial stages of LBEs involving conduction cooldown. This, in turn, is a factor in limiting temperatures of the fuel and metallic components to acceptable levels. Extensive studies of nuclear graphite grades have shown that heat capacity increases with temperature and it does not vary significantly among graphite grades. Measurements on past grades and present candidates confirm the nonvariability of this property between the different grades and its correlation with theoretically calculated specific heat capacity for graphite (see ASTM C781 for example). More importantly, indications are that the specific heat of irradiated graphite varies little from the virgin value (over the applicable temperature range), a factor which is very useful from a practical design perspective, particularly when assessing conduction cooldown heat transfer conditions.

# **Emissivity**

Another important physical property during postulated accident conditions is emissivity, a measure of a material's ability to transport heat via radiation. Thermal radiation is an important thermal energy transport mechanism for LBEs involving conduction cooldown, particularly under depressurized conditions. During such events, heat must be transferred from the region of the core outward, through and between the graphite blocks and, then to the core barrel. Emissivity is defined as the ratio of energy radiated by the material to that radiated by a black body (emissivity = 1) at the same temperature. The emissivity of a given graphite will depend on its surface condition and the environmental temperature. Generally, the duller or blacker the material, the closer its emissivity is to that of a black body. Typical emissivity values for carbon or graphite range between 0.8 and 0.9. Extensive tests of previous and current grades show that machined nuclear grade graphite has an emissivity of about 0.85 at elevated temperatures. The emissivity of nuclear graphite is not expected to change significantly with irradiation.

# **Thermal Expansion Changes with Neutron Irradiation**

The coefficient of thermal expansion (CTE) of graphite is an important consideration in the setting of graphite component dimensional tolerances during both normal operation and accident conditions. This property must be assessed in conjunction with irradiation-induced dimensional changes. Graphite CTE is determined by a combination of the in-crystal CTE, bearing in mind its highly anisotropic nature, and ex-crystal microstructural features such as Mrozowski cracks, which are ultra-fine, interlamellar cracks that lie between crystalline regions of filler grains. Other characteristics, such as the type of coke and, to a lesser degree, grain size, forming method, etc. also play a role in determining the bulk CTE. The Mrozowski cracks play a dominant role in controlling the thermal expansion characteristics of the bulk graphite by accommodating intercrystalline expansion within the bulk, thus contributing to the very low CTE of polycrystalline graphite. This expansion mechanism gives graphite good thermal shock resistance, allowing large crystal expansion in the direction of cracking without leading to intercrystalline cracking.

Nonetheless, the bulk CTE is also strongly influenced by the filler coke CTE and, in this respect, the selection of raw material for the candidate grade requires consideration. There is a competing requirement for relatively low bulk CTE to reduce secondary operational stresses in graphite components (thermal stress) while ensuring sufficient isotropy in the bulk material. ASTM D7219-08 makes specific recommendations regarding the allowable coke CTE range for graphite components exposed to a high fluence regime. The temperature-dependent CTE of reactor grades (past and present) at high temperature has been extensively characterized, and the available data are deemed sufficient for the design of unirradiated graphite components.

More significantly, the graphite CTE will first increase slightly under irradiation, reach a peak, and then drop well below the unirradiated value as the fast neutron fluence increases as shown in Figure B-6. The extent of the peak and irradiation-induced drop in CTE varies with irradiation temperature, tending towards a lower peak value and more rapid drop to values well below the initial value as irradiation temperature increases. While the change in graphite CTE with irradiation has been extensively characterized for a range of graphites (past and present), additional data will be required for the HTGR design conditions and grade of choice. The above mentioned factors will need to be taken into account when determining the projected coefficient of thermal expansion of the selected graphite.

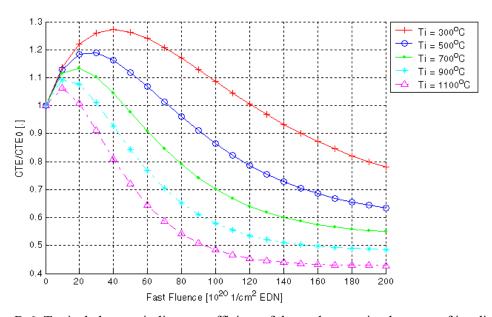


Figure B-6. Typical changes in linear coefficient of thermal expansion because of irradiation.

# Strength and Elastic Modulus

Graphite strength is the most significant property for ensuring the structural integrity of the core components. Graphite is a good choice for core components because its strength increases with temperature up to about 2000°C (3632°F), well beyond the projected peak core temperatures that would be seen under accident conditions. This increase in strength with temperature is largely because of the closure of fine lamellar (Mrozowski) cracks and additional microcracks that form during cooldown from the extreme production (graphitization) temperatures.

The strength of graphite, when subjected to neutron irradiation, increases over an extensive portion of the total fluence range as shown in Figure B-7. The strength increase takes place in two stages. At very low fluences, there is an initial rise in strength that is attributed to dislocation pinning at irradiation-induced lattice defect sites. This effect has largely saturated at doses >1 dpa  $(\sim 7.6 \times 10^{20} \text{ n} \cdot \text{cm}^{-2} \text{ EDN})$ . Above  $\sim 1$  dpa, a more gradual increase in strength occurs. These further increases in strength are more the result of interlamellar and microcrack closure as a result of dimensional changes within the graphite crystallites themselves. (The curves in Figure B-6, as well as Figure B-7, are based on immediate pinning of the crystal structure and, thus, do not show the initial increase.) A strength reduction follows the period of strength increase, with this effect being quite closely matched with the turnaround point in the volumetric change behavior. At this point, the mechanical properties of the graphite begin to decrease with the generation of internal porosity. The compressive strength of graphite also first increases and then decreases with irradiation in a manner similar to the tensile strength. These changes are caused by the same mechanisms described above.

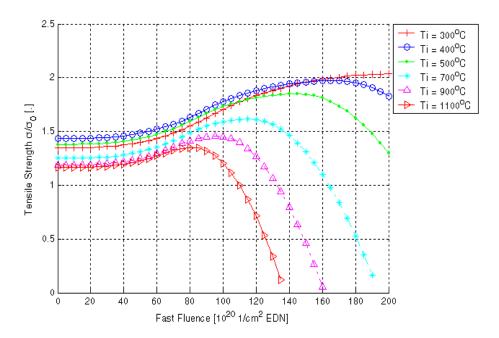


Figure B-7. Typical irradiation-induced strength changes in reactor graphite.

The Young's modulus change with irradiation (Figure B-8) closely resembles the progression of strength change with irradiation. Both increase to a peak value and decline thereafter, and tend to saturate at a value close to the original value. As with other properties described earlier, the progression in both strength and elastic modulus change with irradiation is strongly dependent of irradiation temperature.

# **Irradiation Creep**

Graphite experiences creep deformation under neutron irradiation and stress at temperatures below 1600°C, where thermal creep is normally negligible. The phenomenon of irradiation creep in graphite has been widely studied because of its significance to the operation of graphite moderated fission reactors. The beneficial effect of irradiation creep is to reduce irradiation induced stresses in graphite moderators, thus allowing acceptable service lifetimes to be achieved.

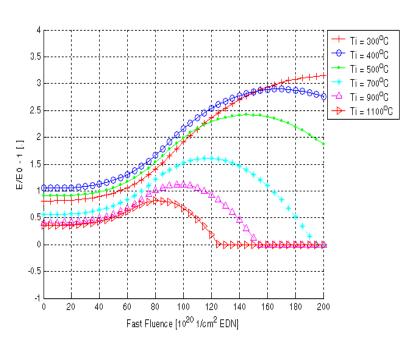


Figure B-8. Typical irradiation-induced modulus changes in reactor graphite.