STATUS OF ONGOING RESEARCH WITHIN THE GIF VHTR MATERIALS PROJECT

William R. Corwin

Oak Ridge National Laboratory (*corwinwr@ornl.gov*)

I. INTRODUCTION

Expanded nuclear energy is a key element necessary to provide an adequate supply of clean, sustainably energy to meet the increasing demands of the world's expanding population and economy. This expansion will require a new generation of nuclear technology to augment the addition of evolutionary light-water-cooled reactors and life extension of the existing nuclear fleet. The Generation IV International Forum (GIF) has developed a technology roadmap for advanced nuclear energy systems that culminated in the selection of the six most promising Generation IV nuclear reactor systems that would best meet broad goals established for sustainability, economic competitiveness, safety and reliability, and proliferation and physical protection.[1]

Among the six advanced nuclear energy systems that were identified as contributing to the Generation IV goals was the Very High Temperature Reactor (VHTR), which employs a thermal neutron spectrum with coolants and temperatures that enable generation of high-quality process heat for hydrogen production or other commercial applications (such as those for the synfuel, petro-chemical, and steel industries), as well as electricity production with high efficiency.

Since the completion of the roadmap, GIF has coordinated worldwide developmental activities for Generation IV reactor systems. A Steering Committee has been formed for the VHTR to help plan and carry out the research and development (R&D), design, and safety studies being conducted by the participating GIF members to establish its viability and optimize its performance. Additionally, system-specific Project Arrangements (PAs) for key VHTR technologies, including structural materials, have been developed, which stipulate what specific international contributions to the advancement of those systems will be made and how information is to be shared. The VHTR Materials PA includes major contributions of both new and protected historical information from its participating partners that currently include Canada, EURATOM, France, Japan, the Republic of Korea, the Republic of South Africa, Switzerland, and the United States.

Materials development and qualification, design codes and standards, as well as construction methodologies, for VHTRs require new investigations for the design and construction of the key components. The development of new material grades, as well as the extended qualification of existing materials, are key issues for meeting the higher temperature and longer lifetime requirements of VHTR normal and off-normal operating conditions, including:

- graphite for the reactor core and internals;
- high-temperature metallic materials for internals, piping, valves, high-temperature heat exchangers, steam generators, and turbo-machinery; and
- ceramics and composites (*e.g.*, C/C,

SiC/SiC, etc.) for control rod cladding and other specific reactor internals, as well as for advanced intermediate heat exchangers for very-high-temperature conditions.

II. GIF VHTR MATERIALS PROGRAM

The key design parameters that will affect the choice of materials and, therefore, the needed R&D include the reactor coolant inlet and outlet temperatures and pressure, as well as the choice of the secondary-side coolant and its associated temperatures and pressures. Expected service conditions include a near-term core coolant outlet temperature between 750 and 900°C, for which existing materials may be used, and a longer-term goal of 1 000°C that will require the development of new materials. The inlet core temperature for such systems could range from about 300°C to 600°C and the primary coolant system pressures from 5 to 9 MPa. Reactors currently being developed, such as the Next Generation Nuclear Plant (NGNP)[2], the Pebble Bed Modular Reactor (PBMR)[3], or the High Temperature Gas-cooled Reactor-pebble-bed Module (HTR-PM)[4], focus on the lower range of core outlet temperatures and will largely utilize existing structural materials, but will serve a vehicle for developing and evaluating the enhanced materials codes and design methods, as well as condition monitoring techniques, needed for their anticipated 60-year lifetimes and design envelops.

To efficiently coordinate the materials development and qualification activities within the VHTR Materials PA, a detailed Project Plan (PP) has been developed under the guidance of VHTR Materials Project Management Board (PMB) that is anticipated to be formally approved and implemented early in 2009. The PP includes three work packages that cover experimental and analytical activities on graphite, high-temperature metallic materials and design methods, and ceramics and composites being conducted from 2007 through 2012 by all partners, as well as identifying their contributions of protected historical information. Deliverables in each of these three areas include both individual technical contributions from the GIF partners (*e.g.*, individual sets of data on mechanical or thermo-physical properties for a particular grade of graphite) and multinational products (*e.g.*, a joint report summarizing experimental data and analysis of the micro-structural stability of Ni-base super alloys in a VHTR helium environment). Contributions with a total value of well in excess of $ 200 M have been identified by the signatories to the VHTR Materials PA.

Materials working groups, comprising technical experts from each GIF signatory, are responsible for coordinating the input to each of the three work packages and advising the VHTR Materials PMB on the technical sufficiency and monetary value of the contribution from each signatory to ensure appropriate progress is made and shared by all partners. Annual review of all work plans and contributions will be made.

III. VHTR GRAPHITE STUDIES

The graphite components of the reactor include the permanent inside and outside reflectors, the core blocks, and the core supports. New graphite grades that are anticipated to show good performance under VHTR in-service conditions are being procured. New fine-grained isotropic graphite types with high strength and low irradiation damage are required to achieve high outlet-gas temperature, long life and continuity of supply. Extensive irradiation and properties test data are needed to qualify the new materials. The reference materials for the side reflectors and core support blocks may be UCAR PCEA or SGL NBG-17 or NBG-18 graphite grades, though several other graphite grades are being considered. At the current time, NBG-18 has been selected for the South African PBMR and IG-110 has been selected for the Chinese HTR-PM, as well as the Japanese GTHTR300C, reactors. Either PCEA or NBG-17 is suitable for use in prismatic reactors, but no vendors or other VHTRs have selected either of these grades at this time.

Participating signatories of the VHTR Materials PA are coordinating the acquisition, management and traceability of candidate nuclear graphites to optimize their overall graphite qualification activities and data generation needed for mechanical, thermo-physical and fracture properties. Such data are being developed as a function of temperature from 25-1 600°C, as well as for graphite oxidation kinetics and the effects of oxidation on relevant mechanical and physical properties in both He-coolant and air. The variations of properties with specimen volume, orientation, position within billet, between billets, and between lots are being addressed.

Effects of neutron irradiation on dimensional changes and properties are being assessed. Data will also be generated for the irradiation-induced creep rate and creep coefficients over relevant dose and temperature ranges. The mechanism of displacement damage in graphite via particle irradiation will be examined in comparative particle irradiation studies to elucidate the differences in behaviour of various graphite grades. Mathematical and mechanistic models are needed to allow interpolation and extrapolation of irradiation effects data. Hence, models for irradiation-induced dimensional changes, thermal conductivity, strength, fracture behaviour, and irradiation-induced creep are being developed, as are stress analysis codes and finite element models for modelling the stress states in components and predicting failure. A particularly valuable example of collaboration among GIF partners is provided in Figure 1, where coordinated individual contributions of irradiation experiments to meet design requirements are collectively displayed.

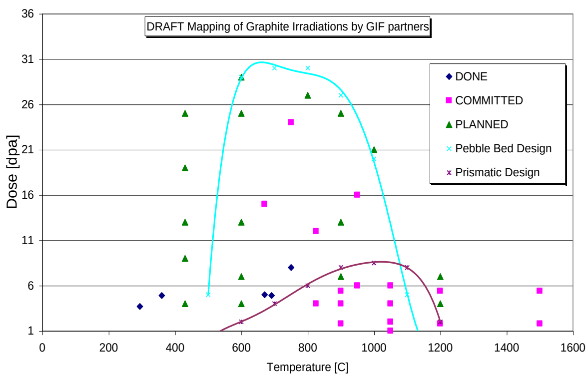

Figure 1: Comparison of graphite irradiations by GIF partners with design needs.

It can be seen that the operating temperatures for graphite in prismatic designs extends to a slightly higher range than for pebble bed designs, but the biggest difference in operating conditions between the two designs is the much higher irradiation dose to which graphite immediately adjacent to the pebbles is subjected. To address the collective set of data needs for both designs, participants in the VHTR Materials PA have jointly agreed to develop data that will cover full range needed. Early results will address the lower doses anticipated for the prismatic designs, since very long irradiation exposures are required to reach the highest doses typical of pebble bed operation.

Consensus design codes [*e.g.*, American Society of Mechanical Engineers Boiler and Pressure Vessel Code (ASME) and Japan Society of Mechanical Engineers Codes for Power Facilities (JSME)] are needed for graphite core structures and consensus test methods [*e.g.*, ASTM International and International Organization for Standardization (ISO)] are needed for nuclear graphite property determinations. The ASME and JSME have begun to establish such design codes including rules for materials selection and qualification, design, fabrication, testing, installation, examination, inspection, and certification and to prepare reports guiding manufacture and installation of non-metallic internal components for fission reactors. Development of ASTM standards for nuclear graphite materials specifications and a wide range of mechanical, thermo-physical, and fracture testing standards is underway. Existing French and German [Deutsches Institut für Normung (DIN)] standards are being also being updated and adopted as ISO standards. Participation in these codes and standards developments is an active component of the GIF VHTR materials program.

IV. VHTR METALS AND DESIGN METHODS STUDIES

Metallic materials will be needed for several reactor sub-systems including: the reactor pressure vessel, high-temperature metallic core internals, hot ducts and other pressure boundary components for the primary coolant system; the

Intermediate Heat exchanger (IHX) coupling the reactor to secondary systems; and the turbo-machinery, heat recuperators, and steam generators used for electric energy production.

This R&D program has been defined according to service temperature conditions.

- Low-temperature materials T<650°C for the reactor pressure vessel and other structural parts, including both qualification of materials typically used in LWR systems under VHTR system conditions, as well as higher temperature alloys suitable for service under conditions where time-dependent processes such as creep and creep-fatigue are significant.

- High-temperature materials, notably for metallic reactor internals, intermediate heat exchangers, and steam generators.

Characterization of materials and welds will be performed for relevant service conditions for each class of materials.

- High-temperature mechanical properties (*e.g.*, tensile, creep, creep fatigue, stress-rupture, high and low-cycle fatigue, fracture toughness) in both air and impure helium environments, as well as following irradiation exposure.
- Environmental degradation processes from exposure to high-temperature helium with contaminants such as CO, CO_2, H_2, H_2O, and CH_4.
- High-temperature metallurgical stability (*i.e.*, thermal aging effects).

Much of the R&D to be performed under the current GIF VHTR Materials PA will be for service conditions between about 350°C<T<900°C and will focus on traditional LWR low-alloy pressure vessel steels, such as A533B and A508, under the longer times and slightly higher temperature required for VHTR pressure boundary applications, as well as super alloys, such as Hastelloy X, 800H, IN617, and Haynes 230 for IHX, steam generator, and high-temperature internals applications. Longer-term R&D will include both development of materials for very high-temperature service beyond 900°C (*e.g.*, oxide dispersion strengthened alloys, refractory-based and advanced super alloys), as well as the qualification of existing materials for nuclear service at intermediate temperatures, such as modified 9Cr-1MoV for higher temperature pressure vessels.

At the current time, LWR pressure vessel steels (*e.g.*, A533B and A508) have been selected as the pressure vessel material of choice for all the VHTRs currently under development. Concerns about availability of large forgings, and the still unproven commercial capability to fabricate the very large ingots they require without macrosegration from advanced pressure vessels steels such as modified 9Cr-1MoV, have led to engineering approaches (*i.e.*, vessel cooling or insulation) to ensure the operating vessel temperatures are low enough to use the LWR steels. The greater challenge for the LWR steel use is their ability to survive potential short-term high-temperature excursions related to loss of coolant flow. Some studies have indicated that the current time-temperature limits for A533B and A508 in ASME Code Case N-499 may be exceeded during such transients. Moreover, the operating temperatures limit of 371°C assumed for time-independent ferritic steel operation may require additional assessments and/or justification of potential creep and creep-fatigue effects to reach the 600 000 hour operating lifetime desired for the vessels.[5]

The research on modified 9Cr-1MoV and its associated weldments illustrate significant synergism in the R&D among signatory members. Many different aspects of this material's behavior and design methods have been addressed by different signatories, resulting in a collectively developed compendium of data and design methods that are critical to its deployment in potential VHTR designs. These efforts include:

- tensile and cyclic mechanical properties;
- creep, creep-fatigue and creep crack-growth data;
- fracture and charpy impact toughness;

- environmental effects due to impure helium, oxidation, and irradiation;
- microstructural evolution and thermal aging for long-term service;
- allowable design and operational stresses, and
- design rules for creep-fatigue interaction and negligible creep conditions.

Candidate materials for IHX and steam generator applications must have a combination of high-temperature strength and corrosion resistance in the impure helium typical of gas-cooled reactor environments. Wrought high-Ni creep-resistant alloys containing 20 to 22 wt% Cr are creep resistant and offer protection against oxidation up to about 900°C by formation of chromia scale, however none are fully qualified in ASME code for HTGR nuclear applications and will require additional qualification data. Only Alloy 800H, is currently ASME Code qualified for high-temperature nuclear service and then only to 762°C.

	Ni	Cr	Mn	Co	C	Fe	Ti	Al	W	Si	Mo
Inconel 617	base	22.0	0.40	12.0	0.10	2.0	0.40	1.2	-	0.40	9.0
Haynes 230	base	22.0	0.65	5.0	0.10	3.0	-	0.30	14.0	0.50	2.0
Alloy 800H	32.0	21.0	1.00	-	0.06	bal	0.40	0.40	-	0.60	-
Hast X	base	22.0	1.00	1.50	0.10	18.5	0.15	0.50	0.60	1.00	9.0

Table 1: Composition of principal high temperature alloys for VHTR IHX and steam generator applications.

Inconel 617 and Haynes 230 are the leading candidates for application above 800°C, as they have greater strength at these temperatures. Haynes 230 appears to have slightly higher corrosion resistance in VHTR helium environments, but the much greater database for Inconel 617 and the existence of a well developed draft ASME Code case have led most designers to favor the use of 617 for higher temperature applications. Below 800°C, the lower cost and Code status of Alloy 800, are advantageous. A special variation of Hastelloy X developed by the Japanese (Hastelloy XR) with tighter controls on some alloying elements appears to offer greater environmental resistance to VHTR He, and has been used in the Japanese High Temperature Test Reactor (HTTR) IHX at operating temperature of 950°C for short periods, and is the current choice for the IHX of their advanced GTHTR300C reactor.

Another area that illustrates the benefits of the combined work from the signatory members is in the R&D efforts on Alloy 617. It is a candidate material for the very high temperature metallic components such as the intermediate heat exchanger and the hot ducting in potential VHTR designs. Progress has been made in the following areas:

- tensile and cyclic mechanical properties;
- fatigue, creep rupture, and creep-fatigue data;
- environmental effects due to impure helium, oxidation, and irradiation;
- microstructural evolution and thermal aging for long-term service;
- implications of deformation mechanisms for long-term service conditions; and
- viscoplastic constitutive models to support design analysis methods.

V. VHTR CERAMIC AND COMPOSITE STUDIES

Ceramics and structural composites are regarded as backup or advanced solutions to metallic materials challenges for several VHTR components because of their superior high-temperature strength or radiation resistance. Key areas for collaborative studies on these materials focus on their use for heat exchangers, control rods, insulation materials, and internals structures such as restraints and fasteners.

V.A. Structural Composites

Carbon fiber reinforced carbon (C/C) composites with useable service temperatures up to 1 800°C and other ceramic composite materials (for example, SiC/C and SiC/SiC) have been proposed for the several internal subcomponents in the near term and for the control rod assembly in the longer term. The C/C and SiC/SiC composite and ceramic materials are relatively new reactor materials for which irradiation and other material properties data are needed. Standardization and codification of materials from within these industries are also major issues that will need to be resolved for use of these materials in reactor safety-related systems.

Mechanical and thermal property, fracture behaviour, and other tests, including oxidation effects and post-irradiation evaluations as a function of fabrication methods are required to establish design guidelines and a design database. The modelling of the material behaviour and stress analyses in these codes will need to consider the anisotropic nature of these materials. Obtaining non-destructive testing data and fracture toughness data is necessary to establish acceptance guidelines.

V.B. Ceramics

High-temperature fibrous insulation may be used throughout the reactor and power conversion systems, notably in the hot duct, upper plenum shroud, shutdown cooling system, helium inlet plenum, and turbo-compressors. Ceramic insulation blocks may be needed under the graphite core support structure. Insulating materials that retain resiliency minimize off-gassing, and do not shed particulate under high gas flows and irradiation damage are needed. All these materials need to be fabricated, tested, and qualified for use under VHTR conditions. Qualification of non-metallic materials will require, in some cases, the development of recognized industry standards and codes for materials and testing.

Current activities include evaluation of composites for in-pile and out-of-pile components including advanced control rods, as well as for stabilizing straps and ties for the core. Evaluation of mechanical and thermal properties and the dimensional stability for both C/C and SiC/SiC composites for un–irradiated materials and at irradiation doses up to about 10 dpa for C/C and over 20 dpa for SiC/SiC composites is ongoing by multiple GIF members.

VI. MATERIALS MODELING

After the introduction of quantitative descriptions of creep and creep-damage mechanisms in metals in middle of the last century (*e.g.*, collective work by Norton, Kachanov, Monkman-Grant, etc.), it took about 25 years to develop a working engineering understanding of creep-fatigue interactions (*e.g.*, collective work by Manson, Coffin, Mowbray, etc.). The introduction of damage mechanics in terms of subcritical crack growth and the introduction of constitutive laws for creep-fatigue interactions (*e.g.*, Chaboche) was a further improvement in lifetime assessments of structures. With the current availability of huge computer clusters operating in parallel mode, numerical solutions of equations for atomistic behavior became very attractive. Although it is well accepted that damage starts at atomistic levels, it is not easy to bridge the gap between atomic and structure levels and requires an understanding of the related physical phenomena on a range of scales from the microscopic level all the way up to macroscopic effects.

Determination of the life-time of components exposed to severe environments such as in VHTRs is very demanding, particularly when damage interactions (like creep-irradiation or strength-microstructure, toughness-irradiation induced phases) must be considered. The simulation of materials behaviour under such extreme conditions needs to encompass broad time and length scales from atomistic descriptions of primary damage formation to a description of bulk property behavior at the continuum limit. This requires a multi-scale, multi-code modelling approach that begins at the atomistic level with *ab initio* and molecular dynamics techniques, moves through the meso-scale using reaction rate theory models, lattice kinetic Monte-Carlo and Dislocation Dynamics, and ends with the macro-scale using Finite Element methods and continuum models.[6] Experimental validation of the modelling results is mandatory. This approach is schematically shown in Figure 2.

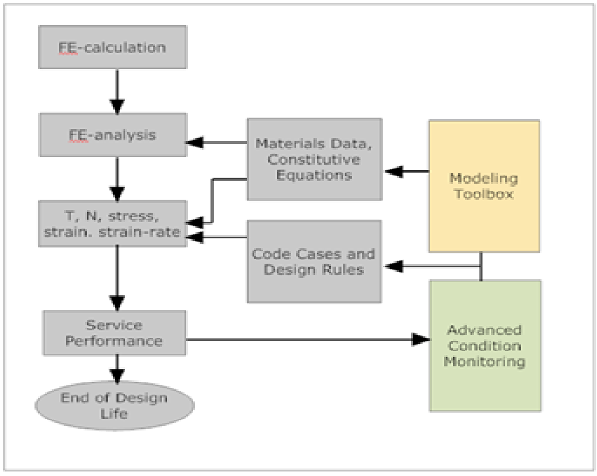

Figure 2: Illustration of approach for interactive support of materials modelling in reactor component design.

The necessary time required for the development of advanced Generation IV systems can provide the necessary lead time to allow such a multi-scale modelling approach to provide useful input to both qualification of existing materials and the development of newer, higher performance ones.

Towards that end, modelling activities will be conducted to support and interact with the experimental and technological part of the studies within the VHTR Materials PA. It is anticipated that this will strongly support the development of a mutual understanding of the design needs and solid-state physics necessary for improved assessments of long-term materials performance.

VII. METHODOLOGY & CODIFICATION OF HIGH-TEMPERATURE MECHANICAL DESIGN RULES

Cumulatively, the results of the materials R&D activities being performed under the VHTR Materials PA will provide input for the improvement of codes and standards needed for VHTR plants. Moreover, modelling and description of materials behaviour and damage development will provide an enhanced scientific basis for the codification work and damage assessments. Improvements of existing high-temperature design methodology, including structural design methods, materials testing and database developments, and nuclear design code and standards (such as the ASME or JSME Codes) must consider the:

- extension of design code approvals for metallic materials at higher operating temperatures and longer service lifetimes;
- development and approval of design code for graphite, composite, and ceramic materials in nuclear service.

Similarly, materials test standard development and approval to obtain qualified advanced materials properties by organizations such as ASTM or ISO must include the development of approved testing standards for thermal, physical, mechanical, and fracture properties of graphite and advanced composites.

VIII. MATERIALS DATABASE

The development of VHTRs requires extensive materials data on metals, graphite, ceramics, and composites. To efficiently manage the materials data and facilitate coordinating international activities, it was recognized that a materials property database that provides an authoritative single source and is internally consistent, validated, and highly qualified is crucial to the success of the program. Hence, a dedicated database has been developed and will be used to retain and assemble data provided by GIF members, data available in the literature, and other resources. It is web-based with highly secure access and will be used to coordinate the extent of testing with minimum redundancy; track and exchange results of various types of testing, test conditions, product forms, and metallographic information; assess and rank quality of test data; and preserve data for current and future use.

IX. CONCLUSION

To address the extensive needs for development and qualification of structural materials to support VHTR systems, a formalized international program for generation, exchange, and coordination of materials data has been established under the GIF framework. Within that framework, collaborative research on graphite, high-temperature metals, and ceramics and composites is being conducted. Results from this research are being exchanged among the participants and form the basis for augmenting the materials and design codes and standards needed for VHTR system deployment.

Nomenclature

ASME	American Society of Mechanical Engineers Boiler and Pressure Vessel Code
ASTM	ASTM International
C/C	Carbon fiber reinforced Carbon Composite
GIF	Generation IV International Forum
IHX	Intermediate Heat exchanger
LWR	Light Water-cooled Reactor
VHTR	Very High Temperature Reactor
PA	system-specific Project Arrangement
PMB	Project Management Board
PP	Project Plan
R&D	Research and Development
SiC/C	SiC fiber reinforced carbon composite
SiC/SiC	SiC fiber reinforced SiC composite

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