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Materials Research Needs for Advanced Reactors

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ABSTRACT

Metallic and graphite components in advanced high-temperature gas-cooled reactors (HTGRs) may experience creep, fatigue, oxidation, aging, corrosion cracking, irradiation damage, and dimensional changes. The safety designs of these reactors, such as the pebble bed modular reactor (PBMR) and the gas turbine modular helium reactor (GT-MHR), depend on the long term integrity of components needed to maintain pressure boundary integrity, core geometry, adequate cooling of the core, and reactivity control and shutdown systems. Failure could result in air, water and/or steam ingress, and accompanying adverse consequences. In Advanced Light Water Reactors (ALWRs), the operating conditions, materials, and coolant environments are similar to those of conventional LWRs. Nevertheless, there may be a need for new research in the materials area specifically for ALWRs. This paper discusses the information gaps that exist in terms of analytical tools and data shortcomings and describes research needed to establish an acceptable technical understanding of the behavior of metallic and graphite components in advanced reactor environments.

KEY WORDS: advanced reactors, high temperature, gas cooled, metals, graphite, inservice inspection, research needs, degradation, environment, impurities, codes and standards, fatigue, creep, creep-fatigue, oxidation, thermal aging, stress corrosion cracking, crevice corrosion cracking, irradiation damage, dimensional changes.

INTRODUCTION

A key research area important to the safety of advanced reactors is the behavior of metallic and graphite components. These components are relied on for structural, barrier, and retention functions during normal and off-normal reactor conditions. Therefore, sound technical bases must be available for evaluating expected lifetime and failure modes of reactor pressure vessel materials and components whose failure would result in loss of core geometry and/or ingress of air, water, or steam to the pressure boundary. In the HTGR design, high-temperature materials are required to maintain core geometry, adequate cooling of the core, access for reactivity control and shutdown systems and, in the case of the Pebble Bed Modular Reactor (PBMR), a defueling route. This paper emphasizes the need for research to establish a technical understanding of the metallic and graphite components under high-temperature operating and accident conditions.

The licensing approach for HTGRs used by the U.S. Nuclear Regulatory Commission (NRC) to independently confirm design and support safety evaluations relies heavily on the use of probabilistic risk assessment (PRA). Information from the materials research area is needed for conducting PRAs. Since failure probability data for components of advanced reactors are not available from experience, the information can be developed from materials research on potential degradation processes and quantification of their progression. Evaluations of component service life, safety margins, and behavior under accident conditions are dependent on environmental factors, such as temperature, pressure, coolant composition, and fluence.

The operating conditions, materials, and coolant environments used in ALWRs are similar to those of conventional LWRs. A large body of research data, from both the U.S. and Japan, has shown a detrimental effect of the coolant environment in reducing the fatigue life of LWR components. Methods have been developed and are widely available in the literature (NRC NUREG reports and Pressure Vessel Research Council (PVRC) report) for taking into account the effects of the operating environment in the fatigue design of components. Although the American Society of Mechanical Engineers (ASME), through its on-going code activities, is addressing the issue of the effects of the environment, it has not yet incorporated changes in its design rules and correlations. Therefore, during design and review of ALWRs, caution must be exercised to ensure that the effects of the environment are appropriately accounted for in the fatigue design and evaluation of components. The ASME should continue to update its rules for fatigue design of components. In addition, design rules for advanced reactor designs may need to incorporate different materials and correspondingly different deformation mechanisms.

Several aspects of the HTGR and ALWR designs raise the potential for the need for improved inservice inspection (ISI) programs and for continuous monitoring. More components are enclosed in pressure vessels making access for inspection difficult, and there are longer operating cycles between scheduled, short-duration, refueling outages when inspections can take place. This suggests a need for evaluating the effectiveness of the less frequent ISIs for timely detection of cracking and degradation of components and the potential for excessive growth of cracks before the next ISI. To maintain safety, the use of continuous online monitoring techniques for structural integrity and leakage detection may be required.

RESEARCH PURPOSE

The advanced reactor designs are different from traditional LWRs in terms of the materials used, such as high-temperature metals and graphite; higher coolant temperatures; a coolant that does not change phase; different degradation mechanisms such as creep; and expected behavior of the components in this environment. This departure thrusts the materials – environment combination into a regime where more information is needed to define long term behavior and safety margins.

In HTGRs, graphite acts as a moderator, reflector, major structural component that will provide channels for the fuel, coolant gas, control and shutdown rods, and a thermal and neutron shield. Additionally, graphite components are employed as supports. Graphite also acts as a heat sink during reactor trip and transients. During reactor operation, physical properties of graphite are significantly modified as a result of temperature, environment, and irradiation. Significant internal shrinkage, bowing, and stresses can develop which may cause component failure, and/or loss of core geometry. When graphite is irradiated to a very high radiation dose, ensuing swelling causes rapid reduction in strength and loss of structural integrity. Additional loss in life time could result from slow crack growth (SCG) effects. In the event of an accident allowing air ingress, subsequent graphite oxidation causes further changes in its physical and mechanical properties.

Regarding metal components, research had progressed through the 1980s on the high-temperature design (creep, fatigue) for the Liquid Metal Fast Breeder Reactor. This research formed the basis for some ASME code cases and requirements for the design of high-temperature components. The NRC staff has initiated a program to review and evaluate this research and that which has progressed since the 1980s/1990s, in particular with respect to the temperatures, coolant environment, and materials to determine applicability to current HTGR designs.

Development of a research capability in the materials area beyond the licensing basis is needed to understand safety margins, failure points, and reduce uncertainties. To conduct independent PRAs of advanced reactors, information is needed on the probability of failure of various reactor components. Because of the lack of operating experience, this information will have to be developed analytically using probabilistic fracture mechanics. Thus, potential degradation mechanisms of metallic and graphite components need to be identified and progression of degradation quantified under the operating reactor conditions. Potential technical issues that need to be addressed are: (1) national codes and standards for design and fabrication of metallic and graphite components for service in HTGR helium environments; (2) appropriate data bases for calculating fatigue, creep, and creep-fatigue interaction lifetimes of components in high-temperature applications; (3) the effects of impurities, including oxygen, in the high-temperature helium on degradation of components; (4) sensitization and aging behavior of alloys during elevated temperature exposures; (5) treatment of pipe as a vessel in a HTGR; (7) degradation by carburization, decarburization, and oxidation of metals in HTGRs; (8) issues related to inspection of HTGR and ALWR reactor components; (9) long term performance and degradation of graphite and new reactor pressure vessel materials under high levels of irradiation; (10) modeling and methodology that predict irradiated graphite properties from non-irradiated properties; and (11) comprehensive understanding of the governing rates and mechanisms for the oxidation of reflector grade graphite, fuel pebble matrix graphite, and graphite dust. Each of these potential technical issues is addressed in the following sections.

CONSIDERATIONS FOR METALLIC COMPONENTS

Research on metallic components will need to be conducted to evaluate and quantify degradation processes, metallurgical aging and embrittlement, carburization, decarburization, nondestructive examination, and ISI. In addition, currently available (international) procedures for design against fatigue, creep, and creep-fatigue will be reviewed and evaluated. The objective of this review is to evaluate current code design rules and procedures and to provide input for improvements as necessary. The best procedures will be updated to incorporate correlations developed from more recent research.

The availability and acceptability of national codes and standards for the design and fabrication of metallic components for service in HTGRs and ALWRs is a key issue. For high temperatures, background studies and activities for eventual development of codes and standards were conducted in the 1980s for application to the liquid metal breeder reactor. Of particular note is the work conducted by the PVRC in their preparation of several technical reports that provided the basis for development of high-temperature design codes by the ASME. These reports give background and procedures for design of components to resist fatigue, creep and creep-fatigue failures. However, the effects of the helium environment, including the presence of impurities such as oxygen, were not addressed. In addition, improved correlations for creep and creep-fatigue have been developed from research of the 1990s. These improvements are not included in the PVRC reports and the procedures need to be updated before they are included in National Codes and Standards.

Another area of codes development has been taking place internationally for the Advanced Candu Reactor (ACR). The pressure tube material for this ALWR is not covered by the ASME codes and standards. The Canadian Standards Association (CSA) has published rules to complement those of ASME. The CSA codes have followed the ASME code where applicable, but augmented the code as necessary to include zirconium alloys for pressure tube in a reactor with on-power refueling capability. Activity should be undertaken to review and evaluate incorporation of the new materials and environments covered by the CSA codes into the ASME code.

Although methodologies could be assembled from existing knowledge for calculating fatigue, creep, and creep-fatigue lives of components in high-temperature applications, appropriate data bases are needed for these calculations. Based on past experience and research, we have found that environmental effects play an important role in reducing fatigue lives and in enhancing degradation of materials. For example, small levels of impurities, such as less than 1 part per million of oxygen in the high purity water coolant of LWRs, can greatly decrease fatigue life and resistance to stress corrosion cracking of metallic components. These effects were not originally addressed in the ASME Code. For example, the design data for fatigue was obtained from materials tests in air. Because helium is inert, there has been a tendency to obtain design data in pure helium; in impure helium, but not all impurities included; or in air. The effects of all important impurities, such as oxygen, in helium need to be taken into account with respect to reductions in fatigue and creep life and such data and understanding need to be developed. Environmental effects on fatigue under ALWR operating conditions need to be addressed as well.

To address these concerns research will be conducted on the effects of an impure helium environment, especially the effects of oxygen, temperature, and strain rate, on the fatigue life of HTGR metallic components. Similarly, the effects of impure helium environments on the creep and creep-fatigue life of HTGR components will be investigated. The objective of this research is to ensure that the design rules and procedures available address reductions in life due to the operating environment. If the codes and procedures do not consider these phenomena, then the data base developed can be used to update the codes and procedures to provide design procedures and rules that avoid failure of HTGR components during service. In addition, research will need to be conducted to quantify the effects of carburization and decarburization on the reduction of fatigue and creep life to ensure that these reductions are accounted for in the design procedures and analyses.

To address degradation and aging of metals in HTGRs, the effects of high-temperature helium with impurities including oxygen at levels present in HTGRs need to be evaluated with respect to stress corrosion crack initiation and growth rate, crevice corrosion crack initiation and growth rate, and cyclic crack growth rate. Low levels of impurities in high-temperature, high purity aqueous environments are known to cause these types of degradation and to accelerate the crack growth rates. Therefore, research will be conducted on the effects of the high-temperature helium environment containing impurities, including oxygen, at levels typical of HTGRs on stress corrosion crack initiation and growth rates, crevice corrosion crack initiation and growth rate, and cyclic crack growth rate. The tests will be conducted on materials in the as-received condition and in carburized and decarburized conditions. The objective of this research is either to confirm that these degradation mechanisms do not occur and crack growth rates are not enhanced in the environments of interest or to quantify the crack initiation times, quantify increases in growth rates, and define the environmental conditions under which these occur.

Many alloys undergo solid state transformation and precipitation during elevated temperature exposures. These transformation reactions are known as aging and can lead to embrittlement of the alloy. Aging and embrittlement occurs, for example, in cast stainless steel components under temperatures and time conditions experienced in operating LWRs. At the operating temperatures of HTGRs, the reaction rates can be much higher, (i.e., the aging and embrittlement would occur sooner). The different alloys and higher temperatures of HTGRs would indicate potentially different aging reactions and mechanisms, some of which could occur relatively rapidly and render the material embrittled and susceptible to cracking. The aging reactions, as a function of time and temperature, in the different alloys used in important components of HTGRs need to be studied to establish the potential for material property degradation and embrittlement during the lifetime of operating HTGRs.

Another solid state reaction that occurs in stainless steels (and austenitic alloys) is called sensitization. Sensitization is caused by the precipitation of chromium carbides at the grain boundaries of the stainless steel. This precipitation normally occurs during slow cooling of the metal through high temperatures such as when cooling from the high temperatures following welding. Formation of the carbides depletes the chromium from the grain boundary areas rendering the stainless steel susceptible to intergranular stress corrosion cracking (cracking along the grain boundaries) in oxidizing and impurity environments. A less well known method for producing sensitization is through low-temperature sensitization. This occurs over long periods of exposures to relatively low temperatures. Low-temperature sensitization in stainless steel has been studied under temperature conditions relevant to LWRs. Under these conditions, low-temperature sensitization would not occur in times less than 40 years. However, the sensitization rate is exponential with temperature, and at the higher operating temperatures of HTGRs, there is a potential for sensitization during the lifetime of these plants thus rendering the stainless steel components susceptible to stress corrosion cracking.

Thermal aging and sensitization research will need to be conducted on high-temperature alloys used in HTGRs on samples in the as-received and the welded conditions. Samples will be exposed for different times to temperatures at and above the operating temperatures of the HTGR components. Exposure to higher temperatures will provide acceleration in the aging and sensitization reactions. As long as the aging mechanisms at the higher temperatures are the same as at the operating temperatures, correlations can be developed for quantifying the times required to reach different levels of aging and sensitization at the operating temperature. Mechanical property testing will be conducted on the aged samples to quantify the degree of embrittlement and other property changes as a function of aging time and temperature. Metallographic and microscopy studies will be conducted to identify the aging and precipitation reactions if they occur, to ensure that the reactions are the same at the operating and higher temperatures, and to evaluate the potential for and degree of low temperature sensitization. The objective of the research is to identify the potential and the degree to which thermal aging, embrittlement, and sensitization can occur during operation of HTGRs and to evaluate the impact of these changes on the structural integrity of reactor components.

In HTGR designs, the connecting pipe which carries hot helium from the core to the power conversion system is treated as a vessel because this pipe is designed, fabricated, and inspected to the same rules as a reactor pressure vessel. The consequence of this assumption is that a design basis double ended break is not considered for the connecting pipe, and therefore, no mitigating systems are incorporated in the design. Considering this pipe as a vessel will require further investigation, because the pipe is of much smaller diameter and therefore, much thinner wall than a reactor pressure vessel designed to the same working pressure. If an unexpected degradation mechanism should initiate in the pipe because of the thin wall, it can propagate through the wall in a relatively short time and possibly not be detected by ISI. Conversely, if an unexpected degradation mechanism were to initiate in a pressure vessel, it would require long times to propagate through the greater wall thickness, allowing enough time to be detected by ISI.

Carburization, decarburization, and oxidation of metals in HTGRs are other phenomena that can lead to degradation caused by the operating gaseous and particulate environment. Carburization is a phenomenon where carbon, either as a particulate or from carbon containing gases, diffuses into steel to form a surface layer with high carbon content. This surface layer may be hard, brittle, and have higher strength than the substrate. Differences in strength and other physical properties between the surface layer and substrate may lead to high stresses in the surface layer when the component is under load. In addition, carbides may form in the high carbon surface layer of stainless steel leaving the matrix depleted of chromium and susceptible to stress corrosion cracking and oxidation. Cracking, stress corrosion cracking, and oxidation can more easily develop in the surface layer which could then propagate into the component.

Decarburization is a process whereby carbon is depleted from the steel depending on the composition of the gaseous environment. Depletion of carbon results in a softer steel and in reduced fatigue and creep lives. The presence of oxygen results in the formation of scale and general corrosion of metallic components, and more importantly, it can oxidize the graphite and render metallic components susceptible to stress corrosion cracking. To control the phenomena of carburization, decarburization, and oxidation, a very careful control of the level of different impurities in the coolant is required. Conditions that lead to avoidance of one of the above phenomena can lead to development of another. For example, to avoid carburization, some HTGRs might use slightly oxidizing conditions by addition of oxygen to the gas stream. However, this can lead to oxidation of graphite, general corrosion of metals and an increased susceptibility to stress corrosion cracking. Some research has been conducted to study the phenomena described above; however, additional confirmatory research is needed to better define the conditions under which the phenomena occur for important metallic components of HTGRs. In addition, much of the available research did not include oxygen in the gaseous environment. Since oxygen will be present in HTGRs at high enough levels to affect the progression of the above phenomena and to reduce fatigue life, creep life, and resistance to stress corrosion cracking, oxygen needs to be included in new experimental studies.

Carburization, decarburization, and oxidation of HTGR high-temperature metals will need to be studied as a function of time and temperature in helium gas with impurities, including oxygen. Different levels and ratios of impurities will be studied. Metallographic studies and mechanical testing will be conducted on the exposed samples to determine the degree of deterioration and loss of strength. The objective is to define the environmental conditions under which the phenomena can occur, to what degree they occur under the different conditions, the potential for occurrence under the operating conditions of HTGRs, and the significance on structural integrity of components.

A number of potential degradation and aging mechanisms in the operating environment of HTGRs have been discussed. There is an opportunity to evaluate and validate these potential degradations by conducting research on components removed from operating reactors. The AVR HTGR operated for over twenty years in Germany. An international research program will need to be conducted on components removed from the AVR, including microstructural studies and mechanical tests. Microstructural studies are called for to determine if solid state changes and precipitation have occurred during operation to produce thermal aging, sensitization, carburization, and decarburization. In addition, metallographic studies will establish if stress corrosion cracking, crevice corrosion, general corrosion, and oxidation have occurred. Mechanical tests on materials removed from the AVR can be conducted to determine if any degradation in materials properties has occurred. Fatigue and creep tests will determine if fatigue and/or creep damage have occurred, if the design codes and methods correctly predict the damage, and if the coolant environment had an effect in reducing fatigue and creep lives. The results will help determine if and how the design codes/procedures need to be changed to take into account the potential degradation mechanisms.

With respect to international cooperation, there is considerable research that has been performed or is ongoing in the European Community (EC) and Japan on high-temperature metals for HTGRs. Through interactions with technical staff in the EC and Japan, the NRC staff identified several areas that address NRC research objectives. Work of interest in the EC is (a) review of RPV materials, focusing on previous HTRs, in order to set up a materials property database on design properties, (b) compilation of existing data on materials for reactor internals and selection of the most promising alloys for further development and testing, and (c) compilation of existing data on turbine disk and blade materials and selection of the most promising alloys for further development and testing. Experimental work in these areas includes a) research on a pressure vessel steel containing 9% Cr (irradiation testing, fatigue, creep-fatigue, tensile, fracture toughness); both heavy-section base metal and weldments are included in the studies; b) mechanical and creep tests of candidate alloys for reactor internals at temperatures up to 1100^{o} C with focus on the control rod cladding; and c) tensile, fatigue, and creep tests from 850^{o} C up to 1300^{o} C for two different turbine blade materials, one forming an aluminum oxide protective layer, the other a chromium oxide layer.

Work of interest that has been conducted by Japan Atomic Energy Research Institute (JAERI) includes development of a high-temperature metallic component design guide, research on high-temperature metal corrosion, and irradiation effects on a 2 1/4 Cr-1Mo reactor pressure vessel steel.

Other international efforts may include determining the long term degradation mode of glass fiber encased insulation components. This phenomenon has been identified in the UK gas-cooled reactors and discussed at the workshop on HTGR safety and research issues (October 2001, US NRC, Rockville). The objective would be to conduct studies of the effects of vibrations and service conditions to determine the reliability of this insulation since it protects the metallic components and pressure boundaries in the HTGR designs from unacceptably high temperatures. With regard to new materials and environments in the ALWRs, international efforts to augment the ASME codes and standards with the CSA codes and standards should be explored.

Some of the key work not fully addressed in the international programs is in the areas of a) effects of the helium environment with impurities on degradation of materials and b) aging and sensitization. NRC research results in these areas would be a valuable contribution to cooperative international programs.

DESCRIPTION OF ISSUES AND RESEARCH FOR INSERVICE INSPECTION AND MONITORING

ISI intervals may be long and the amount of inspection limited because HTGR and ALWR reactors are generally designed to operate for long periods of time between scheduled short-duration shut-downs for maintenance or refueling. Therefore, there is a need to evaluate the effectiveness of various ISI programs as a function of the frequency of inspections and the number and types of components inspected. Additionally, many internal components are not easily accessible for ISI, and the impact of not inspecting these components needs to be assessed. An alternative to conducting periodic ISIs during reactor shut-downs is to conduct continuous online, nondestructive monitoring for structural integrity and leakage detection of the entire reactor or reactor components during operation. Techniques for continuous monitoring have been developed, validated, and codified for use in LWRs. If ISIs of HTGRs and ALWRs cannot be conducted on a frequent enough basis and certain components cannot be inspected, then continuous monitoring may become necessary. The continuous monitoring techniques need to be evaluated and validated for the materials, environments, and degradation mechanisms of the HTGRs and ALWRs.

To address concerns in this area, research will be conducted on the impact of different ISI plans on structural integrity and risk. The key variables in the study will be the length of time between inspections, the reliability of the inspection methods, and the number of components and locations tested for HTGRs and ALWRs. Different degradation mechanisms appropriate to the reactor design and operating environment, along with the inspection variables, will be considered in probabilistic fracture mechanics analyses to evaluate the impact of potential failures on risk. Results of this work will be used to support the evaluation of proposed ISIs and to determine the technical basis for improved, more frequent, or more extensive ISIs. The results will also provide guidance on the need for continuous online monitoring of structural integrity.

Research will need to be conducted to evaluate continuous monitoring of reactor components for crack initiation and crack growth and for leak detection. Acoustic emission techniques will be used for laboratory testing of specimens under simulated HTGR and ALWR conditions (respective temperature, noise sources, coolant flow, etc.) to evaluate fatigue, creep, and stress corrosion cracking. Correlations will be developed for crack initiation and crack growth rates with the acoustic emission signals for the materials and environments of the HTGRs and ALWRs. Similar research was conducted by the NRC in the 1980s and 1990s where acoustic emission techniques were developed, validated, and codified for application to LWRs. The research, methods, and techniques for HTGRs and ALWRs will take advantage of the knowledge gained in earlier work. Similar acoustic emission techniques will be evaluated for detection, location, and quantification of coolant leakage from the pressure boundary and internal components under the operating conditions of HTGRs and ALWRs. Again, similar work was conducted for LWR applications and the research for HTGRs and ALWRs will benefit from this. Once the laboratory research is completed and correlations of acoustic emissions to crack initiation and growth developed, an operating or test HTGR will be instrumented with acoustic emission sensors and monitored during its operation to validate the methods and correlations developed in laboratory testing. The results from this work will provide an alternative to periodic ISIs and the advantages of continuous online monitoring of reactor structural integrity and leakage. The results will also provide technical data bases for incorporating the techniques into codes and standards.

In the case of the ACR design, a gas annulus surrounding each zirconium-niobium alloy pressure tube is continuously monitored for water content. Significant increases in the rate of water increase in the gas are an indication of pressure boundary leakage. The unique Candu reactor design allows this continuous on-line monitoring of the pressure boundary to augment periodic ISI. Experience with detecting water leakage through a tight crack in the pressure tubes prior to unstable crack growth and pressure boundary failure using this on-line monitoring technique may be adaptable to other ALWR designs.

Areas of international cooperation and exchange in this area would involve work planned by the EC on evaluation of ISI methods, and work on risk-informed inspection program evaluation by NRC. Of additional interest would be potential international cooperation on evaluations of online continuous monitoring techniques for structural integrity and leak detection using HTGR test reactors.

CONSIDERATIONS FOR GRAPHITE COMPONENTS

To be able to effectively review the new HTGR designs, there is a need to conduct confirmatory research and establish an information base related to the long-term performance and behavior of nuclear-grade graphite under the temperatures, radiation, and environments expected during normal operating and accident conditions. Potential loss of strength and of resistance to fatigue and creep due to shrinkage, swelling, cracking, and corrosion during operation could impact the performance and function of the graphite core structural elements, reflectors (side and bottom), and moderator balls. Additionally, any potential adverse SCG effects could limit the life time of graphite components. Various graphite variables, including coke source, size, impurity, and structure; manufacturing processes; density; grain size; crystallite size and uniformity, determine the as-received and irradiated properties of the graphite component.

Research will need to be conducted to evaluate graphite for HTGR application. This will involve studies of the performance and degradation of graphite under high levels of irradiation and temperature. A review will be conducted of available high dose irradiation data for nuclear grade graphite. High dose irradiation data on “old” graphites will be evaluated to determine its applicability to “new” graphites. The data will be utilized to determine the behavior of current graphites planned for HTGRs under operating conditions. In general, because there is a lack of data in the high dose, high-temperature regime of HTGR operating environment, additional research will be conducted on current graphites planned for HTGRs to determine high dose material behavior, properties, and degradation. Experiments will be conducted at various temperatures at high doses in a high flux test reactor. Microstructural evaluations, chemical analysis, dimensional measurements, mechanical testing, and physical property testing of the irradiated specimens will determine the effects of high dose and high temperature on property changes in “new” graphites.

Limited irradiation studies have been conducted on older graphites that are no longer available due to loss of raw materials supply and/or manufacturers. In addition, limited results are available at high levels of irradiation exposure. Thus, two key issues are the lack of data on irradiated properties of current graphites, and the lack of data at higher doses of irradiation. As discussed earlier, the irradiated material properties strongly depend on the particular make-up of the graphite and the manufacturing process; therefore, at issue is whether the irradiated materials properties of the “old” graphites can be assumed to be the same as the “new” graphites. Irradiation affects, and in many cases degrades, physical and mechanical properties of the graphite. Important properties that change with irradiation are density, thermal conductivity, strength, and dimensions. These changes have safety implications since they could degrade structural integrity, core geometry and cooling properties. Some of these changes are not linear with irradiation dose.

Strength of graphite initially increases with irradiation dose, then, at higher levels, it begins to decrease. With respect to dimensional changes, graphite initially begins to shrink with increasing dose then, beyond turn-around, graphite begins to swell with increasing dose. During operation, thermal gradients and irradiation induced dimensional and strength changes result in significant component stresses, distortion, and bowing of components. These can lead to loss of structural integrity, loss of core geometry, and potential problems with insertion of control rods. At still higher doses, beyond turn-around, where the swelling makes the volume considerably greater than the original volume, graphite structures will no longer be able to support design load.

To evaluate the suitability of graphite for HTGR application, property change data due to irradiation is needed in addition to the as-received properties. The development of irradiation data on graphite is difficult, expensive, and time consuming. Therefore, reactor designers/vendors have proposed to use radiation data from studies conducted on older graphites and attempt to use graphites produced in a similar manner. However, the as-received and irradiated graphite properties depend strongly on the raw materials and manufacturing processes. Small variations in these could have strong effects on the graphite properties. Since the exact raw materials and processes have changed and could continue to change in the future, the NRC has a need to independently confirm whether a particular graphite will behave in the same manner as the old graphites under operating irradiation conditions. To accomplish this without irradiation testing every time a change occurs in the graphite raw materials or processing, correlations are needed for predicting irradiated graphite properties and changes from the as-received graphite raw materials characteristics, composition, processing, and properties.

Research will need to be performed to determine irradiated graphite properties from as-received graphite properties. As-received graphite properties are determined by the raw materials and manufacturing process. Important parameters will be identified such as coke and pitch characteristics, and graphitization temperature. A number of different graphites will be selected with carefully varied parameters. Studies will be conducted to establish the as-received properties of the graphites. Selected properties to be measured are: x-ray crystallinity, density, open and closed porosity, pore size and shape distribution, grain size and size distribution, grain orientation and orientation distribution, thermal expansion, thermal contraction, thermal conductivity, absorption cross-section, sonic Young's modulus, stress-strain behavior, strength and strength distribution (Weibull modulus), and fracture toughness. In addition, chemistry, including impurities, of the graphites will be established. Due to the anisotropy of manufactured graphite, the materials properties will be determined for two orthogonal directions since graphite exhibits transverse isotropy. The graphites will then be irradiated at systematically varied irradiation doses and temperatures significant to HTGRs. Following irradiation, the materials properties will be reevaluated to determine the effect of irradiation and to establish correlations between the initial as-received properties and the post-irradiation properties that could apply to any particular graphite that may be used in HTGRs.

Corrosion and oxidation of graphite can occur in HTGRs from oxidizing impurities in (or added to) the helium coolant, from in-leakage during normal operation, or from air or water ingress during accidents. The oxidation of graphite is an exothermic reaction, and it is important to know the rate of heat generation particularly during accidents. Oxidation also will remove the surface layers of graphite components resulting in loss of structural integrity. Further, oxidation will change the thermal conductivity and reduce the fracture toughness and strength of graphite components. The oxidation rates vary for different graphites, and can be greatly affected by the impurities in the original graphite. Therefore, oxidation rate data is needed for the graphites proposed for new reactors.

Investigations will need to be undertaken to understand oxidation effects on the microstructural, physical, thermal, and mechanical characteristics of nuclear graphite. There is a lack of data on oxidation kinetics of reflector grade graphite. Experiments will be conducted to determine weight loss and loss of mechanical integrity due to oxidation of graphite samples. The heat generated from oxidation of graphite dust and the potential detrimental effect on surrounding components due to this elevated temperature will be studied.

The PBMR will use advanced gas-cooled reactor type fuel sleeve graphite for the replaceable and permanent structures in the core. The proposed graphite properties used for design, operating, and accident analyses of these structures will have the same values as those for the sleeves. The sleeves are relatively thin structures manufactured differently from the large structural blocks of the PBMR, and the mechanical and other properties could be different. Furthermore, the properties of the large block graphite will vary through the thickness of the block. The difference in properties between the sleeves and large blocks and through-thickness variations need to be established. The potential for different irradiated properties of sleeve graphite and large block graphite also needs to be evaluated.

Research on large blocks of graphite will need to be conducted to characterize the through-thickness variability of key properties in full size blocks and to establish the variability between batches of graphite. Large graphite blocks to be used for reflector material will be sectioned, tested, and evaluated to determine if properties measured on thin graphite components can be extrapolated to large blocks. Graphite materials properties are typically anisotropic and vary with the forming method and size of the final fabricated component. The sectioned large block specimens will be tested to measure important parameters such as strength, fracture toughness, density, thermal conductivity, coefficient of thermal expansion, level of chemical impurities, isotropy, and absorption cross-section. Based on the results obtained, an assessment will then be conducted to determine if the large block bulk properties would vary under high-temperature and high dose irradiation in a manner similar to thin sleeve graphite material.

There is a lack of consensus design standards as well as material specifications for nuclear grade graphite. Designers of HTGRs intend to use measured properties of the particular graphite in their design calculations. However, nuclear graphites should meet certain minimum requirements with respect to important properties, such as strength, density, and thermal conductivity as is the case for materials used in other reactor systems. If a particular graphite has excessively low strength and the designer uses that value in designing various components, that may not result in a suitable component for the intended service. There are underlying reasons why the strength may be excessively low. For example, the graphite might contain excessive cracking and porosity resulting in low strength. Although the component might have been designed using the low strength (resulting in possibly a thicker component), the excessive cracks in the component may grow during service and cause failure. Specific impurities in the graphite might be detrimental to irradiation properties of the component, and they should be limited in nuclear graphites. Other elements, such as halides, which can be released during operation and cause degradation of other components in the reactor, should also be limited in nuclear grade graphite. Thus, there is a need to develop material specification standards to establish the acceptable physical, thermal, and mechanical properties; composition; and manufacturing variables for nuclear grade graphite.

Staff efforts will be directed toward development of consensus standards for nuclear-grade graphite. Design and fabrication codes are also needed. The NRC staff will work with the international community, industry organizations, and professional societies to develop a nuclear-grade graphite material specification consensus standard. The standard will specify requirements on density, strength, fracture toughness, thermal conductivity, coefficient of thermal expansion, absorption cross-section, impurities, and any other appropriate parameter. The staff will also work with the codes and standards organizations to develop the design and fabrication requirements for graphite components to address processes such as strength, fracture, fatigue, creep, slow crack growth, irradiation damage, dimensional stability, oxidation, and any other appropriate design and fabrication considerations for HTGR service.

The EC research effort is currently reviewing the state of the art on graphite properties in order to set up a suitable database. The EC is planning to perform oxidation tests at high temperatures on fuel matrix graphite and on advanced carbon-based materials to obtain oxidation resistance in steam and in air. Recently, the EC began extensive characterization and irradiation testing of eight different graphites (3 from UCAR, 3 from SGL, and 2 from Toya Tanso) that are currently produced and could be used in future HTGRs. The properties of these graphites as a function of temperature and irradiation exposure will be studied. As mentioned above for the high-temperature metallic components, the EC plans to address a considerable amount of work; however, a key area possibly not fully addressed in the EC programs is the correlations of as-received graphite properties and manufacturing parameters to irradiated graphite properties. Exchange of NRC research results in this area could be used for cooperation with the EC HTR-M programs.

The UK is conducting ongoing research on graphite properties and has had experience with operating gas-cooled reactors that may be useful for NRC cooperation. As part of international cooperation with the UK, in 2002, the NRC assigned a staff from RES to the Nuclear Installations Inspectorate (NII) for three months to develop expertise on graphite behavior under high-temperature and irradiation conditions and to develop knowledge of the inspection and monitoring programs of graphite in Advanced Gas Reactors (AGRs). The NRC staff studied the British correlations of as-received graphite properties with irradiated graphite properties, which were primarily empirical. NRC staff work while on this assignment included discussions on important manufacturing parameters, physical and mechanical properties, composition, etc. of the as-received graphite that could have an effect on irradiated graphite properties.

The staff developed a better understanding of ongoing and past research results at the University of Manchester and explored potential cooperation in their program. The NRC staff also developed an understanding of the codes and procedures available for structural and creep analyses for the design of high-temperature graphite components.

APPLICATION OF RESEARCH RESULTS

Results from the research described will provide the necessary information to estimate component probability of failure as input to NRC PRAs to independently confirm and support safety evaluations. Since failure probability data for components of advanced reactors is not available from operating experience, very large uncertainties are inherent in the values selected and in the results of the PRAs. To reduce the uncertainties, information on failure probabilities would be derived from research results of potential degradation mechanisms (fatigue, creep, creep-fatigue, oxidation, thermal aging, stress corrosion cracking, crevice corrosion cracking, irradiation damage, and dimensional changes) of components in the operating environment of advanced reactors and with quantitative information of the initiation times and growth rates.

Due to the high temperatures and environments with which the industry has relatively little experience, careful analysis of the proposed materials needs to be carried out to indicate whether these materials are prone to degradation and provide the technical bases or criteria for materials acceptability. Aging effects and degradation due to the high-temperature helium environment and radiation need to be considered. Evaluation of potential degradation mechanisms and rate of degradation progression for materials used for connecting piping between the reactor pressure vessel and the power conversion systems will provide the NRC an independent basis to determine the validity of the contention that pipe break analysis does not need to be evaluated.

The research on nondestructive examination (NDE) and evaluations of ISI programs for HTGRs and ALWRs is applicable to independently confirm if an applicant's inspection plans are technically sound, or if additional requirements are needed. Currently accepted NDE and ISI programs may not detect materials degradation due to inaccessibility of components and long time periods between inspections. Research in this area may lead to regulatory requirements to modify NDE techniques and/or to use continuous online monitoring of structural integrity for structures and components of advanced reactors.