7. FUNDAMENTAL NUCLEAR SCIENCE

This element addresses the long-term R&D goal of developing new technologies for nuclear energy applications; educating young scientist and engineers, training a technical workforce; and, contributing to the broader science and technology enterprise.

Today's U.S. reactors, which are based largely on 1970's technology, operate under close supervision in a conservative regulatory environment. Although the knowledge base is adequate for these purposes, improvements in our knowledge and reduction of the inherent uncertainties could bring costs savings in current reactor operations and reduce the costs of future reactors. Furthermore, they could enable innovative designs that reduce the need for excessively conservative and costly factors of safety and reliability and significant extension in safe operating lifetimes. Future reactor technologies are likely to involve higher operating temperatures, advanced fuels, higher fuel burnup, longer plant lifetimes, better materials for cladding and containment vessels, and alternative coolants. To implement such features, substantial research in fundamental science and engineering must be carried out to supplement applied research to individual promising design concepts. Such fundamental research need not and should not be directed to any specific design. Although motivated in part by the need for new nuclear reactor system designs, the research would also have far reaching impact elsewhere in engineering and technology.

The five broad topics identified in the Long-Term R&D Plan include:

- The environmental effects on materials, in particular the effects of the radiation, chemical, thermal environments, and aging;
- Thermal fluids, including multiphase fluid dynamics and fluid structure interactions;
- The mechanical behavior of materials, including fracture mechanics, creep, and fatigue;
- Advanced material processes and diagnostics; and,
- reactor physics.

Projects currently selected under this element includes R&D in fundamental science in the fields of material science, chemical science, computational science, nuclear physics, or other applicable basic research fields. Selected research subjects include irradiation, chemistry, and corrosion effects on nuclear plant materials, advanced new materials research, innovative computational models, and the investigation of nuclear isomers that could prove beneficial in civilian applications.

NUCLEAR ENERGY RESEARCH INITIATIVE

Effects of Water Radiolysis in Water Cooled Nuclear Reactors

PI: Simon M. Pimblott, University of Notre Dame

Collaborators: Pacific Northwest National Laboratory, Atomic Energy Canada Ltd.

Project Start Date: August 1, 1999 Projected End Date: August 31, 2002

Project Number: 99-0010

Research Objective

The goal of this research program is to develop a model that describes the chemical effects of radiation on aqueous systems and on aqueous/solid interfaces at temperatures associated with nuclear power plants and the Advanced Light Water Reactors (ALWR). The program has four thrusts:

- Radiation Chemistry Modeling- An experiment-and-calculation based model will be developed to predict yields of the oxidizing and reducing radicals and the molecular species H_2 and H_2O_2 in aqueous systems like those associated with the ALWR chemistry.
- High Temperature and High Linear Energy Transfer (LET) Effects - Experiments will measure the effect of dose on yields of O_2 and H_2O_2 produced in radiolysis with γ-rays, electrons and with H^+, He^{2+} and O^{8+} (C^{6+}) ions.
- Interfacial Effects of Radiation - Experiments will gather information about radiation effects at aqueous/oxide interfaces of importance in fields such as reactor pipe corrosion and in storage of spent nuclear fuel.
- Low Energy Electrons at Zirconia and Iron Oxide Surfaces and Interfaces - ultra high voltage (UHV) experiments performed at Pacific Northwest National Laboratory with low energy electrons and photons will be used to simulate the damage at interfaces caused by the cascade of reactive secondary electrons produced by high-energy radiation.

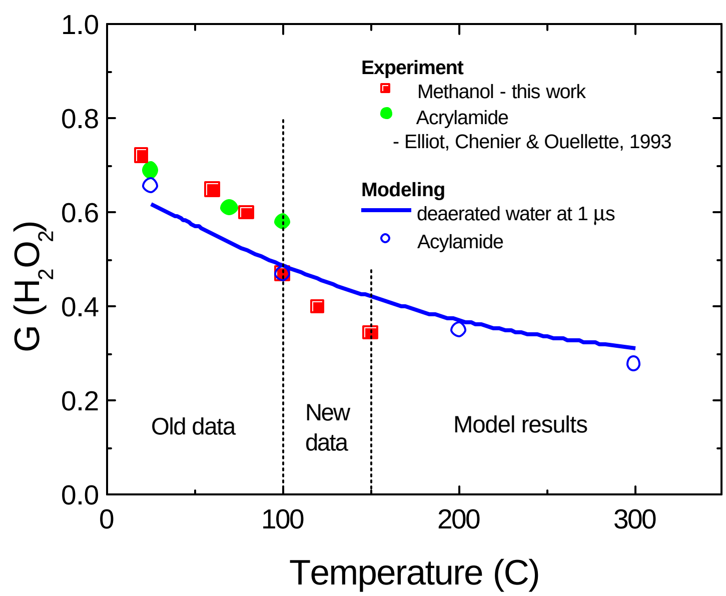
Research Progress

An extensive review of the scientific literature on γ-radiolysis of water and aqueous solutions at room and at elevated temperatures has been performed. There is a large amount of data on the nonhomogeneous track chemistry at room temperature, and the track structure and diffusion-limited kinetics are well parameterized. This wealth of knowledge contrasts with the limited information about the effects of radiation on aqueous solutions (and on aqueous solution/metal oxide interfaces) at elevated temperatures.

The Notre Dame Radiation Laboratory (NDRL) suite of computer codes, TRACKKIN, for the simulation of low-LET track chemistry in water was extended to include the effects of temperature. These codes address two aspect of the radiolysis; the structure of the radiation track and the chemistry of the resulting spatially nonhomogeneous distribution of radiation-induced reactants.

Stochastic radiation chemical kinetic simulations have been made for water over the temperature range 25 °C to 300 °C. The predictions of these calculations were compared with available experimental data for γ-radiolysis. Good agreement is found for all of the radiation-induced species and for the combined yield of H and H_2.

Hydrogen peroxide is a source of corrosion for many structural materials in nuclear reactors and its yield is usually kept to minimum levels with the use of different additives. One common technique is to add various amounts of molecular hydrogen. The yield of hydrogen peroxide in the radiolysis of water has been examined with the goal of understanding the mechanism of its production and how various scavengers can influence its yield. The graph below shows the effect of temperature on the yield of hydrogen peroxide. The range over which experimental data is available has been increased by 50 percent. Modeling has been performed up to the normal operating temperatures of light water reactors.



A literature search of the radiation chemistry of iron-oxide and zirconium-oxide water interfaces was conducted. It showed that charge transfer from aqueous species to solid iron-oxide particles has been studied extensively and that a kinetic model has been developed for that system. There is little data for zirconium-oxide particles.

Pulse radiolysis experiments were conducted on zirconia suspensions at high particle concentrations. The yield of the hydrated electron was measured following a pulse of high-energy electrons in the presence of acceptor molecules.

Experiments measuring escape yield of electrons from zirconia into water employing several different scavengers (zwitterion-viologen and methyl-viologen) were used to determine effect of scavenger charge and its interaction with the surface potential on yield. It was found that adsorption of the electron acceptor onto the surface, due to electrostatic interaction with the surface, enhances yield of electron capture at the surface. This, therefore, leads to the conclusion that: a) escape of electrons from the solid particles can be intercepted by the addition of acceptors, and b) the same acceptors also compete with charge recombination within the particles.

Cubic and monoclinic single crystal zirconia films were grown on yttrium stabilized zirconia (YSZ) substrates by the oxygen plasma-assisted molecular beam epitaxy method. Nominal pure ZrO_2 films were grown by isothermal oxidation of pure (99.94 percent) zirconium metal foils. These unique and well-characterized ZrO_2 materials will be utilized in our future experiments on mechanistic understanding and modeling of radiolysis and radiation-induced corrosion of nuclear reactor fuel surfaces. Current work is focusing on understanding the adsorption and thermal desorption behavior of water at these interfaces and the development of techniques for growing Fe_3O_4 are underway.

Planned Activities

- Development and validation of a stochastic methodology for simulating high-LET, heavy ion track structure, and for modeling kinetics of heavy ion radiolysis. Subsequent calculations will involve the simulation of the energy loss characteristics of heavy ions, and the investigation of heavy ion track structure, including the role of daughter electrons.
- Perform high dose experiments on the effects of added molecular hydrogen on O_2 yields with γ-rays and with high LET heavy ions.
- Measure the temperature dependence of the effects of added molecular hydrogen on H_2O_2 yields with γ-rays.
- Determine effects of particle size and surface charge on the yields of escape for smaller band-gap materials (hematite and magnetite). For these materials, absorption of light may force the use of conductivity or EPR.
- Perform steady-state radiolysis of oxide suspensions to determine hydrogen and hydrogen peroxide yields, as well as dissolution rates if possible in order to obtain long term consequences of irradiation. Because of the Fenton reaction and the Haber–Weiss cycle it may be impossible to obtain both yields of molecular products and dissolution rates. The experimental results, in particular for H_2 and H_2O_2, will be tested against the computational predictions in Year 3.

- Investigate electronic structure of doped ZrO_2 using time-resolved luminescence and electron energy loss measurements.
- Begin controlled irradiation studies of water-covered iron-oxide.
- Perform radiation assisted redox dissolution studies.

Measurements of the Physics Characteristics of Lead Cooled Fast Reactors and Accelerator Driven Systems

PI: P.J. Finck, Argonne National Laboratory (ANL)

Collaborators: Commissariat a l'Energie Atomique (French Atomic Energy Commission)

Project Start Date: August 1, 1999 Projected End Date: September 2002

Project Number: 99-0039

Research Objective

Several recent studies in the U.S. and worldwide have indicated a strong interest in the potential development of lead-cooled critical and sub-critical systems. In order to permit the eventual industrial deployment of these systems, several key technical areas need to be carefully investigated, and solutions for potential technical problems need to be found and implemented.

The neutronic behavior of a lead-cooled fast spectrum system is believed to be relatively poorly known: difficulties arise both from nuclear data uncertainties and from methods related deficiencies. The French Atomic Energy Commission (CEA) has recognized this situation and has launched an ambitious experimental program aimed at measuring the physics characteristics of lead-cooled critical and sub-critical systems in an experimental facility located at the Cadarache Research Center. A complete analytical program is associated with the experimental program and aims at understanding and resolving potential discrepancies between calculated and measured values. The final objective of the two programs is to reduce the uncertainties in predictive capabilities to a level acceptable for industrial applications.

ANL teams are now participating in the experimental design, measurements, and analytical tasks. In exchange for our participation, all experimental data are available to us.

This program will have three critical outcomes: high quality experimental data representative of the physics of lead-cooled cores will be available to the U.S. programs; U.S. neutronics codes will be validated for calculating lead-cooled systems; and potential deficiencies in U.S. nuclear data and codes will be identified.

Research Progress

Efforts this past year were concentrated in two areas: analysis of existing data from the MUSE 3 experiment, and preparation of the MUSE 4 experimental campaign.

- The MUSE 3 experimental program was carried out from 1997 to 1999. The experimental layout consists of a central neutron source with a well characterized spectrum and intensity, surrounded by a buffer zone made out of either sodium or lead, to simulate different design options and to provide the neutronic impact of these different materials. The remainder of the core is built out of MOX fuel with sodium coolant. Several configurations, both critical and sub-critical, have been measured. The complete set of experimental results for this phase has been provided to ANL.

  Very detailed models of the experimental configurations were set up, using both the European code system with European nuclear data (JEF 2.2) and deterministic or stochastic U.S. code systems with U.S. nuclear data.

  In general, reaction rate traverses are calculated with good accuracy, and no difficulties have been observed in the source region. This indicates that the source multiplication and propagation are well estimated at all sub critical levels. Significant difficulties appear close to the core-reflector interface: these discrepancies are attributed to uncertainties in nuclear data for structural materials and are typical in these types of cores. Large discrepancies have been observed in terms of predicted sub-criticality levels. Compared to experimental data, JEF 2.2 provides significantly better results, even though it tends to overestimate reactivity by about half a percent. In general, ENDF/B-VI overestimates reactivity by about one percent. Root causes for this over prediction are still being investigated.

  Kinetic measurements will be an important element of future experiments; the analysis of these measurements is particularly difficult, as it is necessary to employ explicit three dimensional time dependent transport calculations. ANL is hosting a French Ph.D. student to develop the adequate methodology, which will be made available to the French program.

  Significant analytical support has been provided to the French CEA in preparing the MUSE 4 experimental campaign.

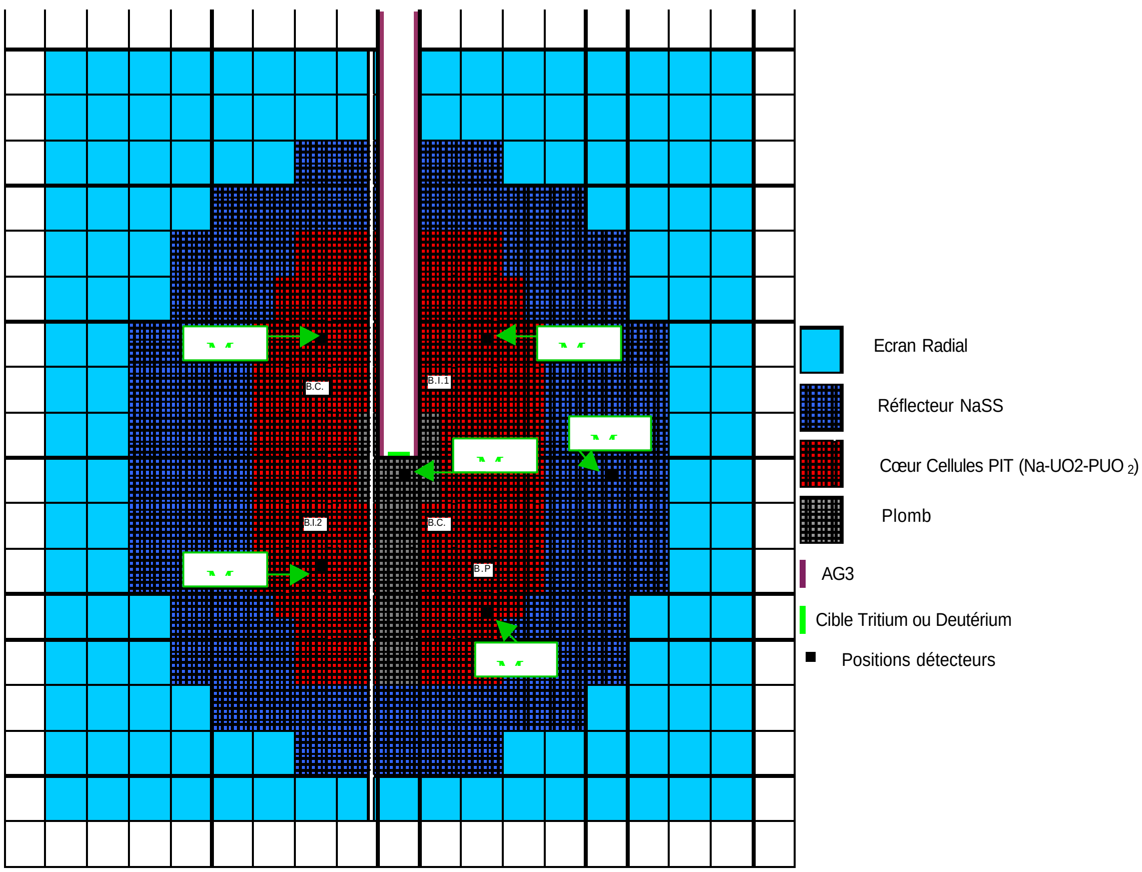
- ANL had a major impact on the preparation of the MUSE 4 experimental campaign. A project researcher has been on attachment in France for several months and has led the effort for developing the experimental techniques for dynamic measurements and for defining the required data acquisition system.

  The dynamic measures to be performed are Feynman/Rossi alpha, single pulse alpha, frequency modulation, and transfer function/noise techniques. Although these are all fundamentally related, different techniques of data acquisition and analysis will be used to yield different kinetic parameters. In addition, analysis over different time scales will lead to different information.

A set of data acquisition equipment and a series of measurements were defined in the Experimental Plan that will yield the most useful information for subsequent analysis.

The full Experimental Plan was developed to provide multiple measures of the parameters that are key to safety (β, l, source importance), as well as measures important to the feasibility of an accelerator driven system (spectrum, reaction rates). It is expected that these measurements will help to significantly reduce the uncertainty in calculated parameters.

The French Safety Authorities have given approval for the MUSE 4 experiments as of November 8, 2000. A series of critical reference measurements are planned until February 2001, at which time the new GENEPI accelerator will be installed and tested. Three sub-critical configurations will be implemented (successive sub-critical levels are: -1.5\$, -10\$, -17\$) and the measurements will last until October 2001. The figure below describes the MUSE 4 configuration.



Planned Activities

Three major tasks are planned for FY 2001:

- Experimental activities: an ANL project researcher will lead the experimental team at Cadarache and realize all measurements described in the Experimental Plan.
- Analytical task: the analysis of MUSE 3 results will be completed by January 2001, and the first MUSE 4 results will be analyzed. Further measurements, in particular those for kinetic parameters, will be analyzed as they become available.
- Support task: the preparations for the following campaign, MUSE 5, will become a top priority, and the choice of options (fuel type and configuration) will be supported by ANL analysts.

Mapping Flow Localization Processes in Deformation of Irradiated Reactor Structural Alloys

PI: K. Farrell, Oak Ridge National Laboratory

Project Start Date: August 1, 1999 Projected End Date: August 31, 2002

Project Number: 99-0072

Research Objective

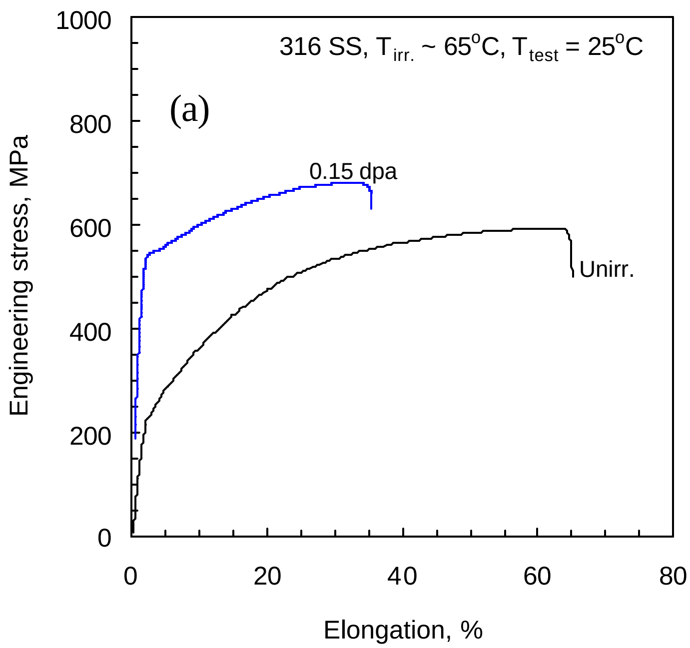
The materials from which nuclear power reactors are constructed, namely ferritic steels for pressure vessels, austenitic stainless steel for core internals and piping, and zirconium alloys for fuel cladding and tubing, are normally quite ductile and workable. They are ductile because they undergo plastic flow, or deformation, by the generation and movement of dislocations on slip planes within the atomic lattice of the metal. Many intersecting slip planes are operative. The dislocations can move from one slip plane to another and they become entangled into a three-dimensional network of dislocation cells. This ability to develop a network of dislocation cells ensures that the material work hardens and deforms in a homogenous manner. That ability is lost when the materials are exposed to the action of penetrating neutrons in the reactor. The neutrons create disturbed regions in the regular arrangement of atoms in the atomic lattice. These disturbed regions, or radiation damage clusters, impede the slip dislocations and inhibit the formation of dislocation cells. Instead, the deformation becomes localized in narrow bands or channels, and sometimes in twin bands. This intensification of strain and stress by dislocation channel deformation (DCD) changes the mechanical properties of the material and causes embrittlement. The degree of embrittlement is related to the nature and the details of the dominant deformation mode, which are functions of the radiation exposure and of the mechanical test conditions.

Most mechanical property data for use in design data banks is derived from tensile tests. Radiation damage raises the tensile yield strength and ultimate tensile strength (UTS), induces yield point drops in materials that do not normally show sharp yield points, reduces the work hardening rate and the elongation, and causes premature plastic instability and failure. All of these changes are now known or suspected to involve DCD but only a few quantitative correlations have been made. If such correlations are made in detail they can allow preparation of deformation mode maps in which the regions and boundaries of the deformation modes are plotted in terms of plastic strain and neutron fluence. Mechanical properties representing the different deformation modes can be overlaid on the maps, and the maps become pictorial repositories of knowledge relevant to the irradiation behavior of the materials. These maps will not only simplify, condense, verify and specify essential properties and applications limits for crucial reactor components, but they will add immeasurably to our understanding of the interplay between radiation damage microstructures, deformation modes and mechanical properties responses. They should bring cohesion and assurance into the processes of selection, assessment, and application of reactor materials. Presently, deformation mode maps for irradiated materials exist only for nickel and gold. The goal of this project is to determine deformation mode maps for A533B ferritic steel, 316 stainless steel, and Zircaloy-4 alloy.

Research Progress

Literature surveys have been made and dossiers on deformation behaviors of the three test materials have been compiled. These files proved to be very appropriate for the principal investigator to present an invited paper on strain localization at the Workshop on Dislocation-Defect Interactions in Irradiated Materials in Toledo, Spain, April 3-5, 2000. The workshop was the first of its kind devoted to flow localization in irradiated materials, and its timing was perfect for assessing and influencing the direction of the present NERI project. Mapping of deformation modes for irradiated materials entails irradiating suitable tensile specimens to prescribed neutron fluences, straining the specimens to predetermined strain levels and recording the tensile properties, cutting small pieces from the gauge sections of the specimens, electrothinning the pieces to make transmission electron microscopy (TEM) samples, and conducting a detailed study of the samples to obtain quantitative measurements of the radiation damage microstructures and the deformation mode microstructures. Each sample involves a great deal of work, and since the project is limited to three years, it was necessary to expedite the process. All of the above steps had to be minimized and honed to save time, cost, and effort, and to reduce radiation exposure to the operators. To those ends, a special, miniature, flat tensile specimen was developed that occupies minimum space in the irradiation capsule, permits excision of tiny TEM pieces without the need for sanding operations, and has reduced radioactivity. To allow remote handling of these small specimens in and out of the tensile machine without inadvertent damage, a sliding cradle that supports and protects the specimen during the test procedure was designed. The tiny TEM pieces are only 1.5 mm square, much smaller than standard 3mm diameter disks, and they required development of refined electrothinning and TEM clamping procedures. These refinements were achieved in parallel with specimen procurement, heat treatment, and encapsulation and documentation for irradiation in the High Flux Isotope Reactor (HFIR).

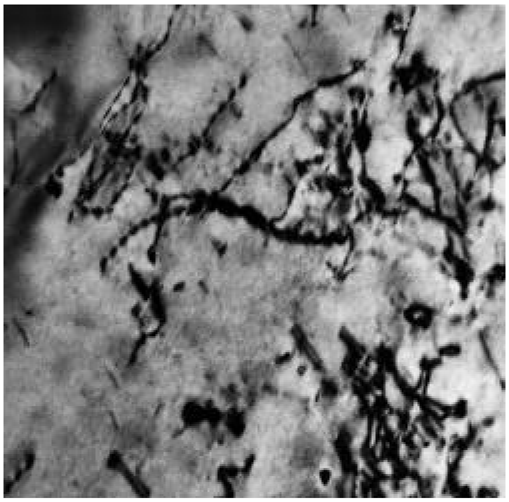
Five capsules for irradiations at 65^{o}C to neutron fluences in the range 6 x 10^{20} $n \bullet m^{-2}$ to 6 x 10^{24} $n \bullet m^{-2}$ at decade intervals were ready for irradiation by February 2000. These irradiations would normally have been treated as routine irradiations and should have been conducted in about four weeks. Unfortunately, several incidents occurred at the HFIR, unconnected with these experiments that provoked a revision of the rules for materials irradiation experiments in the HFIR. Coping with the revised regulations, and difficulties with an overcrowded schedule at the HFIR, delayed execution of our irradiations until August/September. This unexpected long delay threw project plans off schedule and, as a result, the investigators requested and were granted a six month, no-cost extension of the Year 1 phase of the program. To make up lost time and expenses, much of the planned specimen testing and TEM preparation phases has been rerouted from costly and slow hot cells operations to speedier and more cost-effective C zone operations. These moves allowed Year 2 TEM studies to begin on the original schedule. Presently, the specimens have been recovered from the capsules and are undergoing tensile testing, TEM preparation, and TEM examination. The first TEM results show that the deformation mode for irradiated 316 stainless steel is very narrow bands of microtwins. The graph below illustrates the irradiation effects for a dose of 0.15 dpa (9 x 10^{23} n. m^{-2}, E>1MeV). The tensile yield strength is raised from 230 Mpa to 540 Mpa and the elongation is reduced from 60 percent to 30 percent. Correspondingly, the deformation mode changes from random dislocation tangles (left picture below) to narrow twin bands (right picture below).

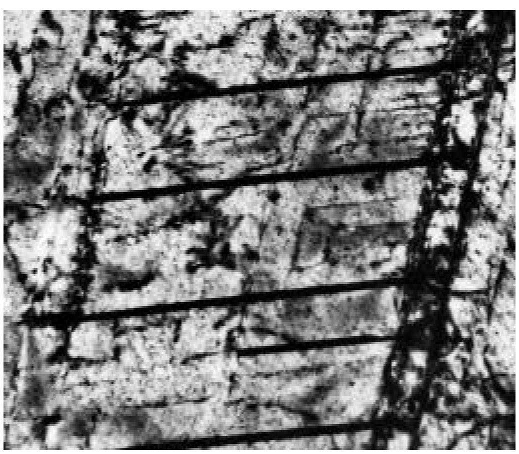


Some setbacks have been encountered in obtaining satisfactory electrothinned samples from the irradiated A533B steel and the Zr-4, however, the problems are surmountable.

Planned Activities

Before the end of Year 2 of the project, the irradiated tensile specimens and the unirradiated control specimens will be tested to five different strain levels and preliminary TEM examinations will be conducted to identify the deformation modes. These observations will be used to construct the deformation maps. Concurrently, selected specimens will be subjected to detailed TEM studies to measure and quantify the microstructural features involved in the deformation processes. This will involve measuring the sizes and wall thicknesses of dislocation cells, and characterization of the dislocation channels in terms of crystallographic planes, channel widths, channel frequency, degree of cross-channeling, amount of strain in the channels, and the effects created when channels impact grain boundaries and other barriers. These data will be analyzed and correlated with the corresponding tensile properties data.





Changes in tensile properties and deformation microstructures of 316 stainless steel after irradiation to 0.15 dpa. The deformation microstructures represent 6% elongation in the picture above, the unirradiated steel and the picture to the left, the irradiated steel.

In Year 3 more irradiations will be carried out on a reduced scale to look briefly at the effects of a higher irradiation temperature and a higher strain rate. An irradiation temperature of 300^{o}C will be used which seems to induce enhanced radiation embrittlement in all three materials.

With regard to higher strain rate, a higher rate is expected to give narrower dislocation channels. Correspondingly, the channel strains and the resultant stress intensification will be increased, and embrittlement and plastic instability will be worsened. The specimens will be irradiated to two fluences, 6 x 10^{23} n•m^{-2} and 6 x 10^{24} n•m^{-2}. Specimens of the A533B steel will be irradiated to a fluence of 6 x 10^{23} n•m^{-2}. These specimens will be tested at two strain rates, 10^{-3}•s^{-1} as in the Year 1 tests, and 10^{-1}•s^{-1}. Several specimens for each condition will be strained to failure. Other tests will be truncated at two strain levels, 5 percent and at the UTS, if sufficient ductility is maintained. Otherwise, the tests will be terminated at whatever minimum strain is achievable. TEM will be performed on each specimen to identify the deformation mode(s). Selected specimens will receive intense TEM scrutiny and extraction of quantitative data. A report of the findings will be published.

A Novel Approach to Materials Development for Advanced Reactor Systems

PI: Gary S. Was, University of Michigan

Collaborators: Pacific Northwest National Laboratory, Oak Ridge National Laboratory

Project Start Date: September, 1999 Projected End Date: August 31, 2002

Project Number: 99-0101

Research Objective

Component degradation by irradiation is a primary concern in current reactor systems as well as advanced designs and concepts where the demand for higher efficiency and performance will be considerably greater. In advanced reactor systems, core components will be expected to operate under increasingly hostile (temperature, pressure, radiation flux, dose, etc.) conditions. The current strategy for assessing radiation effects for the development of new materials is impractical in that the costs and time required to conduct reactor irradiations are becoming increasingly prohibitive, and the facilities for conducting these irradiations are becoming increasingly scarce. The next generation reactor designs will require more extreme conditions (temperature, flux, fluence), yet the capability for conducting irradiations for materials development and assessment in the next 20 years is significantly weaker than over the past 20 years. Short of building new test reactors, what is needed now are advanced tools and capabilities for studying radiation damage in materials that can keep pace with design development requirements.

The most successful of these irradiation tools has been high-energy (several MeV) proton irradiation. Proton irradiation to several tens of displacements per atom (dpa) can be conducted in a short amount of time (weeks), with relatively inexpensive accelerators, and result in negligible residual radioactivity. All of these factors combine to provide a radiation damage assessment tool that reduces the time and cost to develop and assess reactor materials by factors of 10 to 100. What remains to be accomplished is the application of this tool to specific materials problems and the extension of the technique to a wider range of problems in preparation for advanced reactor materials development and assessment.

The objective of this project is to identify the material changes due to irradiation that affect stress corrosion cracking (IASCC) of stainless steels, embrittlement of pressure vessel steels and corrosion and mechanical properties of Zircaloy fuel cladding. Until such changes are identified, no further progress can be made on identifying the mechanisms and solving the problems. An understanding of the mechanisms will allow for the development of mitigation strategies for existing core components and also the development of radiation-resistant alloys or microstructures that are essential for the success of advanced reactor designs.

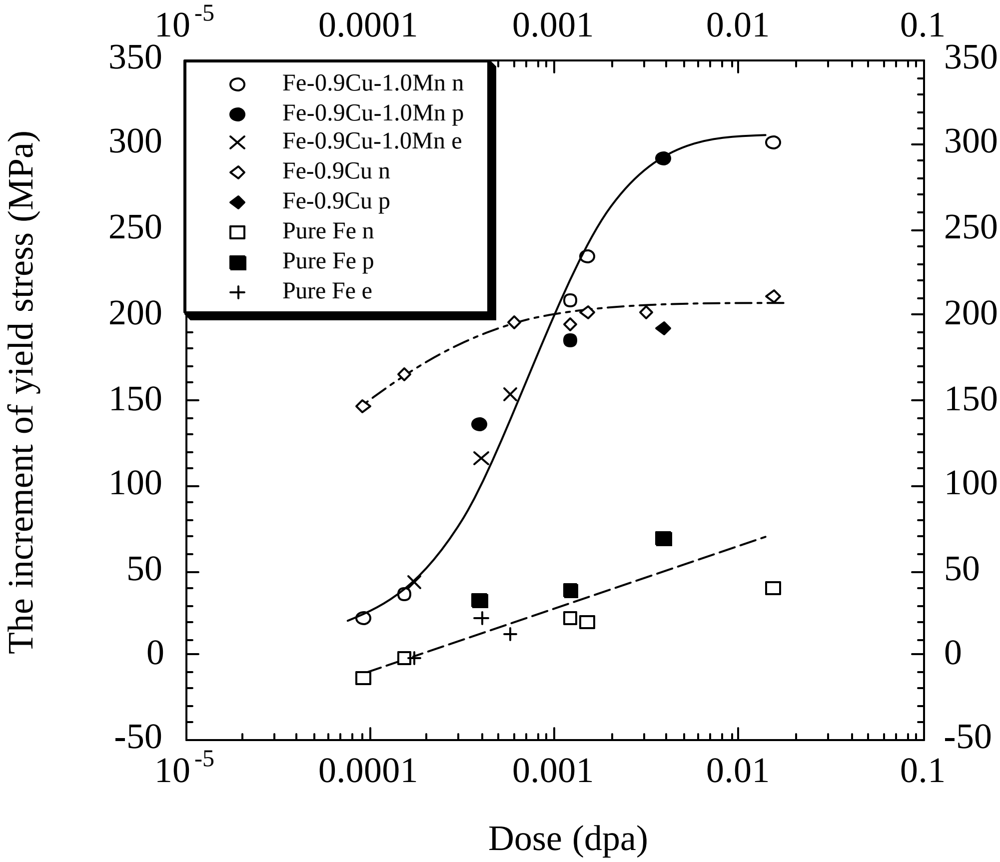
Research Progress

The objective of the first year of research was to understand the microstructure evolution in model alloys of reactor pressure vessel (RPV) steels and also in stainless steels under light water reactor irradiation conditions. The work focused on investigating the irradiated microstructure by using proton irradiation in comparison with neutron irradiation. The following points summarize the results:

- Proton-irradiation of model RPV alloys results in a comparable level of hardening as does neutron irradiation of the same alloys at the same temperature and dose. Annealing of the irradiated microstructure produces a recovery of hardening that also matches that from neutron irradiation.
- Small angle x-ray scattering data and preliminary results from transmission electron microscopy reveal that the precipitate size/density combination is similar to that from neutron irradiation. The agreement in hardening between proton and neutron irradiation is supported by the similarity in the character of the defect clusters. The results also support those from electron irradiation that imply that the character of the damage cascade does not strongly affect the resulting microstructure.
- The irradiation-induced microstructure in proton-irradiated austenitic stainless steels consists mainly of dislocation loops up to 5.0 dpa.
- The dose dependence of the dislocation loop density and size in proton-irradiated Fe-Cr-Ni alloys follows the same trend as in neutron-irradiated alloys. The higher loop density in commercial alloys is probably due to enhanced nucleation of loops by minor constituent elements (phosphorus, silicon).
- Yield strength in proton-irradiated austenitic alloys as a function of dose and temperature are consistent with neutron data. Across a wide dose range, the hardening estimated from micro-hardness measurements agrees with calculations from the dispersed barrier-hardening model.
- The difference in the effect of the character of the displacement cascade on loop nucleation between neutron irradiation (275^{o}C, 7x10^{-8} dpa/s) and proton irradiation (360^{o}C, 7x10^{-6} dpa/s) has little effect on the final irradiated microstructure. The reduced level of loop nucleation by in-cascade interstitial clustering in proton irradiation appears to be balanced by the higher cascade efficiency and higher vacancy supersaturation caused by the higher dose-rate and the lower sink strength at the higher irradiation temperature.

Planned Activities

The research plan for years 2 and 3 will focus on three areas. We will be studying the tradeoff in hardening between cold work and irradiation on the IASCC susceptibility of austenitic stainless steels. We will also be experimenting with pre-segregation at grain boundaries to neutralize the effect of radiation induced stress (RIS) on IASCC. The



Yield strength increment as a function of dose for neutron, electron and proton irradiation of alloys VA (Fe), VH (Fe-0.9Cu) and VD (Fe-0.9Cu-1.0Mn). The trend lines ---, -· and ––– represent VA, VH and VD hardening trend with dose respectively. Hardening with proton irradiation is in good agreement with that from neutrons and electrons.

activity on RPV model alloys will focus on the composition effects of proton irradiation, the development of hardness with dose, and the comparison with neutron irradiation results. The effort is aimed at understanding the role of alloying additions on the hardening behavior of these steels. The last task will focus on the irradiation of Zircaloy–2 and –4 to assess the capability of proton irradiation to emulate neutron irradiation in terms of the dislocation microstructure, precipitate structure and morphology, and corrosion behavior.

Complete Numerical Simulation of Subcooled Flow Boiling in the Presence of Thermal and Chemical Interactions

PI: Vijay K. Dhir, University of California, Los Angeles

Project Start Date: August 1, 1999 Projected End Date: August 31, 2002

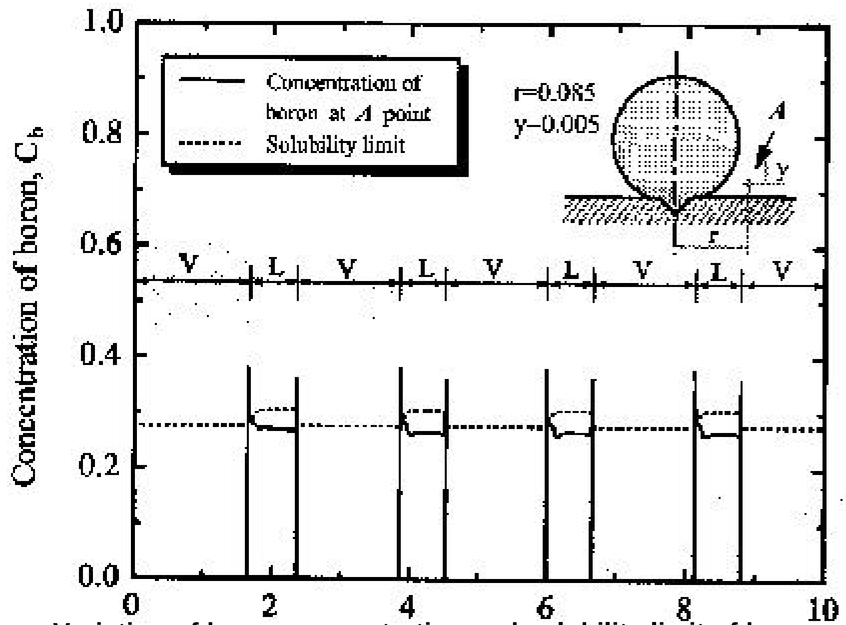
Project Number: 99-0134

Research Objective

The key objective of the proposed research is to develop a mechanistic basis for the thermal and chemical interactions that occur during subcooled boiling in the reactor core. The axial offset anomalies (AOA) are influenced by local heat flux for subcooled nucleate boiling, the nucleation site density on the fuel cladding, and the concentration of boron and lithium in the primary coolant. The approach proposed in this work is very different than what has been employed in the past in that complete numerical simulations of the boiling process along with thermal, hydraulic and concentration fields in the vicinity of the cladding surface will be carried out. This approach is considered to be the only viable one that can provide, simultaneously, a mechanistic basis for the portioning of the wall heat flux among vapor and liquid and the concentration of boron and lithium, at, and adjacent to, the heated surface. The model will be validated with data from detailed experiments. A building block type of approach will be used where, starting with a bubble at a single nucleation site, the complexity of the numerical model and experiments will be increased to include merger of bubbles at the wall as well as interaction of the detached bubbles with the bubbles present on the heated surface. The concentration of boron and lithium in water, pH value of water, and system pressure will be important variables of the problem.

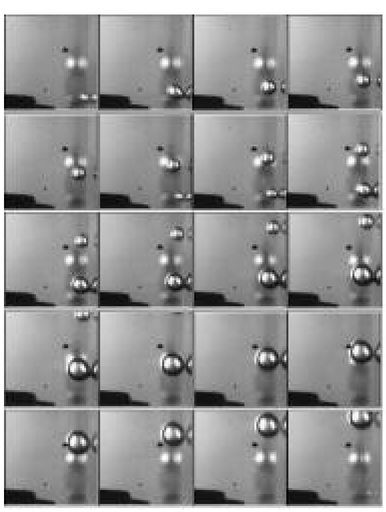
Research Progress

During the past year, an axi-symmetric numerical simulation model of a single bubble formed on a horizontal surface containing a chemical species has been developed. In carrying out the analysis, the conservation equations of mass, momentum, and energy for the two phases along with the conservation of species equations for chemicals present in water have been solved simultaneously. The level set method is used in the numerical simulation. As such, a single momentum equation is solved for both liquid and vapor regions. A function representing distance from the interface is derived and the distance function is advanced at each time step by solving the advection equation. The results of numerical simulation using orthoboric acid as the chemical species present in water reveal that during growth and departure phases of bubble the concentration of orthoboric acid varies both spatially and temporally. As shown in graph below, the local concentration especially in the regions very close to the heater surface can exceed the solubility limit at a given temperature. However, experimental validation of the predicted



Variation of boron concentration and solubility limit of boron in solution with time at point *A*.

behavior is pending. The numerical simulation tool has also been extended to three dimensions with flow along the heater surface. It is found that consistent with experimental observations the bubbles slide along the surface before departure. An experimental apparatus for flow-boiling studies has been developed and experiments using silicon strips made from polished silicon wafer with a microfabricated cavity have been performed. The experiments show consistent with the model predictions that bubble lift off diameter is larger than the bubble departure diameter. Both bubble lift off and departure diameters decrease with flow velocity. Below are photographs of bubbles before and after departure, but prior to lift off on a vertical surface with imposed flow velocity.



Growth Cycle for Vertical Surface, Upflow, (*near field view*); V = 0.025 m/s, ΔT_{wall}= 5.9°C, ΔT_{sub}= 0.3°C.

Planned Activities

During the next two years, the three-dimensional code will be augmented to include the species conservation equation. The code will be exercised more thoroughly to study the effect of liquid subcooling, wall superheat, flow velocity and surface wettability on bubble dynamics and precipitation of the solute in water. The experiments will be continued to obtain data on bubble dynamics and heat transfer with and without the presence of chemical species in the liquid. If possible, bubble-bubble interaction as may occur at high wall superheats will also be investigated. The results of the research will be disseminated to the technical community by presenting papers at conferences and publication in peer reviewed journals.

Developing Improved Reactor Structural Materials Using Proton Irradiation as a Rapid Analysis Tool

PI: T.R. Allen, Argonne National Laboratory-West

Collaborators: University of Michigan

Project Start Date: August 1, 1999 Projected End Date: August 31, 2002

Project Number: 99-0155

Research Objective

The purpose of this program is to design advanced reactor materials with improved resistance to void swelling and irradiation assisted stress corrosion cracking (IASCC) using three principal methods: bulk composition engineering, grain boundary composition engineering, and grain boundary structural engineering. The focus of the first year was bulk compositional engineering in which five different alloying additions were made to a base Fe-18Cr-8Ni-1.25Mn alloy whose bulk composition corresponds to 304 stainless steel. This alloy was chosen as the reference alloy for the program because 304 stainless steel is known to be susceptible to both swelling and IASCC. In addition to the studies on bulk composition engineering, work commenced on the grain boundary structural engineering. Thermomechanical treatments were developed for the Fe-18Cr-8Ni-1.25Mn alloy that increased the fraction of coincidence site lattice (CSL) boundaries.

Each of the bulk composition alloying additions was chosen for a specific purpose. Fe-18Cr-40Ni-1.25Mn was chosen because higher bulk nickel concentration is known to reduce swelling, but its affect on IASCC is unknown. Fe-18Cr-8Ni-1.25Mn+Zr was chosen because Zr is an oversized element that might trap point defects and prevent swelling and grain boundary segregation. Fe-16Cr-13Ni-1.25Mn, Fe-16Cr-13Ni-1.25Mn+Mo, and Fe-16Cr-13Ni-1.25Mn+Mo+P were chosen to determine why 316 stainless steel is more resistant to swelling and IASCC than 304 stainless steel.

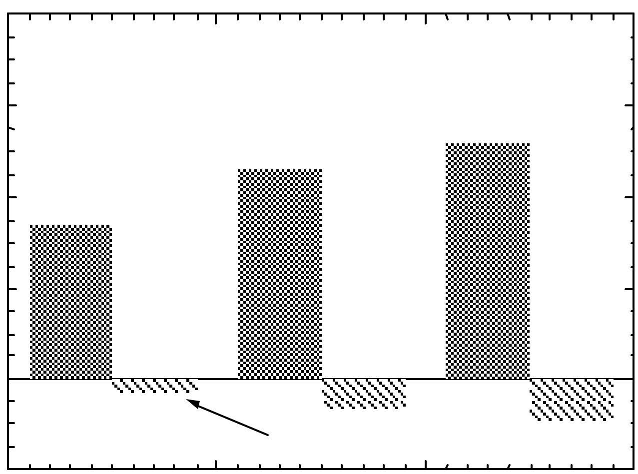
Research Progress

In the first year of the project, Fe-18Cr-8Ni-1.25Mn, Fe-18Cr-40Ni-1.25Mn, Fe-18Cr-8Ni-1.25Mn+Zr, and Fe-16Cr-13Ni-1.25Mn were studied. Samples were irradiated using 3.2 MeV protons at 400°C to 1 displacement per atom (dpa). Swelling was characterized by measuring the void size distribution using a transmission electron microscope (TEM). Radiation-induced grain boundary segregation was measured using a field emission gun scanning transmission electron microscope (FEG-STEM). Microhardness measurements were performed on irradiated and non-irradiated alloys to estimate the effect of irradiation on strength.

New insight on the relationship between IASCC, grain boundary composition, and irradiation hardening was reached in the first year of the program by studying the Fe-18Cr-8Ni, Fe-18Cr-40Ni, and Fe-16Cr-13Ni alloys. Increasing the bulk nickel concentration decreases void swelling, increases matrix hardening, and increases grain boundary chromium depletion and nickel enrichment. Two effects of radiation on microstructure have been hypothesized to contribute to IASCC: Cr depletion leading to decreased grain boundary corrosion resistance and matrix hardening leading to a grain boundary mechanically weakened relative to the matrix. Comparing the Fe-18Cr-8Ni and Fe-16Cr-13Ni alloys, moving toward the 316 composition (and in general to alloys with greater bulk nickel concentrations) causes greater hardening and greater Cr depletion (figure below). Greater hardening (yield strength) and greater Cr segregation should make 316 more susceptible to IASCC. Yet 316 is typically less susceptible than 304 to IASCC.

Alloys with greater bulk nickel concentration have greater radiation induced segregation (RIS), causing a decrease in grain boundary volume. They also have increased hardening and Cr depletion, theoretically making the alloy more susceptible to IASCC. But Cookson et. al. found that increasing Ni content decreased IASCC susceptibility. Decreased IASCC susceptibility may be related to boundary strengthening associated with the grain boundary volume decrease. The lattice parameter and shear moduli decrease with decreasing Cr concentration and with increasing Ni concentration. Cr depletes and Ni enriches at the boundary during irradiation. The smaller modulus and smaller solute atoms caused by RIS may strengthen the boundary, mitigating grain boundary deformation and cracking. While typical studies of IASCC assumed RIS was a contributing factor to cracking, it may be that properly controlled RIS can be used as a mitigating factor.

- •IASCC Causes (Traditional thinking)
  - •Cr depletion weakens grain boundary corrosion resistance
  - •Matrix hardening weakens grain boundary relative to matrix
- •316 (and in general alloys with greater bulk Ni) more resistant to IASCC than 304
- •This study:
  - •316 surrogate (and in general alloys with greater bulk Ni) has greater grain boundary Cr depletion and greater matrix hardening (should be more cracking susceptible)
  - •Increasing bulk Ni changes RIS such that grain boundary volume decreases (decreased lattice parameter, decreased shear modulus)
  - •Properly controlled RIS may reduce susceptibility to IASCC



Effect of Bulk Composition on Grain Boundary Cr Depletion and on Irradiation Hardening

Planned Activities

For year two of the project, the research will focus on all three research areas; bulk composition engineering, grain boundary composition engineering, and grain boundary structural engineering. Bulk composition engineering and grain boundary compositional engineering will be studied on the 316 series of alloys, Fe-16Cr-13Ni-1.25Mn, Fe-16Cr-13Ni-1.25Mn+Mo, and Fe-16Cr-13Ni-1.25Mn+Mo+P. These alloys will be irradiated in the solution-annealed condition and following special treatments to set the grain boundary composition prior to irradiation. The greater resistance of IASCC in 316 stainless steel may also be related to the addition of Mo, which is not present in 304 stainless steel. The study of Fe-16Cr-13Ni-1.25Mn-2.5Mo is planned to address the effect of Mo. Bulk composition engineering and grain boundary structural engineering will be focussed on zirconium containing alloys. These alloys will be irradiated in the solution-annealed state and following special processing to alter the grain boundary structures. The microhardness, void swelling, radiation-induced grain boundary segregation, and the radiation-induced microstructural changes will be determined for each irradiation condition.

An Investigation of the Mechanism of IGA/SCC of Alloy 600 in Corrosion Accelerating Heated Crevice Environments

PI: Jesse Lumsden, III, Rockwell Science Center

Project Start Date: August 1999 Projected End Date: September 2002

Project Number: 99-0202

Research Objective

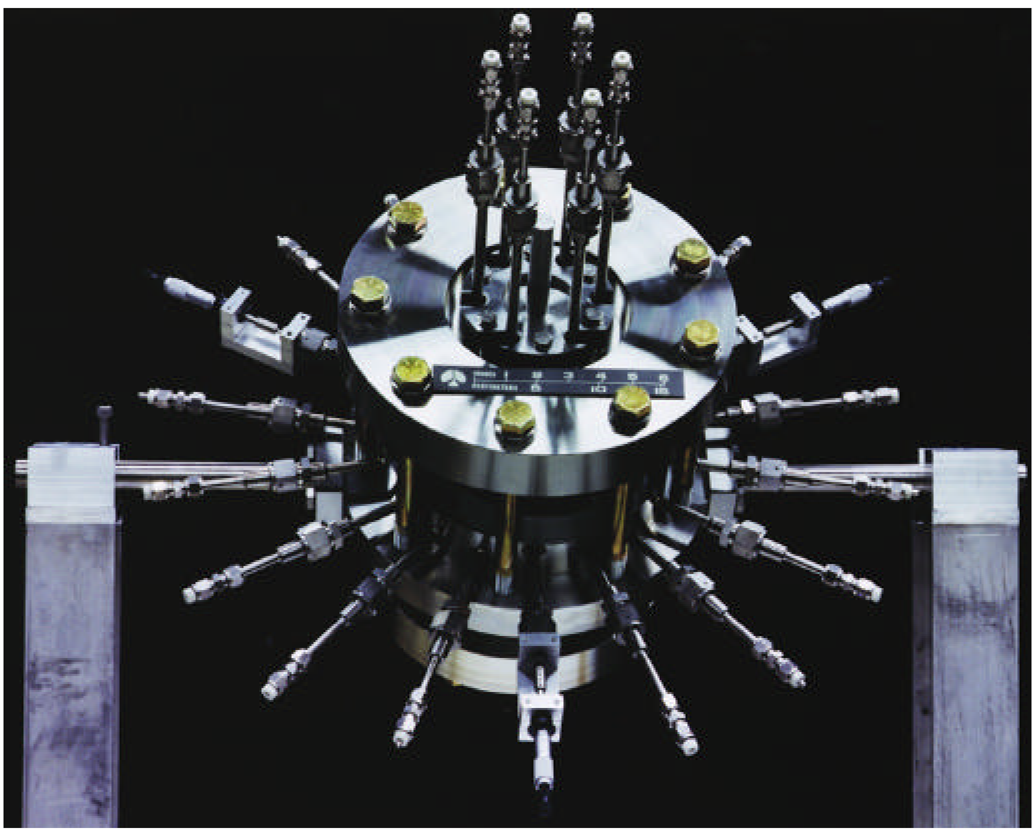
The concentrated solutions and deposits in tube/tube support plate crevices of nuclear steam generators have been correlated with several forms of corrosion on the outer secondary side of Alloy 600 steam generator tubes including intergranular attack/stress corrosion cracking (IGA/SCC), pitting, and wastage. Crevice chemistries in an operating steam generator cannot be measured directly because of their inaccessibility. In practice, computer codes, which are based upon hypothesized chemical reactions and thermal hydraulic mechanisms, are used to predict crevice chemistry. The objective of the Rockwell program is to provide an experimental base to benchmark crevice chemistry models, to benchmark crevice chemistry control measures designed to mitigate IGA/SCC, and to model IGA/SCC crack propagation processes. The important variables will be identified, including the relationship between bulk water chemistry and corrosion accelerating chemistries in a crevice. One important result will be the identification of water chemistry control measures needed to mitigate secondary side IGA/SCC in steam generator tubes. A second result will be a system, operating as a side-arm boiler, which can be used to monitor nuclear steam generator crevice chemistries and crevice chemistry conditions causing IGA/SCC.

Research Progress

The research approach uses an instrumented heated crevice, which is a replica of a PWR steam generator tube/tube support plate crevice (T/TSP). During the first contract year, the construction of this apparatus was completed.

The heated crevice consists of an Alloy 600 steam generator tube inserted through a drilled hole in a 1-inch thick Alloy 600 ring. The gap between the outer diameter (OD) of the tube and the drilled hole is 15 mils. The heated crevice operates at simulated steam generator thermal conditions and pressure. The entire assembly is contained in a 2 liter, Alloy 718 autoclave. The tube, the ring, and the autoclave are structurally independent and electrically isolated from each other. A cartridge heater is inside the Alloy 600 tube, which heats the tube to simulated primary water temperatures. The autoclave is heated by strip heaters and can be maintained at 280°C independent of the tube heater, simulating the bulk water temperature in a steam generator. In addition to thermocouples for temperature control in the bulk water in the autoclave and inside the tube, eight thermocouples are brazed onto the inner diameter (ID) of the Alloy 600 tube. These thermocouples are in a helical pattern at different depths in the crevice, and monitor the temperature profile in the crevice. Capillary tubes are located in the bulk water and extend into the crevice for extracting solution. A silver/silver chloride reference electrode is located in the bulk water for measuring the electrochemical potential (ECP) of the freespan of the Alloy 600 tube. A second silver/silver chloride reference electrode extends through the ring into the crevice for monitoring the electrochemical potential (ECP) of the crevice. Additional electrodes extend through the ring to the crevice for monitoring IGA/SCC of the tube.

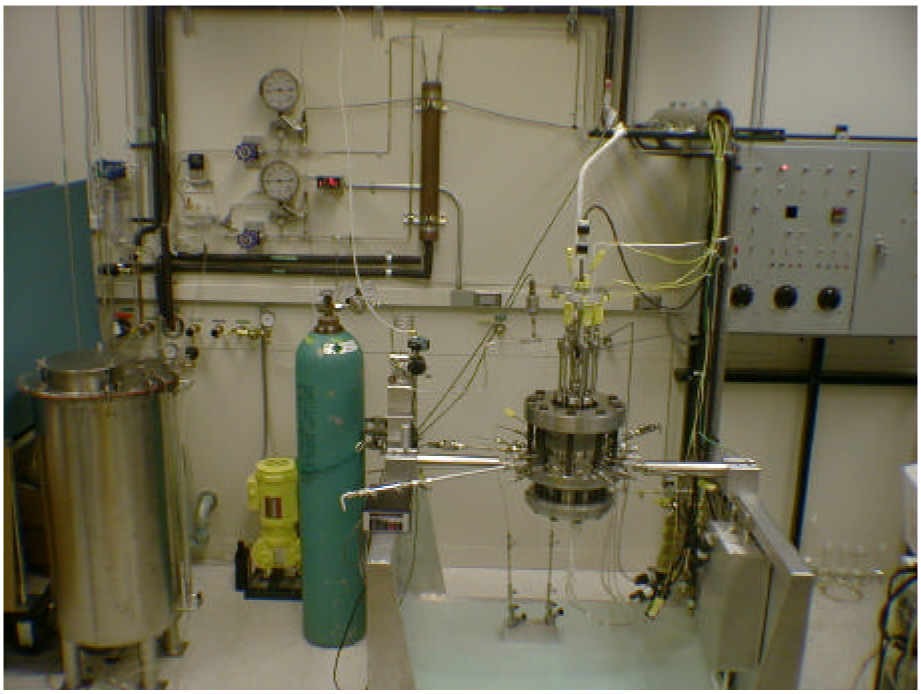
Illustrated below is a photograph of the assembled heated crevice. The radial array of ports for sensors surrounds the center section of the autoclave. An Alloy 600 tube, which does not have the heater assembly attached, is clearly visible in the center of the top section of the autoclave. There are six feedthroughs located in the top of the autoclave. These are for the water system and for inserting probes and thermocouples into the bulk water. Stainless steel rods, inserted into the center clamping ring, support the heated crevice on a stand so that it is suspended above the floor.



Assembled heated crevice before installing the tube heater

A continuously flowing water system is used. A high-pressure pump delivers water from the 500-liter makeup tank through the autoclave. A backpressure regulator allows the pressure in the autoclave to be adjusted to the 280°C boiling point. A system qualification run has been successfully completed, and experiments are now underway.

The photograph below shows the complete layout. The heated crevice assembly is in the center of the photograph. The pressurized Alloy 600 tube containing a cartridge heater extends trough the top. The connectors, surrounding the tube extension, are for thermocouples located at various locations in the tube and in the crevice. Compressed He, in the cylinder to the left of the autoclave, is used to pressurize the tube. A high pressure pump pumps deoxygenated water from the feed tank (located at the far left in the photograph) through the remainder of the water circuit Water exits the system after passing through a condenser. The panel on the wall at the far right contains the circuits and controls for the band heaters on the autoclave and the tube heater.



Complete set-up showing the heated crevice, the water circuit, and the control panel

An electrochemical noise (EN) system was set up for corrosion monitoring. This technique is based on the electrochemical nature of corrosion processes and the distinctive nature of EN signatures for all corrosion processes. The EN signature from SCC of Alloy 600 results from the creation of new surface area when a crack is initiated or advances. When a crack initiates or propagates, there is a transient surge in current associated with the oxidation processes of dissolution and film formation on the new surface exposed to the aqueous solution. Most of the current surge is from dissolution, which decreases rapidly as new oxide film is formed on the newly created surface.

EN was measured from stressed samples (C-rings) in a static autoclave containing a caustic solution at 320°C. These conditions simulate the chemistry and temperature in the heated T/TSP crevice when feedwater is used with a high concentration of NaOH. The length of SCC cracks in the specimens depended upon the magnitude of the applied stress and the time of exposure. A comparison of SCC crack lengths with the EN, indicated that crack propagation was discontinuous and that the cracks remained dormant for long intervals of time between propagation periods. The amplitude of current transients from stressed samples was over an order of magnitude greater than the amplitude from identical samples with no stress.

The EN results obtained using static autoclaves provide a baseline for measurements from the heated crevice. A heated crevice run is underway using deaerated feedwater with 40 ppm NaOH, simulated primary water of 320°C, and a bulk water temperature of 280°C. The heated crevice is packed with diamond powder to simulate the deposits found in steam generator crevices. Stress is applied by pressurizing the tube to 2800 psi. An analysis of solutions extracted from the crevice indicate that the crevice chemistry is 30 percent NaOH at a pH of 10.1. Early results show current transients suggesting the occurrence of SCC. This run will be continued until the tube is depressurized by a through-wall crack. After the run a metallographic evaluation will be performed to identify the SCC and other corrosion damage in the tube.

Planned Activities

The present strategy for mitigating IGA/SCC is based on the assumption that the crack initiation and propagation rate in Alloy 600 steam generator tubes in T/TSP crevices depend only on pH and the electrochemical potential. Planned work will examine the pH and electrochemical potential dependence of IGA/SCC in Alloy 600. The plan is to adjust the pH in the heated crevices from the caustic condition now used to lower pHs. This will be accomplished by replacing the presently used 40 ppm NaOH feedwater with feedwaters having progressively lower Na^+/Cl^- ion ratios. Computer codes indicate that the pH of the concentrated crevice solutions decrease as the Na^+/Cl^- ion ratio in the feedwater decreases. Crevice chemistries will be determined by chemical analysis of solutions extracted from the crevice. The crevice chemistry results will be used to benchmark the EPRI computer codes used by utilities to calculate crevice chemistry and crevice pH.

Planned work will also examine the effectiveness of the present practice of adding hydrazine to the feedwater to inhibit the initiation and propagation of IGA/SCC. The hypothesized rationale for adding hydrazine is that it lowers the ECP of the crevice chemistry to a regime where IGA/SCC does not occur. Measurements will be made of IGA/SCC and ECP at various pH with air saturated feedwater and with hydrazine added to deaerated feedwater.

Signal analysis and other data analysis procedures will be developed for the EN technique. The analysis will model the transition from microcracks to macrocracks and crack propagation processes. This will enable steam generator crevice monitoring for the initiation and severity of IGA/SCC.

Interfacial Transport Phenomena and Stability in Molten Metal-Water Systems

PI:	M. Corradini, University of Wisconsin-Madison (UW)	
Collaborators:	Argonne National Laboratory (ANL)	
Project Start Date:	August 1, 1999	Projected End Date: August 31, 2002
Project Number:	99-0233	

Research Objectives

One concept being considered for steam generation in innovative nuclear reactor applications involves water coming into direct contact with a circulating molten metal. The vigorous agitation of the two fluids, the direct liquid-liquid contact, and the consequent large interfacial area give rise to very high heat transfer coefficients and rapid steam generation. For an optimum design of such direct contact heat exchange and vaporization systems, more detailed knowledge is necessary of the various flow regimes, interfacial transport heat transfer coefficients, and operational stability under reactor relevant operating conditions. This research projects studies these transport phenomena involved with the injection of water into molten metals (e.g., lead alloys) with the following objectives:

- Design, fabricate and operate experimental apparatuses which investigate molten metal-water interactions under prototypic thermal-hydraulic conditions;
- Measure the integral behavior of such interactions to determine the flow regime behavior for a range of conditions and stability of these flow regimes;
- Measure the local interfacial mass and heat transfer behavior to ascertain the interfacial area concentration and heat transport length and time scales; and
- Analyze test results to determine an envelope of operating conditions, which yield optimum energy transfer between molten metal and water and maximizes stability.

Research Progress

In its first task of the project, the research team has done a comprehensive review of past experimental investigations, fabricated experimental apparatuses, and developed an experimental plan. Some of the highlights of this work are detailed here. Over the years, experimental studies have been conducted to provide measurements on direct contact heat transfer and vaporization of cold, volatile liquid drops dispersed in a continuous phase of hot liquid. A key measurement of these experimental studies is the evaporation zone height, *L*, which is the approximate height of the continuous phase of the hot liquid needed for complete evaporation of the injected dispersed liquid. This overall height, *L*, is related to the overall volumetric heat transfer coefficient for the evaporation zone, U_v, by an energy balance:

$$\boldsymbol{L\,)}\,T = (m?)\,h\boldsymbol{)}/\boldsymbol{U_v} \qquad (1)$$

where ***m?*** is the mass flux of the dispersed phase, ***)h*** is the change in enthalpy of the dispersed liquid to vapor (primarily the latent heat of vaporization of the cold liquid), and *)T* is the average temperature difference between the injected volatile liquid and the liquid metal pool. This temperature difference between the two fluids is rigorously the log-mean temperature difference (in these initial estimates we use the arithmetic difference). Basically, equation (1) represents a method to determine $\boldsymbol{U_v}$ by measuring the overall height empirically for volatile liquid vaporization and superheat. These past data indicate that the ***L)T*** product is constant for a low flow of injected volatile liquids (bubbly flow regime). In the direct contact heat transfer application for innovative liquid metal reactors, the flow regime is expected to be higher flow and churn-turbulent. Thus, the challenge is to measure this overall heat transfer coefficient in this flow regime and use X-ray diagnostics to measure the local area concentration to determine the local heat transfer coefficient.

Two experimental apparatuses were designed and constructed during the first year of the research contract as illustrated below. The ANL and UW facilities, while sharing this common goal, are designed to provide mutually complementary information. The ANL experiments focus on the heat transfer and flow stability behavior of water injected into molten metal and provide measurements on the evaporation zone length and associated volumetric heat transfer coefficients. The UW experiments focus on two-dimensional mixing behavior and provide real-time X-ray imaging of the multiphase structure of vaporizing water in the molten metal. The ANL 1-D and UW 2-D test sections are also expected to generate complementary aerosol data on the geometry effect of the molten metal pool.

Argonne One-Dimensional Test Apparatus

UW Two-Dimensional Test Apparatus and X-ray Imaging System

Planned Activities

Current analysis using a drift-flux model has verified the review of the past investigations and has supported the conclusion for experiments in the churn flow regime. Experimental work is clearly required to determine whether the predicted transition from bubbly to churn flow is correct or not. Moreover, tests would determine whether past data indicate any churn flow behavior are in the transition zone, as well as any potential differences in the observed heat transfer coefficients. The following test matrix is proposed to study such parameters and investigate them more deeply.

Water mass flow rate (g/s): 0.5 to 5.0
Water Subcooling (^{o}C): 0 to 70
Melt Superheat (^{o}C): 50 to 400
System Pressure (MPA): 0.1 to 1.0

Near term experimental activities also include test preparatory experiments and calibration of the X-ray imaging system for local area and heat transfer measurements:

- Assemble test section, support structure, water suppression tank, metal reservoir.
- Conduct imaging tests with X-ray system to calibrate void detection system.
- Setup power control and data acquisition systems for the first experimental test series.

The initial experimental test series is to begin this calendar year and will be compared to past liquid metal experiments from Japanese investigators.

Fundamental Thermal Fluid Physics of High Temperature Flows in Advanced Reactor Systems

PI: Donald M. McEligot, Idaho National Engineering and Environmental Laboratory (INEEL)

Collaborators: Iowa State University, University of Maryland, General Atomics, University of Manchester (UK), University of Podgorica (Montenegro), Kyoto University (Japan), Toyama University (Japan)

Project Start Date: August 1, 1999 Projected End Date: September 5, 2002

Project Number: 99-0254

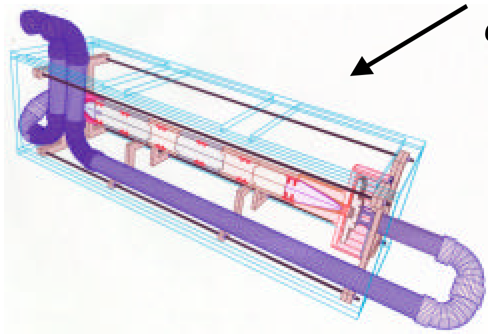
Research Objective

This laboratory/university/industry collaboration of coupled computational and experimental studies addresses fundamental science and engineering in an effort to develop supporting knowledge required for reliable approaches to new and advanced reactor designs for improved performance, efficiency, reliability, enhanced safety and reduced costs and waste. For small reactors, it addresses remote power and hydrogen generation. This research will provide basic thermal fluid science knowledge to develop increased understanding for the behavior of fluid systems at high temperatures, application and improvement of modern computation and modeling methods, and incorporation of enhanced safety features. The project promotes, maintains and extends the nuclear science and engineering base to meet future technical challenges in design and operation of high efficiency reactors, low output reactors, and nuclear plant safety.

The unique INEEL Matched-Index-of-Refraction (MIR) flow system, the world's largest such facility, is being applied to obtain fundamental data on flows through complex geometries important in the design and safety analyses for advanced reactors for the first time. A graphical representation of the MIR system is shown below. Successful completion of the study will provide the following new fundamental and engineering knowledge, which is not presently available in the literature:

- Time-resolved basic measurements of turbulence quantities (e.g., turbulence kinetic energy and Reynolds stresses) in internal gas flows with large property variation.
- Time-resolved data plus flow visualization of turbulent and laminarizing phenomena in accelerated flow around obstructions (spacer ribs) in annuli.
- Separation of effects of phenomena – buoyancy, property variation, and acceleration – occurring in strongly-heated gas flow to evaluate their individual importance and consequent flow behavior (by application of Large Eddy Simulation (LES) and Direct Numerical Simulation (DNS)).

World's Largest Matched-Index-of-Refraction Flow System



Ribbed annulus

Ribbed annulus

mixing and

impingement

- Application of DNS and LES for the first time to complex turbulent flows occurring in advanced reactors.
- Fundamental data of internal turbulence distributions for assessment and guidance of Computational Thermal Fluid Dynamic codes proposed for advanced gas cooled reactor (AGCR) applications.

Research Progress

- <u>Heat transfer and fluid flow in advanced reactors</u>: Six areas of thermal hydraulic phenomena in which the application of Computational Fluid Dynamic techniques can improve the safety of advanced gas cooled reactors have been identified.
- <u>Complex flow measurements</u>: Conceptual experimental models were developed for laser Doppler velocimeter measurements in the MIR flow system to examine flow in complex core geometries (ribbed annular cooling channels and control rod configurations) and in the transition from cooling channels to formation of jets issuing into a plenum. The initial experimental model is a ribbed annulus forming an annular jet exhausting into the MIR flow system. This model was designed, fabricated, and tested with the MIR system. The second experiment addresses two-

stage jet transition from generic coolant channels to a plenum; preliminary design of experiment and auxiliary flow control system has been completed.

- DNS development: DNS of laminarizing gas flow has been completed; subturbulent case initiated.
- LES development: Preliminary LES results have been obtained for vertical upward flow of air in a channel heated on one side and cooled on another; such a channel flow corresponds closely to the flow in an annular passage with a large radius ratio. This work is the first known LES study of a vertical flow accounting for buoyancy and variations in fluid properties.
- Multi-sensor probe development: Completed the design and construction of a four-sensor miniaturized probe. A calibration and testing facility has been designed and constructed for testing these probes.
- Mixed convection: Circular tube test facility in operation with a vertical, heated section providing either specified distributions of heat flux or temperature along the tube with air flow in either the upward or downward direction. Measurements of mean velocity and temperature profiles in the radial direction have been obtained near the exit for the upward flow case under buoyancy-influenced conditions at Reynolds numbers from 6,000 to 20,000.

Planned Activities

- DNS and LES development: Work will continue.
- Complex Flow Measurements: laser doppler velocimeter (LDV) measurements of velocities and turbulence will be conducted with the initial model. The second experiment will be fabricated.
- Mixed Convection and Multi-sensor Probes: Three experiments emphasizing buoyancy effects are planned. The first obtains data on velocity in strongly heated air flow through a heated pipe under conditions of mixed convection to determine the effects of buoyancy forces combined with property variation on local mean velocity and turbulent fluctuations. A second experiment would measure the influences of the temperature dependence of fluid properties and buoyancy for air flowing in an annulus. The third will measure local mean velocities and turbulent fluctuations.

An Innovative Reactor Analysis Methodology Based On a Quasidiffusion Nodal Core Model

PI: Dmitriy Y. Anistratov, Texas A&M University

Collaborators: Oregon State University, Studsvik Scanpower, Inc.

Project Start Date: August 1999 Projected End Date: September 2002

Project Number: 99-0269

Research Objectives

The present generation of reactor analysis methods uses few-group nodal diffusion approximations to calculate full-core eigenvalues and power distributions. The cross sections, diffusion coefficients, and discontinuity factors (collectively called "group constants") in the nodal diffusion equations are parameterized as functions of many variables, ranging from the obvious (temperature, boron concentration, etc.) to the more obscure (spectral index, moderator temperature history, etc.). These group constants, and their variations as functions of the many variables, are calculated by assembly-level transport codes. The current methodology has two main weaknesses that this project will address. The first weakness is the diffusion approximation in the full-core calculation; this can be significantly inaccurate at interfaces between different assemblies. This project will use the nodal diffusion framework to implement nodal quasidiffusion equations, which can capture transport effects to an arbitrary degree of accuracy. The second weakness is in the parameterization of the group constants; current models do not always perform well, especially at interfaces between unlike assemblies. The project will develop a theoretical foundation for current models and use that theory to devise improved models. The new models will be extended to tabulate information that the nodal quasidiffusion equations can use to capture transport effects in full-core calculations.

Research Progress

Homogenization and Group Constants Functionalization: During the first year, part of the research efforts were concentrated on studies of the problem in one-dimensional geometry in order to develop basic ideas of functionalization and homogenization procedures. To model reactor physics phenomena of interest, first a code was developed for solving one dimensional (1D) one-group eigenvalue neutron transport problems based on the quasidiffusion (QD) method. Then a code was developed for solving 1D multigroup transport problems by means of the QD method.

Certain approaches have been formulated for spatial assembly homogenization that are based on consistent discretization spatially averaged QD low-order equations and their fine-mesh discretization. The homogenization procedure must preserve the averaged reaction rates, surface-averaged group currents, and eigenvalue. The homogenization fits naturally into the framework of the QD method that is based on the idea of successive averaging of the transport equation over angular and energy variables. The averaging over spatial variables is the next logical step. An approach has been developed for exact spatial averaging of the discretized QD low-order equations and generating a coarse-mesh discretization that is exactly consistent with the given fine-mesh discretization. The developed technique can be applied to any number of spatial zones. This is a rigorous mathematical result. The proposed method uses the quantities that are similar by their definitions to discontinuity factors, however, the resulting solution preserves continuity of both the scalar flux and the current on interfaces. The procedure of spatial decomposition based on albedo boundary conditions was formulated. The proposed methodology creates a theoretical background for homogenization of spatial regions. The presented approach of consistent coarse-mesh discretization can be extended to multigroup and multidimensional problems, as well as to different kinds of discretization methods. These results were presented at the Winter ANS Meeting 2000 in Washington D.C. and published in its transactions.

<u>Numerical Method for Solving Two Dimensional (2D) QD Low-Order Equations</u>: During the first year of this project, effort has been focused on developing nodal solution techniques for the quasidiffusion low-order equations (QDLO) in x - y geometry. There are several important differences between these equations and the standard neutron diffusion equation. First, the neutron current is equal to the gradient of the QD tensor times the scalar flux, rather than using a standard diffusion Fick's law representation and, second, this QD tensor is a complex function of space, in that it is computed directly from the solution of the transport equation. The partial differential equation (PDE) associated with the QD low-order equations is much more challenging to work with than standard diffusion, especially in the context of advanced nodal discretizations.

Initially, the focus was on using standard polynomial based nodal techniques (namely the QPANDA method) with the QDLO equations. In the QPANDA approach, the diffusion equations within a node are transverse-integrated to generate coupled one-dimensional diffusion equations. These transverse-integrated fluxes are then approximated by a fourth-order polynomial expansion, and the resulting systems of equations are then solved iteratively. Project researchers attempted to apply the QPANDA technique to the QDLO equations in two ways. First, all the leakage terms were treated implicitly; i.e., leaving the equations in coupled x - y form and second, by treating the leakage terms semi-implicitly, causing each direction to be decoupled, but with an extra level of iteration on the transverse leakage terms. Then the nodal discretization techniques were fundamentally changed by adopting instead the combination polynomial-analytic nodal method that was shown to provide the accuracy needed in the simulation of mixed oxide-uranium oxide fueled reactor cores.

The non-linear marching method was also evaluated—a solution technique designed for use with the QDLO equations with a finite volume discretization, as a potential solution strategy for the nodal QDLO equations. It was discovered that the added complexity of the nodal discretization made the non-linear marching method less desirable than standard iterative techniques.

Planned Activities

Research activities will focus upon two areas—the full-core few-group diffusion-like calculation and the assembly-level many-group transport calculation—and upon the interface between them. Work will proceed on developing homogenization procedures as well as methodologies for functionalization of data based on single-assembly calculations. The effects of an unlike neighboring assembly upon a given assembly's group constants must be accurately estimated without knowledge of the neighbor. Capturing such "interface effects" is one of the major issues studied on this project. The developed methodologies will be applied to multidimensional geometries. Research will proceed on advanced discretization schemes for the multidimensional quasidiffusion low-order equations that will enable researchers to efficiently solve reactor physics problems of core designs with MOX fuel.

Radiation-Induced Chemistry in High Temperature and Pressure Water and Its Role in Corrosion

PI: David M. Bartels, Argonne National Laboratory

Collaborators: Atomic Energy of Canada LTD - Chalk River Laboratories

Project Start Date: August 1999 Projected End Date: August 2002

Project Number: 99-0276

Research Objective

Commercial nuclear reactors essentially provide a source of heat, used to drive a "heat engine" (turbine) to create electricity. A fundamental result of thermodynamics shows that the higher the temperature at which any heat engine is operated, the greater its efficiency. Consequently, one obvious way to increase the operating efficiency and profitability for future nuclear power plants is to heat the water of the primary cooling loop to higher temperatures. Current pressurized water reactors run at roughly 300^{o}C and 100 atmospheres pressure. Designs under consideration would operate at 450^{o}C and 250 atmospheres, i.e., well beyond the critical point of water. This would improve the thermodynamic efficiency by about 30 percent. A major unanswered question, however, is: What changes occur in the radiation-induced chemistry in water as the temperature and pressure are raised beyond the critical point, and what does this imply for the limiting corrosion processes in the materials of the primary cooling loop?

The direct measurement of the chemistry in reactor cores is extremely difficult, if not impossible. The extreme conditions of high temperature, pressure, and radiation fields are not compatible with normal chemical instrumentation. There are also problems of access to fuel channels in the reactor core. For these reasons, theoretical calculations and chemical models have been used extensively by all reactor vendors and many operators, to model the detailed radiation chemistry of the water in the core and the consequences for materials. The results of these model calculations can be no more accurate than the fundamental information fed into them, and serious discrepancies exist between current models and reactor experiments. The object of this research program is to generate the necessary radiation chemistry data (yields and reaction rates) needed to accurately model chemistry in both existing water-cooled reactors, and the higher temperature reactors proposed for the future. This will allow engineers to define the optimum chemical conditions conducive to long life of the primary heat transport system.

Research Progress

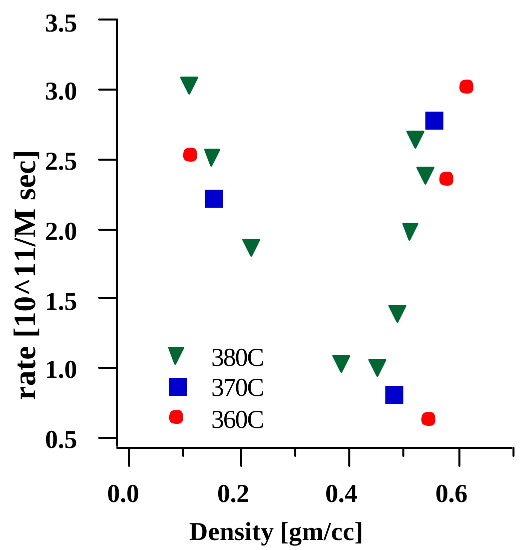
Irradiation of water produces mostly H_3O^+, OH radicals, and solvated electrons—$(e^-)_{aq}$. Using the Argonne Chemistry Division's 20MeV electron linac, a short pulse can be used to create a high concentration of these species for kinetic studies. The solvated electron absorbs intensely in the visible and near-IR, making it easy to detect even on subnanosecond time scales. Reaction rates are measured by monitoring how fast the absorption disappears in the presence of a scavenger. In the first year of the project, researchers have focussed on measuring the spectra and some reactions of this important species.

Cell Design: As the necessary first step, a flow system was constructed for the experiment in order to allow signal averaging of solutions, and also to allow fast analysis of products after the irradiation. The intention is to analyze products with liquid chromatography methods, using the pulse radiolysis as the "injector." Typically two HPLC pumps provide flow of pure water and a scavenger solution, which is mixed in a "T" before the cell. The flow rate of 2-10 ml/minute is sufficient to flush the cell quickly and also maintain the pressure drop in a back pressure regulator or capillary after the cell. The most difficult aspect of the experiment is to maintain stable flow conditions inside the cell. If the temperature of water entering the cell is not the same as water already inside, schlieren effects scatter the analyzing light very severely. Details of the entire apparatus have been published as the first "deliverable" of the project.

Solvated Electron Spectrum: The strongest absorption induced by irradiation of supercritical water is that of solvated electrons, and it is natural that efforts begin with a study of its spectrum. Extinction coefficients of this species will be needed to determine second order recombination reaction rates in a later study. A great deal can already be learned about the electron solvation environment and energetics just from the shape of the spectrum.

Solvated electron spectra were recorded in heavy water at various temperatures from 300 to 450 degrees. The temperature shift of the maximum absorption, roughly –.0028eV/deg K, is similar to that found by other workers. Above 300^{o}C, the spectrum becomes sensitive to the pressure (i.e., density) as well as the temperature. Data from a series of spectra recorded at different densities at 380^{o}C (just above the critical temperature) showed the spectrum shifting to the red as density (pressure) is decreased, just as observed for solvated electron spectra in other liquids.

Reactions with O_2: The figure below shows the first measurements of reaction rate for electrons with the oxygen molecule in supercritical water. At temperatures below 300^{o}C in liquid water this reaction obeys a "normal" Arrhenius law, but close to and above the critical temperature the rate is a strong function of density. A qualitative explanation of this behavior might be found in the hydrophobic nature of the O_2 molecule. At low density it is expected that the negatively charged $(e-)_{aq}$ will be clustered with water molecules, but the hydrophobic O_2 molecule will be found in the voids between clusters. As density is increased the clusters around $(e-)_{aq}$ should become larger. The clustered water molecules may simply prevent the hydrophobic O_2 from approaching the electrons, presenting a "potential of mean force" barrier, so the reaction rate goes down as clusters get larger. At some point the density becomes large enough that transient clusters merge into a more continuous medium. Then the reaction rate increases again to approach the high-density liquid limit as extrapolated from lower temperature Arrhenius plots. Based just on this initial study, it is clear that simple extrapolation of lower-temperature reaction rate data for the purpose of making model predictions in reactor cores will not be possible.



Rate constant for reaction of solvated electrons with O_2 at several temperatures as a function of water density.

Planned Activities

Additional experiments in progress include final precise measurements of the electron spectra at selected temperatures and pressures, some further O_2 reaction rates, and a similar investigation of SF_6 reaction rates with the solvated electron. (Similar density effects appear to apply to this reaction.) Publications on the spectra and oxygen reactions are in preparation. A survey of reactions of the hydrated electron with numerous solutes including NO_3^-, NO_2^-, N_2O, Fe_3^+, and Ni_2^+ can now be carried out in a routine fashion. A very important reaction to measure is the solvated electron with hydronium ion (acid), which cannot be introduced into the normal high pressure cell for fear of corrosion. The project team has built a second cell with special acid-resistant coating for the purpose of measuring this reaction.

A reaction of critical importance to understanding the kinetics at elevated temperatures is the bimolecular recombination of hydrated electrons:

$(e-)_{aq} + (e-)_{aq}$? $H_2 + 2\ OH^-$

Project researchers will be building equipment to allow high-pressure hydrogen-saturated water to be pumped into the cell in order to measure this reaction, and also the reaction of OH radicals with H_2. The latter reaction is the key to suppression of corrosion effects in the primary heat transport system of a nuclear reactor.

Novel Concepts for Damage-Resistant Alloys in Next Generation Nuclear Power Systems

PI: S.M. Bruemmer, Pacific Northwest National Laboratory

Collaborators: General Electric Corporate Research & Development, University of Michigan

Project Start Date: August 1, 1999 Projected End Date: August 31, 2002

Project Number: 99-0280

Research Objective

The objective of the research is to develop the scientific basis for a new class of radiation-resistant materials to meet the needs for higher performance and extended life in next generation power reactors. New structural materials are being designed to delay or eliminate the detrimental radiation-induced changes that occur in austenitic alloys, i.e., a significant increase in strength and loss in ductility (<10 displacements per atom (dpa)), environment-induced cracking (<10 dpa), swelling (<50 dpa), and embrittlement (<100 dpa). Non-traditional approaches are employed to ameliorate the root causes of materials degradation in current light water reactor (LWR) systems. Changes in materials design are based on mechanistic understanding of radiation damage processes and environmental degradation and the extensive experience of the principal investigators with core component response.

Research Progress

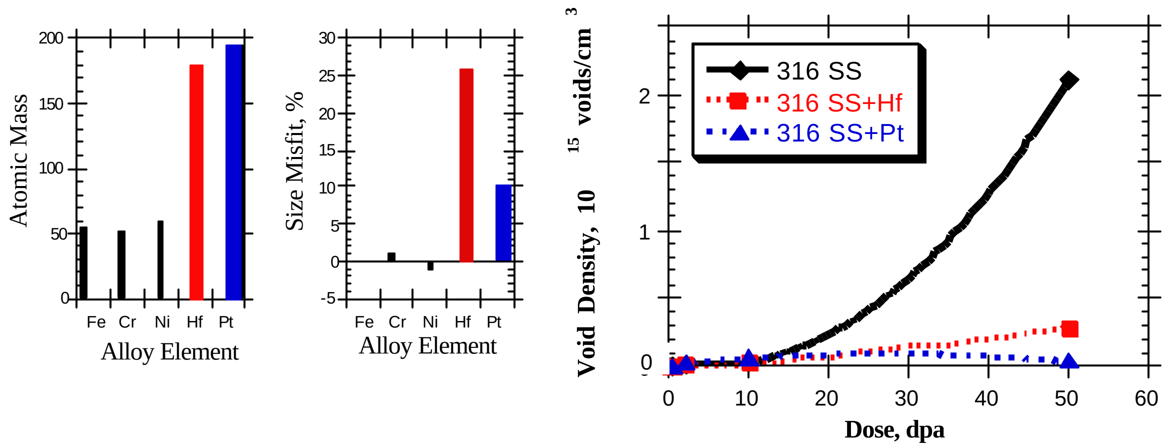
Phase 1 research has focused on the lattice perturbation mechanism for damage resistance. Alloy fabrication for heavy-ion, proton and neutron irradiations, and for stress corrosion cracking (SCC) testing, was completed. Three sets of Ni-ion irradiations (to various dose levels) were conducted and microstructures characterized by transmission electron microscopy. These experiments are being used to establish direction for further studies using protons and neutrons. The proton irradiation source was upgraded for this high-dose study and the first series of samples were irradiated. Stress corrosion crack growth rates have been established in high-temperature water environments for strengthened, non-irradiated stainless steels. The tests on the intermediate strength stainless steel were completed.

<u>Materials Processing</u>

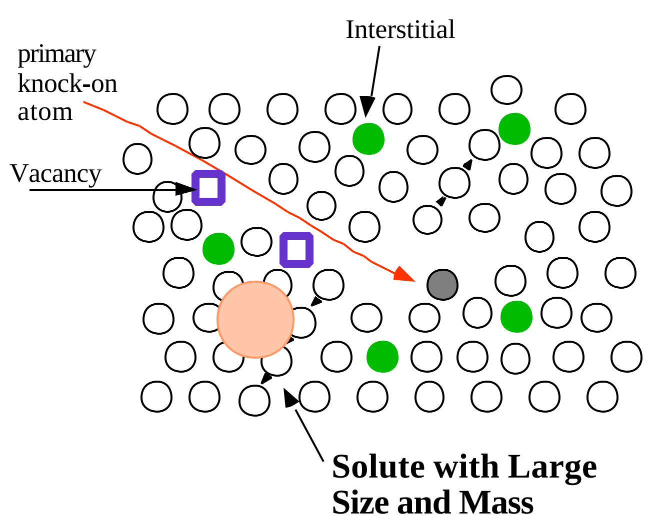
- Alloys were successfully fabricated having the optimum small grain structure with grain interiors of low network dislocation density. Research alloys were fabricated from button melts of pure metals. Alloy samples were processed to achieve the desired sheet form and grain microstructure for subsequent irradiations.

<u>Irradiations</u>

- Charged particle irradiation techniques were enhanced to enable the high-dose conditions needed for the project. In particular, the Ni^{++} ion irradiation technique was improved to produce uniform temperatures and beam flux over a large sample area and reach very high radiation doses. Stainless steel samples were irradiated to a series of irradiation doses from 0.5 to 50 dpa. The proton accelerator system was upgraded to increase the maximum beam current by more than 3 times. Initial irradiations were completed at 5 dpa.



Solute additions of large mass and size misfit have been shown to suppress radiation damage in stainless steels. Similar behavior between Pt and Hf alloys suggests that atomic mass (cascade production) is more important than atomic misfit (point defect trapping) for nucleation of voids.



Minor additions of massive oversized solute are being explored to delay or eliminate detrimental radiation-induced material changes. Next generation water reactors will require damage resistance to very high irradiation doses.

Materials Characterization

- Techniques for reliable examination of near surface regions were optimized for the Ni^{++} ion samples and defect microstructures were characterized. Transmission electron microscopy revealed distinctly different microstructural evolution paths for the base stainless steel alloy compared to the alloys with small platinum or hafnium additions. Dislocation loop development and void development were affected indicating that large misfit solute atoms can alter the formation of detrimental microstructures by modifying defect recombination, migration, and aggregation.

Mechanical Behavior and Stress Corrosion Cracking

- Reliable stress corrosion crack growth rate measurements on stainless steels in high-temperature water (290 to 340^{o}C) were established at low growth rates. Unexpectedly, an increase in matrix strength alone was demonstrated to promote intergranular stress corrosion cracking in both oxidizing and non-oxidizing environments. Detrimental changes in grain boundary composition were not required for cracking. This result is the first step toward isolating radiation strength effects on stress corrosion cracking from other radiation-induced material changes.

Planned Activities

In Phase 2, property evaluation of lattice-perturbation alloys studied in Phase 1 will be continued with an emphasis on the proton-irradiated materials. The ability of the large misfit solutes to alter the evolution of detrimental microstructures and effects on irradiation-assisted stress corrosion cracking will be evaluated. Additional charged particle irradiations will be performed as needed to establish radiation resistance to very high dose levels. Strength effects on stress corrosion crack growth will be quantified in the non-irradiated alloys and compared to cracking for irradiated stainless steels. Research will begin on the metastable precipitate alloys that are tailored to alter radiation-induced material changes and improve properties. New alloys will be fabricated and heavy-ion irradiations will be conducted.

Advanced Ceramic Composites for High-Temperature Fission Reactors

PI: Russell H. Jones, Pacific Northwest National Laboratory

Project Start Date: August 1, 1999 Projected End Date: August 31, 2002

Project Number: 99-0281

Research Objective

This research seeks to develop the understanding needed to produce radiation-resistant SiC/SiC composites for advanced fission reactor applications. Structural and thermal performance of SiC/SiC composites in a neutron radiation field depends primarily on the radiation-induced defects and internal stresses resulting from this displacement damage. The researchers are working to develop comprehensive models of the thermal conductivity, fiber/matrix interface stress and mechanical properties of SiC/SiC composites as a function of neutron fluence, temperature, and composite microstructure. This model will be used to identify optimized composite structures that result in the maximum thermal conductivity and mechanical properties in a fission neutron field.

Research Progress

Task 1 (Develop a Model of Radiation Effects on the Dimensional Stability of Monolithic SiC) accomplishments are as follows:

- Development of a method for calculating spectral-averaged displacement cross sections for SiC using the SPECOMP code to integrate the numerically determined displacement functions in SiC over the spectrum of recoil atom energies for a given neutron energy. The spectral averaged displacement cross section for a specific neutron field is then obtained by integrating over the spectrum of neutron energies.

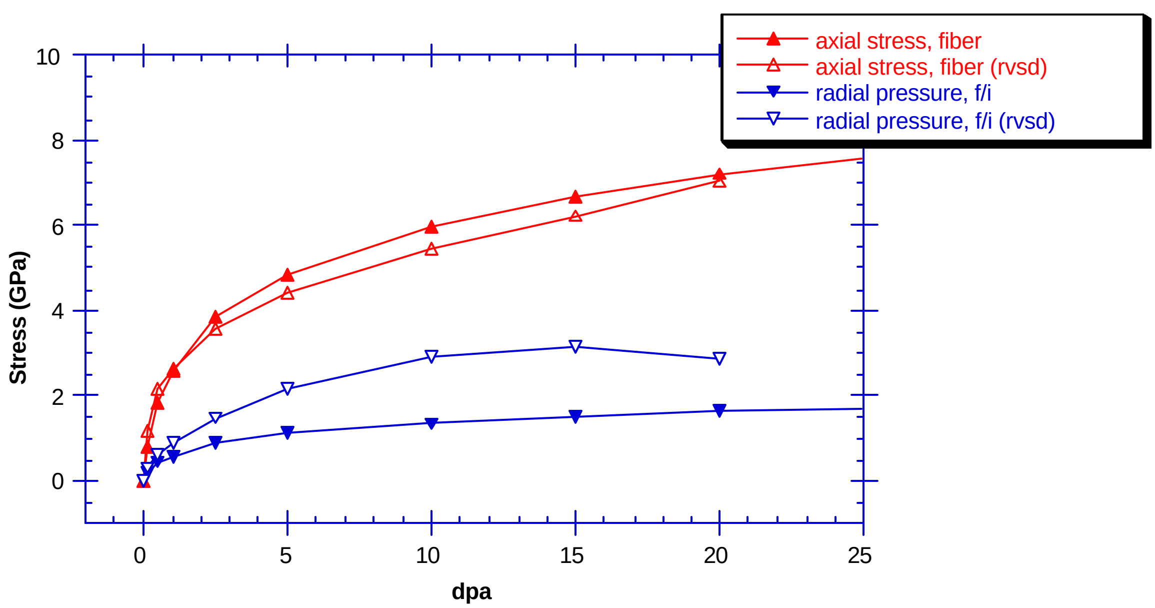
- The transmutation of atoms under neutron irradiation can significantly affect material properties. Transmutation can lead to production of gaseous impurities, especially He and H through (n, α) and (n,p) reactions, respectively. Helium may play an important role in the dimensional stability of SiC. Transmutation also leads to production of substitutional and interstitial impurity atoms, and the burnout of the original material. Transmutation calculations were performed for pure SiC irradiated in the neutron spectra of the fast reactor EBR-II and the mixed spectrum reactor HFIR using the REAC-3 code with FENDL-2.0 nuclear cross sections.

Task 2 (Develop a Model of Radiation Effects on the Dimensional Stability, Internal Stress and Thermal Conductivity of SiC/SiC Composites) accomplishments include:

- SiC fibers are either prepared from polymer precursors or SiC powders so their dimensional stability is not identical to that of monolithic SiC. All existing data on the dimensional change due to neutron irradiation for all available SiC fibers has been collected and reviewed. A mathematical description of the dimensional change of these fibers has been derived as a function of neutron fluence, temperature, and fiber type.

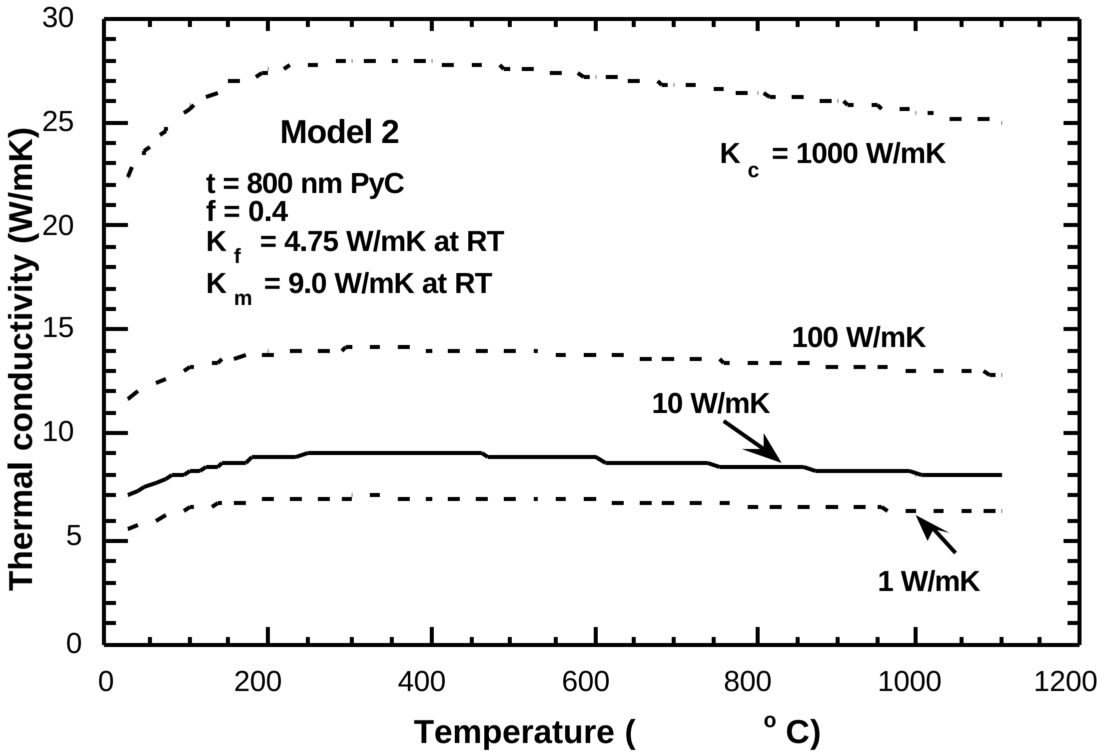
- Interphases between the fiber and matrix are critical to the dimensional, thermal, and mechanical performance of SiC/SiC composites. Evaluation of the response of various phases of carbon to neutron irradiation was carried out. Carbon interphases exist as amorphous, turbostratic or graphitic forms. The data for the irradiation effects on dimensional changes for all forms of C have been reviewed with consideration of anisotropic effects. It is known that the graphitic form of C exhibits considerable anisotropic swelling from the formation of defect clusters on the basal plane. Concepts for beneficial use of this anisotropy to minimize the stress between fibers that shrink and a matrix that swells were evaluated.

- Preliminary formulation of a 3-cylinder dimensional change/interfacial stress model was developed to examine the dissimilar dimensional changes between the fiber, interphase and matrix resulting from neutron irradiation that leads to internal stress between these composite components. The preliminary results for the stress between the fiber and matrix that develops during irradiation are demonstrated below.



A comparison of stresses predicted for Hi-Nicalon fibers in a silicon carbide matrix composite with a carbon interphase.

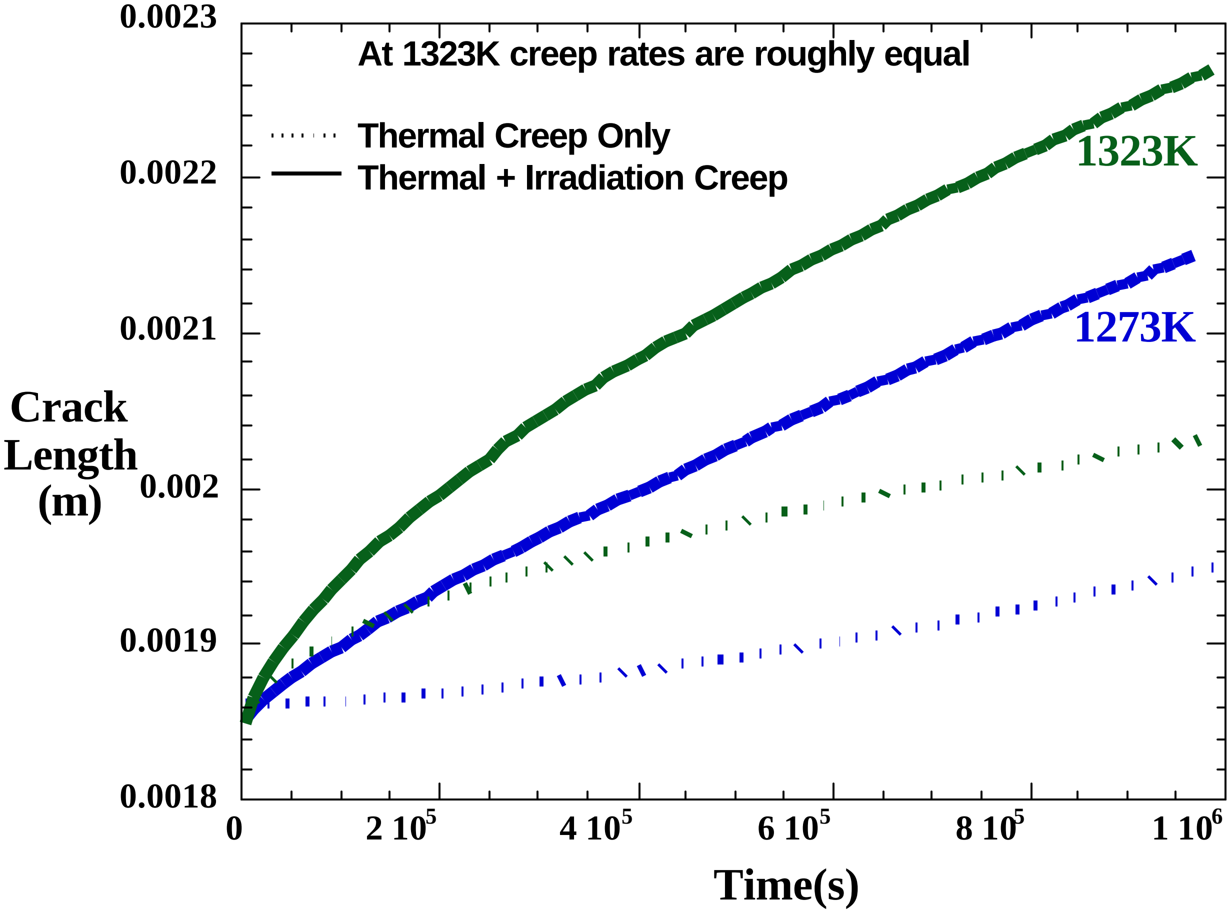
- Models for predicting the effective-transverse, thermal conductivity of SiC/SiC composites were evaluated and compared to experimental data. The modeling effort demonstrated the strong dependence of the composite thermal conductivity on the interphase conductance, K_c, which are in turn dependent on the interphase thermal conductivity, bonding to the matrix and phonon coupling between the matrix/interphase and interphase/fiber. An example of the model calculations is shown in the graph below.



Model predictions of K_{eff} for a 2D-Hi-Nicalon/PyC/SiC composite when the PyC fiber coating has a 800 nm thickness.

Task 3: (Model the Mechanical Properties of Irradiated SiC/SiC Composites) accomplishments include:

- A dynamic crack-growth model has been developed to predict crack growth in ceramic composites containing creeping fibers in an elastic matrix. Mechanics for frictional bridging and both linear and nonlinear fiber-creep equations are used to compute crack extension dynamically. Thermal creep of polymer derived ceramic fibers is often nonlinear, or viscous-like, in time and stress, but irradiation creep of these same fibers appears to be linear in both time (dose) and applied stress. Accordingly, we used a standard thermal creep equation and a simple irradiation creep equation suitable for SiC-based fibers and generated some model results for two test temperatures, 1273 and 1373K (1000 and 1100 Centigrade). The thermal creep equation has an activation energy of about 600 kJ/mol. The irradiation creep equation assumes a temperature independent regime below 1173K (900 C) and an activation energy of 50 kJ/mol for temperatures greater than 1173K. The creep rate is linear in dose rate and stress. We obtain the following results from the model.



Model result in terms of crack length as a function of time for the indicated test temperatures. We observe that irradiation creep of the fibers dominates the fiber deformation process for temperatures below 1273K (1000 C) but thermal creep dominates at higher test temperatures. This implies that we will need to further understand and model irradiation creep processes in SiC-based fibers since apparently that process is important in applications, such as fission reactors, that operate at or below 1000 C.

Planned Activities

The relationship between radiation damage, displacements per atom, chemical transmutations, and fluence will be established for an HTGR neutron spectrum. The thermal conductivity model will be used to identify composite microstructures with optimized thermal conductivity. A concentric cylinders model was used to determine the stresses in a three constituent system: fiber, interphase, and matrix but the preliminary model needs further development to include dimensional change with various C interphase microstructures. The concentric cylinders model and data on the irradiation creep of fibers will be utilized with PNNL's creep crack growth model to predict the creep behavior of SiC/SiC composites in a neutron field with the combination of thermal, irradiation and oxidation enhanced creep crack growth.