INL/CON-11-23186
PREPRINT

U.S. Department of Energy Accident Resistant SiC Clad Nuclear Fuel Development

Enlarged Halden Programme Group Meeting 2011

George Griffith

October 2011

Title

U.S. Department of Energy Accident Resistant SiC Clad Nuclear Fuel Development INL/CON-11-23186

Author

George Griffith
Idaho National Laboratory
Light Water Reactor Sustainability Advanced LWR Nuclear Fuel Pathway

Abstract

A significant effort is being placed on silicon carbide ceramic matrix composite (SiC CMC) nuclear fuel cladding by Light Water Reactor Sustainability (LWRS) Advanced Light Water Reactor Nuclear Fuels Pathway. The intent of this work is to invest in a high-risk, high-reward technology that can be introduced in a relatively short time. The LWRS goal is to demonstrate successful advanced fuels technology that suitable for commercial development to support nuclear relicensing. Ceramic matrix composites are an established non-nuclear technology that utilizes ceramic fibers embedded in a ceramic matrix. A thin interfacial layer between the fibers and the matrix allows for ductile behavior. The SiC CMC has relatively high strength at high reactor accident temperatures when compared to metallic cladding. SiC also has a very low chemical reactivity and doesn't react exothermically with the reactor cooling water. The radiation behavior of SiC has also been studied extensively as structural fusion system components. The SiC CMC technology is in the early stages of development and will need to mature before confidence in the developed designs can created. The advanced SiC CMC materials do offer the potential for greatly improved safety because of their high temperature strength, chemical stability and reduced hydrogen generation.

Keywords: silicon carbide, SiC, ceramic matrix composite, cladding

Introduction

The goal of the LWRS Advanced LWR Nuclear Fuel Pathway is to develop advanced nuclear fuels that will support the continued safe and economic operation of the current generation of nuclear power plants operating in the United States. Additional benefits include the fundamental scientific understanding of nuclear fuel behavior and advanced computational predictive tools. The testing and development program will also help speed the development of additional fuel technologies.

The development of new technical features in the current generation of nuclear fuel takes approximately 10 years. On-going technical advances in nuclear fuel are providing are being made with greater effort and lower performance gains. The Department of Energy is developing technical solutions that provide a higher benefit at a higher risk than industry would on their own develop.

After the Fukushima Daiichi nuclear accident a strong emphasis on increasing safety has developed. The intent is to delay core damage and reduce the extent of the damage in severe station black out or LOCA events. The LWRS mission also has the additional requirement that nuclear power plant economics improve making current nuclear power plant operation desirable. The improved economics could come from increased fuel cycle lengths, burn-up or higher power density.

The approach taken in the Advanced LWR Fuels pathway is to emphasize one flagship technology centered on SiC CMC nuclear fuel cladding. This cladding technology offers the potential for rapid technical demonstration because there is an established non-nuclear industry and known nuclear properties from fusion system applications. The high strength at high temperatures, stable properties under irradiation, and low chemical activity will allow significantly improved safety margins. Improvement in margin and improved behavior will also allow economic improvements at the nuclear power plant.

LWRS mission

The LWRS mission centers maintaining the safe and efficient operation of nuclear power plants beyond the current 60 licensing period. Strategic needs that exceeded industrial capabilities were reviewed and four areas were selected for development. These include material aging, instrumentation and controls, improved plant risk evaluation and advanced LWR fuels. Each area is being developed as part of a Pathway in the larger LWRS program. Each Pathway is optimized to assist in the plant reinvestment choices that would be needed to maintain nuclear power plant operation after 60 years.

The Advanced LWR Nuclear Fuel Pathway is intended to demonstrate SiC CMC cladding technology by 2015. Fuels rods will be fabricated and operated with technology that can be upscaled economically to commercial requirements. The advanced fuel performance will be understood well enough that a licensing application can be confidently created. The Pathway will also have developed computational models predicting fuel performance over the entire operating regime.

Advanced LWR Nuclear Fuel Pathway

A flagship technology was selected to maximize the return on the research funds and allow for significant progress before the 2015 LWRS mission date. Multiple technologies including, unique fuel composition and fuel geometry were evaluated before moving ahead with SiC CMC nuclear fuel cladding development. Industry was already evaluating and producing prototype SiC CMC rodlets creating an intrinsic industry interest. High strength at high temperatures and low chemical activity, including no exothermic reaction with water as zirconium demonstrates, at elevated temperatures were the primary reasons to select SiC CMC for development. SiC CMC structure technology was funded by the aerospace and defense industries allowing the nuclear applications to advance an existing mature specialized technology. SiC CMC structures properties are also understood because they have been evaluated for use in fusion reactor first wall applications. Focus on a single development activity has allowed the program to make rapid progress.

The high temperature properties of SiC CMC imply that the fuel system can retain its geometry and fuel protective function even during an accident. Removal of the exothermic zirconium and water reaction also increases the temperature at which the fuel can operate. Eliminating the generation of free hydrogen would also lower type of risks created during an accident scenario. These properties tend to reduce the severity of the severe accidents similar to Fukushima-daiichi.

The Advanced LWR Nuclear Fuel Pathway includes multiple university, laboratories, and commercial interests. The use of many knowledgeable resources and inputs has assisted in rapidly moving forward. The growing interaction with fuel vendors and supplies is helping set the strategic goals. A figure describing the program is shown in Figure 1.

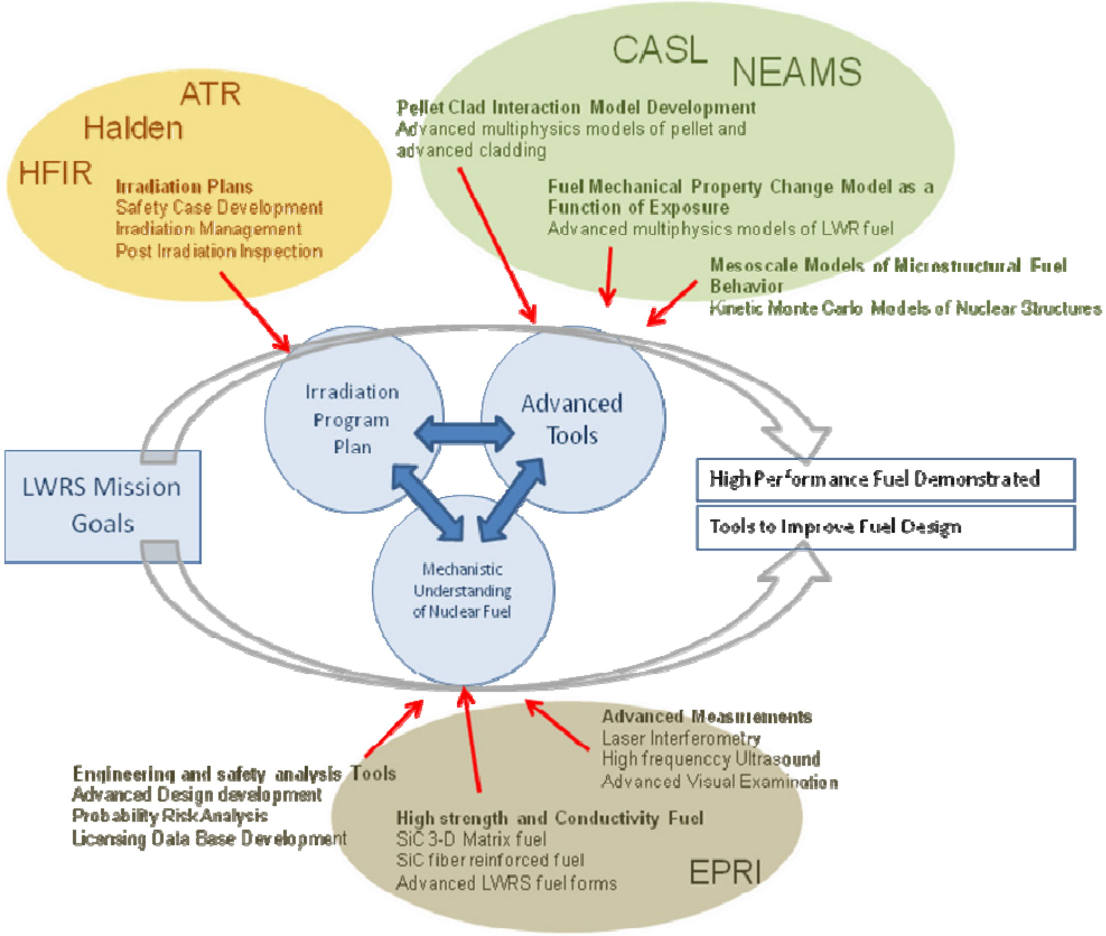

Figure 1: LWRS Advanced LWR Nuclear Fuel Pathway Structure

SiC CMC Cladding

The properties of SiC CMC that make it attractive for nuclear fuel cladding include minimal chemical activity, reasonable thermal conductivity, high strength at high temperatures, SiC decomposes at 2700C, and established fabrication techniques. [i]SiC CMC show little chemical reaction at elevated temperatures compared to zirconium. Even at very high temperatures the major reaction is exchange of carbon for oxygen in the SiC structure. This reaction produces a less reactive silicon dioxide layer. The conductivity of SiC can exceed the value of zirconium before irradiation. Extended irradiation tends to lower the conductivity to a value half to one-third that of zirconium. Material property optimization and design details, final SiC CMC thickness, can help mitigate the penalty created by the reduced thermal conductivity. The ultimate strength of a CMC is established by the SiC fibers which have a very high modulus. The soft but bonding interface layer between the matrix and fibers allows cracking in the matrix and stress distribution to the included fibers. This property allows the CMC to display a more graceful failure and increased apparent ductility compared to a pure ceramic. The SiC hardness of SiC will also reduce susceptibility to fretting failures that currently affect the reliability of nuclear fuel in operation. The details of SiC CMC fiber orientation, interface layer design and matrix properties effect the final properties of the engineered cladding.

The programmatic needs to rapidly develop test rodlets that can be scaled up to commercial and economic production are driving the development program within the Advanced

LWR Nuclear Fuel Pathway. The basic development of CMC structures is a mature technology from the aerospace and turbine engine development. Turbine engine and rocket applications demonstrate that SiC CMC can operate at high temperatures in an oxidizing environment. There are also applications with severe thermal cycling and thermal shock. The program is integrating multiple individual technologies to develop advanced nuclear fuel cladding.

The integration is being guided by the need to demonstrate commercial potential to support the LWRS mission. This has lead to utilizing radiation stable high purity commercial fiber technology, HI-NICALON^{TM} ceramic fibers. The matrix created after the interface layer is applied to the woven fibers with a polymer infiltration process (PIP). The polymer is infiltrated into the woven fibers then reacted at high temperatures leaving. Multiple infiltration and high temperature processing can be required to achieve high SiC density. This process appears to scale up to full length LWR nuclear fuel cladding more economically than using chemical vapor deposition (CVD) to create the SiC matrix.

Accelerating the development of the cladding has also lead to utilizing a zirconium inner liner in the rodlets. The liner allows the matrix to remain fully sealed even if the ceramic matrix cracks through. The choice of zirconium also creates a known chemical environment against the UO2 fuel. There is a technical risk that fission product or heavy metal reaction would create undesirable reactions with a SiC inner layer. The inner zirconium layer also allows a reliable welded end cap join to be created. The current design does allow zirconium to face the coolant. Later designs are very likely to include a SiC structure to fully enclose the inner layer. Design optimization will minimize the thickness of the layer helping minimize issues with differential thermal and irradiation expansion. A schematic and image of the test rodlet is shown in Figure 2.

The extension of the SiC CMC technology to LWR nuclear fuel is aided by applications in fusion reactors and TRISO fuel forms. These applications have established the properties of SiC under extended neutron irradiation and at high temperatures. Recent work is demonstrating the effect of a very hot steam environment on SiC CMC materials. These efforts are being performed at multiple research and industrial sites.

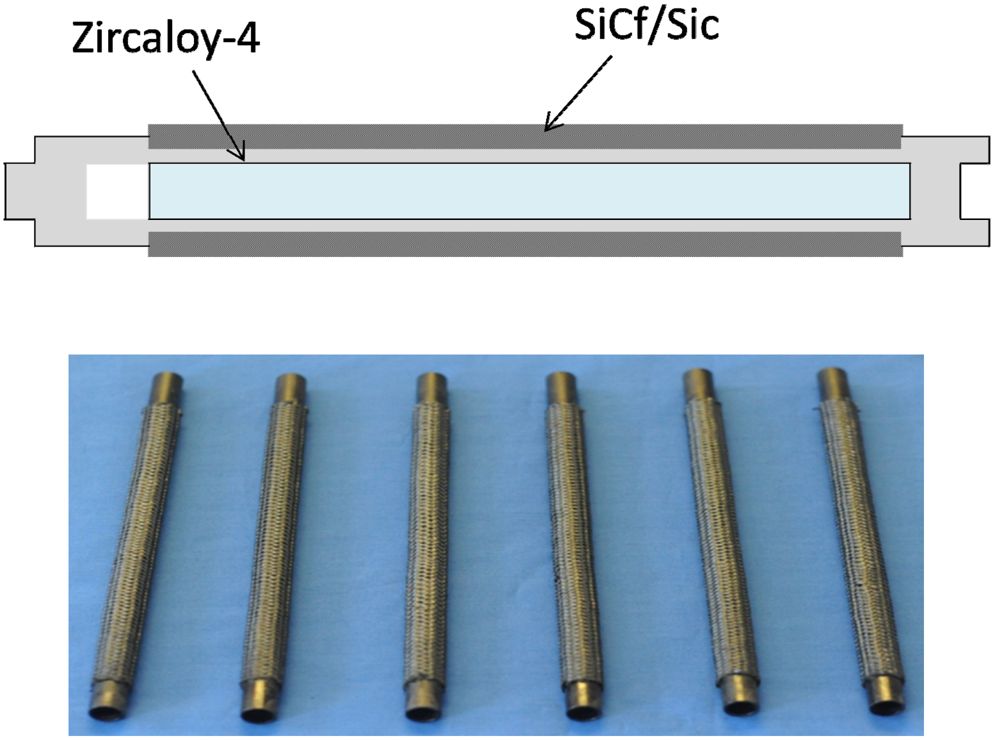

Figure 2: SiC CMC fuel rod cladding with metal liner

Pathway Development

The LWRS Advanced LWR Nuclear Fuel Pathway is broken into three research tasks. The first research task includes the irradiation and testing of different design samples. Development of predictive computational models is the second task and development of all the required supporting technologies is the final task.

Irradiation and Testing Campaign

Multiple reactors are being used to optimize the testing of prototype samples. The different reactors will provide demonstrations of specific performance in the increasingly sophisticated test rods.

Currently the Oak Ridge National Laboratory (ORNL) High Flux Isotope Reactor (HFIR) is being used to irradiate commercially developed SiC CMC rodlets. These rods are multilayer rodlets that are fabricated entirely from SiC and SiC CMC. The rods are fueled with uranium dioxide and uranium nitride fuel. The rods are the most advanced rods being irradiated. A pair of test rods will be removed before the end of 2012 for visual and non-destructive examination. The test plan will progress to destructive testing. The HFIR irradiations will include the LWRS SiC CMC hybrid rods as testing space is created in the test assembly. A drawing of the HFIR irradiation rodlets and insert capsule are shown in Figure 3.

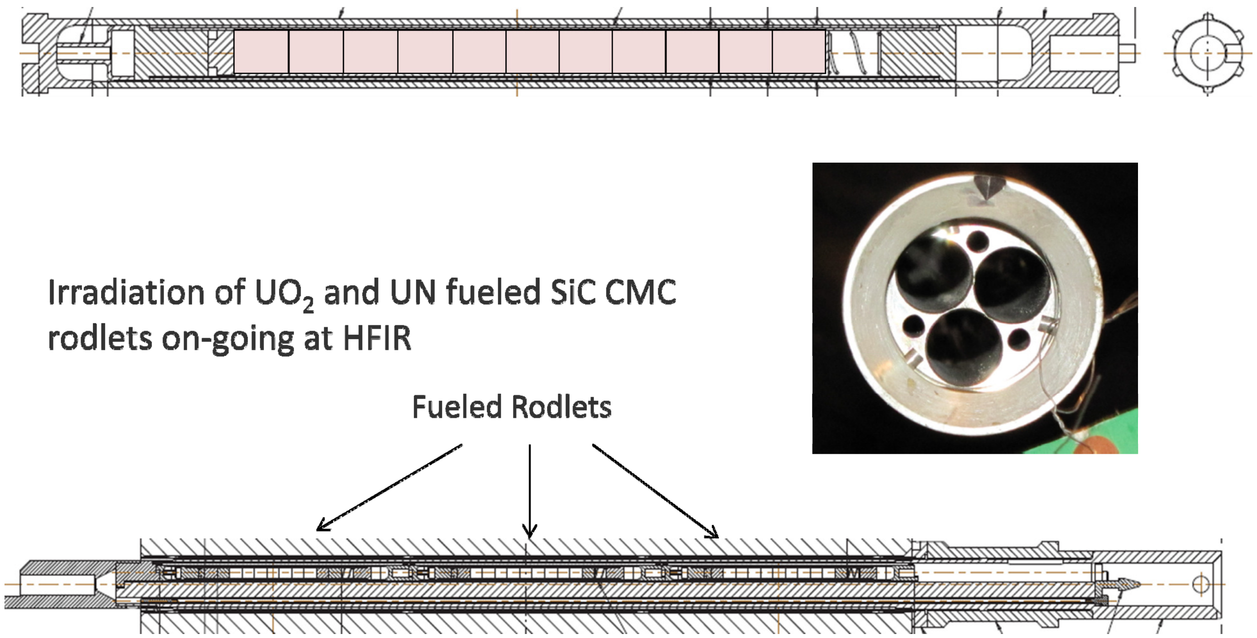

Figure 3: Test sample and Reactor Capsule used at HFIR

The Idaho National Laboratory (INL) Advanced Test Reactor will begin irradiation in 2012. The test rods are the LWRS SiC hybrid rods. Initial testing will be done on unfueled rods. This simplifies the safety analysis of the fuel rod allowing earlier insertion into the reactor. Collecting the data required for the safety analysis has driven several small scale pre-irradiation tests on corrosion, strength and thermal properties. The ATR will be utilized to create high fluence in a short time in the test samples. Access to the advanced post irradiation systems will also help move the program quickly. Operating transient performance will also be tested. ATR will be (re)installing a pressurized loop that can simulate PWR reactor conditions. This is a potential source of more realistic testing than is currently available. A pre and post irradiation measurement campaign has been drafted to optimize the measurements performed. The measurement plan is maximizing the understanding of the SiC CMC materials and evaluating the test rod performance. ATR test rodlet and irradiation capsule are shown in Figure 4.

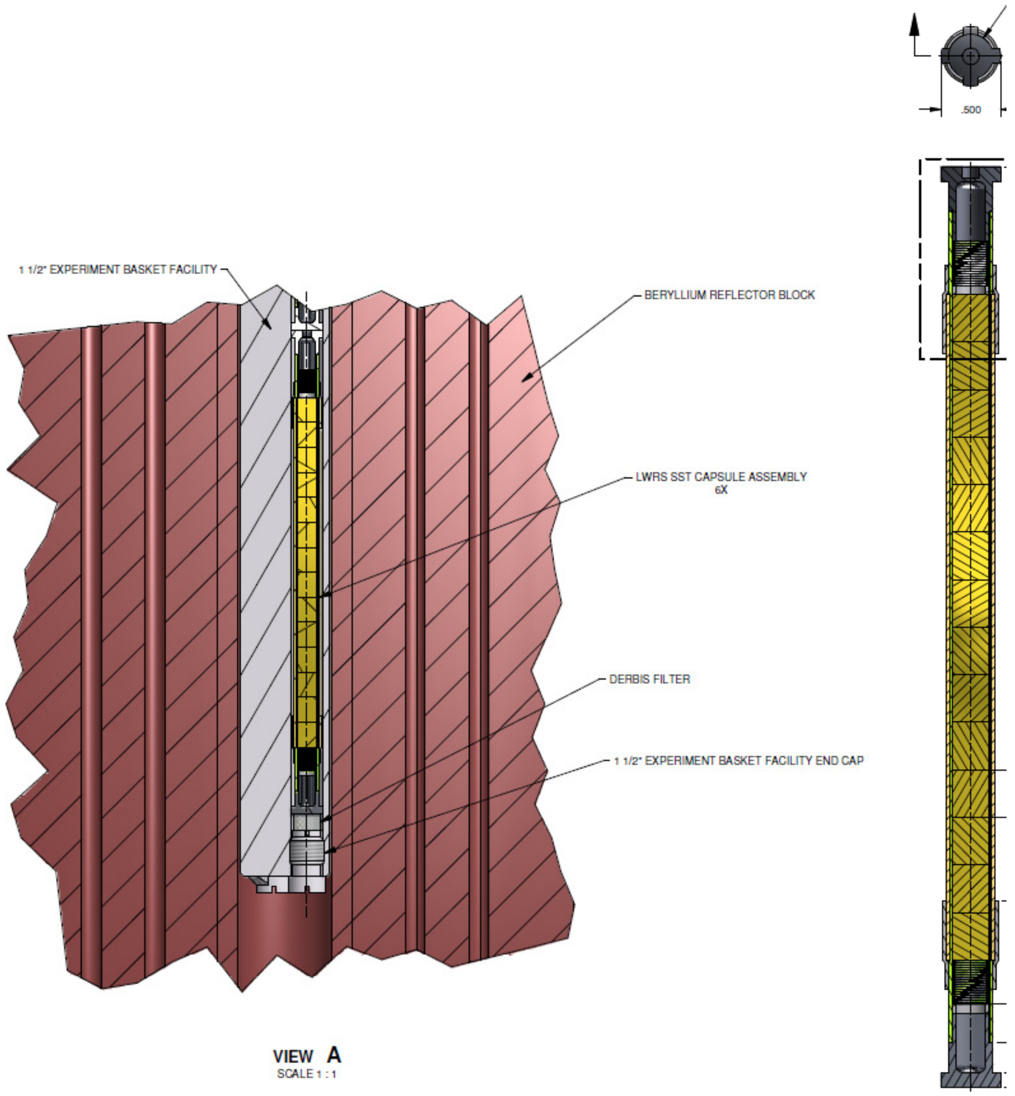

Figure 4: ATR Nuclear Fuel Rodlet

A contract is being established to utilize the Halden Reactor Project (HRP) Research Reactor to demonstrate basic fuel performance and widen the know fuel performance to include severe transients and accident events. Initial analysis will start in 2012 with unfueled testing following. Further contracts will be created to perform further fueled testing. The sophisticated measurement techniques available at HRP will allow unique understanding of the in-core performance. The ability of HRP to simulate severe reactor transients will be used to benchmark computational models.

Developing Advanced Modeling

This is a research task will create detailed mechanical benchmarked nuclear fuel models and allow optimization of complex multilayer silicon carbide (SiC) cladding. Computing the behavior of SiC CMC structures in a reactor is not provide the detailed understanding of nuclear fuel performance required for design activities. This activity will need to develop increasingly complex models to allow prediction of cladding properties before irradiation and eventually include thermal and repeated stress models. This knowledge can be used to improve cladding performance and simplify the experimental campaign.

The product from the research is analytic tools and understanding to allow design optimization. The combination of SiC CMC and metallic components will be optimized using the developed tools. The balance of stress, thermal properties and volumes can all be done with sophisticated models of the SiC CMC hybrid fuel rod cladding.

This task will also involve modeling and benchmarking of specific aspects of SiC CMC LWR cladding, fuel, and coolant behavior. Important examples include pellet cladding interaction between ridged fuel pellets and very stiff ceramic based cladding, fission gas release (FGR), and fuel swelling. Improved understanding of the fuel system behavior can be used in fuel design, licensing, and performance prediction.

An improved fundamental understanding of phenomena that impose limitations on fuel performance will allow fuel designers, fabricators, and code developers to optimize the performance of current fuels and the designs of advanced fuel concepts. A life-cycle concept will be applied so that optimization will be applied to fabrication, in-reactor use, and performance as spent fuel in storage. Fundamental mechanistic models will provide a foundation for supporting the LWRS R&D Program strategic objectives in developing advanced fuels.

The mesoscale, microstructural model development and refinement must include methods to introduce irradiation defects, temperature, and a power profile. Fundamental materials parameters and phenomena will be included to allow validation of fission gas transport, diffusion coefficients, grain restructuring, bubble size, and distribution. The FGR mechanism and a more accurate radial power profile will be part of the integrated model.

The computational portion of the pathway is heavily dependent on the larger computational effort being supported by DOE-NE. There are major fuel model development activities performed by Nuclear Energy Advanced Modeling and Simulation (**NEAMS**) and Consortium for Advanced Simulation of LWRs (CASL) programs.

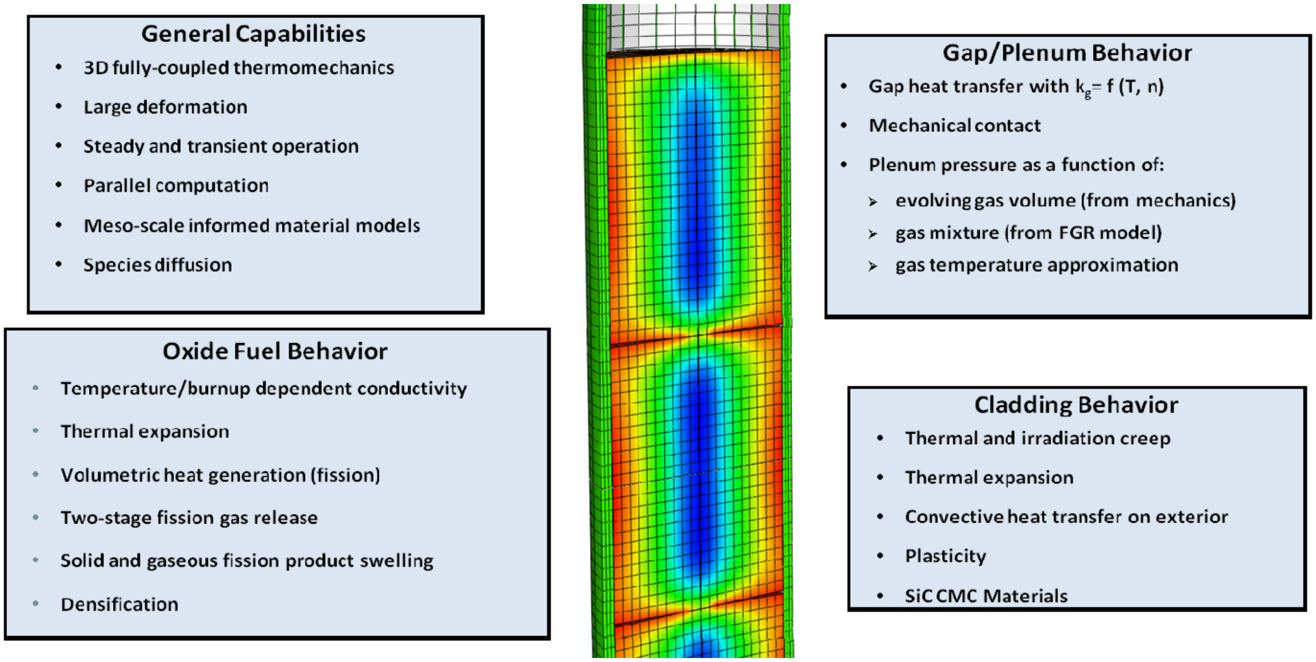

Figure 5: MOOSE/BISON planned fuel modeling capabilities

The NEAMS program is supporting the development of the MOOSE, BISON and MARMOT codes used to model conventional nuclear fuel. These codes are computational advanced and work across multiple scales and can include multiple physics models. These codes allows for multiscale simulation of fuel and nuclear materials and how they interact. Capabilities and results from the advanced fuel modeling codes are shown in Figure 5 and Figure 6.

Figure 6: Demonstration of MOOSE/BISON/MARMOT code system results

The CASL program will be providing a virtual reactor to evaluate the performance of advanced nuclear fuel. The ability to evaluate full core fuel performance will greatly assist the the development of advanced nuclear fuel. NEAMS Hub is providing detailed nuclear fuel models and the integration of sophisticated micro, meso, and engineering scale computer models. The inclusion of multiple scales and advanced physics will provide more accurate results that can be applied over a greater range than is currently utilized in fuel performance codes.

Utilizing the improved fundamental understanding of phenomena that impose limitations on fuel performance will allow fuel designers, fabricators, plant chemists, and code developers to optimize the performance of current fuels and the designs of advanced fuel concepts. A life-cycle concept will be applied so that optimization will be applied to fabrication, in-reactor use, and performance as spent fuel in storage. Fundamental mechanistic models will provide a foundation for supporting the LWRS R&D Program strategic objectives in developing advanced fuels.

The Advanced LWR Nuclear Fuel Pathway supports the development of SiC CMC specific modeling features. These features include the stiff SiC CMC material, advanced cracking models and the interaction of ceramic fuel pellets and ceramic based cladding.

Advanced Tools

This task will create the specific fuel technology required to support the development of SiC CMC nuclear fuel cladding. These advanced tools are the tasks required to fill critical development needs to succeed in developing advanced nuclear fuel. In addition, the advanced tools developed will be used to minimize the time required to realize the gains made through this R&D effort by decreasing the amount of time needed for materials development and fuel qualification.

The engineering design and safety analysis tool will create engineering-scale models and prototype samples that integrate the multiple aspects of fuels performance from the mechanistic understanding of fuel behavior. This task will allow prediction of fuel behavior during steady-state, transient operation and severe event transients. The goal is to create models that include multiple fuel performance effects and starts to predict increasingly complex. The focus on engineering-scale results will require that other tasks are integrated and provide practical results that can be used for design analysis.

The design development activities and technology integration occur within this task. Design activities feed all the other development tasks. The development of concepts requires unique testing and evaluation before and after irradiation. The ability of fabricate, design and license new technologies must also be included in the evaluation of the program.

The design work has centered on the development of robust SiC CMC utilizing a metallic liner. Detailed working designs have been created within the fabrication technology available. This has created a design that addresses research reactor safety concerns and can be extended to commercial applications. Material properties under thermal stress between SiC CMC and metallic liner have evaluated to provide needed performance. Material optimization is on-going to maximize the radiation stability of the finished SiC CMC structure.

INL has a balance of experience in SiC CMC materials that has greatly improved the design of the test rodlets. INL has a significant experience base in the development of very advanced armor systems that utilize CMC and advanced metals. The insights from those programs have allowed rapid access to the commercial CMC industry and understanding of the complex engineered material that make up the test rodlet.

A critical need for the program is measurement techniques that allow study on the scale that the performance of the SiC CMC system is created. Small scale measurements of SiC fibers and interface layers are required. Larger quality scale survey measurements are also needed. Advanced measurement techniques will also be developed to evaluate advanced measurement methods to derive SiC CMC cladding properties. The unique properties of SiC, very hard, high speed of sound, low cross sections, require unique measurement techniques. The measurements will be able to evaluate SiC CMC properties at very small scales. The scale of the SiC fibers, interface layer and damage features are very small. The critical properties of SiC CMC structures are determined by these small scale features.

This research will introduce needed measurement technologies to allow the needed information on SiC at very small scales to be generated. Laser-based resonant ultrasound spectroscopy will be used to measure dimensional properties, cracks, voids, elastic properties, microstructure-mediated plastic properties, and internal friction. Micro-raman spectroscopy will be used to measure localized mapping of microstructure, chemical composition, phase transformation, stress distribution, and temperature; thermal conductivity measurements. The laser-based thermometry using nanosecond pulse pyrometry and laser ultrasonics will be developed; and measurements of pulse laser heating temperatures, including temperatures of phase transitions. An example test using SiC CMC samples of Eddy current testing is shown in Figure 7. The results of X-ray tomography of a SiC CMC hybrid cladding sample is shown in Figure 8.

Figure 7: Eddy Current Survey Measurement of SiC Sample

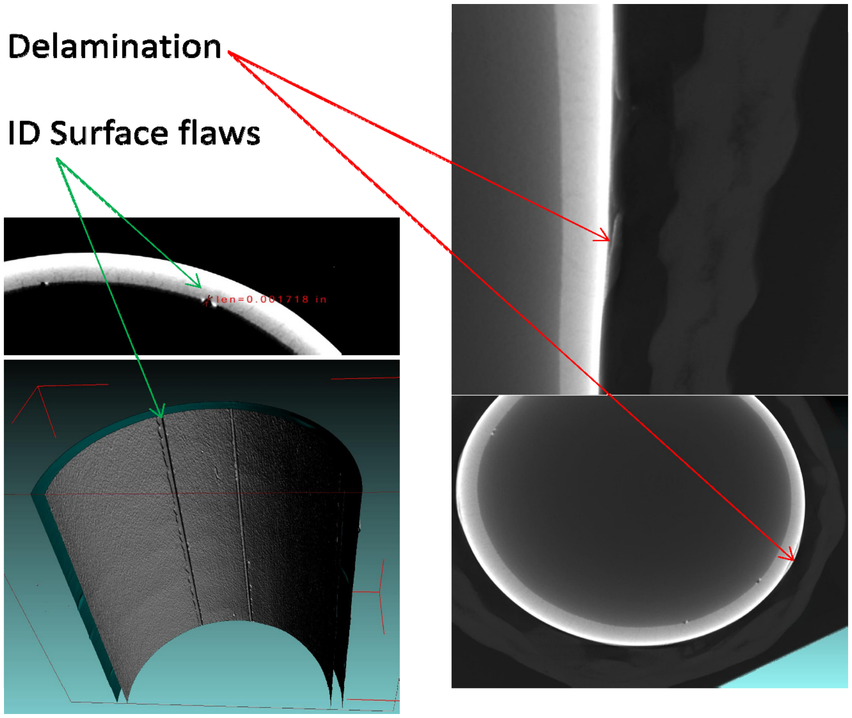

Figure 8: Tomography image of SiC CMC hybrid cladding

Sophisticated visual examinations are also being developed to evaluate larger scale structures for quality evaluations. Unique analysis of digital images will be used to scan the rodlets as a whole for fabrication consistency. This technique will also be applicable to the irradiated samples allowing new evaluations to be performed.

Future Plans

The primary focus of the program is currently starting irradiation to create performance results, improving the accuracy of the fuel performance modeling, developing a radiation resistant SiC matrix and developing a design that more accident resistant. There will be three reactors irradiating samples with planned post irradiation examination in 2012 greatly expanding the test results being generated. The fuel modeling is advancing with specific SiC properties included for 2012. The next iteration of the fuel performance modeling will include specific SiC CMC mechanical performance properties. Radiation resistant fibers currently exist commercially. Initial tests have shown that a polymer that contains fewer trace elements that will contaminate or create voids in the final SiC matrix would be desirable. An effort has started to develop a nuclear grade SiC processing polymer.

Development of an end cap or cover for the metal liner is becoming a very desirable result. The covering of the metal liner and eliminating the potential for water and metal reactions will improve the cladding performance. It will also eliminate complexity in performance estimates during a reactor safety evaluation.

---

[i] http://www.ioffe.ru/SVA/NSM/Semicond/SiC/, “Properties of Silicon Carbide (SiC)". Ioffe Institute. Retrieved 2011-08-20.