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Development of a Modeling Approach to Describe Thermal Conductivity of Prototypic SiC/SiC Tubes for LWR Fuel Cladding

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SUMMARY

SiC/SiC composites are a promising material intended to be used as a fuel cladding for light water reactors. This material has exceptional steam oxidation resistance, outstanding high temperature strength, and small neutron cross sections. Thermal conductivity is one of the most important physical properties for modeling and assessment for the performance of SiC/SiC composite-cladded fuels. However, there is a significant lack of knowledge for thermal conductivity of tube materials because of the variability of thermal conductivity with the architecture and manufacturing quality of SiC-SiC composites, the challenge associated with measurement with small diameter tubes, and the complex nature of the effects of neutron irradiation on transport properties of composite materials. Development of a constitutive model of the composite thermal conductivity incorporating the effects of temperature, neutron irradiation, and potential matrix damages is essential toward a comprehensive performance modeling for the SiC-SiC composite-cladded fuels. As such, the current effort focuses on establishing a high-fidelity thermal conductivity model for SiC-SiC composite cladding.

This report summarizes the modeling methodology, experimental methodologies, and computational software tools that will be constitute the thermal conductivity modeling for the SiC/SiC composite cladding tubes. The proposed experiments are directed to establish a baseline of thermal conductivity values and to validate experimental simulations. To obtain the thermal conductivity of these materials laser flash analysis will be implemented to obtain the thermal diffusivity of flat specimens and sections of round shape specimens. Thermal diffusivity experiments will be complemented by finite element method simulations and a branch of this methodology known as image-based finite element method. The FEM simulations will help to identify the possible limitations of experimental measurements in round shape specimens and verify the results of experiments in flat shape samples. The results of the image-based finite element method will help quantify the possible exact effects of porosity and other micro- to meso-structural features in SiC/SiC composites. The findings of the aforementioned activities will be also applied to SiC/SiC neutron-irradiated tube samples to capture the effects of irradiation on constitutive thermal conductivity and possible matrix cracking damages produced due to the complex stress state evolutions as the result of irradiation – heat flux synergism.

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ACRONYMS

CAD	computer aided design
CVI	chemical vapor infiltrated
EDS	energy-dispersive -ray spectroscopy
FEM	finite element method
IBFEM	imaged-based finite element method
LWR	light water reactor
SEM	scanning electron microscopy
SiC	silicon carbide
UMAT	user material subroutine
XCT	x-ray computed tomography

DEVELOPMENT OF A MODELING APPROACH TO DESCRIBE THERMAL CONDUCTIVITY OF PROTOTYPIC SIC/SIC LWR FUEL CLADDING

1. INTRODUCTION

Silicon carbide (SiC)–based composites, specifically the continuous SiC fiber-reinforced SiC-matrix (SiC-SiC) composites, are being considered as a prospective material for fuel cladding for light water reactors (LWRs) [1]. SiC intrinsically exhibits very slow oxidation kinetics in high temperature steam, high strength retained to ultra-high temperatures, small neutron cross sections, and outstanding stability under neutron irradiation to high doses. In addition, composite materials like SiC/SiC composites are damage tolerant, highly predictable for mechanical properties, can be tailored for desired anisotropic properties and be designed to have high performance under the most demanding environments. One of the critical performance-limiting factors for SiC-SiC composite cladding is the decrease in thermal conductivity under neutron irradiation [2-5].

Establishing high fidelity modeling capability for the thermal conductivity of SiC/SiC composite cladding is critically important to qualify these materials because their thermal conductivity significantly affects temperature distribution of fuel and cladding [6]. Thermal conductivity is obtained by calculating the product of thermal diffusivity, density, and specific heat capacity. However, there is a significant lack of data for the thermal diffusivity of tube materials because it is challenging to measure due to the specimen geometry; this is in contrast to plate materials, which have been systematically investigated [3]. We have conducted thermal diffusivity experiments of curved SiC/SiC composite coupons machined from a tube using a laser flash apparatus. The analysis found that thermal conductivities of both nonirradiated and irradiated tubes were similar to those of flat specimens in the literature [7]. However, the accuracy of the measurement can be improved.

Modeling thermal conductivity of a SiC/SiC composite tube is a great complementary effort to support the experimental measurement. A combination of modeling and experimentation was successful to evaluate thermal transport of components and material with complex microstructures. More specifically, modeling transient heat transport of the material based on experimentally obtained material microstructure is a promising approaches to advance thermal conductivity modeling and improve analysis of the thermal diffusivity experiment [8].

The overarching goal of this task is predicting the thermal conductivity of SiC/SiC composite cladding based on its microstructure using a high-fidelity model. In this report we describe the steps that will be followed to model the thermal conductivity of SiC/SiC composite tubes. Figure 1 shows a diagram that presents the different activities that will be undertaken to measure, model, validate, and determine the effective thermal conductivity of SiC/SiC composites. As can be seen from Figure 1, the strategy to model the thermal conductivity of these composites is divided into experimental methodology and modeling using the finite element method (FEM) and a branch of the FEM known as imaged-based finite element method (IBFEM). IBFEM uses images or a 3D reconstruction obtained with an imaging technique that accounts for the complex microstructure of SiC/SiC composite materials. A natural progression of this work will be to model the thermal conductivity of neutron irradiation on SiC/SiC composites tubes. Novel thermal conductivity data, dimensional inspections, simulations, and x-ray computed tomography (XCT) scans will help to understand the influence of irradiation in this material at different dose and temperature conditions. This research will support the manufacturing of SiC/SiC composites for fuel cladding in two different thrusts. First, improving the understanding of neutron irradiation effects on thermal conductivity of SiC/SiC composites will help determine if this material is suitable as a fuel cladding component. Second, this project will investigate the effect of irradiation in the

SiC/SiC microstructure. Neutron irradiation may produce cracks that may reduce the thermal conductivity and sealing capacity of SiC/SiC composites [1]. Together, these efforts will help to determine the possible stress concentration produced by low thermal conductivity areas, neutron irradiation effects, and porosity in SiC/SiC composites.

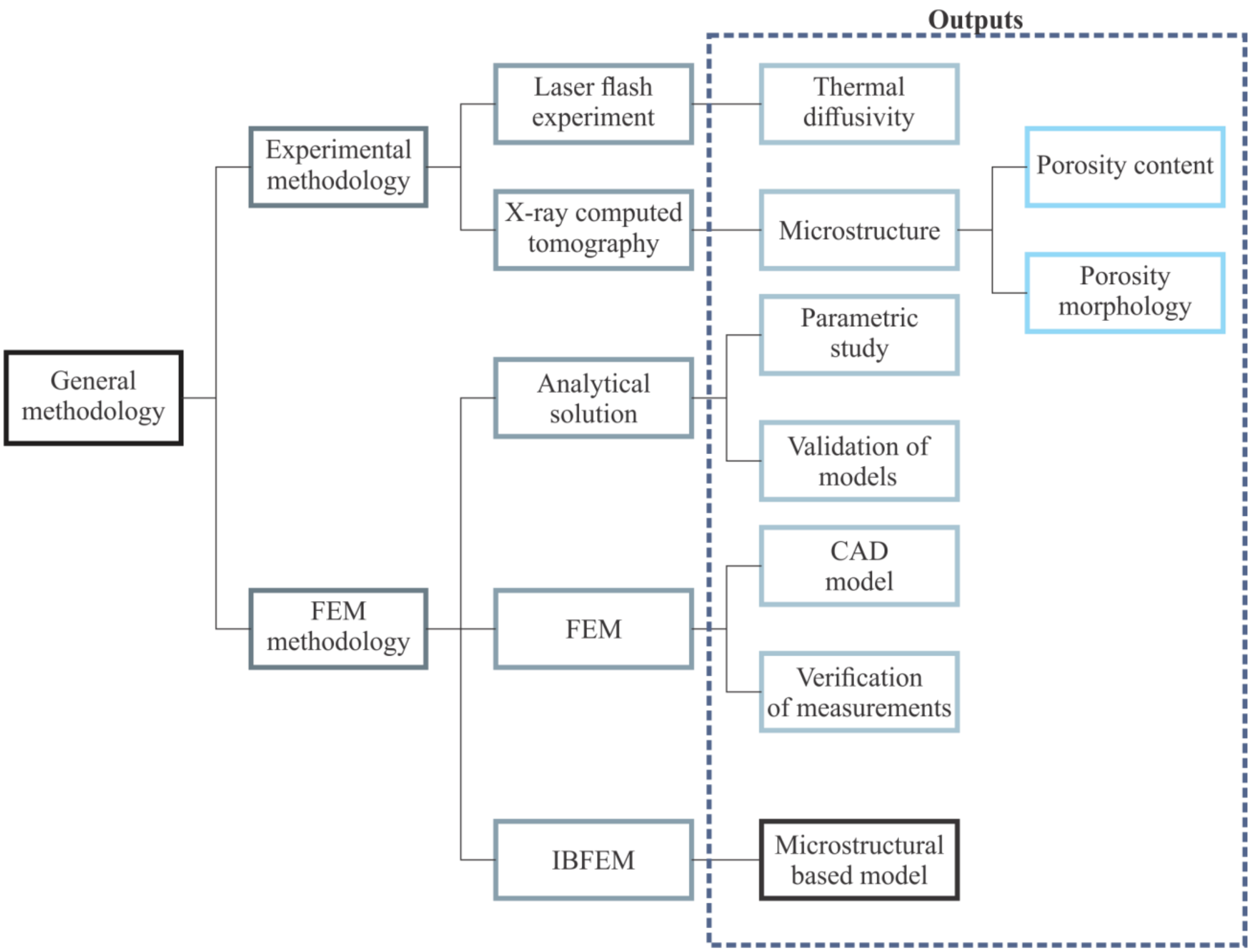


Figure 1. General methodology to determine the effective thermal conductivity of SiC/SiC composites.

Figure 1 shows a scanning electron microscopy (SEM) micrograph of the microstructure of a polished SiC/SiC composite sample. From the micrograph (Figure 2) we can see the wide range of porous geometries (from 1 mm to >100 µm) and different phases (matrix SiC, SiC fibers, and carbon interphase) found in this material. The proposed framework (Figure 1) will help determine the influence of the geometry and microstructure on the effective thermal conductivity of SiC/SiC composites.

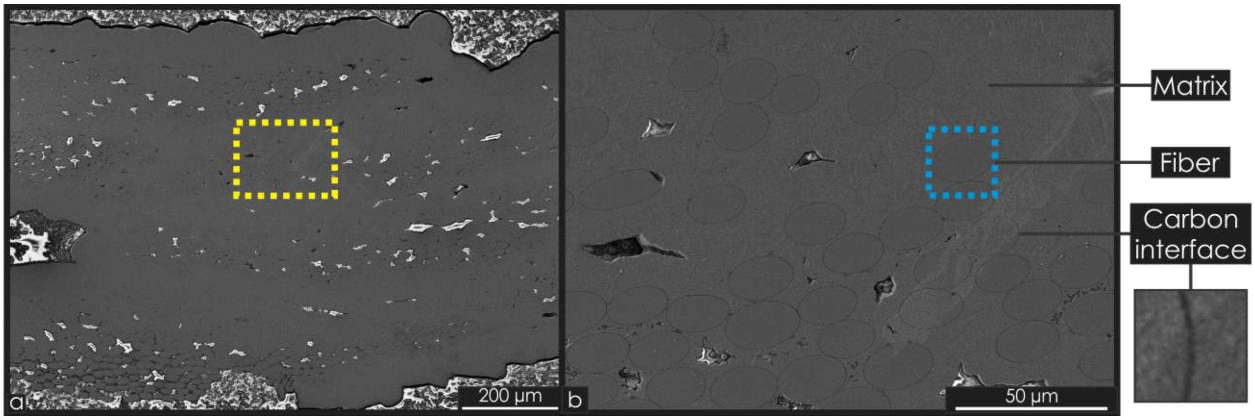


Figure 2. SEM micrographs of the SiC/SiC composite. (a) Low resolution micrograph of the surface of a polished SiC/SiC composite. (b) High resolution of the yellow highlighted area of micrograph (a). Both images were collected with an acceleration voltage of 2 kV.

2. EXPERIMENTAL METHODOLOGY

This section presents the overall experimental methodology planned for this study. Two basic approaches are currently being considered to characterize the microstructure and thermal conductivity of the SiC/SiC composites. The following subsections describe the instruments and experimental methodologies that will be used to study the SiC/SiC composites.

2.1 Thermal Diffusivity Measurements

A light flash apparatus (LFA467 HyperFlash Netzsch) will be used to conduct thermal diffusivity measurements of SiC/SiC samples. This instrument's pulse capabilities have a range of 20–1,200 µs, which allows measurements on samples that are thin or are highly thermally conductive, and it is capable of measuring the thermal diffusivity at temperatures close to 1250°C. This instrument uses laser flash analysis to obtain the thermal diffusivity at the top of a surface by applying a heat pulse generated from a laser while at the same time measuring the temperature difference. By applying this pulse and measuring the temperature, it is possible to obtain the thermal diffusivity of the material. In addition, the LFA467 system has the unique capability of monitoring the temperature of a selected area on the specimen using a ZoomOptics system, which limits the thermal diffusivity signal to the specimen (no noise from sample fixture) and consequently provides high quality data for comparing the experiment and model.

As part of these experiments, two types of samples will be characterized: flat disks and sections of tubes (Figure 3). These measurements will serve as a validation of the FEM simulations and as a baseline for future measurements. In this case, the SiC/SiC composites thermal diffusivity measurements can be classified into two types by the type of geometry and objective:

- Flat samples: This type of geometry is the most common used in thermal diffusivity measurements because of its repeatability. With flat specimens, the main purpose of using this technique is to have reference measurements as well as validation for the measurement.
- Round section of a tube: This is the actual geometry of interest; however, the laser flash method is optimized for flat specimens. A direct comparison between FEM simulations will help identify the possible limitations of this technique in round shape specimens of SiC/SiC composites.

A total of 16 samples will be characterized using the laser flash method. The details of these samples are described in Table 1. The materials include chemical vapor infiltrated (CVI) SiC matrix reinforced with Hi-Nicalon Type S and Tyranno SA3 SiC fibers coated with single-layer carbon interphase.

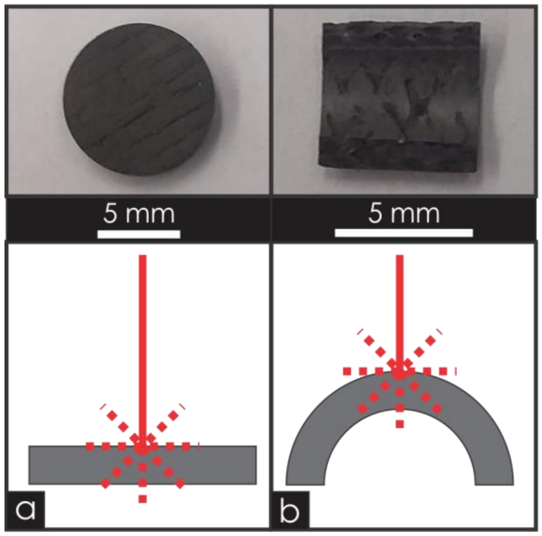


Figure 3. SiC/SiC composites methodology and geometry of specimens for thermal conductivity measurements. (a) Flat specimen, (b) round section of tube.

Table 1. Selected samples for thermal conductivity measurements

Material type	Geometry	No. samples
CVI SiC/SiC composite (Hi Nicalon Type S fiber)	Flat specimen	5
CVI SiC/SiC composite (Tyranno SA3 fiber)	Flat specimen	5
CVI SiC/SiC composite (Hi Nicalon Type S fiber)	Round section of tube	4
CVI SiC/SiC composite (Tyranno SA3 fiber)	Round section of tube	2

2.2 X-ray Computed Tomography Characterization

Some of the same samples used for thermal diffusivity measurements (Table 2) will be scanned with Zeiss Xradia Versa 520 XCT equipment, which can achieve a pixel resolution of 0.9 μm in a 2 × 2 × 2 mm volume. Scans of 5–10 hours are necessary to achieve the required resolution for these experiments. Examples of typical XCT images of SiC/SiC composites are provided in Figure 4. These images can be processed to study the morphology of pores, solid material, and other features of these materials. Microstructural features found to be influencing the thermal properties of SiC/SiC composites can be analyzed by segmenting XCT reconstructions, and this step will be done using Avizo Fire software. Important features such as pore morphology, connectivity, distribution, orientation, and content can be easily obtained from XCT segmented data. Even though XCT can provide valuable information about the pore content, it cannot resolve the individual fibers of the composite. However, preliminary segmentation analysis has shown that the fiber bundles contain distinctive pore size distributions, elongated shapes, and preferential orientation. These characteristics will help to differentiate the different types of pores found in the matrix and fiber-rich areas of the composites.

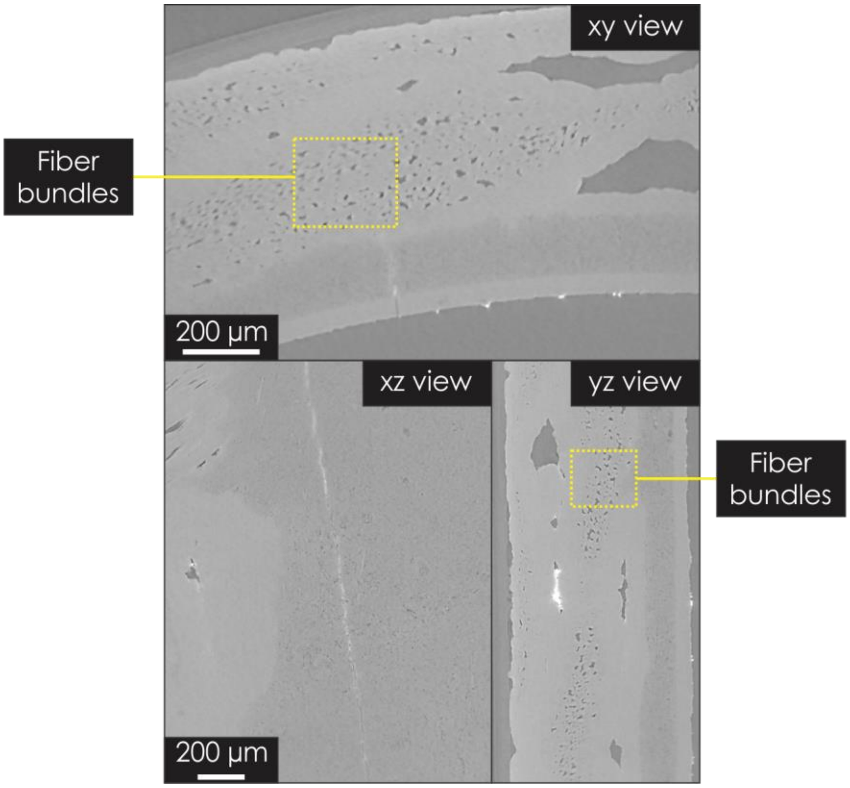


Figure 4. Examples of views of typical XCT reconstructed tomographic images of SiC/SiC composite tube.

Table 2. Selected sample for XCT scans

Sample ID	Geometry	No. samples
CVI SiC/SiC composite (Hi Nicalon Type S fiber)	Flat specimen	2
CVI SiC/SiC composite (Tyranno SA3 fiber)	Flat specimen	2
CVI SiC/SiC composite (Hi Nicalon Type S fiber)	Round section of tube	2
CVI SiC/SiC composite (Tyranno SA3 fiber)	Round section of tube	2

2.3 Serial Polishing of SiC/SiC Composite Tubes

An automatic grinder and polisher will be used to prepare flat surfaces and samples for SEM imagining and energy-dispersive x-ray spectroscopy (EDS) elemental maps of the surface of a SiC/SiC composite sample. By alternating the polishing and mapping of the surface of the sample with SEM and EDS some microscopic features that cannot be resolved through XCT (i.e., individual fibers or carbon interface) will be captured (Figure 5). These experiments are intended to detect all the phases of the material and use this information in FEM simulations.

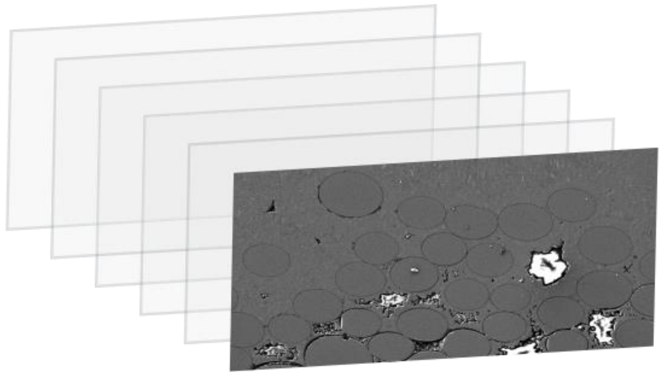


Figure 5. Serial polishing combined with SEM and EDS will help to identify each phase of SiC/SiC composites.

3. FINITE ELEMENT METHDOLOGY

3.1 Analytical Solution

An analytical solution of the flow of heat in a rectangular parallelepiped that was found in literature for parametric studies and to verify and validate the results from FEM models. The analytical solution was taken from Carlaw and Jaeger [9] and can be directly compared with 3D heat transfer problems for a cuboid geometry calculated by the FEM models.

3.2 Finite Element Method Model

In the past, 2D FEM models have been proposed by Youngblood [10] to analyze the thermal conductivity of SiC/SiC composites. These results will be used as a starting point for the determination of thermal conductivity of SiC/SiC composites. After this, a computer aided design (CAD) model will be created in the FEM software Abaqus to verify the physics and boundary conditions that simulate the conditions of the thermal diffusivity experiments proposed in Section 2.1. The thermal diffusivity (or conductivity) of SiC composite constituents is available in literature [11, 12]. Similar boundary conditions and modeling assumptions adopted by Evans [8, 13] are going to be implemented in this research. In general, these boundary conditions consist of applying a thermal load at the top of the geometry that corresponds to the laser power. At the same time the temperature values at the opposite side of the sample (bottom), will be consistent with the temperatures of the experiment. Because of the discretization of the geometry into finite elements, the thermal load must be distributed into the nodes of the elements instead of the surface like what happens in the experiment. Other important considerations result from the differences between the experiment and simulations. Because the measurements are conducted in a plane surface (two-dimensional measurement) and the model is performed in a three-dimensional space, the thermal load will be distributed in a way that compensates these differences.

Two sets of models will test two different geometries: (a) CAD model of a flat disk that represent the thermal properties of SiC, (b) CAD model of a round section of a SiC/SiC composite. Determining the influence of the curvature of the sample will be possible by modeling a flat and a round section of an idealized SiC layer.

3.3 Image-Based Finite Element Method Model

IBFEM models will be generated from the XCT data collected from both flat and round sections of SiC/SiC composites. To generate the FEM meshes the XCT data will be segmented with the Avizo Fire software. After this, the resultant segmented volume will be meshed with the software Avizo Wind. An example of these processes (i.e., segmentation and FEM mesh of a SiC/SiC composite) is shown in Figure 6. Next, the same boundary conditions as proposed in Section 3.2 will be applied to the IBFEM model. The final step is to compare the FEM and IBFEM to see the influence of porosity and curvature in the heat transfer of SiC/SiC composites.

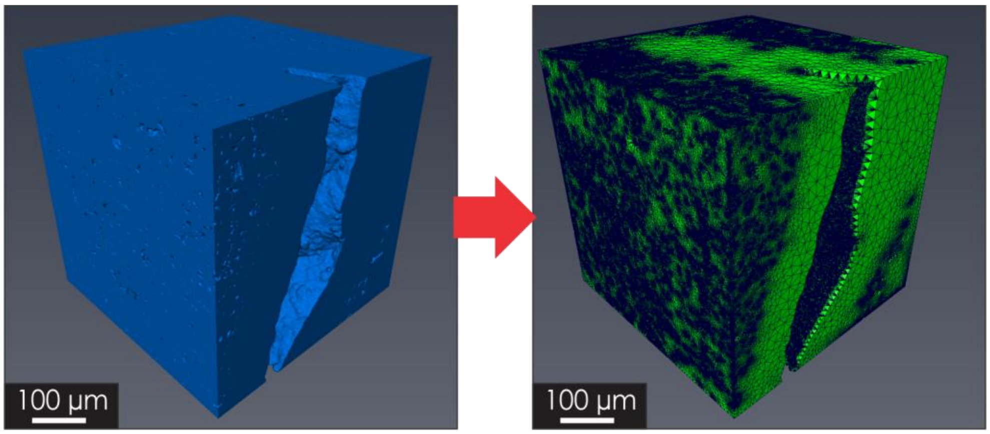
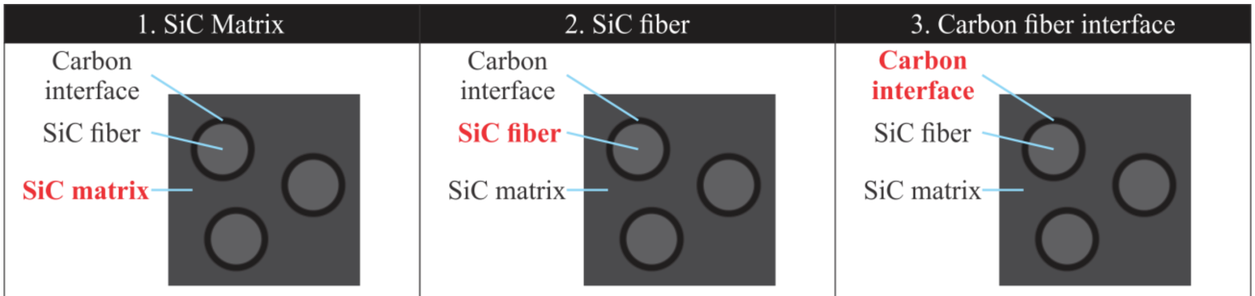


Figure 6. Workflow and examples of IBFEM of a segmented XCT SiC/SiC composite data set and corresponding mesh.

In addition to the XCT data, a combination of SEM micrographs, EDS maps, and serial polishing of the sample will be implemented to obtain more detail microstructure models of a sample as described in Section 2.3. Even though the field of view of SEM and EDS technique is smaller than XCT, these techniques combined are capable of identifying each phase of the material. IBFEM models will be created from the SEM and EDS images in a fashion similar to XCT data. These types of models will help us study the impact on the different thermal conductivity properties of the SiC matrix, SiC fibers, and carbon interface (Figure 2). However, some of the thermal conductivity properties of SiC fibers and carbon interface are not well known. To tackle this issue, a FEM parametric study will be pursued to investigate the influence of uncertainties in the thermal conductivity values of SiC/SiC phases. Overall three cases will be included into the FEM models and be classified as follows (Figure 7):

1. SiC matrix study
2. SiC fiber study
3. Carbon interface study



Phase of interest	Variations of thermal conductivity values for parametric study (%)	Phase of interest	Variations of thermal conductivity values for parametric study (%)	Phase of interest	Variations of thermal conductivity values for parametric study (%)
SiC matrix	-20, -10, +10, +20	SiC fiber	-20, -10, +10, +20	Carbon interface	-20, -10, +10, +20

Figure 7. Proposed FEM parametric models for thermal conductivity values

In all these scenarios or FEM models the thermal conductivity value that correspond to the phase of interests will be changed from a baseline value by ±20% and ±10%. This will show which material phase thermal conductivity value i.e. SiC matrix, SiC fiber or carbon phase have the most significant effect on the effective thermal conductivity of bulk SiC/SiC composites. This type of parametric study using the FEM simulations has been implemented for nuclear graphite stress analysis [14]. Refinement of constituent thermal conductivities will be conducted through comparison of modeling and experiment of laser flash diffusivity tests of SiC/SiC composites with controlled microstructures (e.g. different fiber volume fraction but same carbon interface thickness). Such materials will be produced using CVI processing furnace at ORNL.

3.4 Model for As-Received and Neutron-Irradiated SiC/SiC Tubes

The models and findings of the previous sections will be combined to study the thermal conductivity of SiC/SiC tubes under the operation conditions of a nuclear reactor. Comparing the CAD models to the IBFEM models will help us understand the impact of pores and stress distributions in these types of components. Moreover, XCT data sets of as-received and neutron-irradiated samples will be used to further understand the impact of irradiation on the thermal conductivity of SiC/SiC tubes. Neutron irradiation and high temperatures reduce the thermal conductivity of SiC/SiC composites {Katoh, 2014 #12}. As a consequence, complex stress concentrations can be expected by these reduction on thermal conductivity values. Due to this reason it is necessary to account the changes produced on the thermal properties of SiC/SiC tubes by neutron irradiation. As such, a user material subroutine (UMAT) will be created in the software ABAQUS to take into account the thermal conductivity changes as a function of neutron fluence and temperature. A robust model that includes the microstructure and changes of thermal properties of SiC/SiC tubes will help to determine if the microscopic features of SiC/SiC composites and degradation of mechanical properties will lead to different temperature distribution of the fuel and cladding.

4. SUMMARY

This report provides an overview of the different steps that will be taken to model the thermal conductivity of SiC/SiC composite tube. Each experimental procedure and numerical calculations are summarized below:

- **Thermal conductivity measurements**: A total of 16 SiC/SiC composite samples will be measured using laser flash analysis. Of the 16, 10 correspond to flat specimens and 6 correspond to round cross sections of SiC/SiC composites.

- **XCT**: Scans will be performed in flat and round cross sections of SiC/SiC composites to characterize the microstructure of different SiC/SiC composites.
- **Validate**: An analytical solution was found in literature to validate the computational calculations of thermal analysis.
- **FEM:** Several CAD-based FEM models will be created and used as a baseline for more complex simulations as well as for analyzing the influence the curvature of the sample on the heat transfer of the sample.
- **IBFEM:** Data obtained from XCT scans will be processed to obtained faithful geometric models of the microstructure of SiC/SiC composites. These models will help analyze the possible hotspots created by pores and features of SiC/SiC composites. Another significant aspect of the thermal conductivity of SiC/SiC composites will be investigated by combining serial polishing, SEM micrographs and EDS elemental mapping. By combining this techniques and procedure it will be possible to assign individual material properties to each phase as the SiC matrix, SiC fibers and carbon interface will have different thermal properties.
- **Models of SiC/SiC tubes:** The modeling techniques and experiments will serve as the foundation for the analysis of SiC/SiC tubes. This will enable the production of detailed FEM simulations that predict the performance of SiC/SiC composite tubes under the neutron irradiation and thermal loads found in a nuclear reactor. This model will gather the outcomes of previous activities and expand upon them by considering the microstructural features and adding the relationships between the thermal conductivity, neutron dose and temperature gradients. To capture these relationships a UMAT subroutine will be programmed in the software Abaqus.

Taken together, these activities will provide important insights into the influence of microstructure of SiC/SiC composites on their corresponding thermal properties. Furthermore, the analysis of XCT experiments will provide a better understanding of the porosity shape, size distribution, and shape of these composites. In addition to this, the XCT data will help to down-select the most promising SiC/SiC architectures and improve the thermal conductivity of this materials. On the other hand, the modeling aspect of this project will help identify the types of features that reduce the thermal conductivity of these materials. These findings will help to determine which fiber and matrix architecture will help to improve the bulk thermal conductivity of SiC/SiC composites. All the modeling and experiments proposed in this document will support the development and design of SiC/SiC composites used for fuel cladding for LWRs. Finally, the modeling capabilities develop in this report will be extended to full 3D thermal transfer models of as-received and neutron-irradiated SiC/SiC tubes. Understating and developing a modeling tool for the thermal conductivity in SiC/SiC tubes is of critical importance to evaluate these materials as the thermal property changes and their microstructure will affect the temperature of fuel and cladding materials.

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