3.3 EMTA THERMAL CONDUCTIVITY PREDICTIONS FOR UNIRRADIATED AND IRRADIATED SiC/SiC COMPOSITES

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OBJECTIVE

The objective of this work is to achieve a predictive engineering tool to assess and tailor the thermophysical properties of unirradiated and irradiated SiC/SiC composites. Towards this objective, first, PNNL's EMTA (Eshelby-Mori-Tanaka Approach) software was successfully applied to predict the thermal conductivity of unirradiated 2D SiC/SiC composites [1]. Next, we have extended the EMTA model reported in [1] to predict the thermal conductivity of these composites subjected to neutron irradiation at elevated temperatures and irradiation doses leading to defect saturation [2]. As EMTA thermal conductivity predictions compared well with the experimental results [1-2], in the future, a unified EMTA for SiC/SiC composites will be developed that addresses both thermal and mechanical properties.

SUMMARY

This study first applied an Eshelby-Mori-Tanaka approach implemented in PNNL's EMTA software to predict thermal conductivities of unirradiated 2D SiC/SiC composites. The EMTA homogenization method is structured in three steps to capture the effects of the constituent materials including the pyrolytic carbon (PyC) fiber coating on the thermal conductivities of as-formed composites [1]. EMTA also applies an iterative reverse engineering procedure to identify an unknown constituent material thermal conductivity. EMTA reverse engineering was shown to be efficient and accurate to compute the thermal conductivities of the SiC matrix and of the fiber coating as functions of temperature, and consequently, to achieve better agreement between the predicted and experimental thermal conductivities for the composites. Next, a new method that builds on EMTA to estimate the thermal conductivity of SiC/SiC composites subjected to neutron irradiation at elevated temperature up to a dose of 10 dpa was developed [2]. This method considers the irradiation-induced conductivity degradation behavior of chemical vapor deposited (CVD) SiC and defines an equivalent CVD-SiC material with an equivalent pore content to estimate the thermal conductivity of a SiC/isothermal-chemical-vapor-infiltration (ICVI)-SiC composite. It was shown that the proposed method effectively mimics the irradiation effect that induces the reduction of composite thermal conductivity [2].

PROGRESS AND STATUS

Introduction

Due to their excellent high-temperature fracture, creep, corrosion, and thermal shock resistance in addition to their high thermo-chemical stability, low-induced radioactivity, and low-radiation-induced afterheat, SiC/SiC composites are being developed as structural materials for first wall and breeder blanket applications in advanced fusion power plants. In such applications these materials will be subjected to a high heat flux that imposes extremely stringent requirements on the minimum thermal conductivity needed. Recently, an EMTA method [1, 3-6] was applied to compute the thermal conductivity of unirradiated SiC/SiC composites with excellent results. EMTA has been shown to provide accurate predictions of thermal conductivity for typical SiC/SiC systems based on knowledge of the constituent thermal conductivities and the

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composite microstructural features (i.e., fiber coating, fiber volume fraction, fiber orientation, etc.). Although the transverse thermal conductivities of unirradiated SiC/SiC are in an acceptable range (~ 20 – 30 W/m K), the corresponding values of irradiated SiC/SiC for a large irradiation temperature range may be too low and thus present an important issue for the use of SiC/SiC for fusion power reactors since it appears that much higher thermal conductivity values are required [7]. The following step of our work was therefore to develop a computational model to predict and tailor the thermal conductivities of SiC/SiC composites subjected to neutron irradiation. To this end, we extended the previous EMTA model reported in [1] to predict the thermal conductivity of these composites subjected to neutron irradiation at elevated temperatures and irradiation doses leading to defect saturation. The extended approach makes use of a reasonable surrogate material by replacing irradiated SiC with an equivalent porous SiC that exhibits the same thermal conductivity decrement as does the irradiated SiC using experimental data. Given that a complete microstructural evolution description of radiation damage in SiC is lacking, the approach taken here was a reasonable first step in creating a more comprehensive and fundamental thermal conductivity model for SiC.

Computational Procedure

The EMTA computational procedure to predict the thermal conductivity of unirradiated SiC/SiC composites was described in [1] and summarized in our previous report [8]. This section gives a summary of the extended EMTA method to estimate the thermal conductivity for irradiated SiC/SiC composites [2].

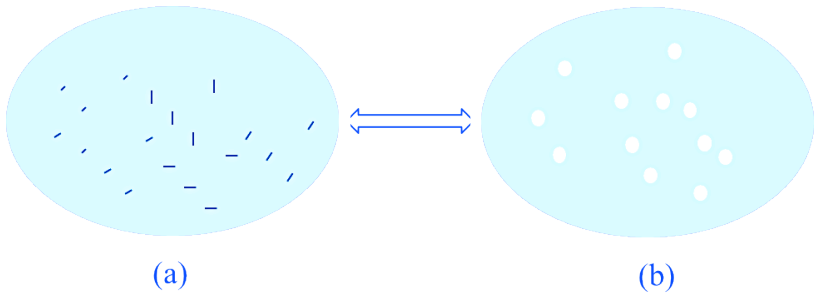

Figure 1. The equivalence between the CVD-SIC with a concentration of irradiation-induced defects (a) and the CVD-SiC containing a pore volume fraction (b) producing the same overall thermal conductivity.

Figure 1 schematically illustrates the equivalence between the irradiated CVD-SiC with a concentration of saturated irradiation-induced defects (Figure 1(a)) and the unirradiated CVD-SiC containing a fictitious spherical pore volume fraction (Figure 1(b)) that leads to the same thermal conductivity as the irradiated CVD-SiC material. The problem consists of determining the equivalent pore volume fraction for the irradiated equivalent material. To this end, the measured thermal conductivity data as a function of temperature and irradiation temperature for the unirradiated and irradiated CVD-SiC materials, respectively, are used in the EMTA reverse engineering to solve for this parameter. Next, the computation continues by assigning the same pore volume fraction representing the radiation damage to the ICVI-SiC matrix of an ICVI SiC/SiC composite subject to the same irradiation conditions as the CVD-SiC reference material. Such an assignment explicitly assumes that subject to the same irradiation conditions,

the CVD-SiC and ICVI SiC/SiC composite would contain the same concentration of defects at saturation; and that the defects that concentrate in the ICVI-SiC matrix and at the fiber/matrix interface are captured by the equivalent pore volume fraction affecting the thermal conductivity of the ICVI-SiC matrix. It also assumes that the fibers are unaffected by neutron irradiation, and this is a reasonable assumption for an irradiation dose up to about 10 dpa [9].

RESULTS

The first typical EMTA application examines the 2D SiC/SiC composite studied in [7]. DuPont processed this material from plain weave Hi-Nicalon fabric using an IVCI processed matrix. Two versions of this composite were examined in [7] including one with a 0.11-micron-thick and one with a 1.044-micron-thick PyC fiber coating layer, termed *thin* and *thick*, respectively. Constituent data for EMTA were taken in [7], however, the thermal conductivity of the PyC coating $k_c(T)$ as a function of temperature is not known, and therefore must be determined numerically by EMTA reverse engineering using measured conductivity data for the thick coating composite version. The as-identified $k_c(T)$ was then used by EMTA to refine the prediction of thermal conductivity for the thin fiber-coating version, and the transverse thermal conductivity results predicted for both thin and thick versions are presented in Figure 2 [1]. Accounting for the temperature-dependent $k_c(T)$ gives very good agreement between the predicted transverse thermal conductivity and experimental results for the whole temperature range studied, and for both versions of the DuPont composite.

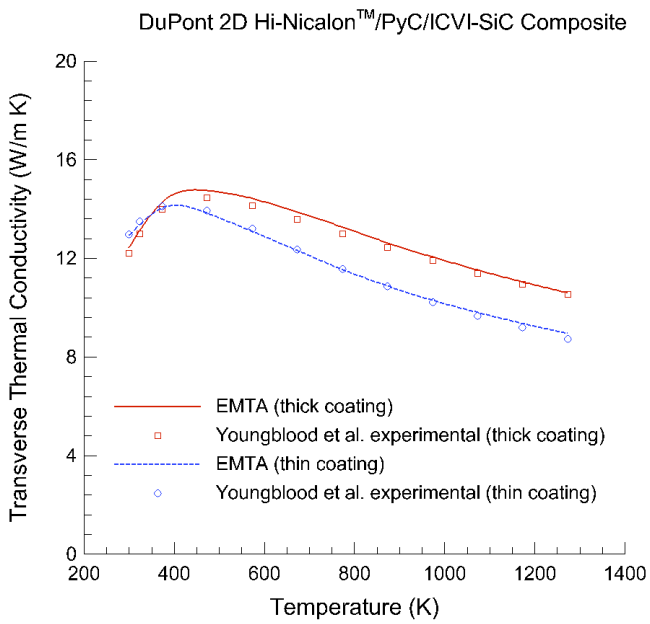

Figure 2. Transverse thermal conductivities predicted by EMTA for the DuPont 2D SiC/SiC composite with the thin and thick fiber coating layers accounting for the temperature-dependent fiber coating thermal conductivity, $k_c(T)$. The symbols denote Youngblood et al.'s experimental results from [7].

The second application illustrates the extended EMTA method to compute the thermal conductivity for the 2D Tyranno^{TM}-SA3/ICVI-SiC composite studied by Katoh et al. [10]. This composite was irradiated according to the following temperature/irradiation dose conditions: 713 K/0.8 dpa, 723 K/2 dpa, 773 K/3 dpa, 1123 K/3.5 dpa, and 1133 K/5.3 dpa [10]. These conditions are assumed to have led to saturation of irradiation-induced defects in the composite. Using the equivalent pore volume fraction calculated from the data determined for CVD-SiC at room temperature [10] and at each irradiation temperature [11], the ambient thermal conductivity of the ICVI-SiC matrix as a function of the irradiation temperature was computed by EMTA and reported in (Figure 3(a)), which shows the effect of irradiation-induced defects at the irradiation temperatures on the ambient thermal conductivity of the ICVI-SiC matrix. Finally, the thermal conductivity values predicted for the ICVI-SiC matrix were used by EMTA to calculate the ambient thermal conductivity of the irradiated Tyranno^{TM}-SA3 composite as a function of the irradiation temperature Figure 3(b).

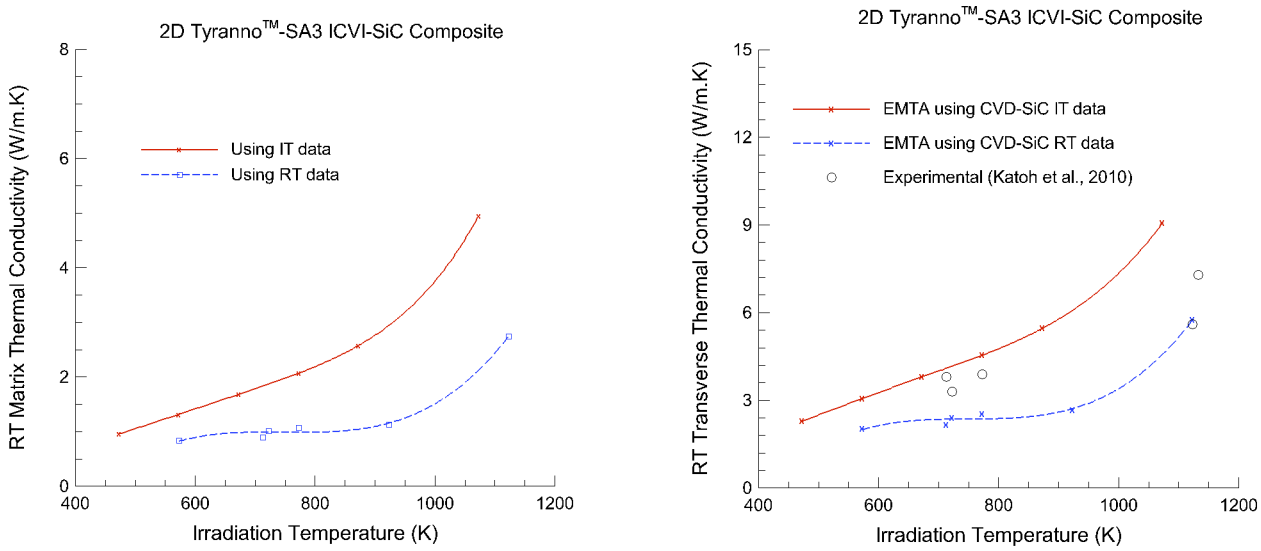

Figure 3. Ambient thermal conductivity vs. irradiation temperature predicted for the ICVI matrix (a) and 2D Tyranno^{TM}-SA3/ICVI-SIC composite (b) using the irradiation temperature (IT) and room temperature (RT) data for the CVD-SiC based on [10-11].

The prediction reported in Figure 3(b) globally agrees in trend and values with the measured data by Katoh et al., [10] presented on the same figure. The extended EMTA method appears to fairly estimate the thermal conductivity of irradiated SiC/SiC for a wide range of irradiation temperatures. As this method depends on the data taken from an irradiated CVD-SiC, its prediction also depends on data variance. The equivalent pore volume fraction approach can be completed by a more fundamental approach using appropriate defect densities and phonon scattering models. The difficulty here is that currently there is no detailed radiation damage evolution model for CVD-SiC that can be used to make these predictions [12]. The power of the EMTA approach, on the other hand, lies in the coupling of the matrix thermal conductivity analysis with the composite architecture and interfacial analysis appropriate to SiC/SiC composites.

Current and Future Work

As the transverse thermal conductivity of 2D woven SiC/SiC composites subjected to neutron irradiation is reduced to small values (several W/m K) although high thermal conductivity fibers (i.e., Tyranno^{TM} SA or Hi-Nicalon^{TM} Type-S fibers) are used, there have been interests in 3D SiC/SiC composites that were processed from 3D preforms directly woven with these fibers and an ICVI-SiC matrix. Currently, we are using EMTA to predict the thermal conductivity of unirradiated 3D woven SiC/SiC composites with different 3D fiber architectures characterized by the relative fiber content in the thickness direction. EMTA's predictions will be compared with experimental data found in literature to validate EMTA for 3D SiC/SiC composites. Next, our extended EMTA method to irradiated composites [2] will be used in a sensitivity analysis to determine the neutron irradiation effect on the thermal conductivity of various 3D woven SiC/SiC composites subjected to the same irradiation conditions leading to defect saturation. The 3D fiber configurations for these composites are varied by varying the relative fiber content in the thickness direction. Such an analysis will show the benefit of through-thickness reinforcement to improve the transverse thermal conductivity of SiC/SiC composites.

Toward achieving a predictive engineering tool to assess and tailor the thermophysical properties of SiC/SiC composites for fusion reactor applications, a unified EMTA for these composites will be developed that addresses both thermal and mechanical properties.

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