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Status Report on Advanced Cladding Modeling Work to Assess Cladding Performance Under Accident Conditions

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Introduction

Scoping simulations performed using a severe accident code can be applied to investigate the influence of advanced materials on beyond design basis accident progression and to identify any existing code limitations. In 2012 an effort was initiated to develop a numerical capability for understanding the potential safety advantages that might be realized during severe accident conditions by replacing Zircaloy components in light water reactors (LWRs) with silicon carbide (SiC) components. To this end, a version of the MELCOR code, under development at the Sandia National Laboratories in New Mexico (SNL/NM), was modified by replacing Zircaloy for SiC in the MELCOR reactor core oxidation and material properties routines. The modified version of MELCOR was benchmarked against available experimental data to ensure that present SiC oxidation theory in air and steam were correctly implemented in the code. Additional modifications have been implemented in the code in 2013 to improve the specificity in defining components fabricated from non-standard materials. An overview of these modifications and the status of their implementation are summarized below.

Description of MELCOR

The MELCOR code is the primary code used by the Nuclear Regulatory Commission (NRC) to model and analyze the progression of severe accidents. It has been developed and maintained for the NRC by the Sandia National Laboratories (SNL). MELCOR is a systems level severe accident code which includes the major phenomena of the system thermal hydraulics, fuel heat-up, cladding oxidation, radionuclide release and transport, fuel melting and relocation, etc.

MELCOR is presently designed for current LWR core material configurations. As such, the code contains material property definitions for "UO_2", "Zircaloy", "ZrO_2", "steel", "steel oxide" and "Inconel" for the fuel, cladding, spacer grids, support plates and channel boxes. The user can change the defined properties of one of these materials, such as Zircaloy, through user input. Internally, however, the code assigns material composition according to core component. As a result, a material property change to one of the core components, e.g. the cladding, will also change the material composition and properties of other core structures, e.g. channel boxes. This assumption can only be changed by modifying and recompiling the code to consider additional materials for any given core component.

Initial MELCOR Modifications: Material Substitution

Modifications to include SiC as an additional cladding material in MELCOR are presently ongoing at the Idaho National Laboratory (INL). INL has focused on applying the modified MELCOR code to the Three Mile Island Unit 2 (TMI-2) loss-of-coolant accident (LOCA) in a pressurized water reactor (PWR). The TMI-2 accident was caused by a small-break LOCA in a two-loop Babcock and Wilcox (B&W)

PWR. The MELCOR input model used for this accident analysis is that developed by SNL-NM and Innovative Technology Solutions Corporation, Albuquerque, NM.[1]

Initial modifications were made by directly substituting all Zircaloy components with silicon carbide. The properties and behaviors of monolithic chemical vapor deposited (CVD) SiC were used in place of Zircaloy in the MELCOR reactor core oxidation and material property routines. A summary of these properties is included at the end of this report. Much of what is considered for nuclear applications (cladding or other components) would use SiC-based ceramic matrix composites (CMC) rather than or in addition to monolithic CVD SiC, a brittle ceramic. However, the property data for SiC CMCs, particularly oxidation behavior, is extremely limited. Many of these data gaps are being addressed by current research in several Department of Energy programs (e.g., Fusion Materials Program, Fuel Cycle Research & Development Program, and the LWR Sustainability Program), but property data was insufficient for the oxidation of SiC CMCs at the time the MELCOR model was benchmarked. Hence, properties for monolithic CVD SiC were applied in these initial studies; updates to the oxidation behavior and failure temperatures can be made when additional data becomes available.

The initial version completed to analyze the TMI-2 event substitutes the properties of CVD SiC for Zircaloy in MELCOR's reactor core oxidation behavior (for oxidation in air and steam) and material property routines (including properties such as thermal conductivity, specific heat, density, emissivity, and heat of formation). Details of the SiC oxidation model and results of these initial analyses have been summarized in Merrill (2012)[2] and Merrill and Bragg-Sitton (2013).[3] Similar analyses could be performed for alternate cladding materials by overwriting the Zircaloy properties (defined as the SiC properties in this version) with the properties of the alternate material, recognizing that this would replace all structures and components in the core that are currently Zircaloy with the alternate material. The oxidation behavior (i.e. linear or parabolic oxidation rate) would determine which version of the code would be most applicable as a starting point for analysis of the potential safety benefit of the new candidate material.

Ongoing MELCOR Modifications: Definition of New Materials

Following the initial study that directly substituted SiC for Zircaloy in the PWR simulation, additional work has been conducted to define SiC as a unique material in addition to Zircaloy in the core physics package and the data materials package. In this version SiC is defined as a new core material using the known properties of CVD SiC and all the necessary links between the code's core physics and materials packages have been set. This version required modification to the MELCOR code (beyond simple user input) and recompilation. Hence, the modified version will now allow a user to assign both Zircaloy and SiC in a reactor core model for the cladding via user input. If necessary, the user can modify the material properties database through the input deck without requiring the code to be recompiled. Hence, this modified version could be made available for users less familiar with the MELCOR code structure.

Several modifications have been made to the core physics package in this second modified version. The SNL version of MELCOR allows for cladding to be Zircaloy and ZrO_2. The initial modification made by INL overwrote the kinetics for Zircaloy, such that Zircaloy and SiC could not be coexistent in

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[1] R. O. GAUNTT, K. ROSS, and K. WAGNER, "MELCOR 1.8.5 Simulation of TMI-2 Phase 2 With an Enhanced 2-Dimensional In-Vessel Natural Circulation Model," *10th International Conference on Nuclear Engineering*, Arlington, Virginia, USA, April 14–18, Vol. 3, pp. 487-494, American Society of Mechanical Engineers (2002).
[2] B.J. MERRILL and S.M. BRAGG-SITTON, "SiC Modifications to MELCOR for Severe Accident Analysis Applications," in proceedings of the *LWR Fuel Performance Meeting, Top Fuel 2013*, Charlotte, NC, September 15-19, 2013, paper 8546.
[3] B. J. MERRILL, "MELCOR SiC Modifications for Severe Accident Analysis Applications," Idaho National Laboratory Report, INL/LTD-12-27961, Rev. 0, December (2012).

the core. The second modification now adds SiC and SiO_2 to the list of possible cladding materials. SiC has also been identified in MELCOR's oxidation logic as a cladding material. If SiC is entered by the user as a cladding material, the new oxidation logic oxidizes SiC first, and then all other user defined cladding materials, such as Zircaloy, once the SiC has been completely oxidized. As modified, this code version allows the user to define multi-layer cladding materials. There has been significant interest in using SiC CMC as an overwrap on a Zircaloy cladding tube to reduce the oxidation rate and total oxidation extent of the underlying Zircaloy. Other researchers are investigating the application of coatings to the external surface of a Zircaloy cladding tube to reduce oxidation. The modifications made for SiC in this code version could easily be duplicated for another material, allowing the user to assess the safety potential of these design options as well. As discussed, this version allows cladding to be defined as Zircaloy or SiC while all other structures and components are individually defined as MELCOR default LWR materials: Zircaloy, stainless steel, or Inconel.

Some additional modifications are necessary to complete this second stage of MELCOR modifications. First, MELCOR defines a zero thickness for Zircaloy as a failure point. Definition of a purely SiC cladding (without a Zircaloy liner) would be translated by MELCOR as a failed core even if all of the SiC cladding is still present in its original configuration. Hence, a very thin layer of Zircaloy must still be defined in a core that contains both SiC and Zircaloy to allow the simulation to proceed. This limitation can be overcome given availability of additional time and funding. Second, SiC has been defined in the core physics package, allowing for its use as a cladding material. However, SiC has not yet been defined as a known material in MELCOR's debris bed model, which is necessary for material tracking and performing energy balance calculations. Initial steps have been taken to verify global conservation of energy, but this verification step has not yet been completed. Additional modifications need to be made to the print and plot routines within MELCOR to allow analysis of results from this second modified version.

Requirements for Definition of New Materials in MELCOR

To define additional materials in MELCOR one can choose to either overwrite properties for Zircaloy or to define a new material. The work has been done to determine how to implement new materials using either method, such that definition of additional materials will be more straightforward than it was for SiC. The preferred option may depend on whether or not the material is proposed for all structures and components that are currently Zircaloy, or only for the cladding material. Additionally, the oxidation behavior will define whether the material is closer in behavior to SiC (parabolic SiC oxidation kinetics limited by a linear SiO_2 volatilization process) or Zircaloy (parabolic oxidation kinetics limited by a stable oxide film). Key material properties and behaviors necessary for MELCOR simulation include:

- Properties of the base material (e.g. SiC) and its oxide (e.g. SiO_2), as a function of temperature and irradiation:
  - Melting temperature (T_{melt} [K]) of the base material, oxide and any eutectics that may form
  - Thermal conductivity (k [W/m-K])
  - Specific heat (c_p [J/kg-K])
  - Density (ρ [kg/m^3])
  - Emissivity (ε [W/m^2-K^4])
- Oxidation reactions, including oxidation rate, heat of reaction, reaction products, etc.
- Arrhenius relationship for parabolic oxidation rate behavior

Appendix A
Properties of Silicon Carbide

Data presented in Figures A-1, A-2 and in Tables A-1, A2 were applied in the presented MELCOR analysis.

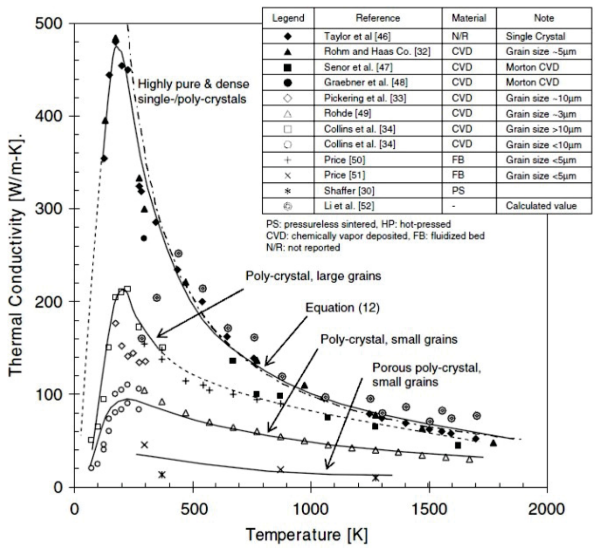

Figure A-1. Thermal conductivity of SiC (Snead et al. 2007).

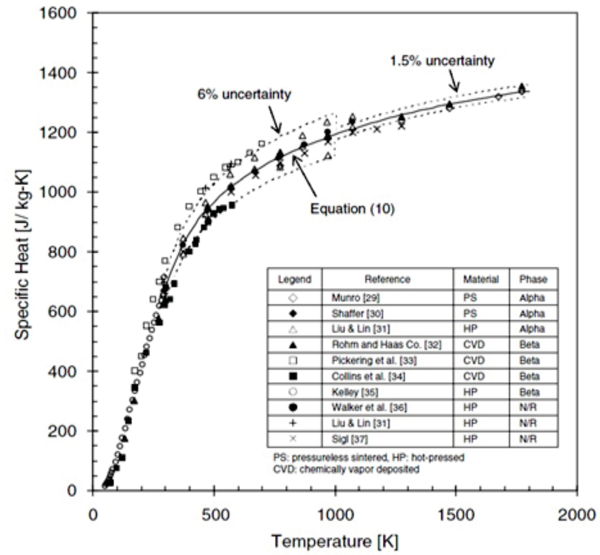

Figure A-2. Specific heat of SiC (Snead et al. 2007).

Table A-1. Chemical vapor deposition properties of SiC.

Thermal Conductivity		Specific Heat	
Temp (K)	**W/m-K**	**Temp (K)**	**J/kg-K**
273	389.6	273	594.08
350	296.3	400	864.19
400	256.4	500	966.65
500	202.0	600	1034.70
700	141.8	700	1085.66
900	109.3	900	1161.50
1300	74.91	1100	1218.26
1500	64.72	1300	1263.16
2000	48.31	1500	1298.92
2500	38.54	1800	1337.95
2818	34.14	2300	1367.89
5000	19.16	2500	1368.17
		2818	1355.17
		3300	1304.35
		5000	1262.76

Source: Snead et al. 2007

Density = 2900 kg/m^3 (Meyer 2012)

Emissivity = 0.75
(Touloukian and DeWitt 1972)

Evaporation temperature = 2900 K
(Byrappa and Ohachi 2003)

Complete disassociation
temperature = 3150 K
(Byrappa and Ohachi 2003)

Table A-2. Typical properties of silica (SiO_2).

Thermal Conductivity[a]		Specific Heat[b]		Heat of Formation[c,d]	
Temp (K)	W/m-K	Temp (K)	J/kg-K	Material	kJ/mol
273	1.33	273	682.32	SiC	−73.22
500	1.62	300	724.70	SiO_2	−910.86
700	1.92	500	952.14	CO	−110.525
900	2.48	700	1084.89	H_2O (g)	−241.818
1100	3.36	900	1165.82	$Si(OH)_4$	−1340.68
1300	4.82	1100	1215.56		
1400	6.20	1300	1245.28		
5000	6.20	1500	1261.61		

a. Source: Touloukian et al. 1971.
b. Source: Touloukian and Buyco 1970.
c. Source: Lide 1997.
d. Source: Plyasunov 2012.

Melt temperature = 1873 K (Wikipedia 2013)

Density = 2000 kg/m^3 (Fox 1998)

Table A-3: Typical mechanical and lifetime properties of chemical vapor infiltration SiC/SiC.[a]

Property	Unit	2D 0/90°	2D ±45°	Braiding, Bi-axial ±55°	Braiding, Tri-axial
In-plane tensile proportional limit stress	MPa	100–150[b]	100–150[b]	70–150[b]	≈100[b]
In-plane tensile strength	MPa	250–400[b]	≈200[b]	100–300[b]	≈150[b]
In-plane tensile modulus	GPa	200–300[b]	200–300[b]	200–300[b]	200–300[b] 300[b]
Poisson's ratio		0.2[b]	0.2[b]	0.2[b]	—
Trans-thickness tensile strength	MPa	≈20[c]	≈20[c]	≈20[e]	≈20[e]
In-plane flexural proportional limit stress	MPa	300–500[e]	—	—	—
In-plane flexural strength	MPa	500–800[e]	—	—	—
In-plane shear proportional limit stress	MPa	≈40[f]	≈40[f]	—	—
In-plane shear strength	MPa	≈100[f]	≈120[f]	—	—
Interlaminar shear strength	MPa	≈40[f]	≈4[0f]	—	—
Fracture toughness	$MPa\text{-}m^{1/2}$	20–30[c]	≈20[c]	20–30[b]	≈20[b]
Cyclic fatigue run-out stress (1000ºC in argon, tension-tension)	MPa	≈70[f]	—	—	—
Critical rupture stress (1200ºC in air, tension)	MPa	<50[f]	—	—	—
Mass density	g/cm^3	2.6–2.8[b]	2.6–2.8[b]	2.6–2.8[b]	2.6–2.8[b]
Thermal conductivity, in-pane	W/m-K	≈30 (RT)[c] ≈18 (800ºC)	≈30 (RT)[c] ≈18 (800ºC)	≈40 (RT)[b] ≈25 (800ºC)	≈40 (RT)[b] ≈25 (800ºC)
Thermal conductivity, transverse	W/m-K	≈15 (RT)[c] ≈10 (800ºC)	≈15 (RT)[c] ≈10 (800ºC)	≈35 (RT)[b] ≈20 (800ºC)	≈25 (RT)[b] ≈18 (800ºC)
Thermal expansion (0–1000ºC)	ppm/K	≈4.5[d]	≈4.5[d]	≈4.5[d]	≈4.5[d]

a. Source: Katoh, Wilson, and Forsberg 2007. Materials are Hi-Nicalon™ Type-S, 150 nm pyrocarbon interphase, chemical vapor infiltration SiC matrix composites unless otherwise noted. RT = room temperature.

b. Data obtained in U.S. fourth generation (Gen-IV) Next Generation Nuclear Plant Composite Research and Development Program and affiliated Small Business innovative Research programs.

c. Data obtained in Fusion Advanced Materials Program for materials in the same class.

d. Intrinsic properties for crystalline, stoichiometric SiC. Confirmative data exist.

e. Values guessed by the authors based on data for similar materials.

f. Data for CG-Nicalon™, pyrocarbon interphase, chemical vapor infiltration SiC matrix composites.

Table A-4: Summary of neutron irradiation effects on mechanical properties of advanced fiber CVI SiC/SiC.[a]

Reference	Material	Irradiation Condition	Observation[b]
Snead et al. 2000	Hi-Nicalon S / CVI	400ºC, ≈1 dpa	▪ Slight increase or no change in flexural strength
Hinoki et al. 2002	Hi-Nicalon S / CVI Tyranno-SA1 / CVI	300ºC, 6 dpa 800ºC, 7.7 dpa	▪ Slight increase or no change in flexural strength ▪ Slight increase or no change in flexural PLS ▪ Slight decrease in flexural modulus
Nozawa et al. 2004	Hi-Nicalon S / CVI	800ºC, 7.7 dpa	▪ Slight increase in UTS ▪ Slight decrease in tensile PLS ▪ No significant change in tensile modulus
Hegeman et al. 2005	SA-Tyrannohex Hi-Nicalon S / CVI	600/900ºC, ≈2 dpa	▪ Little change in flexural strength ▪ No or slight decrease in dynamic modulus
Nozawa et al. 2005	Hi-Nicalon S / CVI	800/1000ºC, 1 dpa	▪ Slight increase in UTS ▪ No or little change in tensile PLS ▪ Decrease in tensile modulus
Ozawa et al. 2007	Hi-Nicalon S / CVI Tyranno-SA3 / CVI	750ºC, 12 dpa	▪ No change in UTS ▪ Slight increase or no change in tensile PLS ▪ Slight decrease in tensile modulus
Newsome et al. 2007	Hi-Nicalon S / CVI	300/500/800ºC, < ≈3.4 dpa	▪ Slight decrease in flexural strength, flexural PLS, and flexural modulus

a. Source: Katoh, Wilson, and Forsberg 2007
b. PLS = proportional limit stress
UTS = ultimate tensile strength

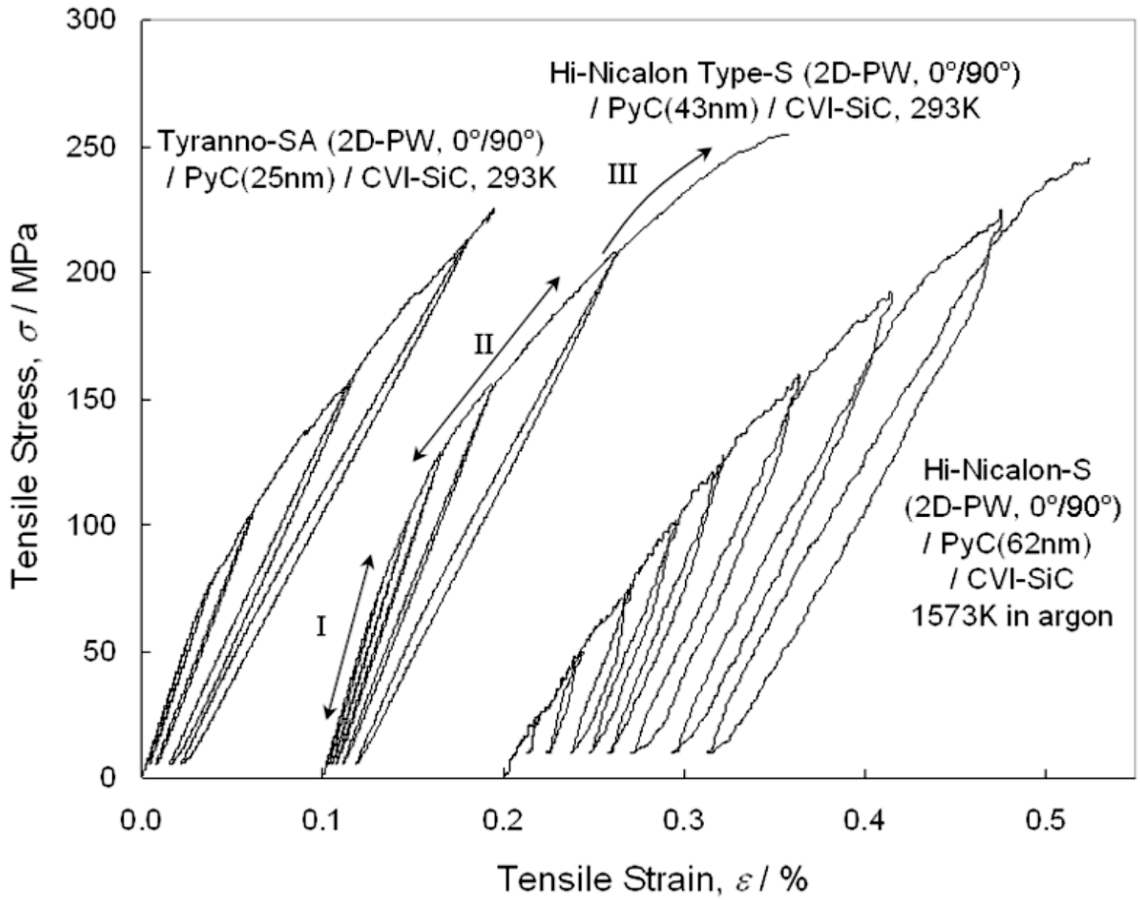

Figure A-3. Examples of tensile stress-strain behavior of chemical vapor infiltration SiC/SiC (testing procedure involves incremental unloading/reloading sequences to determine the extent of matrix damages and interfacial properties). The labels I – III indicate stages of tensile fracture of the composites in this class; namely I: elastic/reversible deformation, II: matrix micro-crack accumulation, and III: progressive fiber failure. The horizontal shifts are intentional for clarity. Source: Katoh, Wilson, and Forsberg (2007).

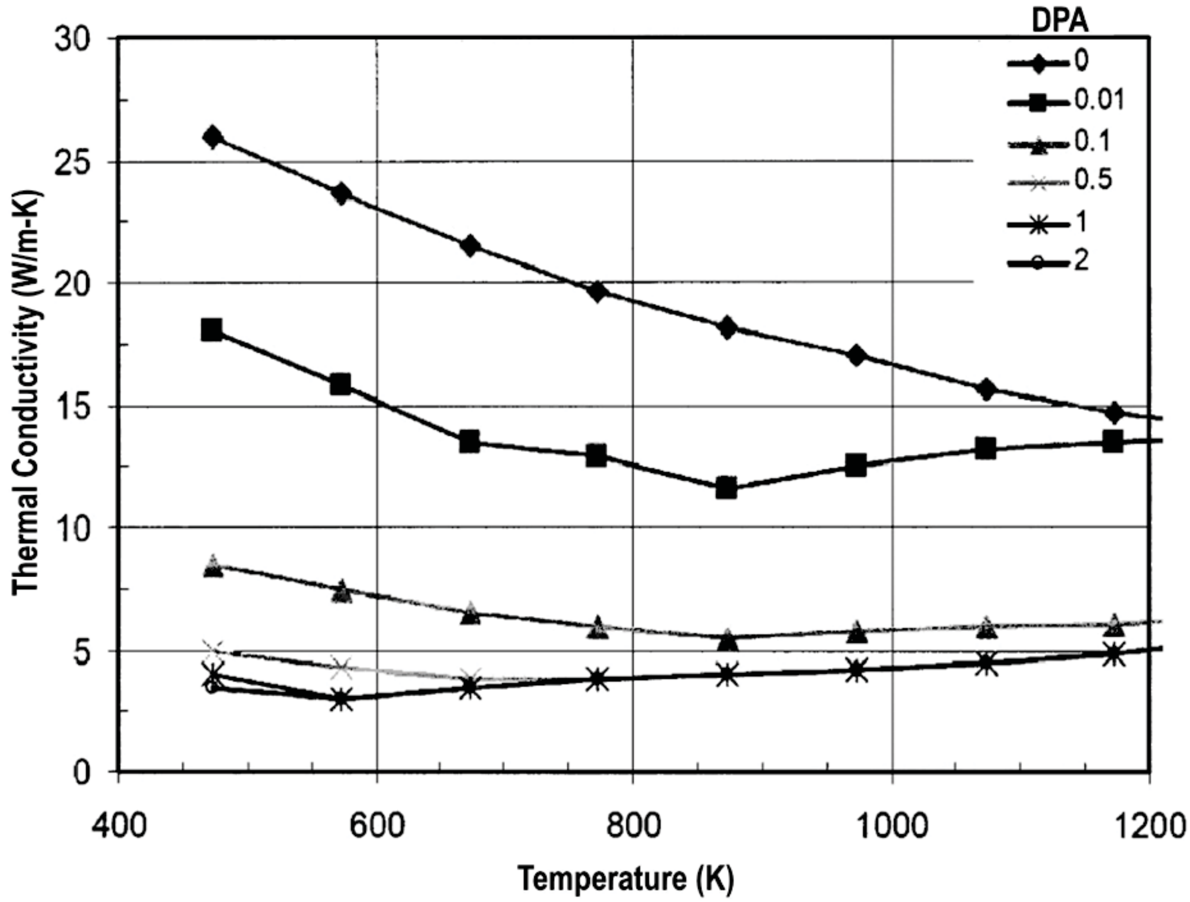

Figure A-4. Variation of thermal conductivity with temperature and irradiation for SiC composite (high-Nicalon Type S with chemical vapor infiltration SiC). Source: Carpenter (2006).

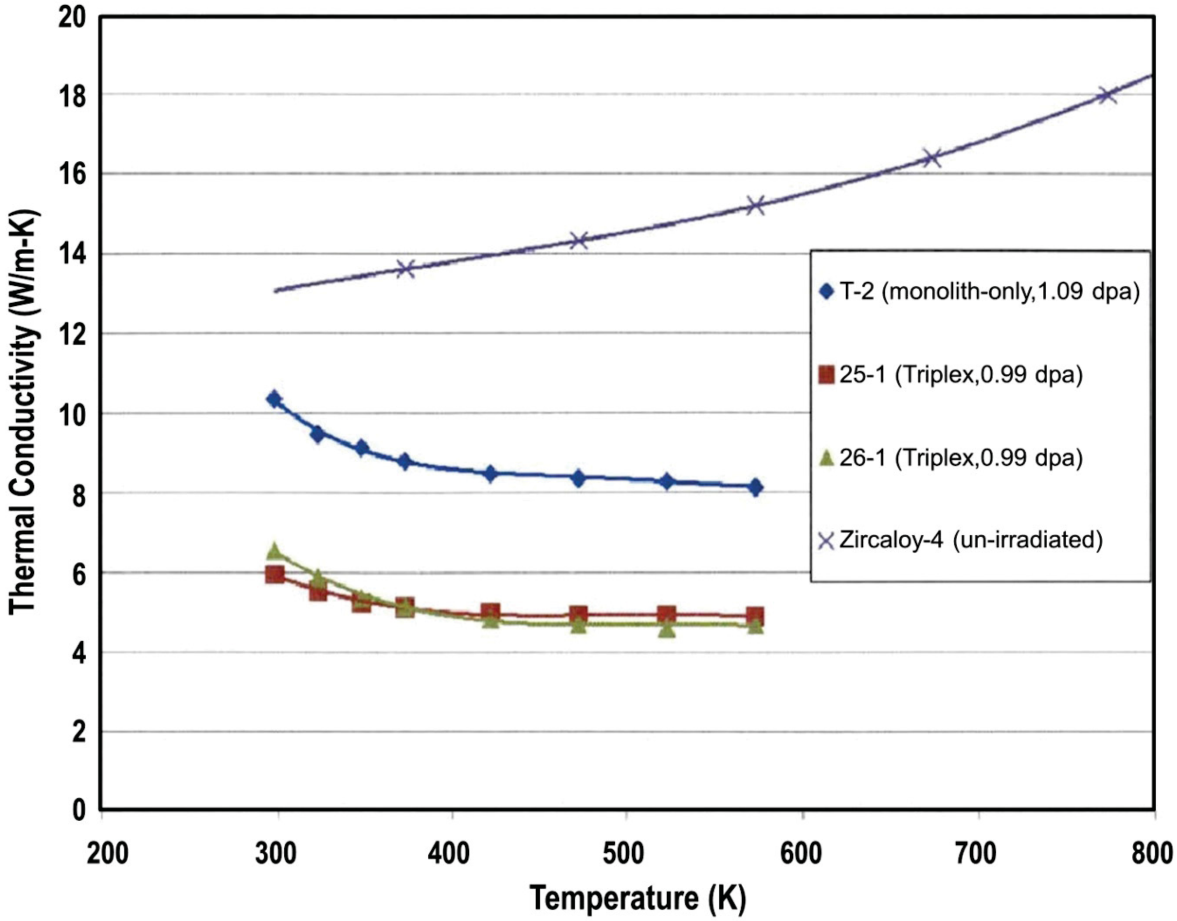

Figure A-5. Thermal conductivity of monolith-only and Triplex SiC vs Zircaloy (Stempien 2011).

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