

Nanocomposite Materials Design Optimization with Experimental Validation for Engineered Microstructure at Multiple Length-Scales

Modeling Contributions

Hongsuk Lee, Vikas Samvedi, Gilberto Mejía-Rodríguez, Vikas Tomar

University of Notre Dame, Indiana
Purdue University, West Lafayette, Indiana

Supported By

The Air Force Office of Scientific Research- AFOSR FA9550-09-0633
Program Manager: Dr. Ali Sayir

Nanocomposite Materials Design Optimization with Experimental Validation for Engineered Microstructure at Multiple Length-Scales (AFOSR FA9550-09-1-0633)

Co PI: Vikas Tomar

Purdue University, West Lafayette, IN

Material Models and Experiments

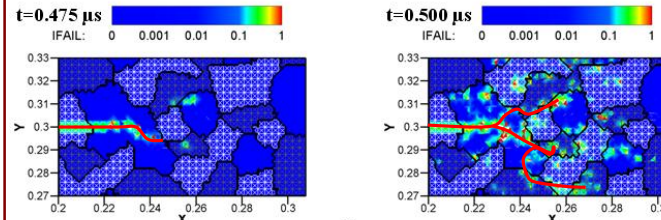
Multiscale Modeling and Experiments

1. Molecular dynamics (MD) framework for complex nano-phase composites.
2. Novel cohesive finite element method (CFEM) to analyze nanocomposite fracture.
3. Prediction of SiC-Si₃N₄ microstructure failure by numerical simulation.

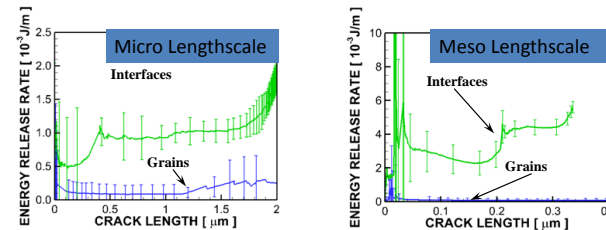
Multiscale Modeling and Experiments

1. MD simulations have provided fundamental insights in SiC-Si₃N₄ composite failure mechanisms
2. A universal fracture energy-material strength relation to predict microstructural fracture has been developed
3. New nanoscale and microscale creep test protocols provide new insights into high temperature small scale material properties

MAIN ACHIEVEMENTS:



Microstructure Dependent Crack Propagation and Fracture Resistance Analyses



$$CE = 50 \cdot \frac{\Phi^2 \cdot (a \times b)}{T_{\max} \cdot h^2} \cdot CD$$

Journal of Engineering Materials and Technology, July 2011, One of the top 10 downloaded papers in 2011-12

HOW IT WORKS:

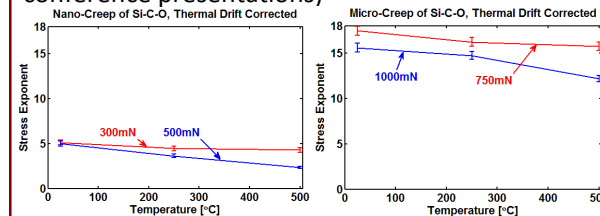
• Experimental microstructure is photographed and imported into finite element analyses while properties are calculated using a combination of multiscale high temperature tests and molecular scale computations.

ASSUMPTIONS AND LIMITATIONS:

• Visualization needed for high temperature experiments and accurate potentials are required for molecular analyses

Current Impact

• A new ceramic nanocomposite microstructure dependent property calculation framework established with identifiable relations and parameters to feed into multiscale design models. Experiments point to new high temperature deformation mechanisms operating at nanoscale and micron scale not yet modeled using simulations. Group Publications: (9 International Journal Articles, 8 invited talks, 2 conference proceeding articles, multiple conference presentations) Collaborative Work: (3 International Journal Articles, 2 invited talks, 4 conference proceeding articles, multiple conference presentations)



Planned Impact

• A robust design optimization capable of predicting optimal SiC-Si₃N₄ microstructures under uncertain processing and operating environments. • Experimental validation of models and design procedure.

Research Goals

• Development and experimental validation of a numerical tool to optimally design multiscale nanocomposites based on direct correlation with processing and experiments.

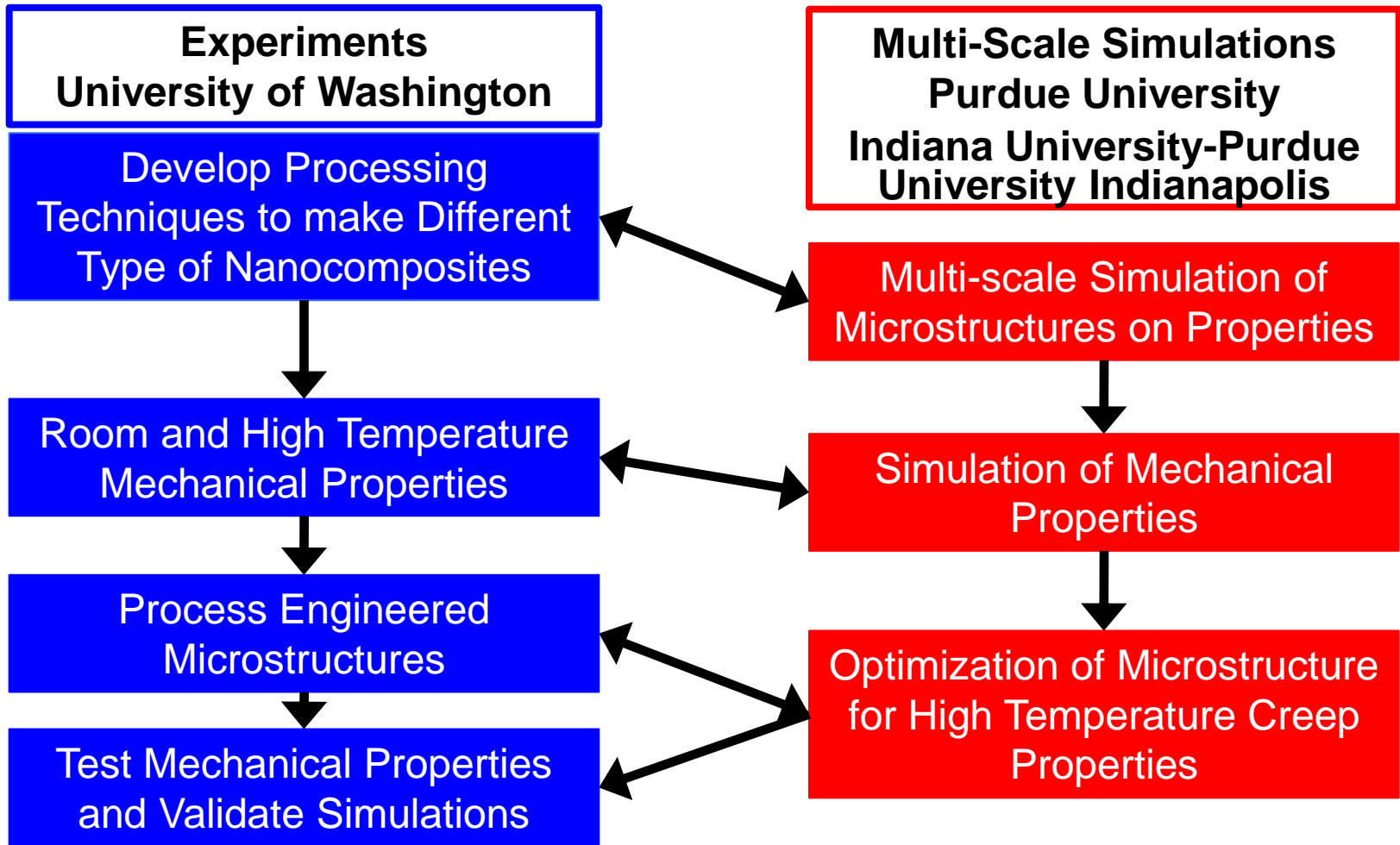
QUANTITATIVE IMPACT

END-OF-PHASE GOAL

STATUS QUO

NEW INSIGHTS

Simulation Guided Materials Development

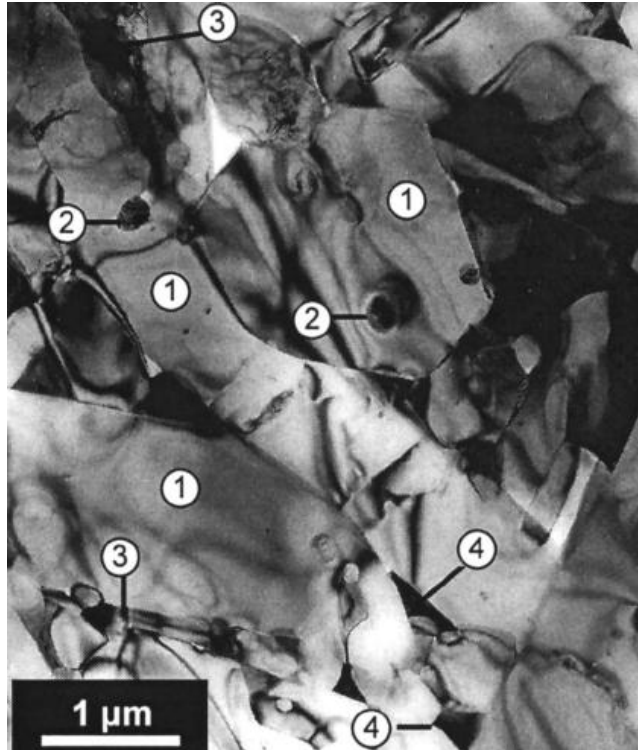


Predictive tool to design composite microstructures with optimized properties

The Big Picture

Molecular

1. Effect of nanoscale variations in particle sizes
2. Effect of changes in grain boundary composition, placement of particles with respect to grain boundary etc.
3. Effect of temperature, development of new constitutive models
4. BUT, length scale and time scale are out of reach. At best, it is mechanistic information



Continuum

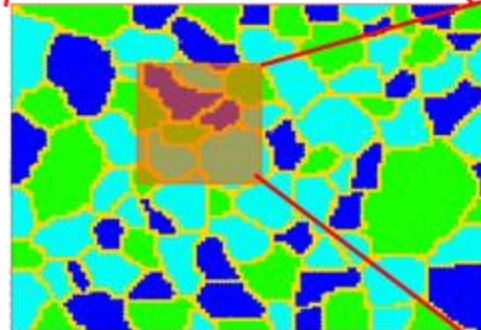
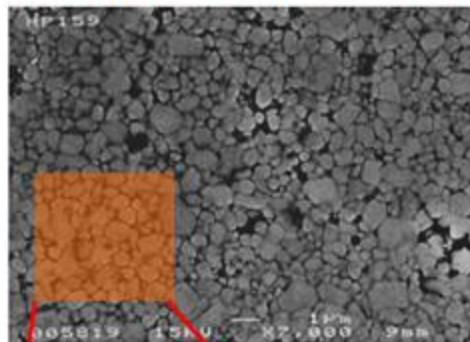
1. Overall combined effect of different morphology factors present simultaneously or separately on the fracture resistance of a composite morphology
2. Analyses at experimentally accessible loading rates
3. BUT, there is a lack of mechanistic information

Experiments

Bridging using experiments

Set-UP

Experimental Microstructure

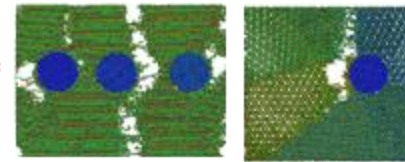


Finite Element Representation Based on Experimental Microstructural Characterization

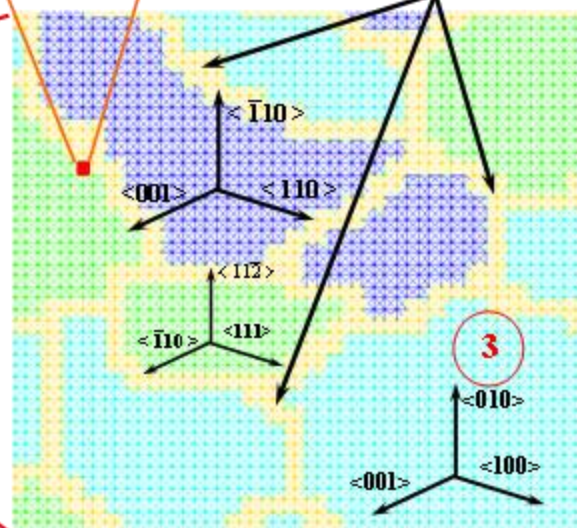
Finite Element



Molecular Models of Interfaces To Derive Finite Element Properties

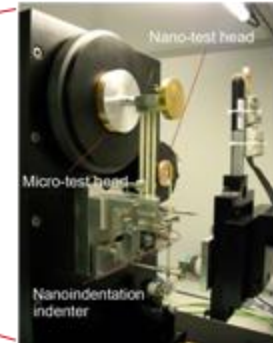
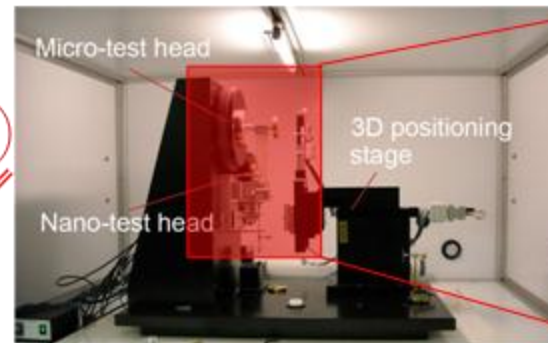


Grain Boundary

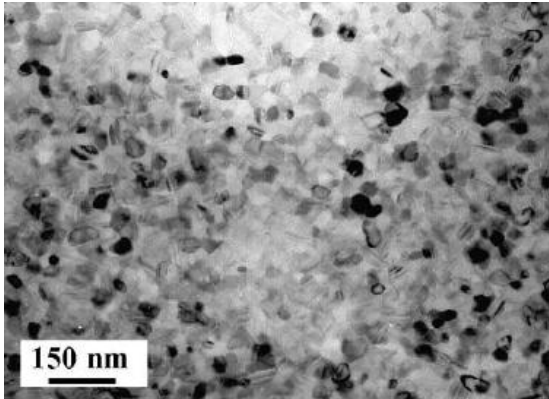


Detailed View of Finite Element Mesh with Explicit GB and Orientation Representation

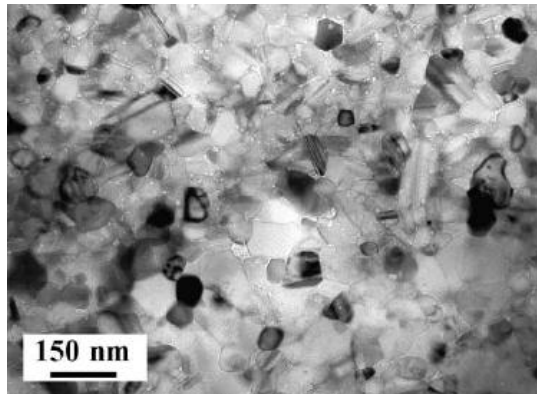
Experiments



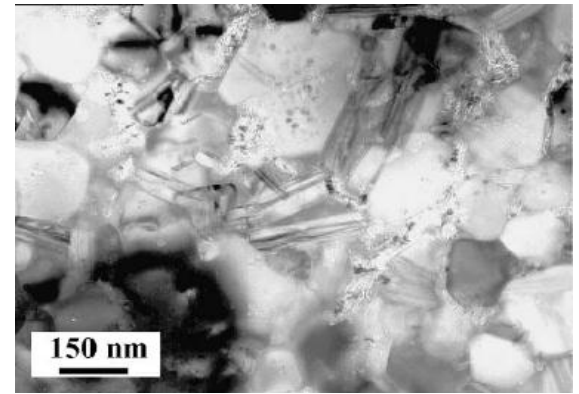
Few Micrographs



30 SC

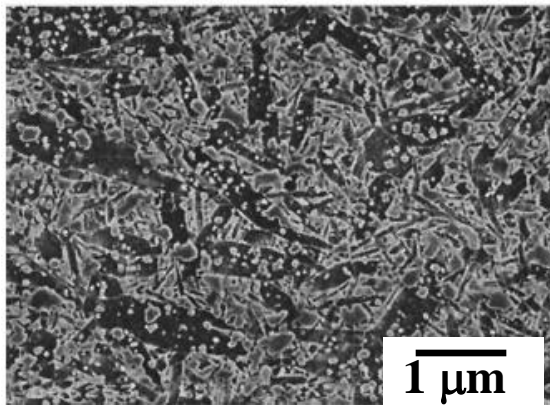


20 SC

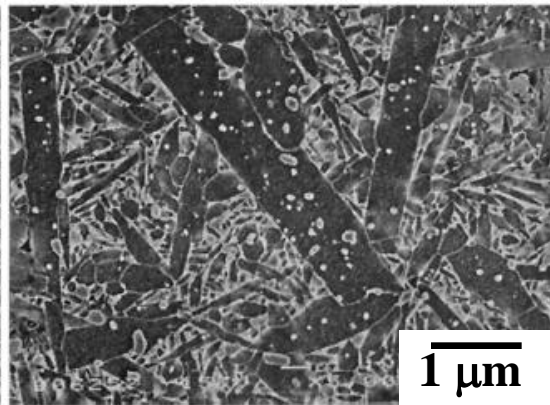


10 SC

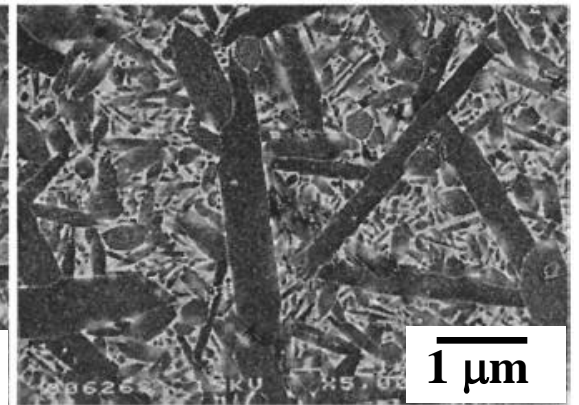
TEM



20SC



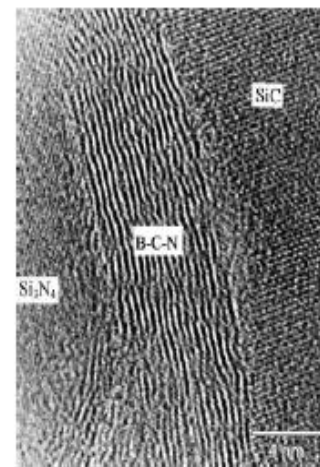
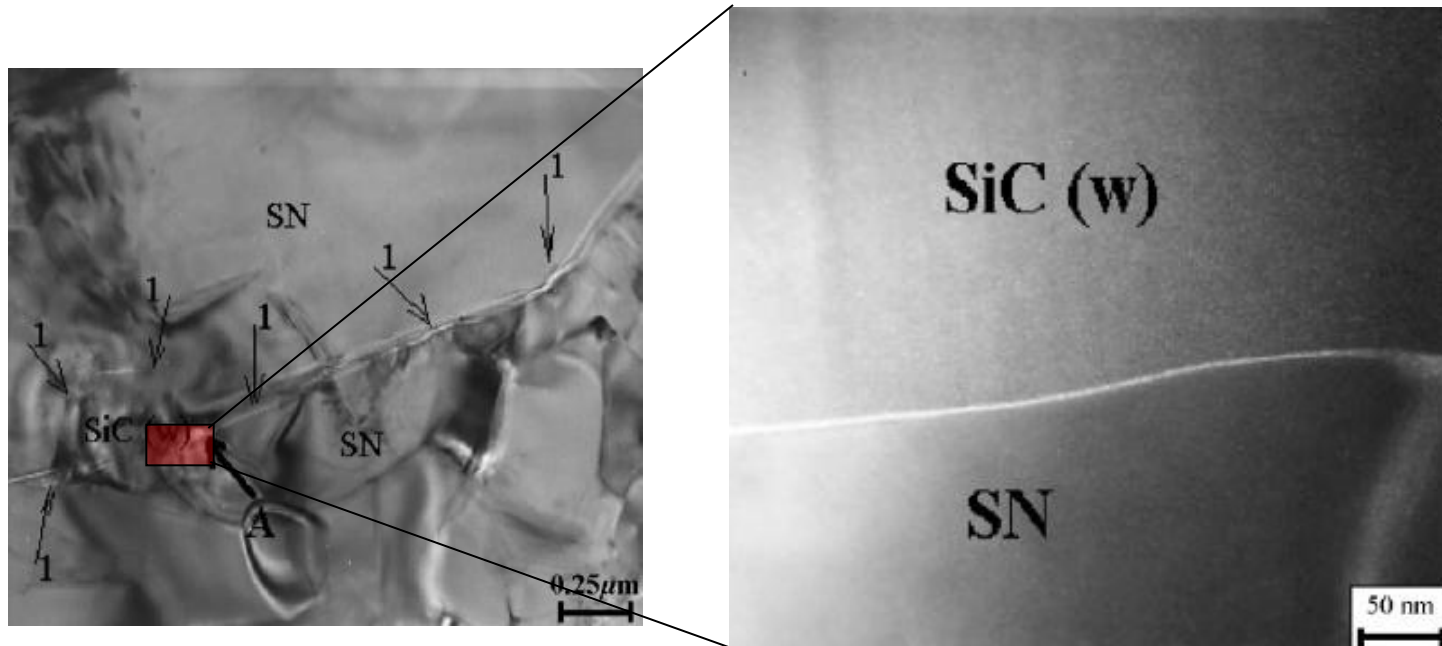
5SC



100SN

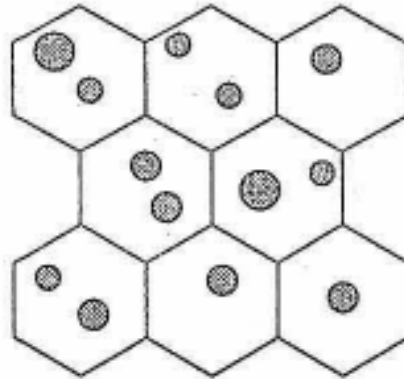
SEM

Interfaces

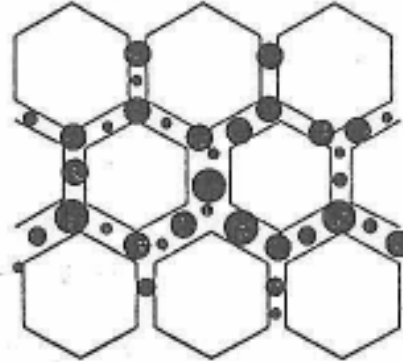


Classes of Nanocomposite

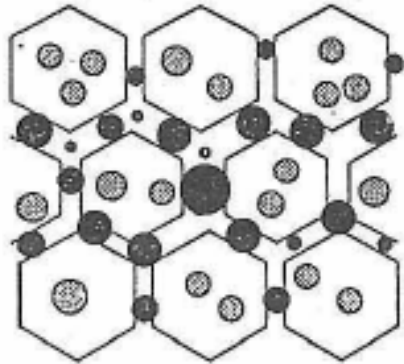
Intra-type



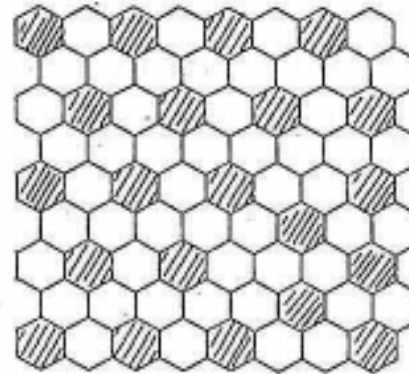
Inter-type



Intra/inter-type

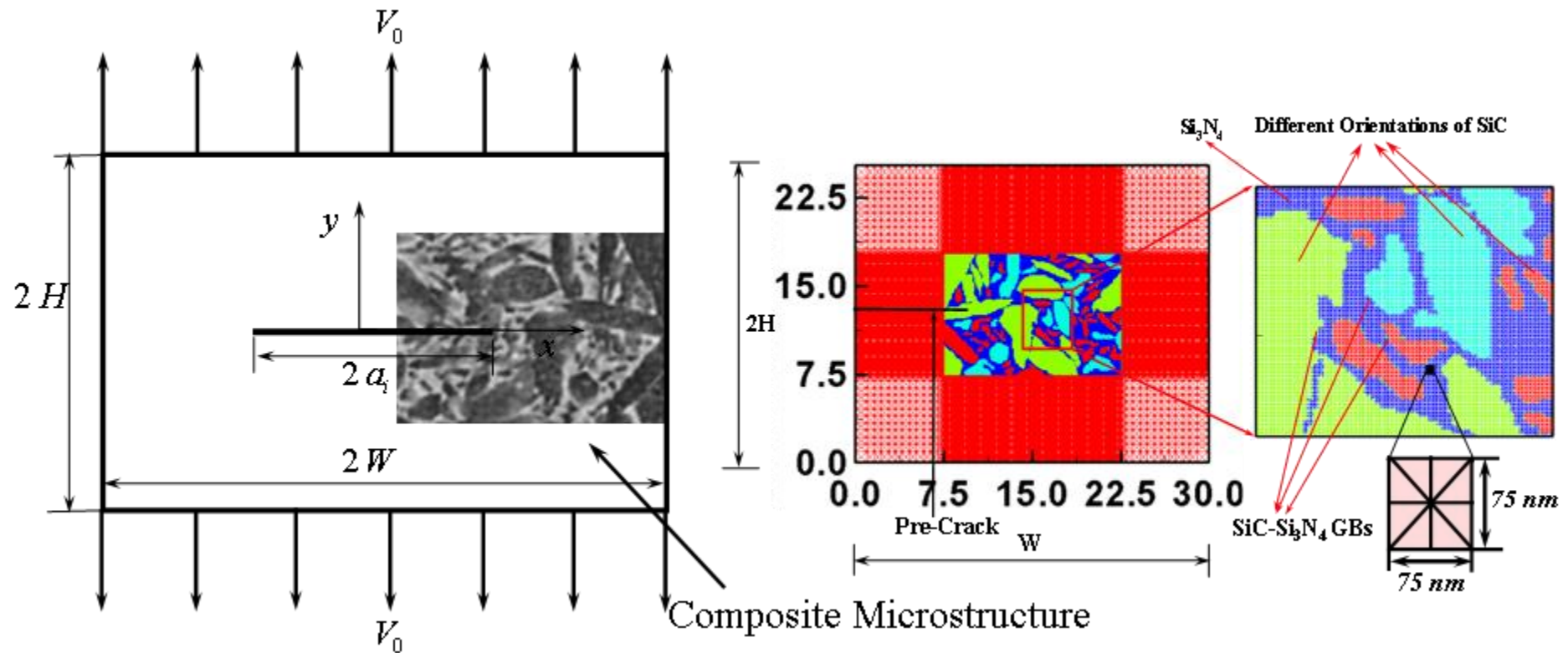


Nano/nano-type



Niihara, *J. Cerm. Soc. Jap.*, 99
[10] (1991) p. 974.

Modeling Framework



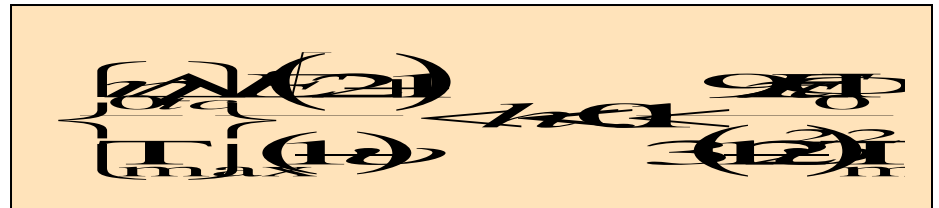
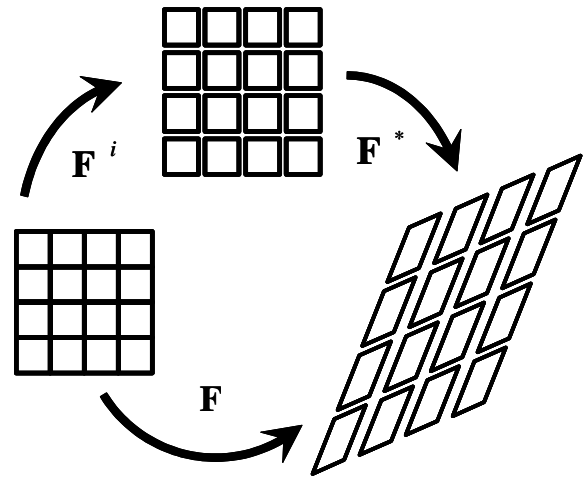
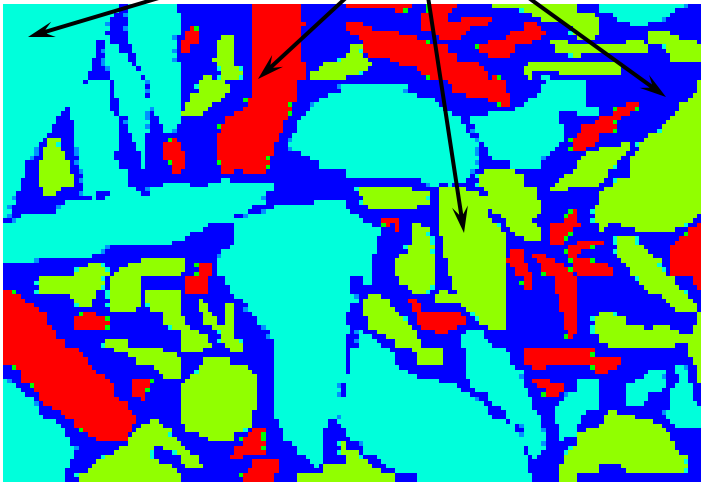
Issue of Fracture Lengthscale In a Nanocomposite

- LaGrangian Kinetics Description

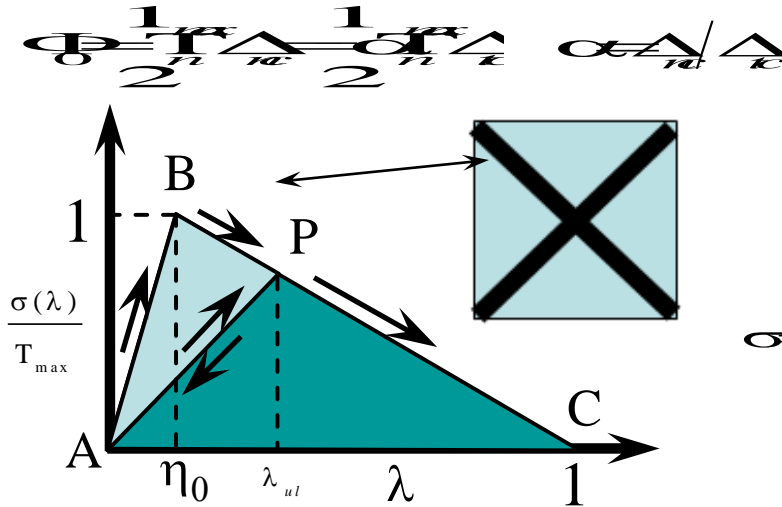


- Cohesive Zone Size

$$d_z = \frac{9\pi E \Phi_0}{32(1-\nu^2)T_{max}^2}$$



Bilinear Mixed-Mode Cohesive Law (Tomar and Zhou, 2004)



$$\sigma = \sqrt{\left(\frac{T_n}{T_{\max}}\right)^2 + \left(\frac{T_t/\alpha}{T_{\max}}\right)^2} = \begin{cases} \left(\frac{T_{\max}}{1-\eta_0}\right)^{\frac{1-\eta}{\eta}} \lambda^{\frac{1-\eta}{\eta}}, & \text{if } 0 \leq \lambda \leq \eta_0 \\ \left(\frac{T_{\max}}{1-\eta_0}\right)^{\frac{1-\eta}{1-\eta_0}} (1-\lambda)^{\frac{1-\eta}{1-\eta_0}}, & \text{if } \eta_0 < \lambda \leq 1 \\ 0 & \text{if } \lambda > 1. \end{cases}$$

Effective separation

$$\lambda = \sqrt{\left(\frac{\Delta_n}{\Delta_0}\right)^2 + \left(\frac{\Delta_t}{\alpha \Delta_0}\right)^2}$$

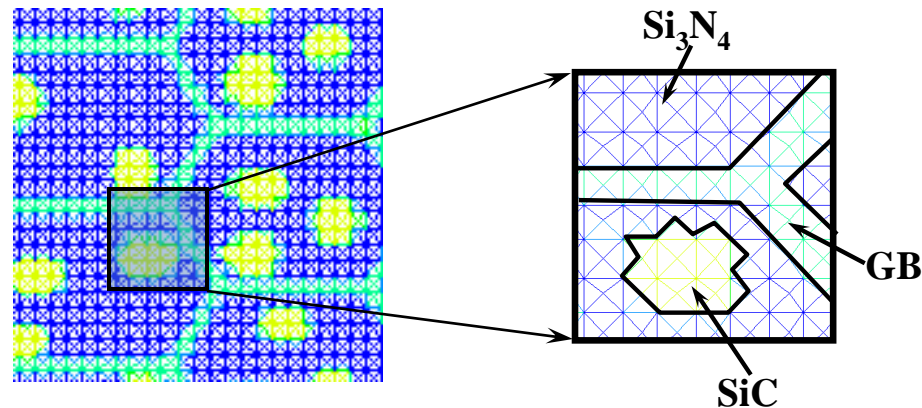
Effective traction

$$T_n = T_{\max} \left(\frac{\Delta_n}{\Delta_0}\right)^{\frac{\eta}{1-\eta}} \quad \text{and} \quad T_t = \alpha T_{\max} \left(\frac{\Delta_t}{\Delta_0}\right)^{\frac{\eta}{1-\eta}}$$

Damage Parameter,

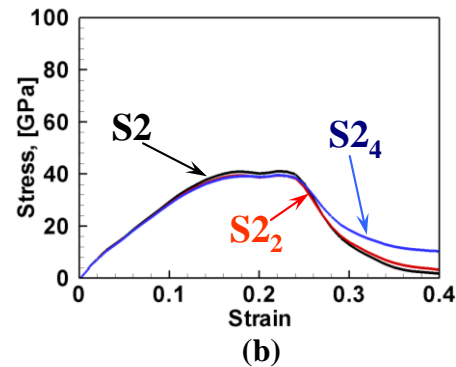
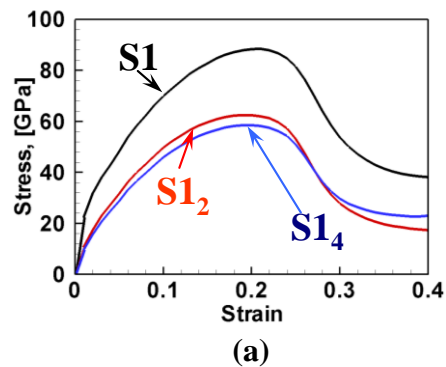
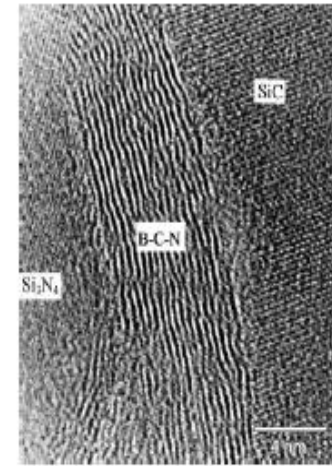
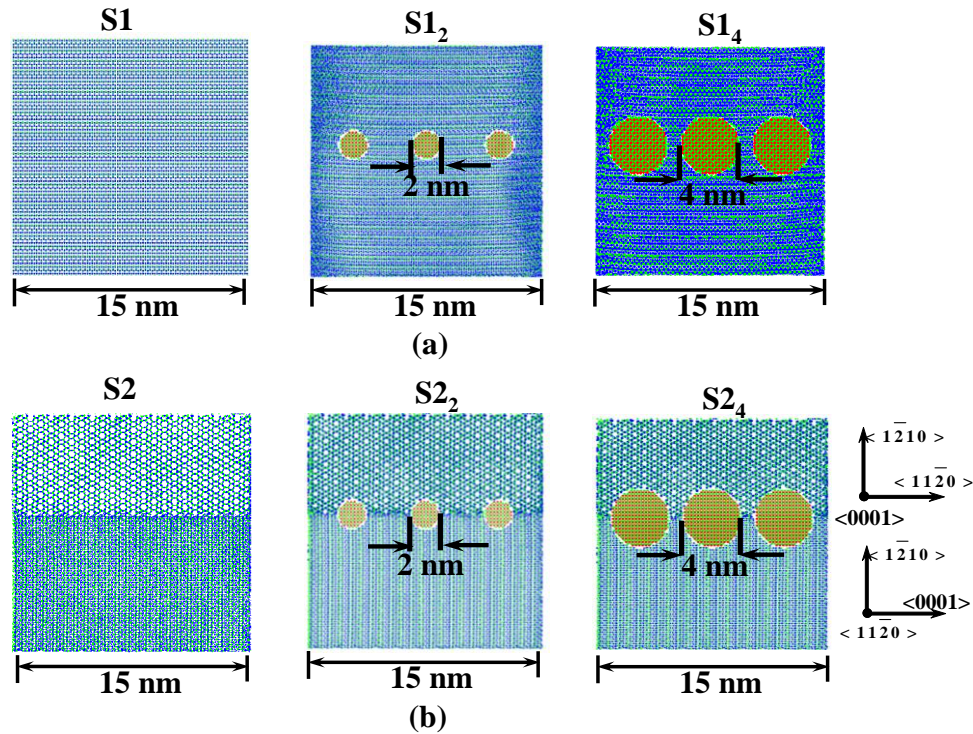
$$D = \frac{\Phi_d}{\Phi_0}, \quad 0 \leq D \leq 1$$

Material Properties

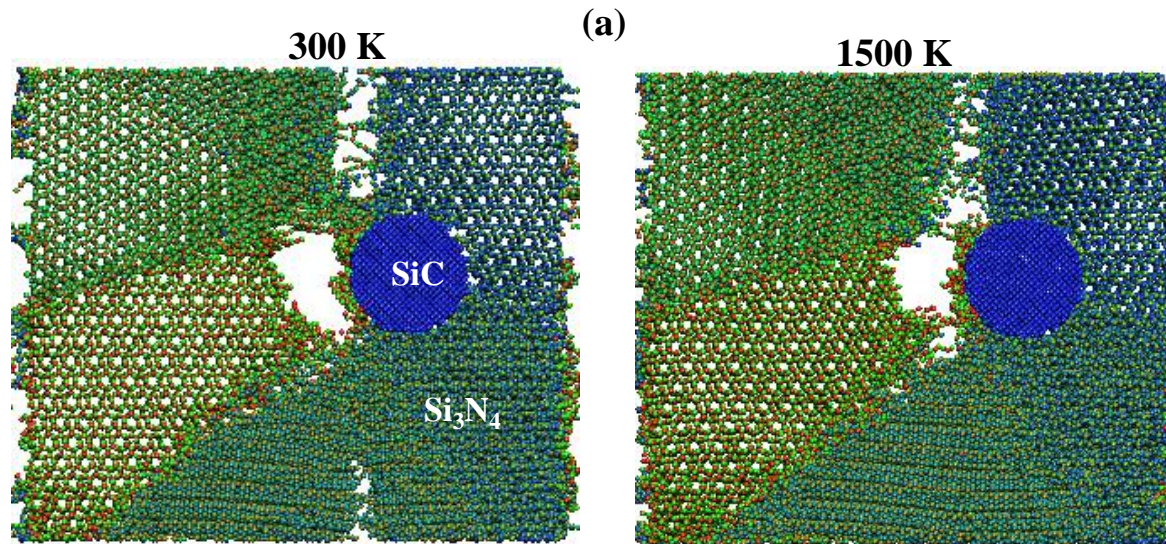
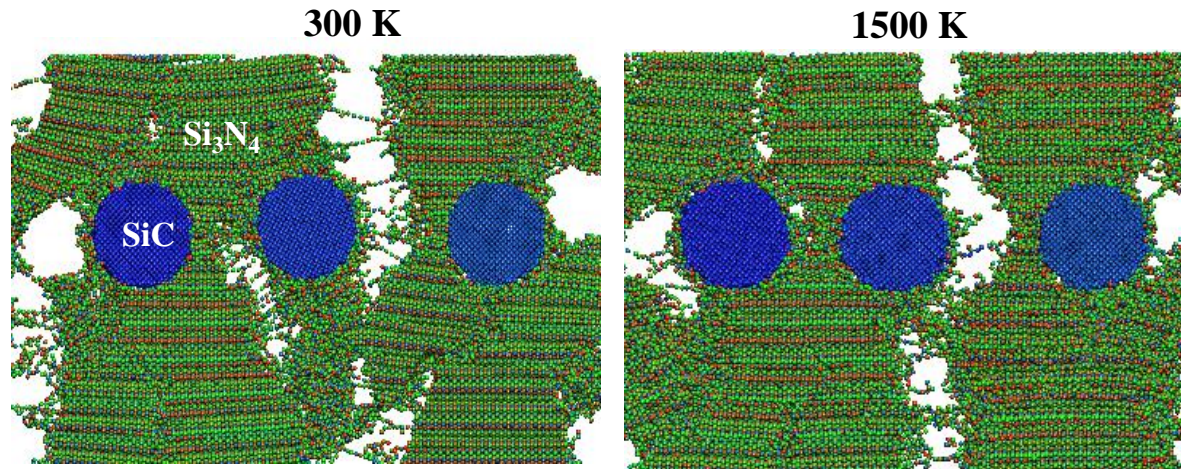


Component	Φ (N/m)	T (GPa)	Δ_c (nm)	E (GPa)	ν	ρ (kg/m ³)
SiC (sc)	19.53	1.02	38.3	449	0.16	3215
Si ₃ N ₄ (sn)	191.5	2.3	166.5	210	0.22	2770
GBs (g)	238.7	2.38	200.6	200	0.16	4000
(sc-g)	19.53	1.02	38.3	--	--	--
(sc-sn)	19.53	1.02	38.3	--	--	--
(sn-g)	191.5	2.3	166.5	--	--	--
Homogenized (H)	127.8	2.03	125.9	256.8	0.202	2982
H-sc	19.53	1.02	38.3	--	--	--
H-sn	127.8	2.03	125.9	--	--	--
H-gb	127.8	2.03	125.9	--	--	--

Effect of Particle Size Change



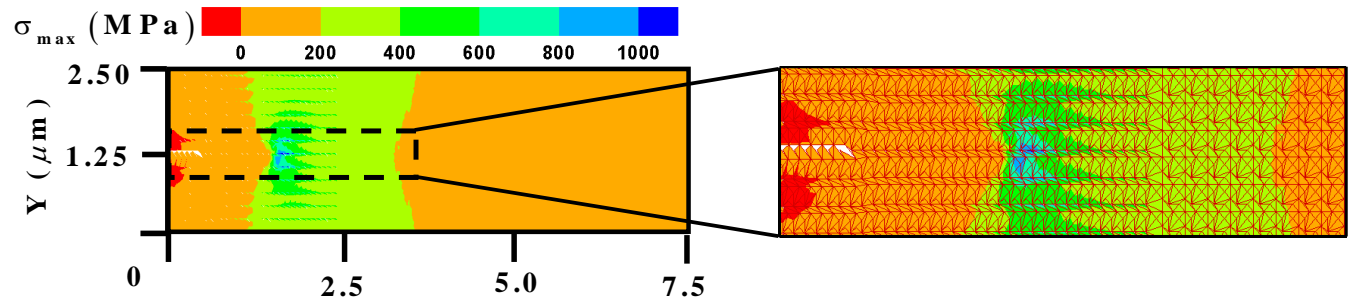
Effect of Temperature on Deformation Mechanism



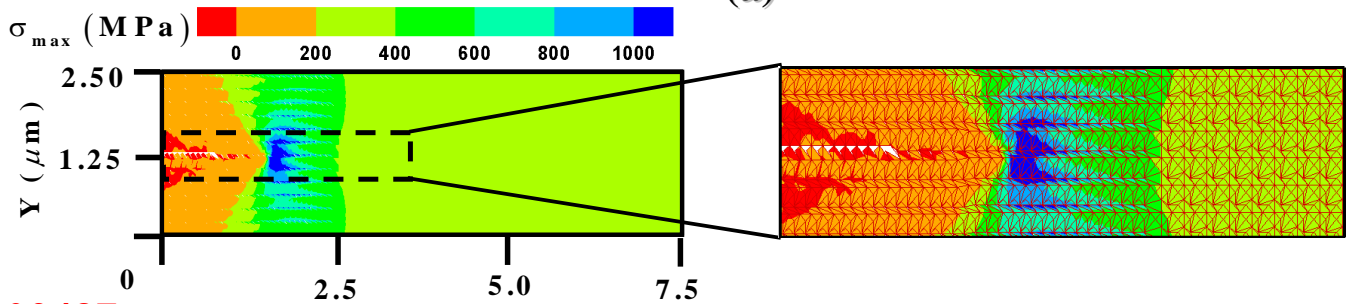
(b)

Propagation of Crack-Local View

0.01312 μsec

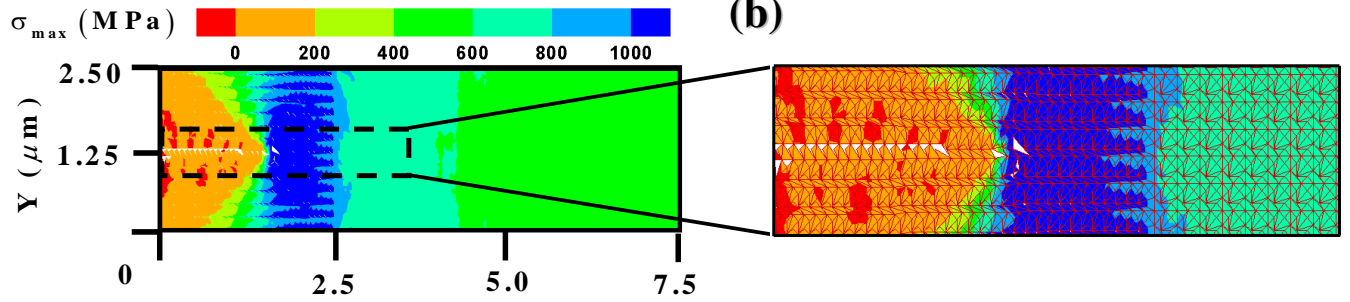


0.01688 μsec



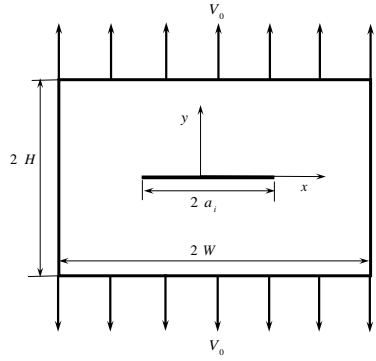
(a)

0.02437 μsec



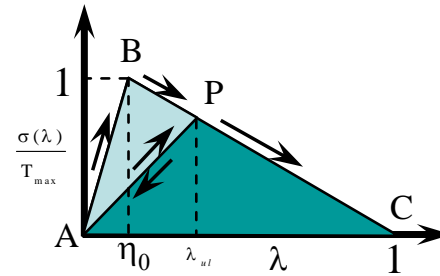
(b)

(c)



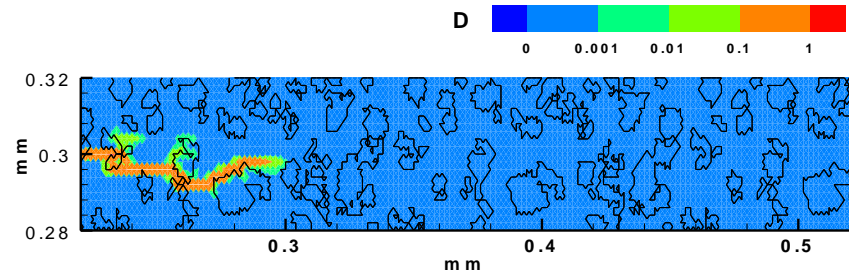
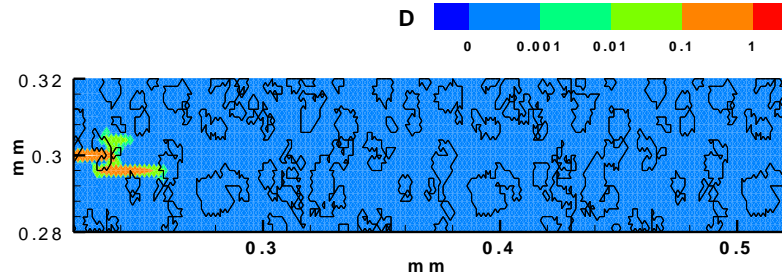
Microstructure Damage Pattern

$$D = \frac{\Phi_d}{\Phi_0} = \frac{ABP}{ABC},$$



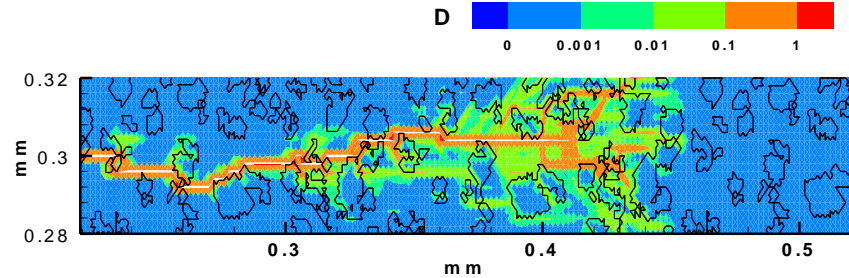
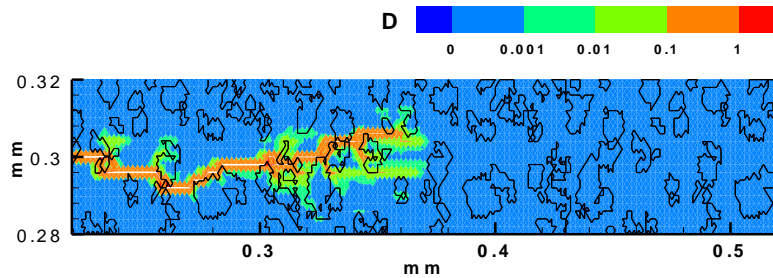
Time = 0.09 μ s

Time = 0.11 μ s

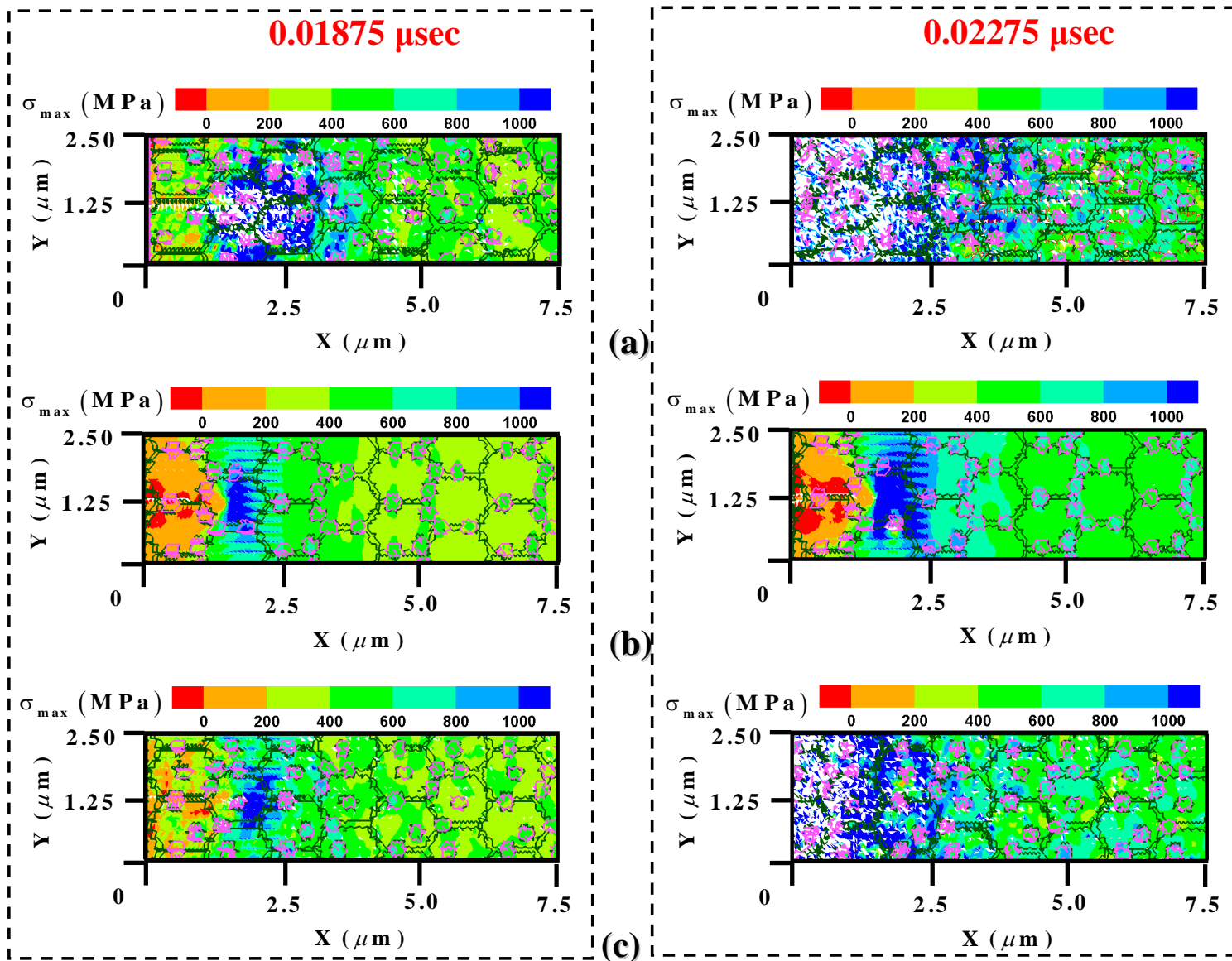


Time = 0.13 μ s

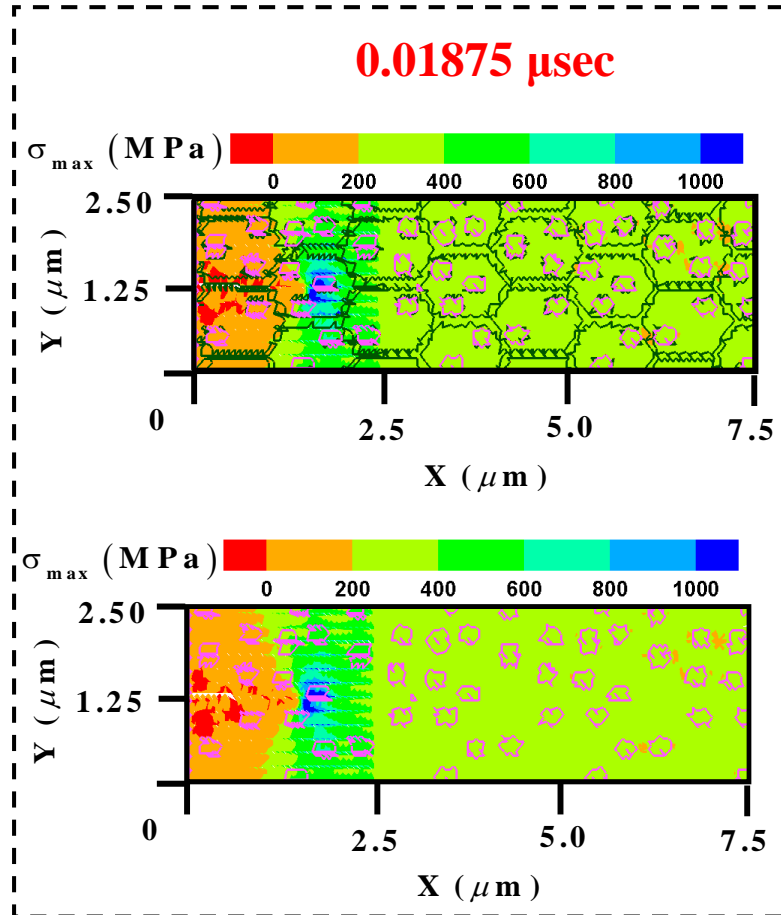
Time = 0.15 μ s



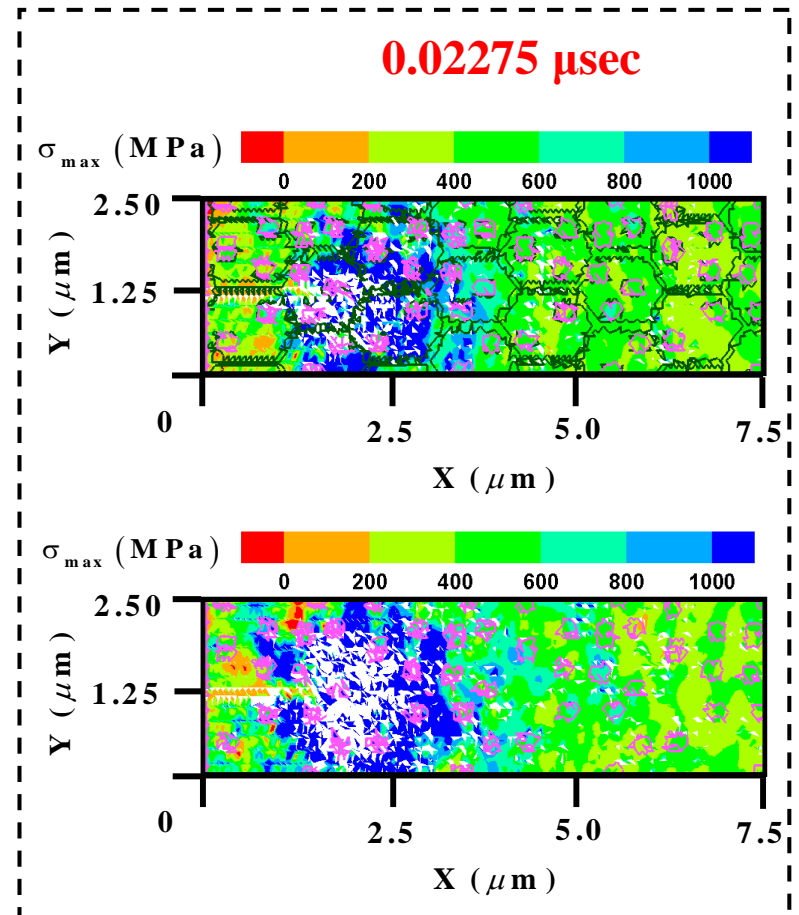
Damaging Effect of Second Phase Particles



Damage Control Effect of GBs

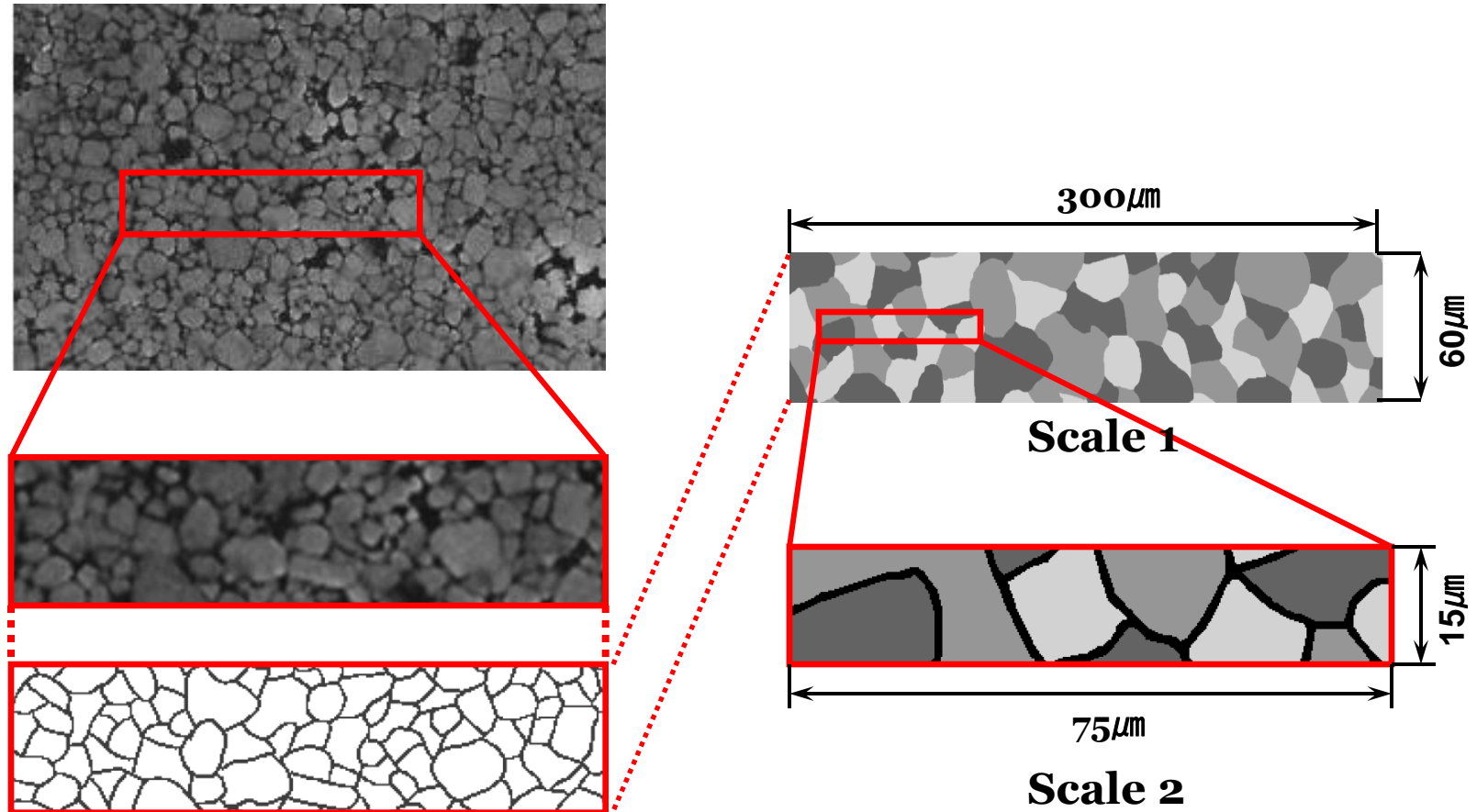


(a)



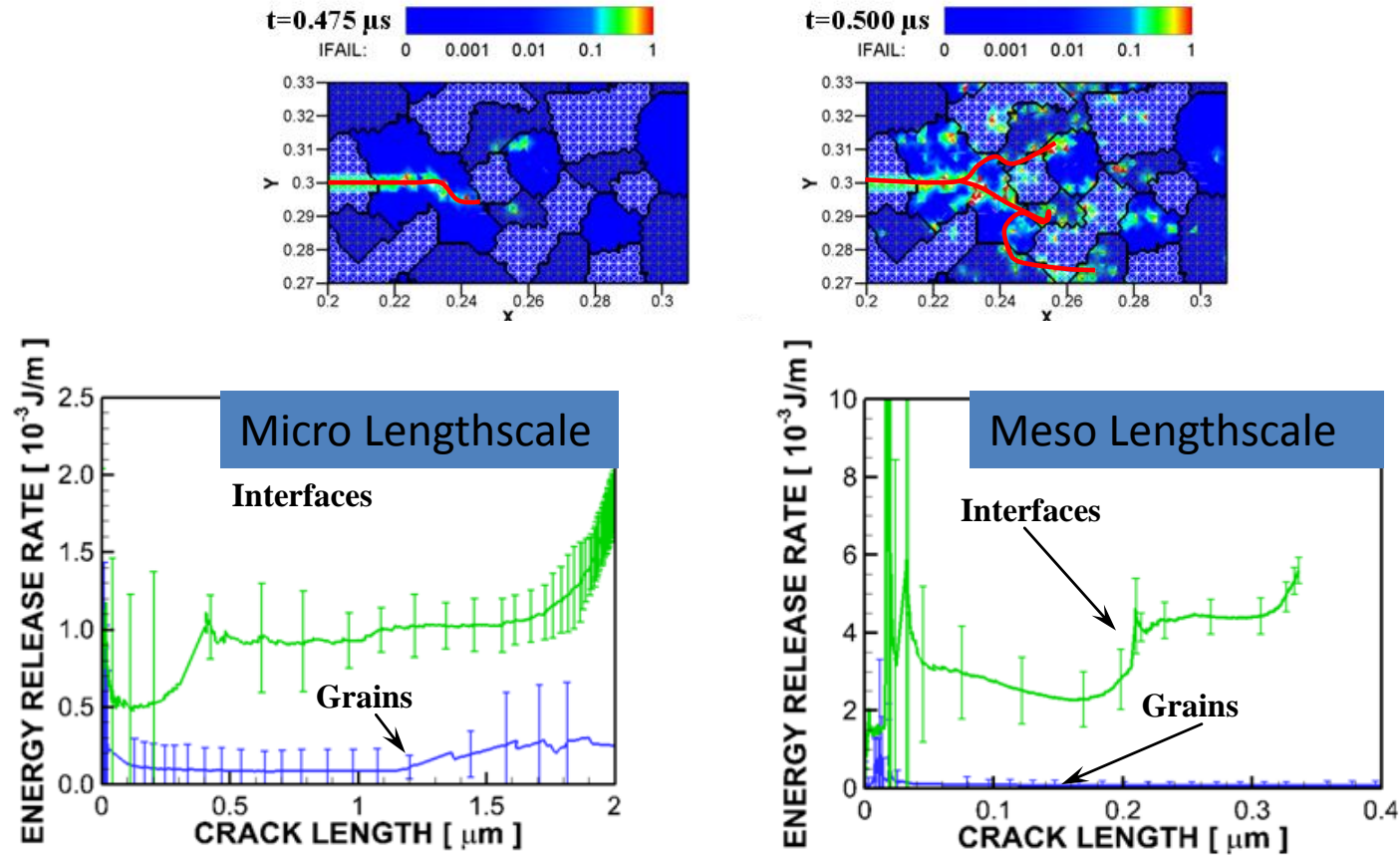
(b)

Finite Element Modeling Based on Nanoscale Experiments and Classical Molecular Dynamics Models of Ceramic Experimental Microstructures for Design Models



Woetting, G., Caspers, B., Gugel, E. and Westerheide, R. (2000).

Average Energy Release Rate and Variations vs. Crack Length: A New Relation Has Been Proposed to Link Grain Boundary Properties with Microstructural Failure



$$CE = 50 \cdot \frac{\Phi^2 \cdot (a \times b)}{T_{\max} \cdot h^2} \cdot CD$$

Journal of Engineering Materials and Technology, July 2011, One of the top 10 downloaded papers in 2011-12

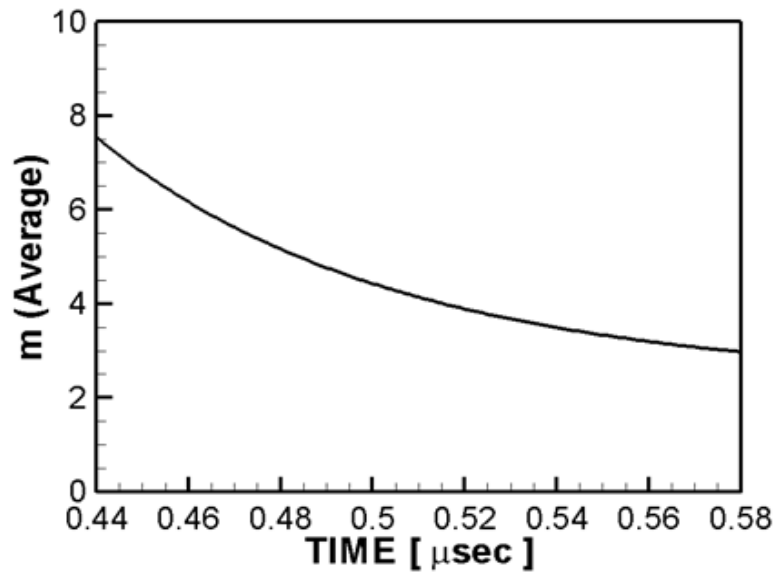
Surface energy relation

$\gamma = \gamma_{pc} [1 - (G / G_s)]^m + \gamma_{\mu} \rightarrow \gamma_{pc}$ and γ_{μ} are polycrystalline surface energy and micro crack formation energy, respectively.

Term, $\gamma_{pc} [1 - (G / G_s)]^m$, is used to denote contribution of a polycrystalline microstructure's grain size dependence on primary crack propagation fracture energy.

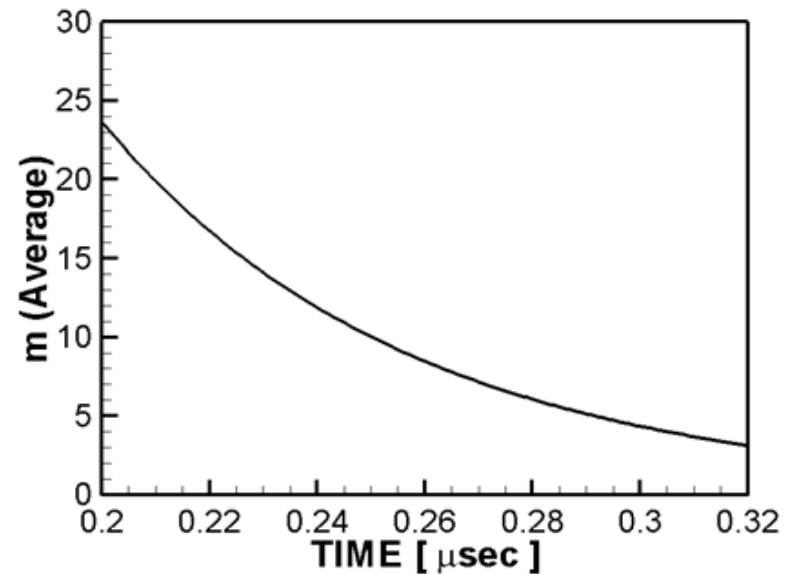
The parameter **m** indicates the sensitivity of the primary crack formation and extension energy on average microstructure grain size.

1st Scale



$$m = 6292 e^{-16.31 \times t} + 3.671 e^{-0.6735 \times t}$$

2nd Scale



$$m = 666.4 e^{-22.43 \times t} + 333.5 e^{-15.14 \times t}$$

Energy Dissipation vs. Time relation

The form of developed relation is

$$\text{Energy dissipation} = A \cdot \exp(B \cdot t) + C \cdot \exp(D \cdot t)$$

It appears to have A, B, C, D with 95% confidence bounds with,

$$A = 3.391\text{E-}6$$

$$B = 8.804\text{E}4$$

$$C = -2.278\text{E-}5$$

$$D = -3.719\text{E}6$$

$$CE = \frac{100}{T_{\max}} e^{(\rho \cdot \Phi \cdot t)} - \frac{2}{\rho \cdot \Phi} e^{\left(-\frac{E}{10000} \cdot t\right)}$$

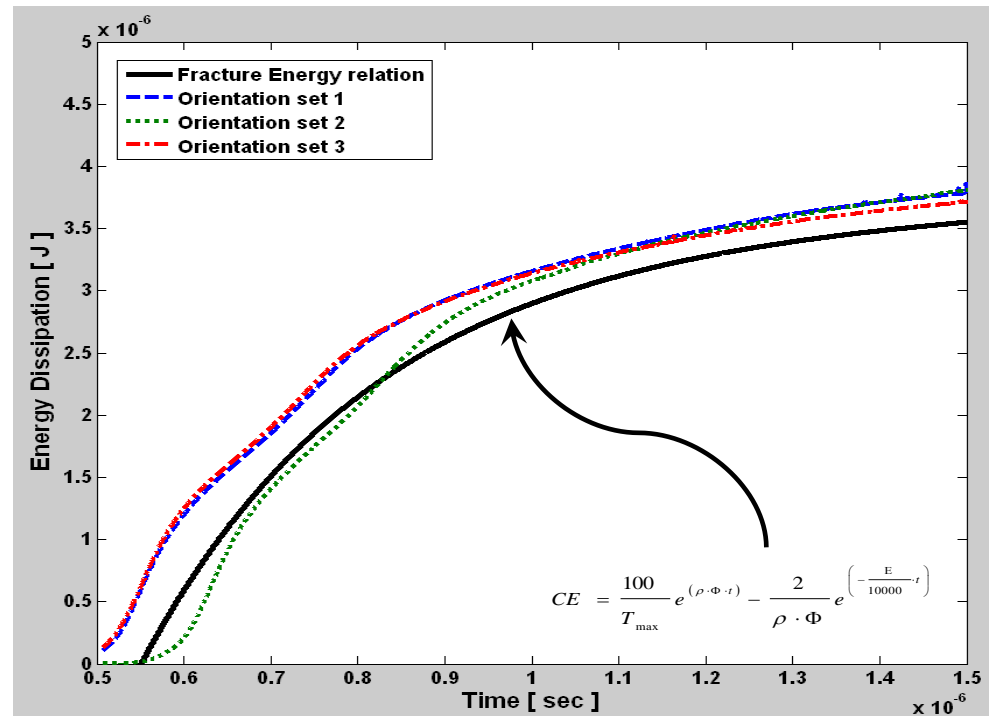
$$E = 379.91 \times 10^9 \text{ pa}$$

$$\nu = 0.184$$

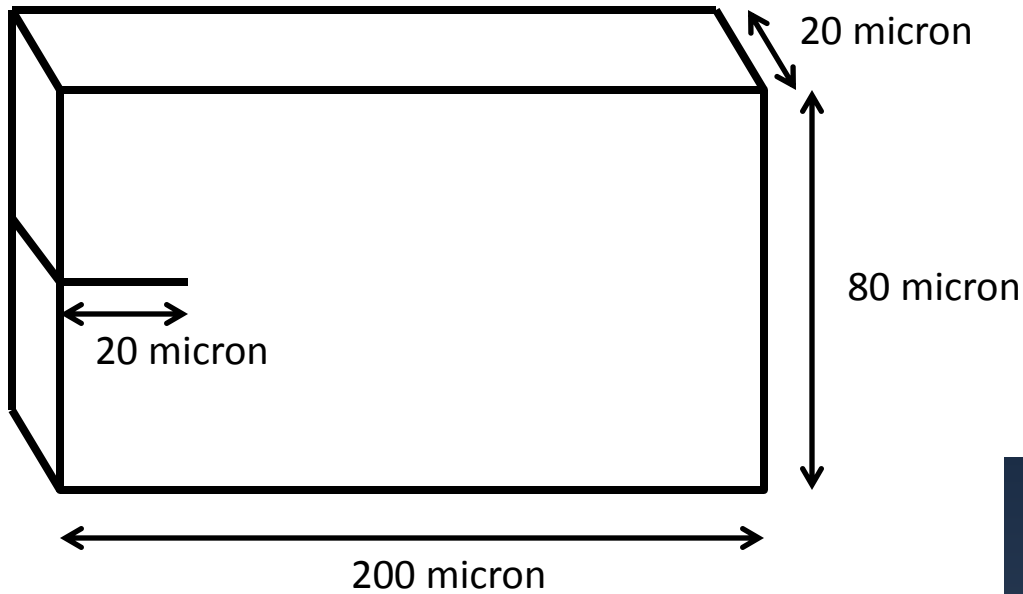
$$\rho = 3536.5 \text{ Kg} / \text{m}^3$$





$$\Phi = 20.51 \text{ J} / \text{m}^2$$

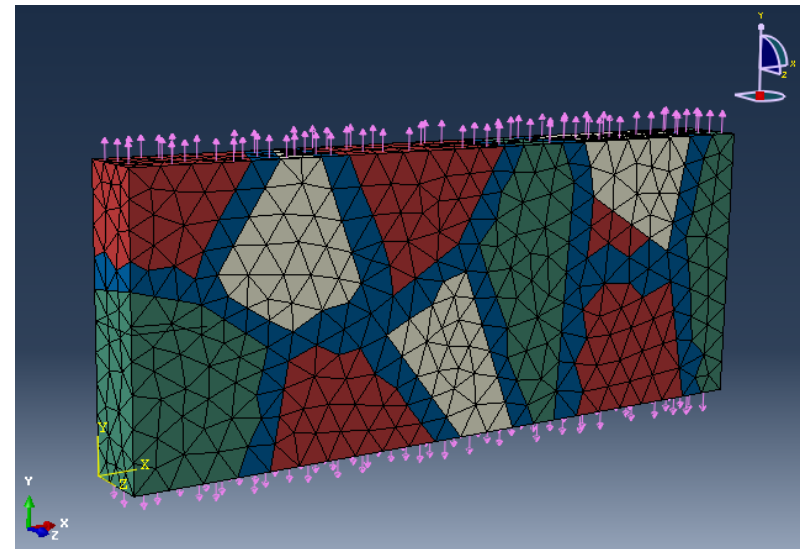
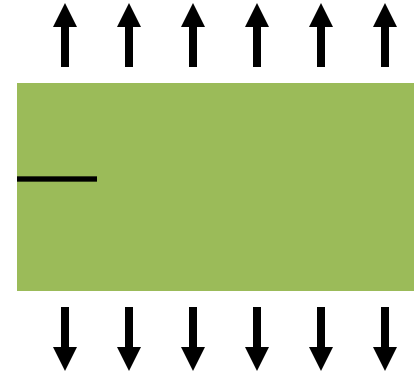
$$T_{\max} = 326.8 \times 10^6 \text{ pa}$$



3-D POLYCRYSTALLINE MICROSTRUCTURE MODEL



- Material 1:  $E=300\text{Gpa}$, $\nu=0.28$, $T=250\text{Mpa}$
- Material 2:  $E=400\text{Gpa}$, $\nu=0.28$, $T=200\text{Mpa}$
- Material 3:  $E=500\text{Gpa}$, $\nu=0.28$, $T=150\text{Mpa}$
- Interface :  $E=15\text{Gpa}$, $\nu=0.28$, $T=3000\text{Mpa}$
- Displacement at failure : 0.1 microns

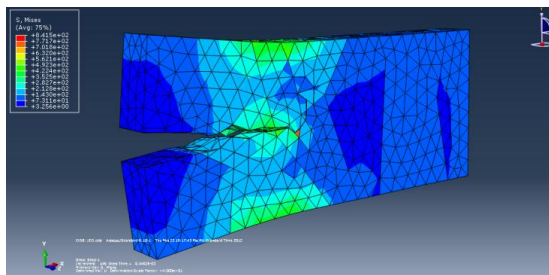
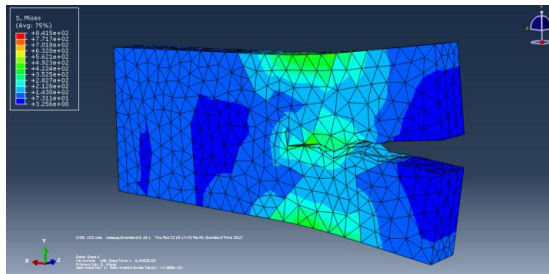
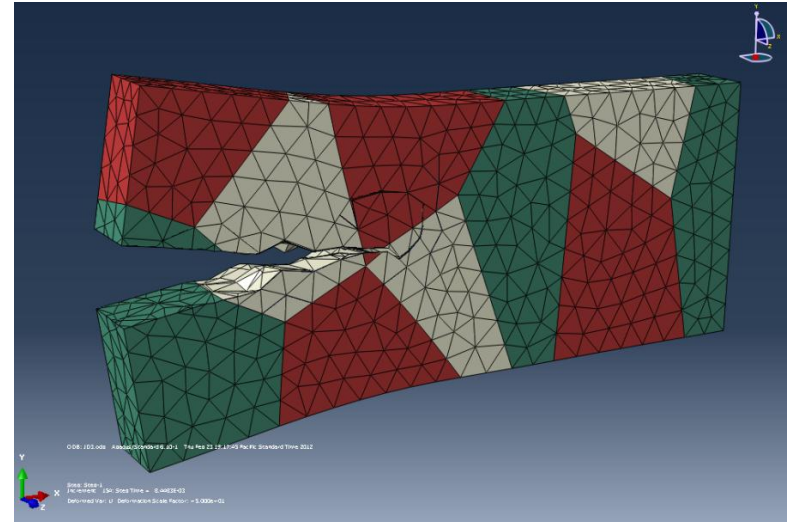
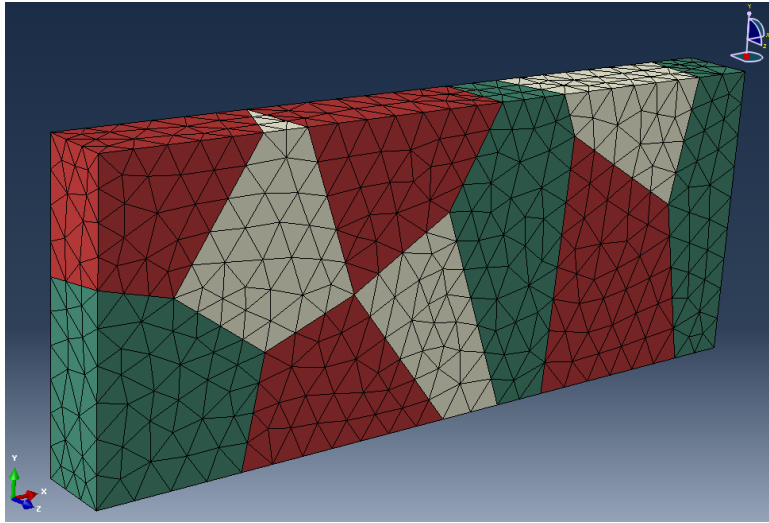


LOAD : SURFACE LOAD

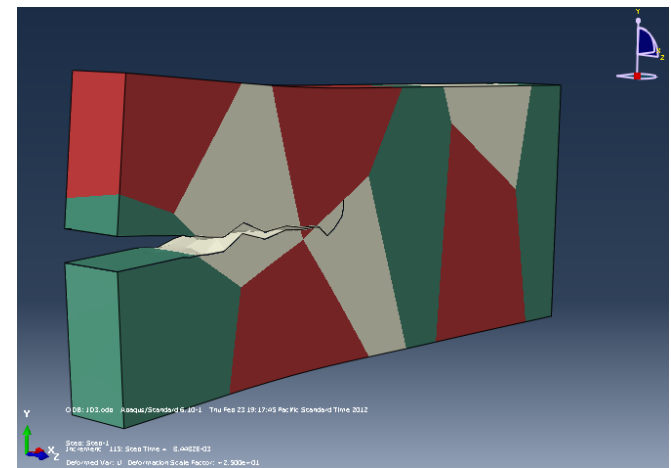
$F = 800\text{N}$

SIMULATION

WITHOUT INTERFACE (GRAIN BOUNDARY)



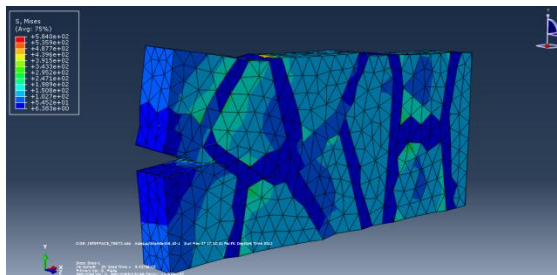
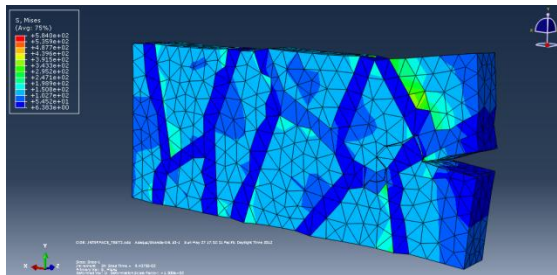
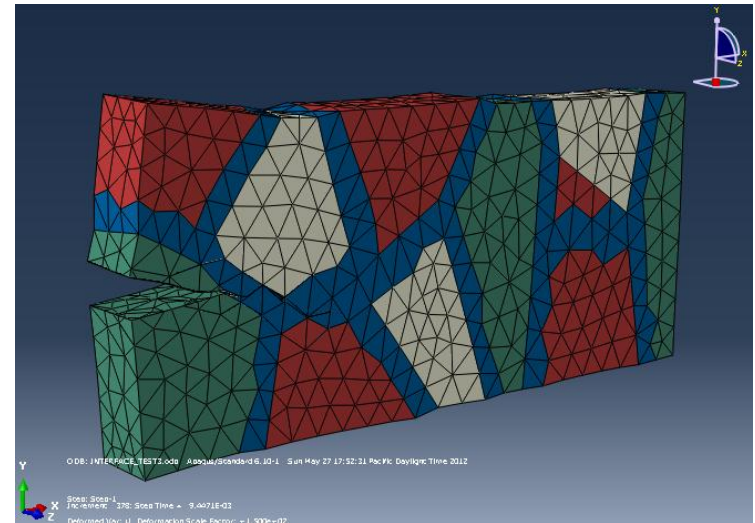
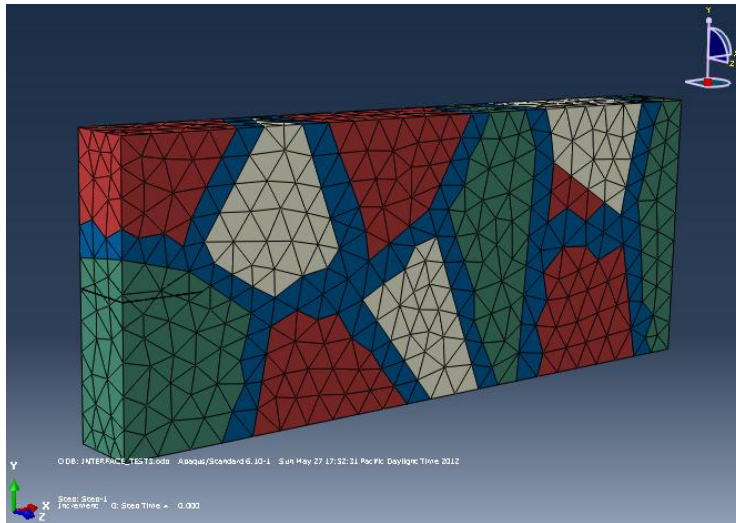
Mises stress contour



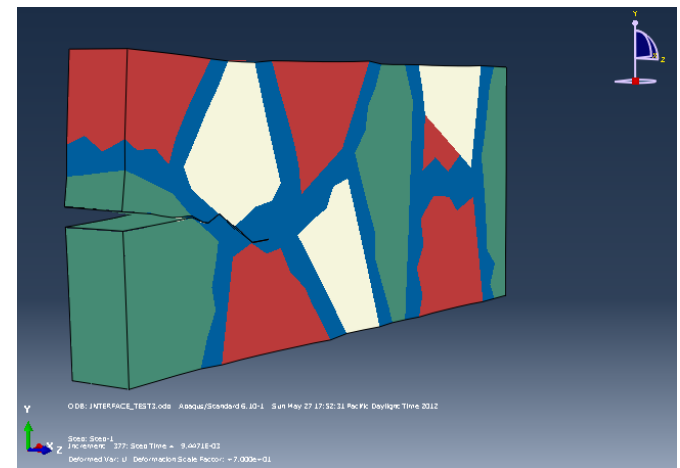
Mesh view disabled

SIMULATION

WITHOUT INTERFACE (GRAIN BOUNDARY)



Mises stress contour



Mesh view disabled

Creep and Strength Models for SiC and Si₃N₄

❖ SiC: Diffusional controlled creep. $\dot{\epsilon}_c = A\sigma^n \exp\left(-\frac{Q}{RT}\right)$
 Krause R.F. *et al.*, 1999

The constants A , Q and n are determined from a fit of the last equation to experimental data taken at 150 MPa and at temperatures ranging from 1500°C to 1600°C. $A = 6.2\text{e}6 \text{ seg}^{-1}\text{Pa}^{-1}$, $n = 1.6$, $Q = 980 \text{ KJ/mol}$, $R = 8.31447 \text{ J/mol}^\circ\text{K}$

❖ Si₃N₄: Mechanisms of creep that involve the formation of cavities. Krause R.F. *et al.*, 1999

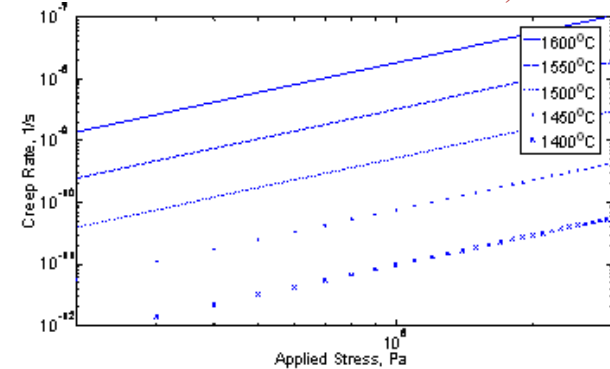
The constants A , Q , and α are determined from a fit of the last equation to experimental data taken at 1150°C, 1200°C, 1250°C, 1300°C, 1350°C and 1400°C; and at stresses ranging from 10 to 400 MPa. $A = 3.89\text{e}6 \text{ seg}^{-1}$ $\alpha = 0.0197 \pm 0.0008 \text{ MPa}^{-1}$ $Q = 715.3 \pm 22.9 \text{ KJ/mol}$ $R = 8.31447 \text{ J/mol}^\circ\text{K}$

❖ Yield function for strength (Si₃N₄ & SiC)

$$S = S_0 \left(1 + \frac{e^{-\frac{Q}{RT}}}{e_0} \right)^{\frac{1}{n}} \exp\left(\frac{Q}{RT}\right)$$

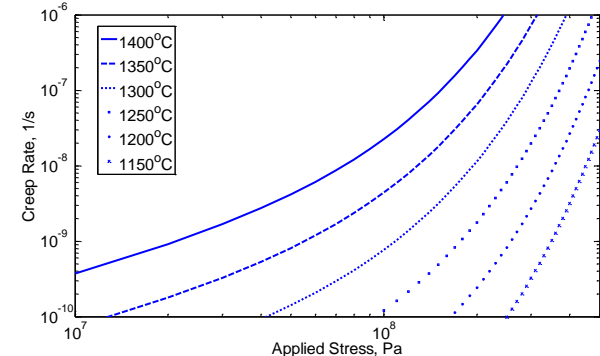
Lei *et al.*, 2000

A fit to tensile data collected on Hexology
 Wiederhorn *et al.*, 1999

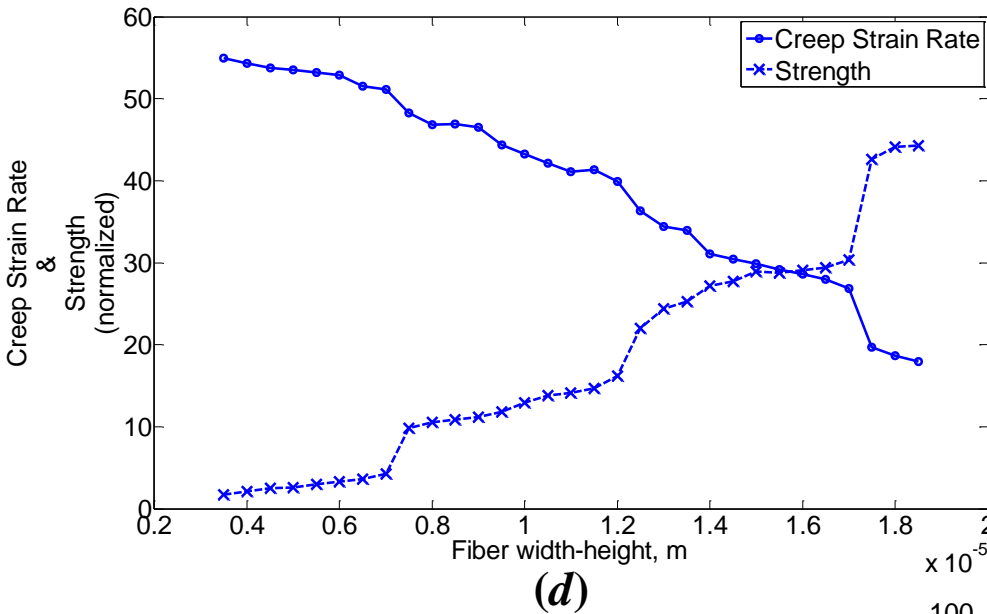


Fit of tensile data collected on SN-88
 Yoon *et al.*, 2000

$$\dot{\epsilon}_c = A\sigma \exp(\alpha\sigma) \exp\left(-\frac{Q}{RT}\right)$$

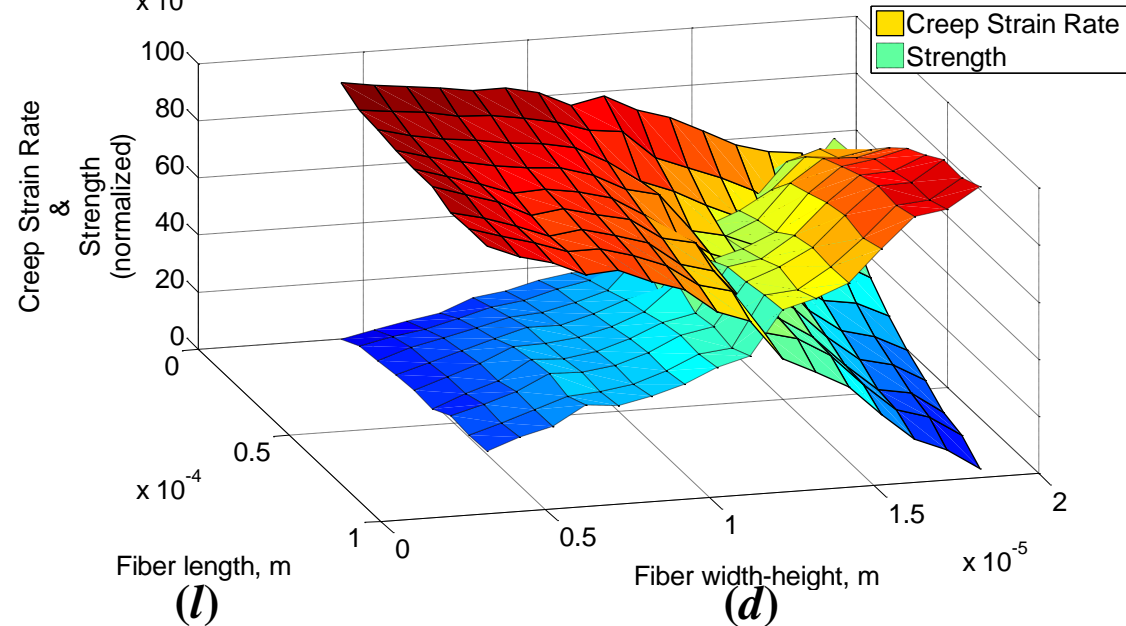


Design Variables: Strength and Creep strain



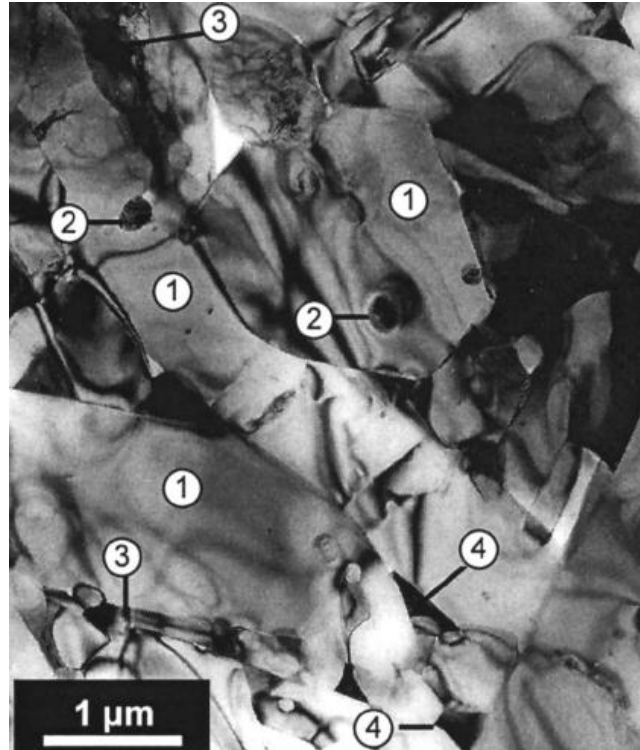
Strength and creep strain rate at 1500°C as a function of the design variable fiber width-height (d) for the 2-D low fidelity model.

Strength and creep strain rate at 1500°C as a function of the design variables fiber width-height (d) and fiber length (l) for the 3-D high fidelity model.



Molecular

1. Effect of nanoscale variations in particle sizes
2. Effect of changes in grain boundary composition, placement of particles with respect to grain boundary etc.
3. Effect of temperature, development of new constitutive models
4. BUT, length scale and time scale are out of reach. At best, it is mechanistic information



Continuum

1. Overall combined effect of different morphology factors present simultaneously or separately on the fracture resistance of a composite morphology
2. Analyses at experimentally accessible loading rates
3. BUT, there is a lack of mechanistic information

Experiments

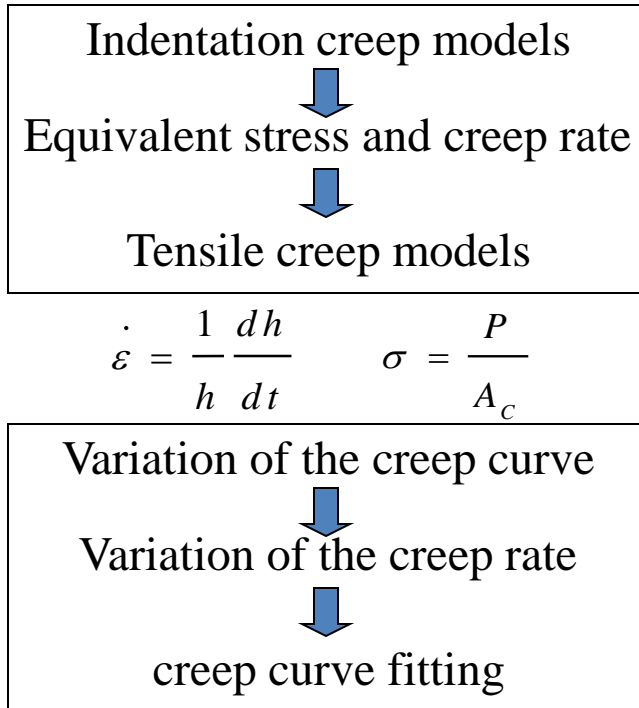
Bridging using experiments

Introduction to Indentation Creep

➤ Aspects

- ❖ Less stringent requirements for specimens
- ❖ Thin films
- ❖ High-melting-point material at room temperature

➤ Indentation Creep Models



$$h(t) = h_i + a(t - t_i)^b + kt$$

• Developed by Li and Ngan in 2004 for Ni₃Al, Al, and fused quartz at room temperature.

• Used by Ma et al. (2008), and Cao et al. (2009).

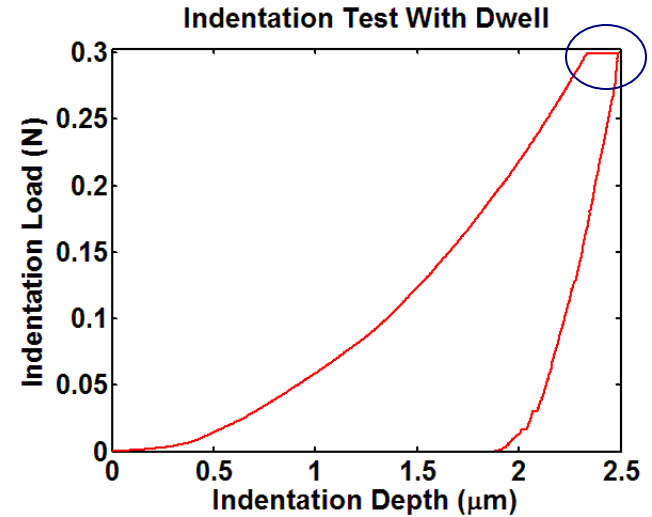


Fig. 1 Combined indentation test and creep test

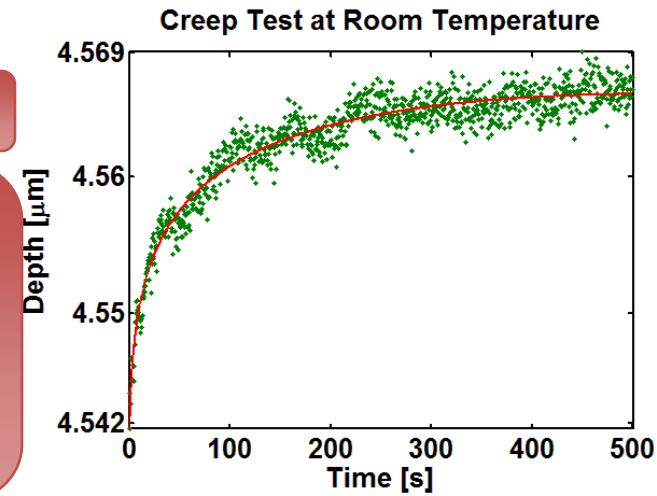
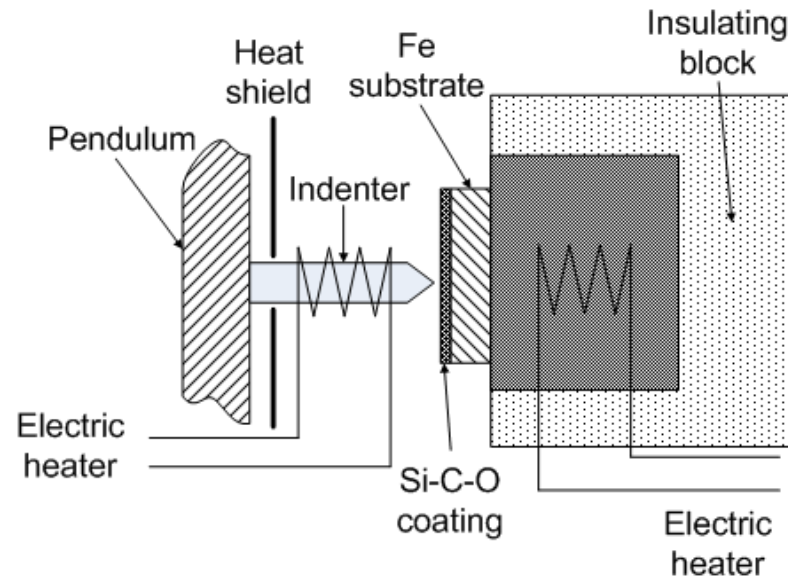
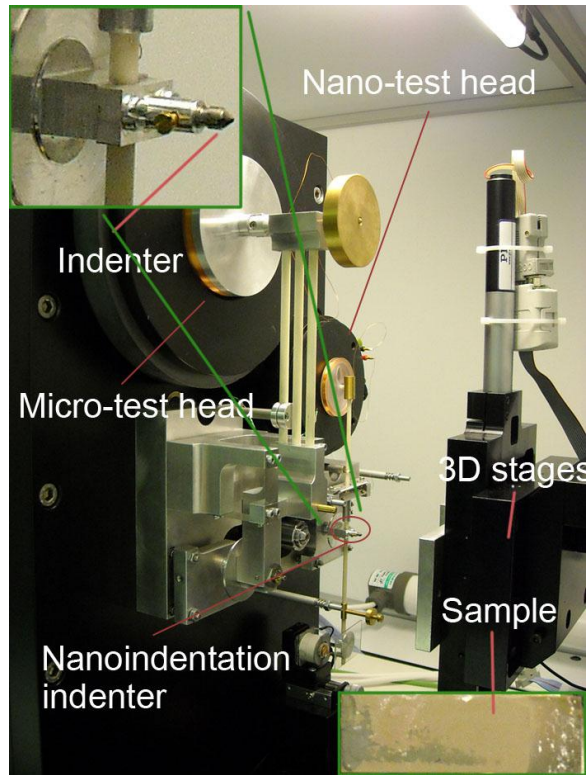


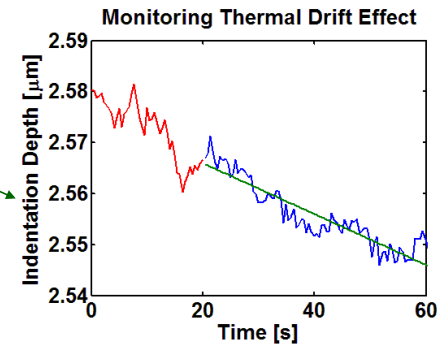
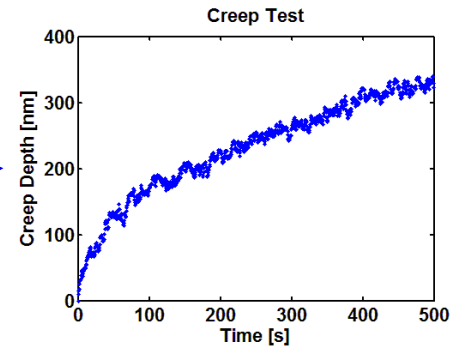
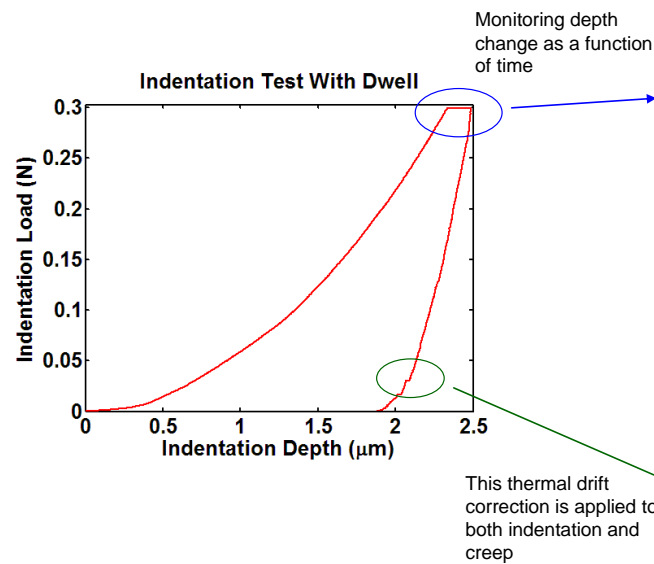
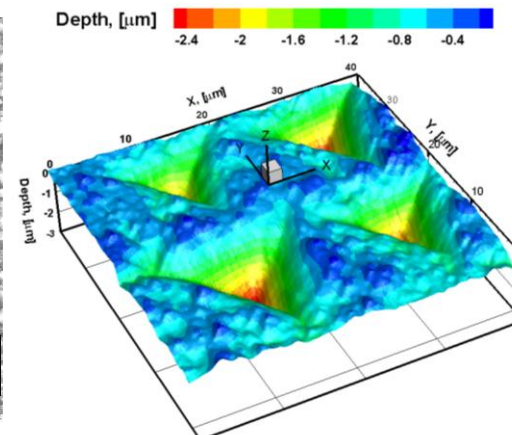
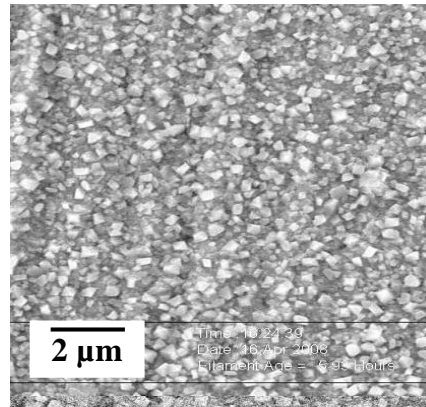
Fig. 2 Curve fitting for indentation creep

Setup

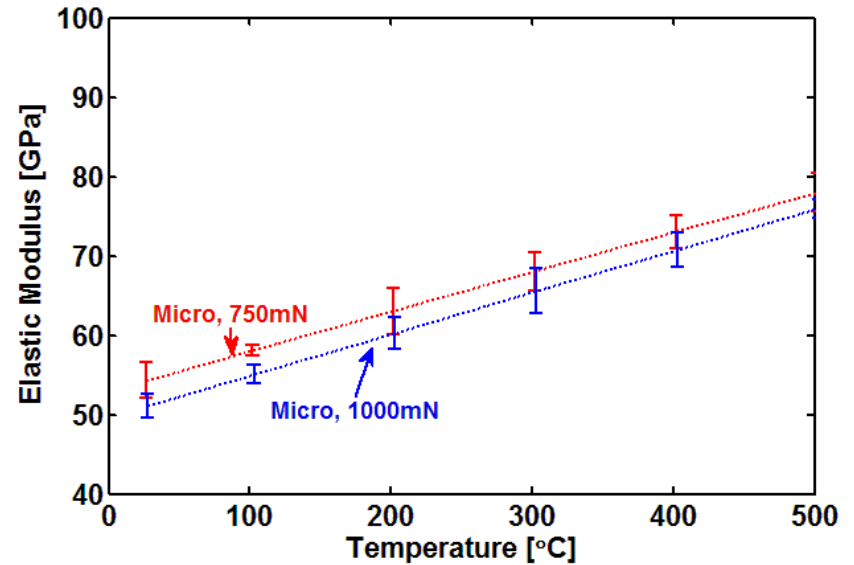
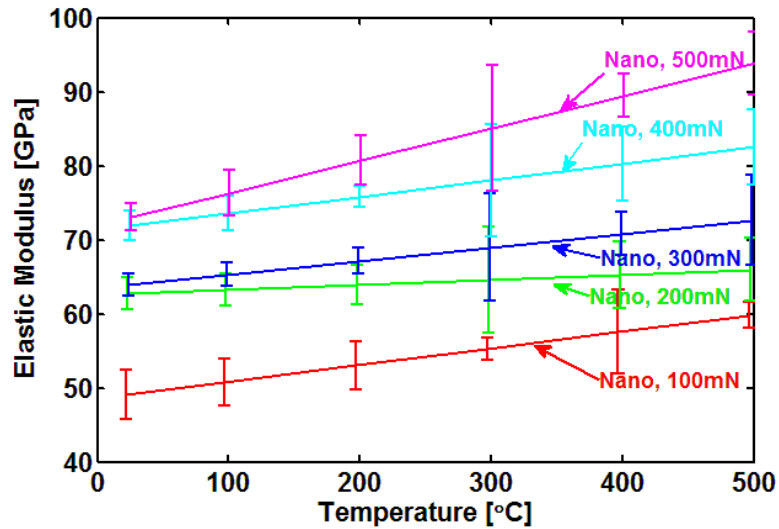


Gan, M, and Tomar, V., 2010, Role of length scale and temperature in indentation induced creep behavior of polymer derived Si-C-O ceramics, *Materials Science and Engineering-A*, vol. 527 pp 7615–7623

Sample Description and Testing Approach

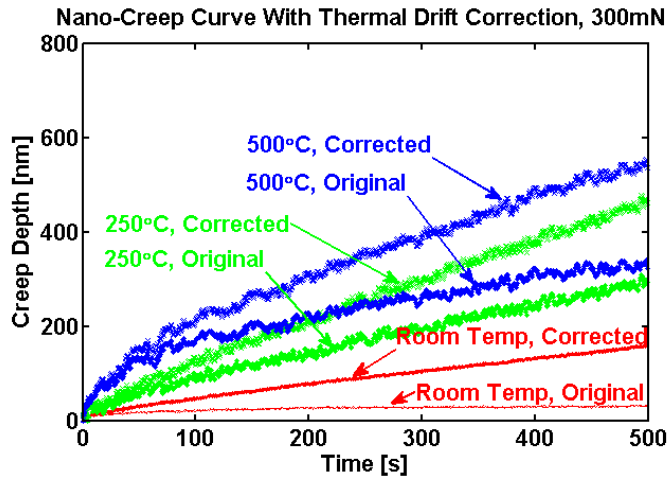


Elastic Moduli

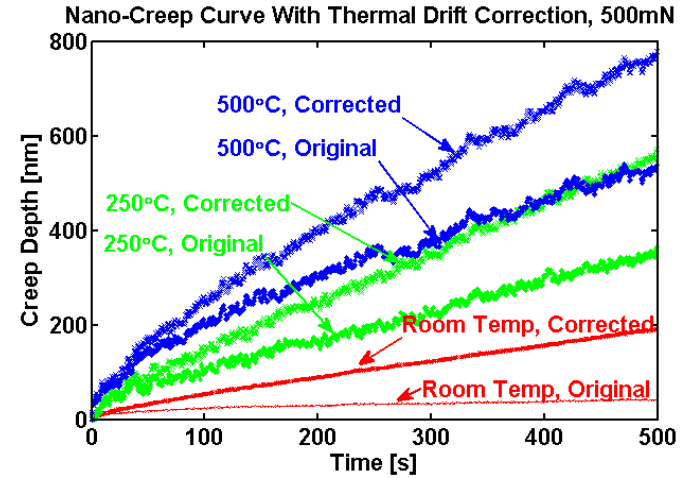


Analyses take into account creep recovery and softening at the end of dwell period in calculating the data based on Li and Ngan Creep Factor Relations, 2002

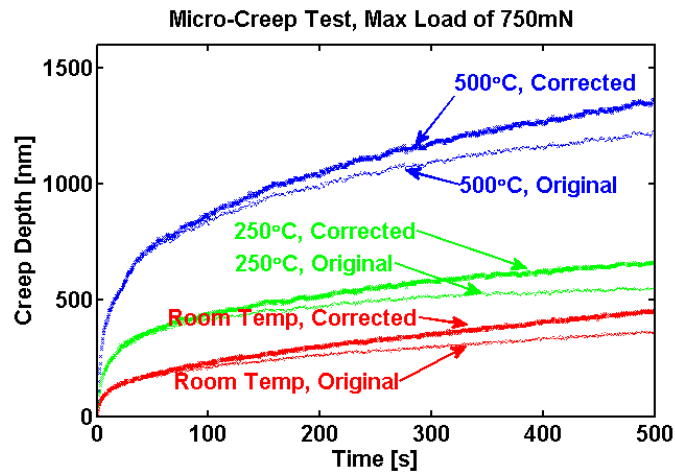
Creep Curves



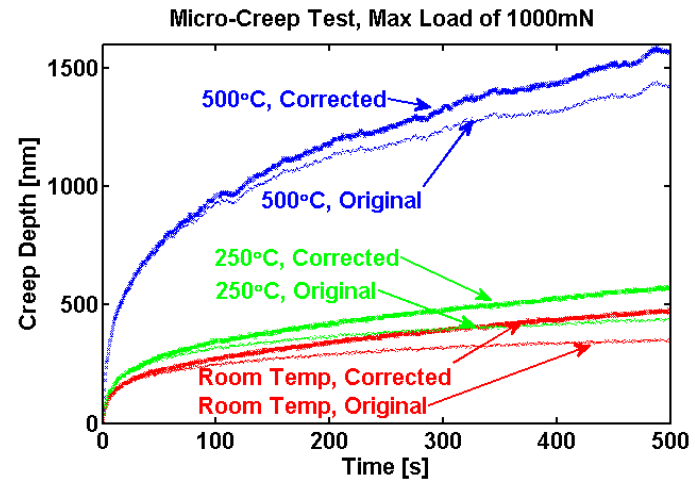
(a)



(b)

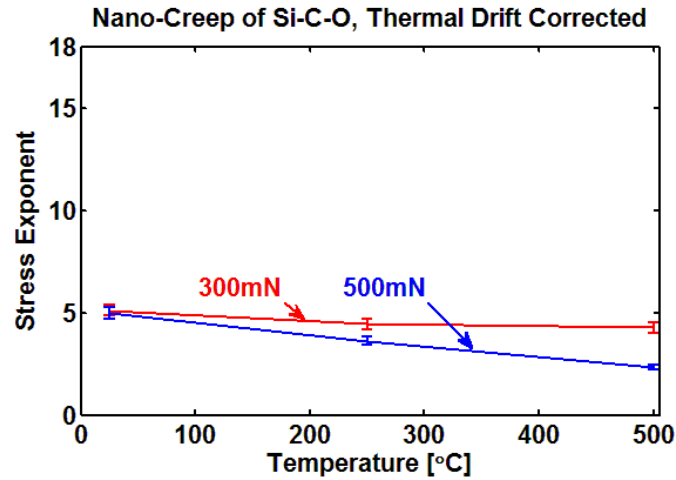


(c)

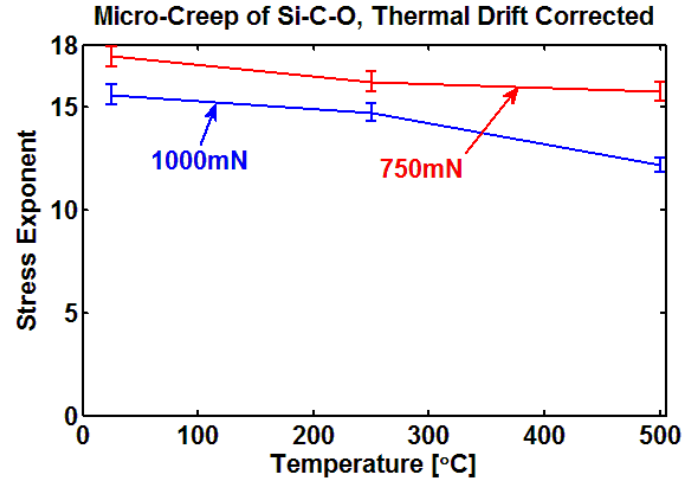


(d)

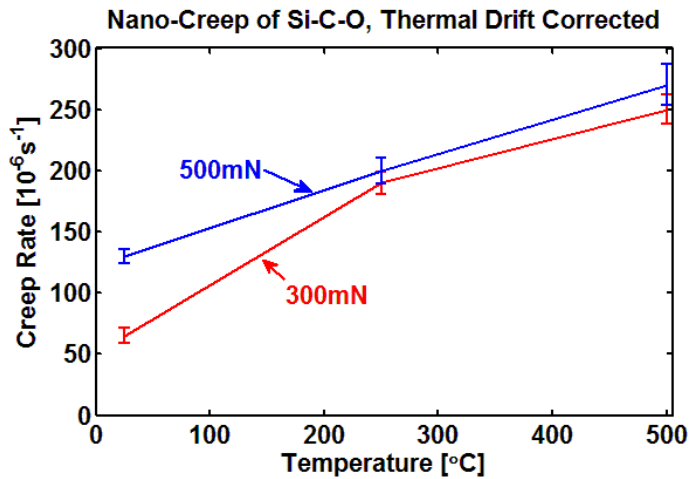
Exponent and Creep Rate



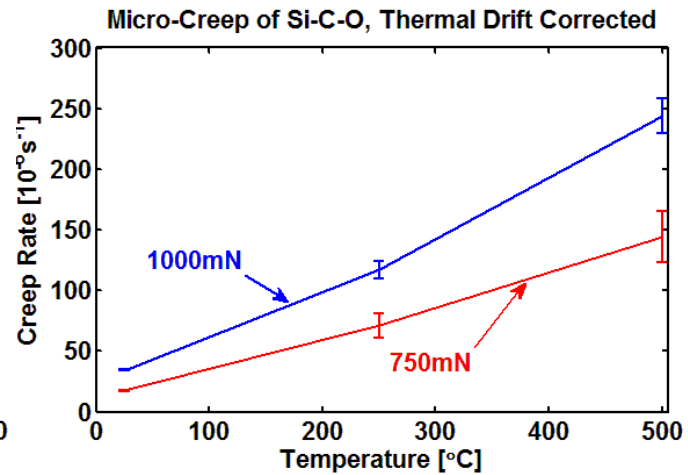
(a)



(b)

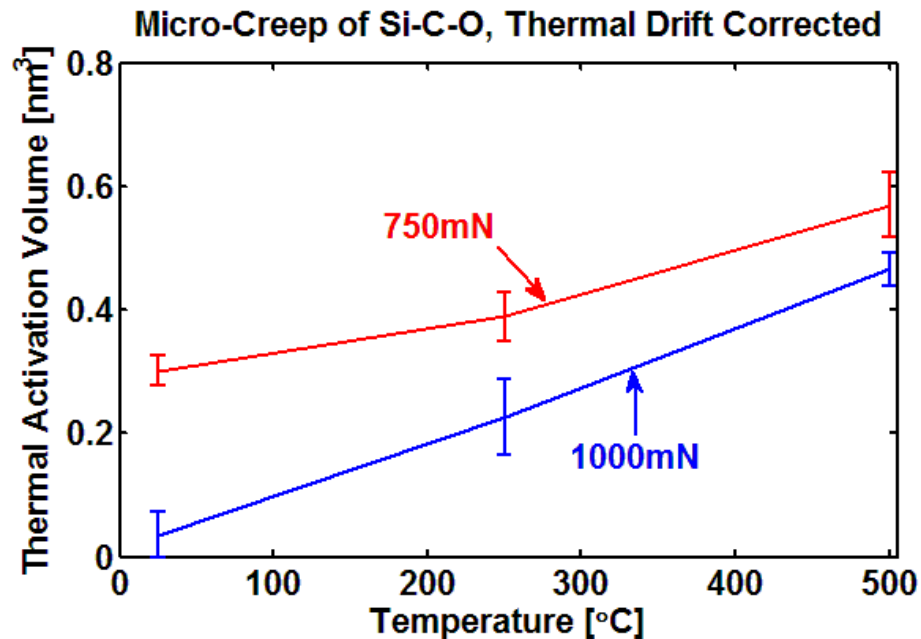
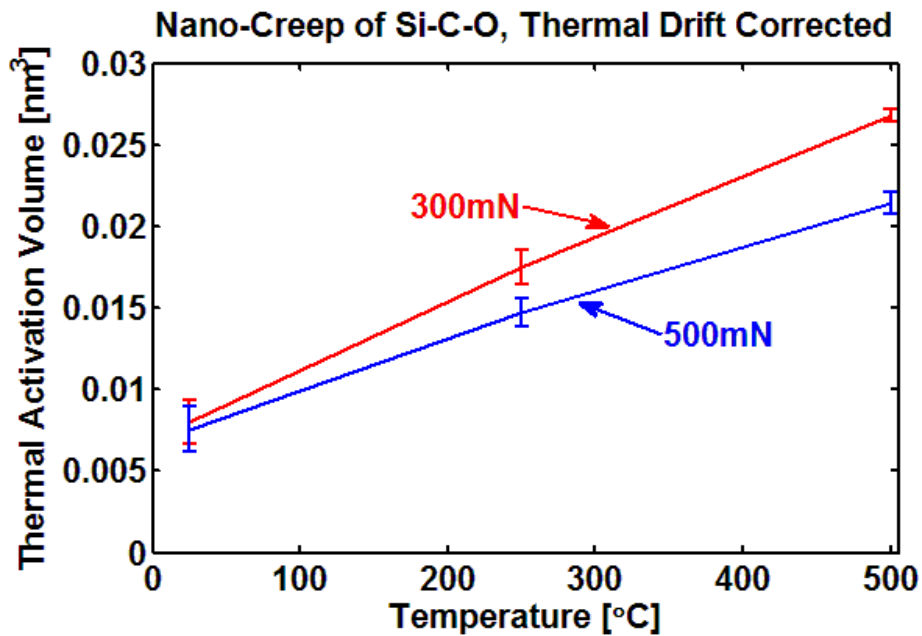


(c)

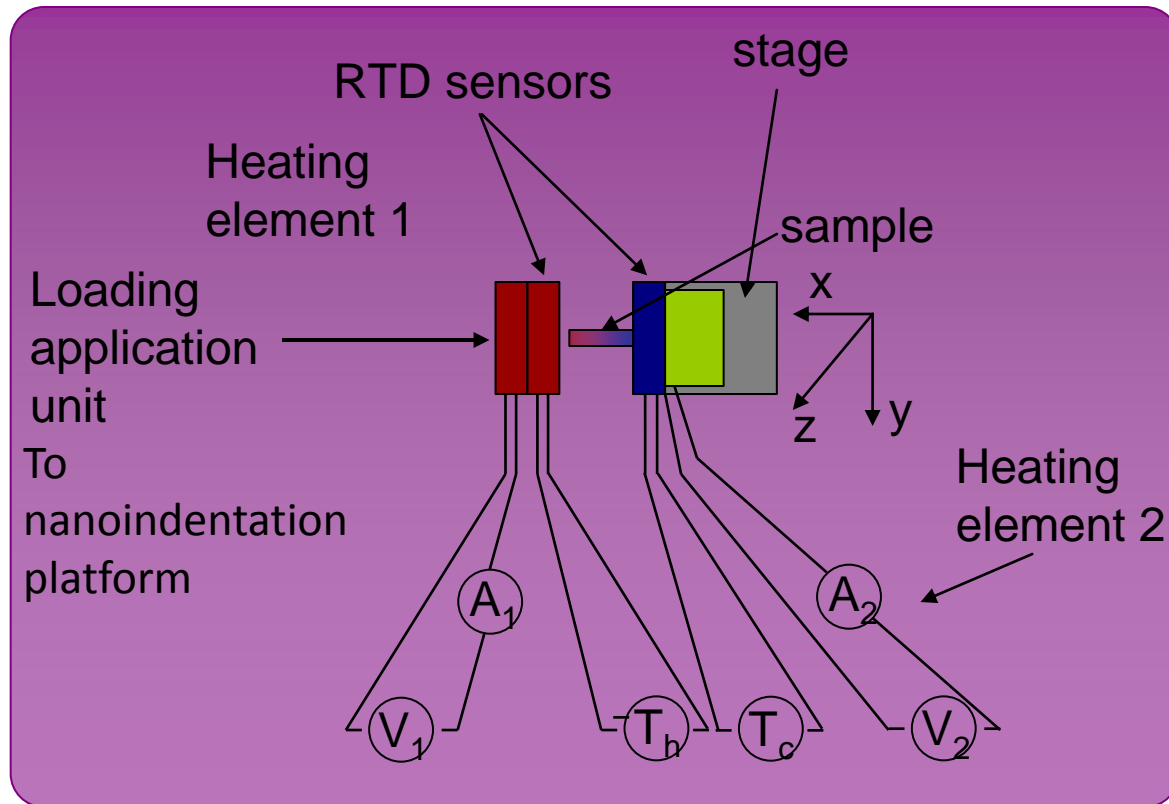


(d)

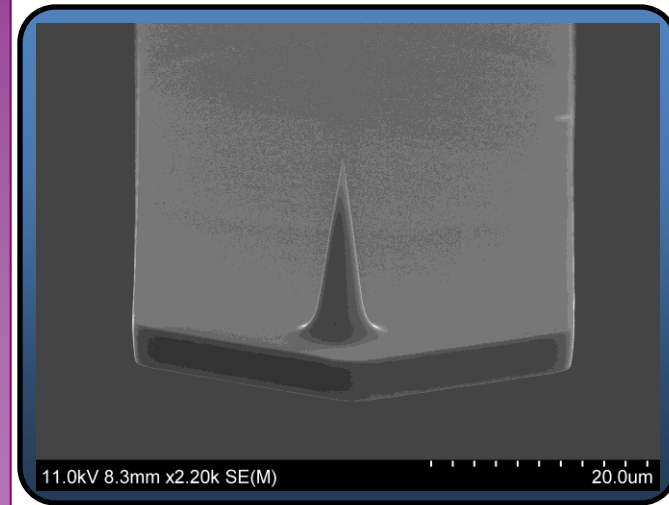
Thermal Activation Volume



Experimental Setup



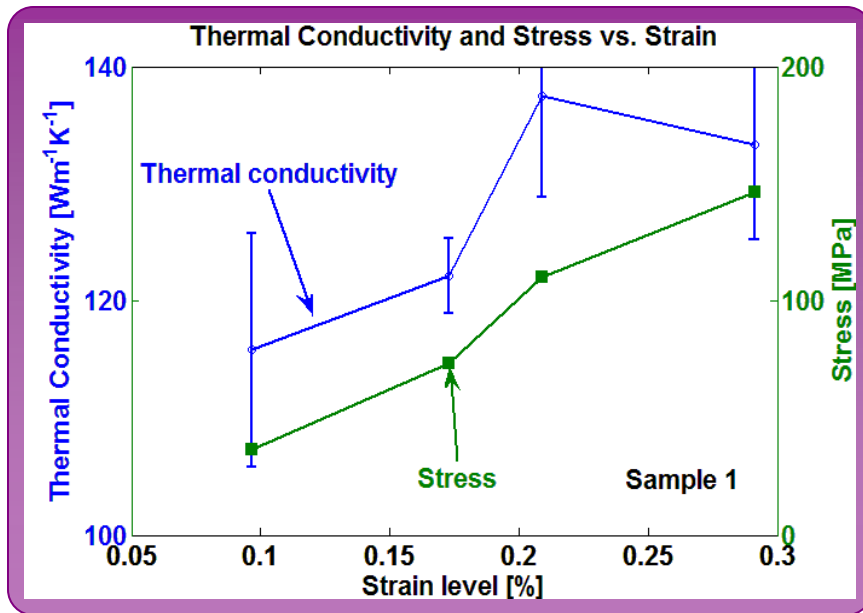
Experimental setup



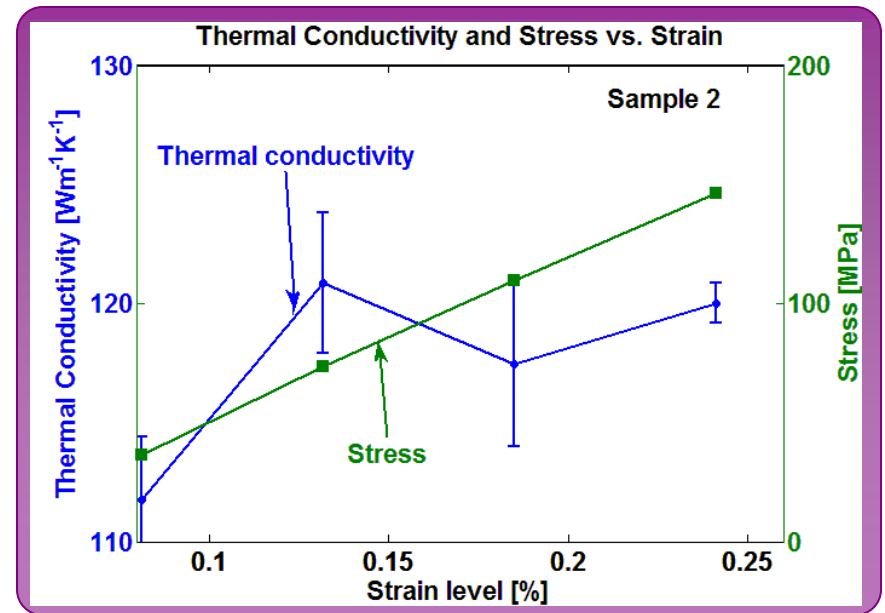
AFM cantilever

- **Heater & temperature sensor: resistance temperature detector Pt100 – most accurate temperature recorder 0°C ~200°C**
- **Sample: atomic force microscope (AFM) cantilever**
- **Provisional Patent 65680-PRF, Gan and Tomar, JEMT-2011**

Brief Results



(a)



(b)

Thermal conductivity as a function of strain level for
(a) sample set 1 and (b) sample set 2

- The trend matches simulation results in literature, Klimeck et al.
- Bulk Si 156W/(m·K) at 300K, 105W/(m·K) at 400K
- Our measurement temperature range 300K ~ 330K

Prof. Vikas Tomar Contributions as a Sub-awardee

Total Project Sub-Contract Cost: \$159,000 including subcontract (10/01/2009-09/29/2012)
The project cost at Purdue was \$53,000/year which supported one student partially including provision for supplies, Dr. Tomar's salary, and travel to review meetings and conferences.

NO-COST-EXTENSION was not accepted at Purdue.

Achievements: (9 International Journal Articles, 8 invited talks, 2 conference proceeding articles, and multiple conference presentations)

- A multi-scale material design approach that encompasses computations and experiments in a simple framework has been established.
- Computational methods to analyze fracture in experimental SiC-Si₃N₄ morphologies have pointed out important phase based fracture mechanics relations that can be used to guide experiments and design models for improvement in the composite properties.
- Computational fracture analyses are ably supported by computational molecular simulation codes to understand mechanisms behind failure.

Collaborative Work: (3 International Journal Articles, 2 invited talks, 4 conference proceeding articles, and multiple conference presentations)

- An experimental work regarding SiCO composites was published in Mat Sci Eng. A and JOM. The samples were supplied by Dr. Raj Bordia
- Collaborative modeling work has been published in Engineering Optimization in 2010 and 2012.
- Collaborative talks are underway with Dr. Andres Tovar on how material models developed can be incorporated into his design models. Correspondingly a paper is under preparation to be submitted shortly.

Student Supported:

Hongsuk Lee (graduate student: completing PhD Fall 2011)

Vikas Samvedi (graduate student: partially in 2009-10)

Special Recognition

One of the papers co- published by supported student and Dr. Tomar was top 10 most downloaded paper at ASME in 2011-2012.

International Journal Publications:

The following international journal publications acknowledge full or partial support of the work.

1. Mejia-Rodriguez, G., Kim, H., Renaud, J.E., and Tomar V., 2012, "Sequential Approximate Optimization Based Robust Design of SiC-Si₃N₄ Nanocomposite

- Microstructures", Accepted for publication in *Engineering Optimization*, manuscript ID GENO-2011-0188. (Project Contribution of time by Dr. Tomar)
2. Lee, H., and **Tomar, V.**, 2011, Effect of Meso to Micro Transition in Morphology Dependent Fracture of SiC Ceramics, to appear in *Journal of Engineering Materials and Technology-ASME Transactions*. (Project Contribution of time by Dr. Tomar and student supported Hong Suk Lee)
3. Gan, M., and **Tomar, V.**, 2011, Scale and Temperature Dependent Creep Modeling and Experiments in Materials, *JOM-Special Issue on Combining Multiscale Models and Experiments*, September 2011. (Project Contribution of time by Dr. Tomar)
4. Gan M. and **Tomar, V.**, 2011, "Combining experimental and multiscale computational perspectives in complex ceramics mechanical behavior description", *JOM*, September 2011, ed Vikas Tomar. (Project Contribution of time by Dr. Tomar)
5. **Tomar, V.** Special Issue Guest Editor: "Issues in Combining Multiscale Modeling and Experiments", *JOM* (flagship publication of TMS), September 2011. (Project Contribution of time by Dr. Tomar)
6. Gan, M., and **Tomar, V.**, 2010, Role of length scale and temperature in indentation induced creep behavior of polymer derived SiCO ceramics, *Materials Science and Engineering-A*, vol. 527 pp 7615–7623, DOI: 10.1016/j.msea.2010.08.016 (Project Contribution of time by Dr. Tomar)
7. **Tomar, V.**, and Samvedi, V., 2010, Correlation of thermal conduction properties with mechanical deformation characteristics of a set of SiC-Si₃N₄ nanocomposites, to Appear in *Journal of Engineering Materials and Technology* (Accepted June 02, 2010). (Project Contribution of time by Dr. Tomar and Vikas Samvedi)
8. **Tomar, V.** and Gan, M., 2010, Temperature dependent nanomechanics of Si-C-N nanocomposites with an account of particle clustering and grain boundaries, in press in the *Int. J. Hydrogen Energy*, doi:10.1016/j.ijhydene.2010.03.070. (Project Contribution of time by Dr. Tomar)
9. Mejia-Rodriguez, G., Renaud, J.E., and **Tomar V.**, 2010, Multiobjective composite material design using the variable fidelity model management optimization framework, *Engineering Optimization*, Vol 42, No. 11, , pp. 1055-1078. (Project Contribution of time by Dr. Tomar)
10. **Tomar, V.**, Samvedi, V., and Kim, H., 2010, Atomistic understanding of the particle clustering and particle size effect on the room temperature strength of SiC-Si₃N₄ nanocomposites to Appear in *Int. J. Multiscale Comp. Engg.* special issue on Advances In Computational Materials Science, Volume 8 issue 4, 2010. (Project Contribution of time by Dr. Tomar and Vikas Samvedi)
11. **Tomar, V.**, Gan, M., and Kim, H., 2010, Effect of temperature and morphology on mechanical strength of SiCO and SiCN nanocomposites, the *Journal of European Ceramic Society*, Volume 30, 2223-2237. (Project Contribution of time by Dr. Tomar)

Besides the above mentioned publications multiple conference presentations (20), 4 conference proceedings, and 10 invited talks in universities and international conferences presented the supported work.

International J. Manuscripts *In Preparation*:

12. Lee, H., and Tomar, V., 2012, Understanding temperature dependent fracture resistance of polycrystalline SiC in 3-D with an account of grain boundary properties, *In Preparation*. (Project Contribution of time by Dr. Tomar and student supported Hongsuk Lee)
13. K. Khadke, V. Tomar, A. Tovar, "Improved nanocomposite properties for high temperature application using multiobjective and variable fidelity optimization", *In preparation* for J. computer aided material design. (Project Contribution of time by Dr. Tomar)