Computer Aided Multiscale Design of SiC-Si_3N_4 Nanoceramic Composites for High-Temperature Structural Applications

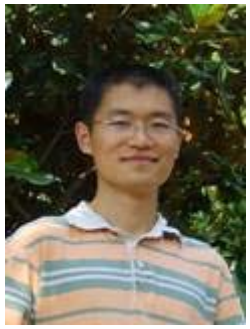
PIs: Vikas Tomar and John E Renaud

Aerospace and Mechanical Engineering
University of Notre Dame, Notre Dame-Indiana, USA, 46556

Program Manager: Dr. Patricia Rawls

DoE-NETL UCR Grant DE-FG26-07NT43072

Students



Gilberto Meja-Rodriguez, Devendra Dubey, Ming Gan, Vikas Samvedi

Collaborators

John Renaud (Material Design), Raj Bordia (Material Processing and Manufacturing)

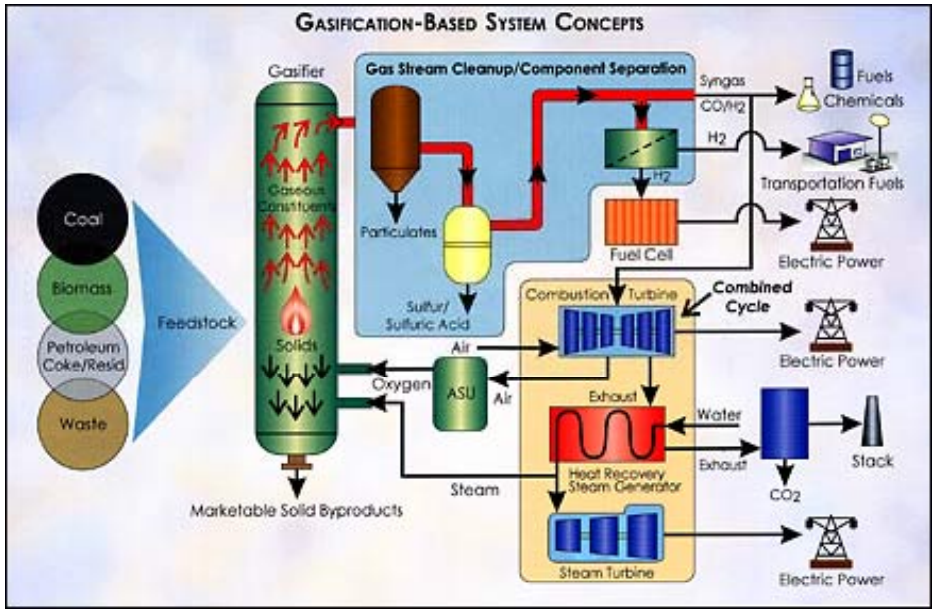
Lab Facilities

52 Processor Opteron dual core supercomputing cluster, 8 Terabytes storage server, Veeco AFM, MTS Instron mechanical hydraulic testing machine, Hopkinson bar for dynamic high strain rate testing

Clean Coal Utilization

- **The Energy Department's Office of Fossil Energy is working on coal gasifier advances that enhance efficiency, environmental performance, and reliability as well as expand the gasifier's flexibility to process a variety of coals and other feedstocks (including biomass and municipal/industrial wastes).**

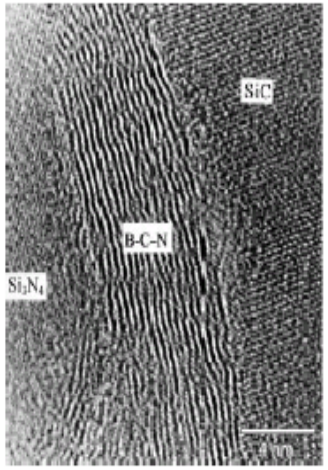
- **Coal gasification is one of the most promising technologies for energy plants of the future.**
  - **Need materials for high temperature applications.**
    - **Oxidation, corrosion, creep and fracture resistance.**
  - **$SiC-Si_3N_4$ composites.**



Outline

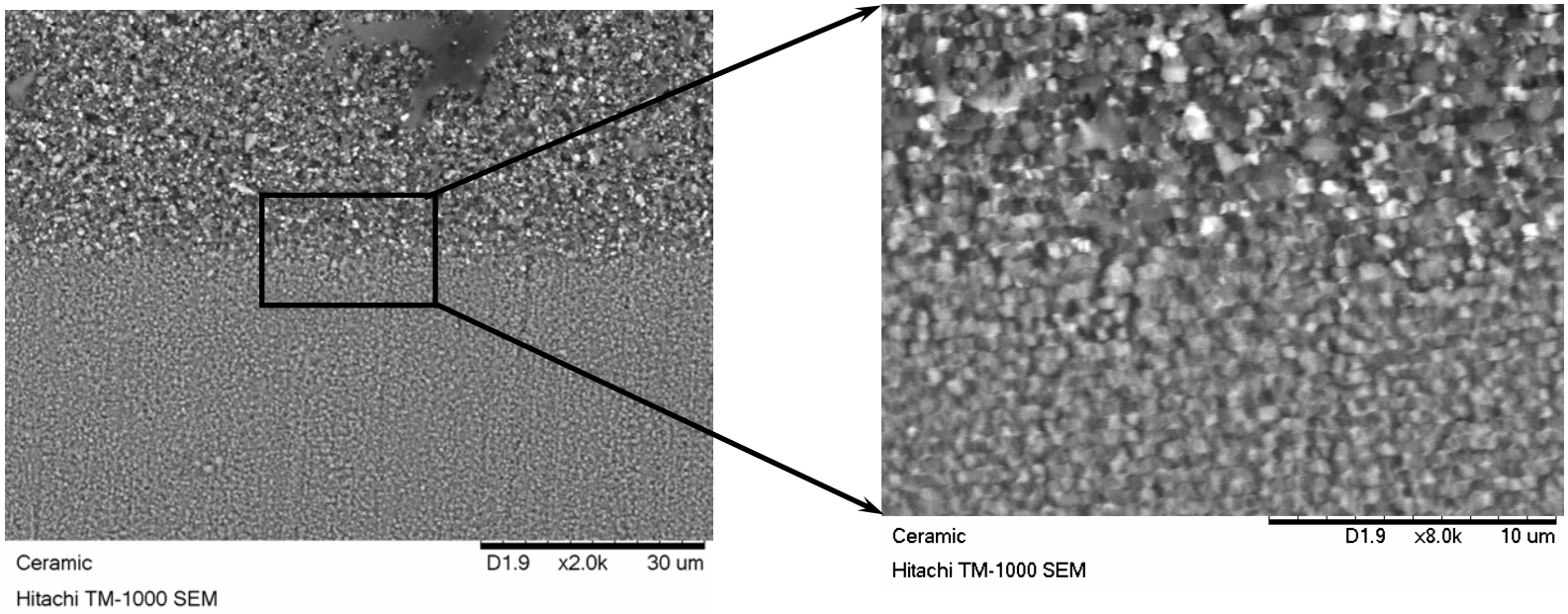
- **High temperature creep and nanoindentation experiments in high temperature composites**
  - **Scale dependent nanoindentation upto 773 K**
  - **Nanoscale creep at different strain rates and maximum load**
- **Cohesive finite element based modeling of dynamic fracture failure in SiC-Si_3N_4 nanocomposites**
  - **Cohesive analyses accounting for second phase particle distribution and grain boundary distribution effects**
  - **High temperature constitutive models and trade-offs in designing these materials for high temperature performance**
- **Insights from molecular analyses**
  - **Nanoscale fracture and strength at room temperature**
  - **Role of temperature in strength upto 1500 K**
- **Designing materials in an optimization framework**

Nanocomposite Material System

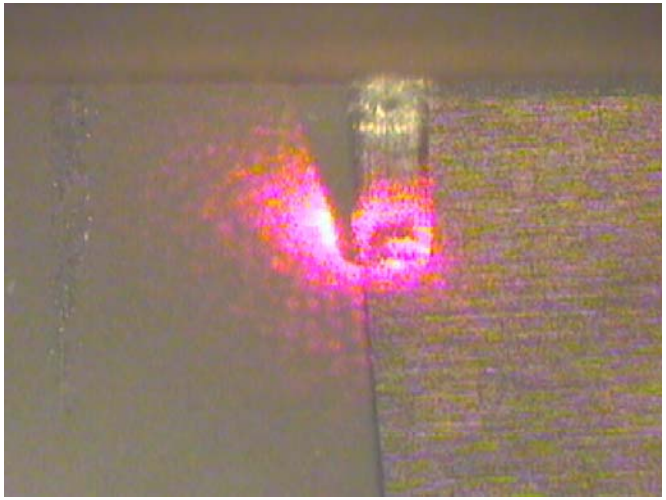


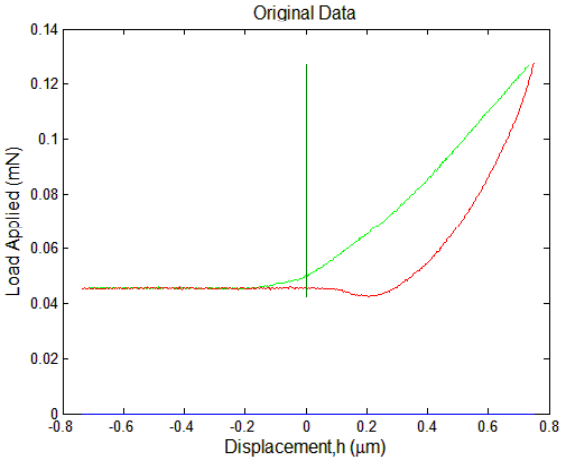
- Grain boundary thickness of the order of 50 nm
- SiC particle size of the order of 200-300 nm
- Si_3N_4 grain size of the order of 0.8 to 1.5 µm
- Spherical SiC particles distributed in Si_3N_4 matrix in the form of intergranular as well as intra-granular dispersions
- SiC volume fraction varies between 10% and 50%

Experimental Investigations

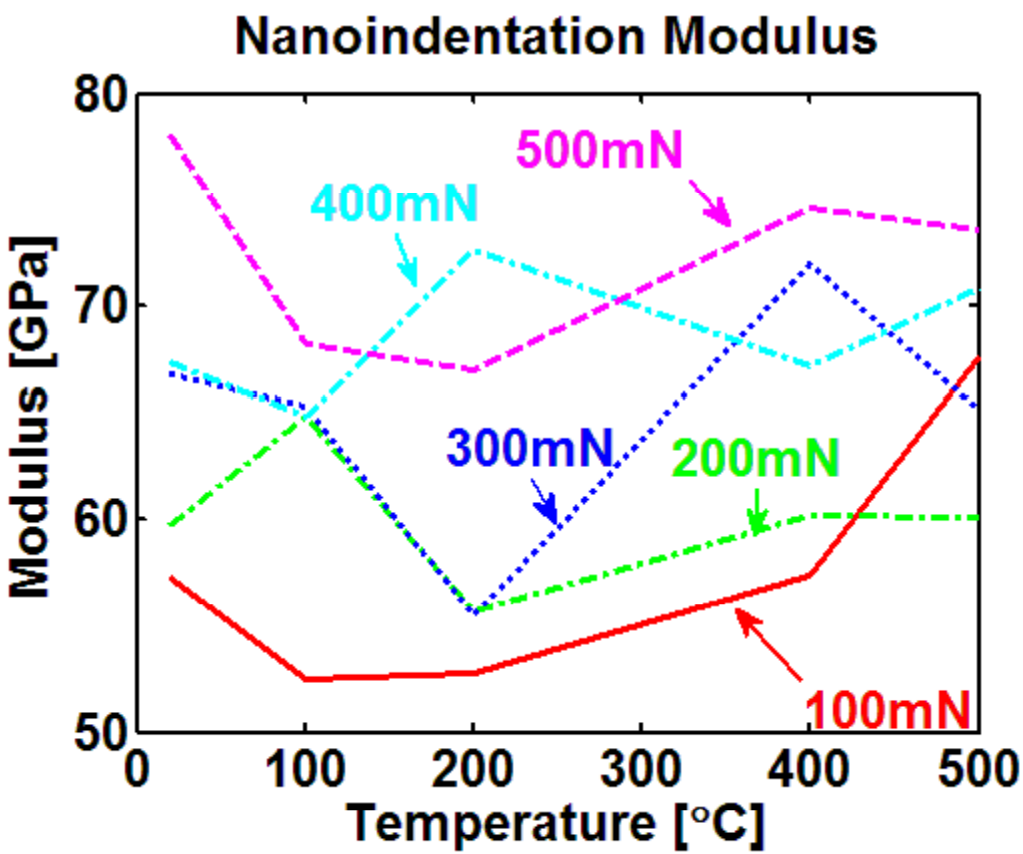


Nanoindentation



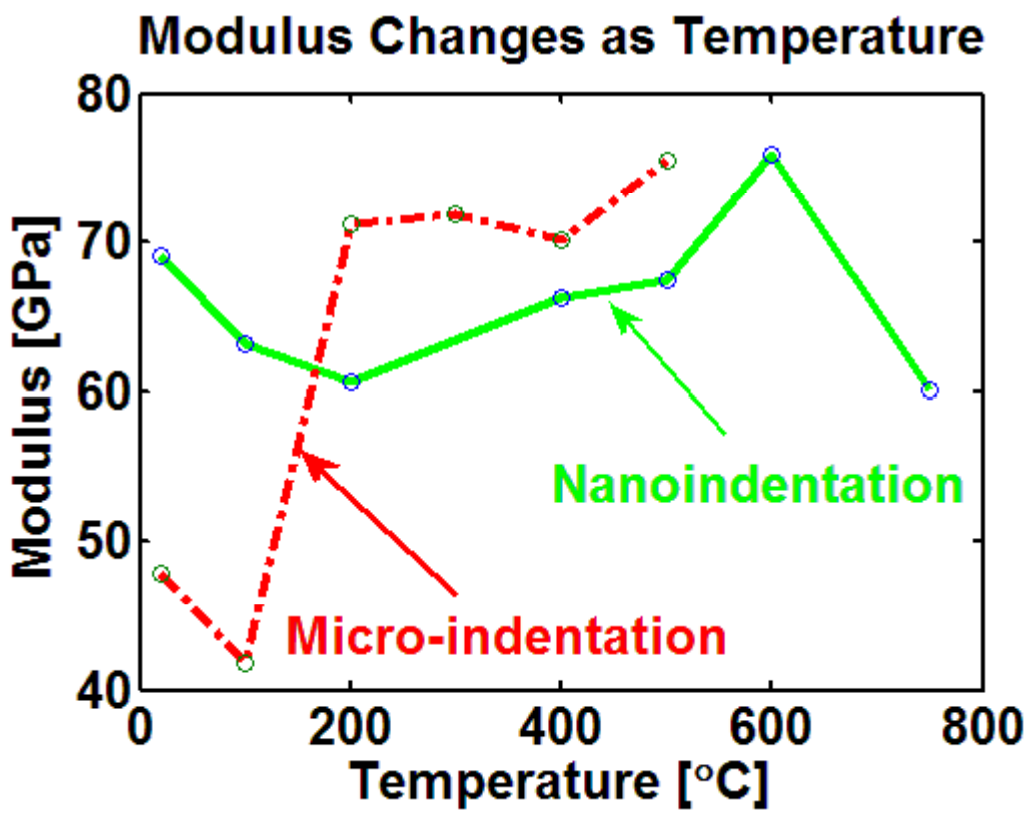


Elastic Modulus From Nanoindentation



he elastic modulus of ceramic coating varies as temperature and maximum dentation load. The max load varies from 100mN to 500mN, with 100mN crement. The testing temperature varies from 100℃ to 500℃, with 100 ℃ crement. Room temperature is also included.

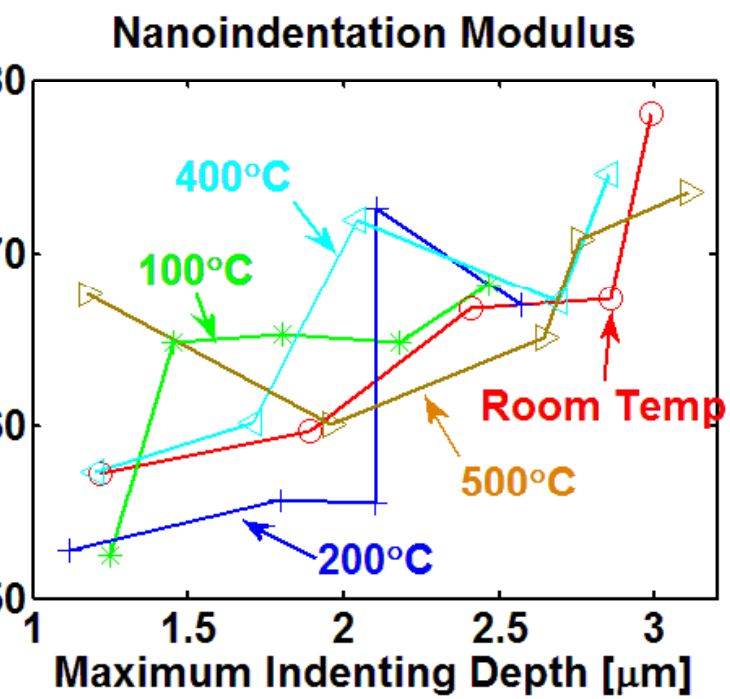
Elastic Modulus Changes as Temperature

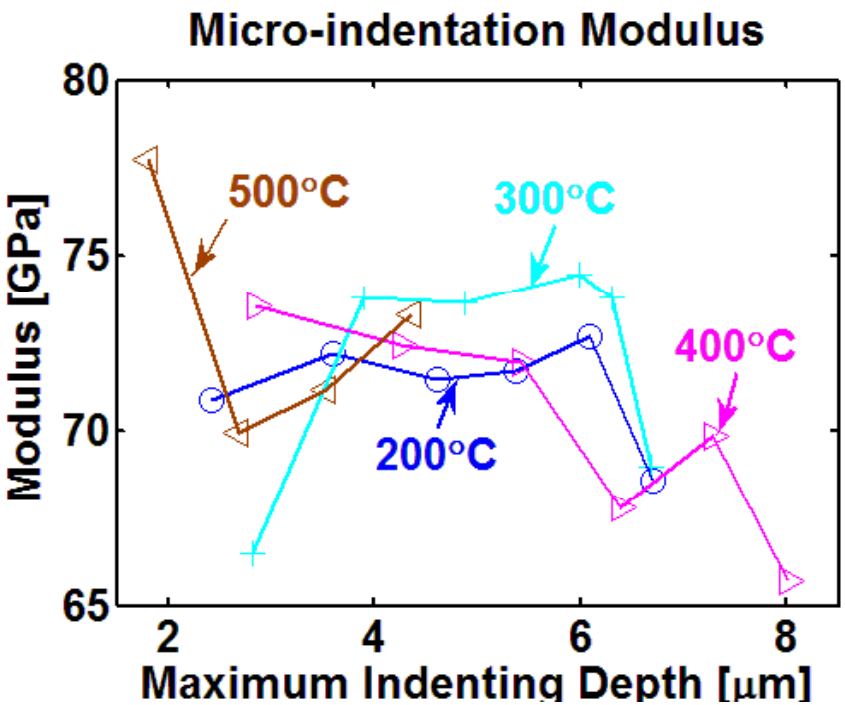


For the micro-indentation, we did not change the max load. It was set to 1N.

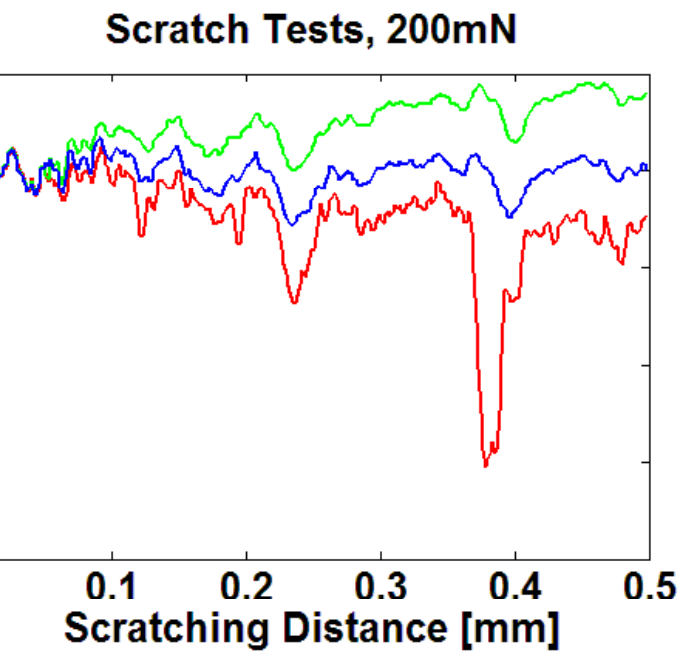
e elastic modulus of ceramic coating varies as temperature. Data are collected om both nanoindentation and micro-indentation. The testing temperature varies om 100℃ to 500℃, with 100℃ increment. Room temperature is also included for th tests. Tests at 600℃ and 750℃ were conducted for nanoindentation only.

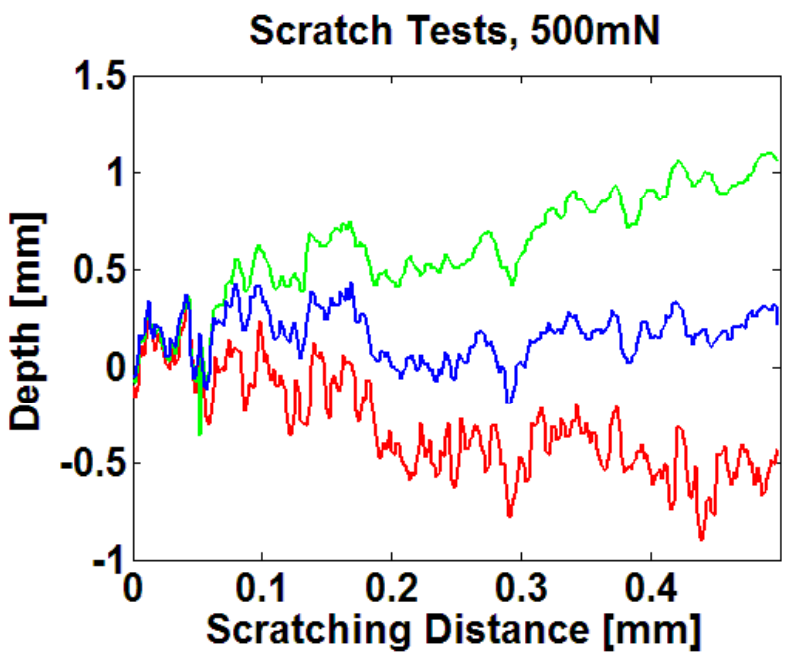
Modulus from Nanoindentation

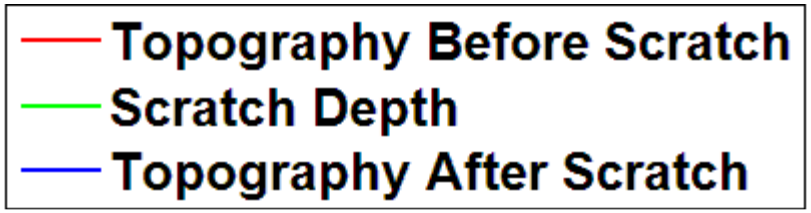




Nano-scratch Tests







Creep Models

rom classical mechanisms of creep deformation, creep is a onsequence of dislocation motion or diffusion.

$$\dot{\varepsilon}_s = \dot{\varepsilon}_0 \left(\frac{\sigma}{\sigma_0} \right)^n \left(\frac{D}{D_0} \right)^m \exp\left(-\frac{Q}{RT} \right)$$

islocation controlled creep: *n*= 3 to 5, *m*=0
iffusional controlled creep: *n* ≈1,*m*= -3 to -2

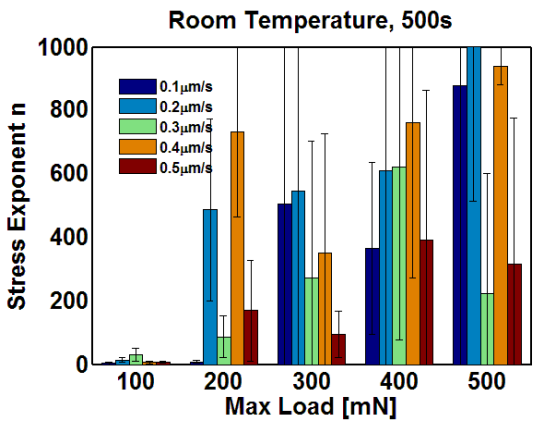
$$\dot{\varepsilon}_c = A\sigma^n \exp\left(-\frac{Q}{RT} \right) \qquad \dot{\varepsilon}_s = \frac{\dot{\varepsilon}_0}{\eta_0} \frac{V_f^3}{\left(1 - V_f\right)^2} \sigma \exp(\alpha\sigma) \exp\left(-\frac{Q}{RT} \right)$$

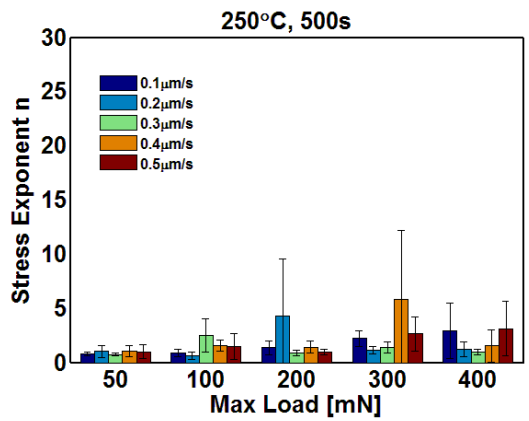
diffusion **Cavities**

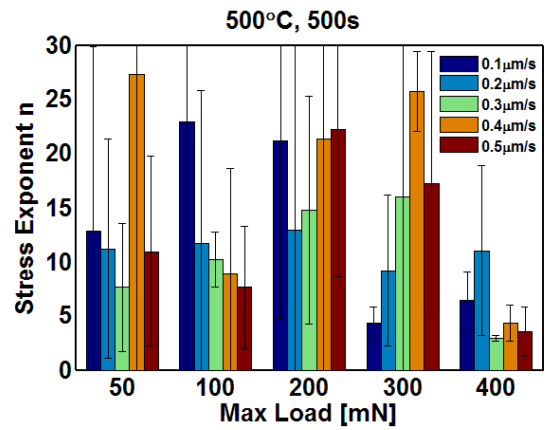
Stress Exponent Changes as Maximum Load and Loading Rate

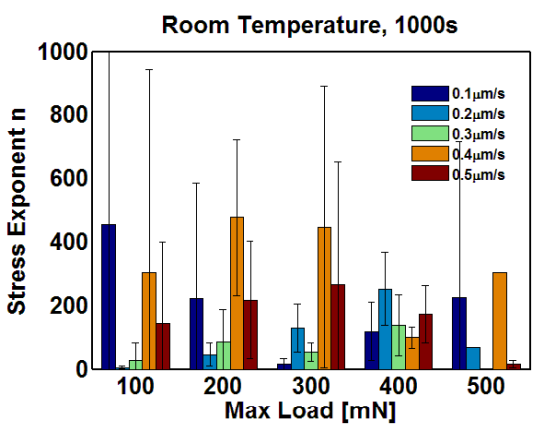
ng
e

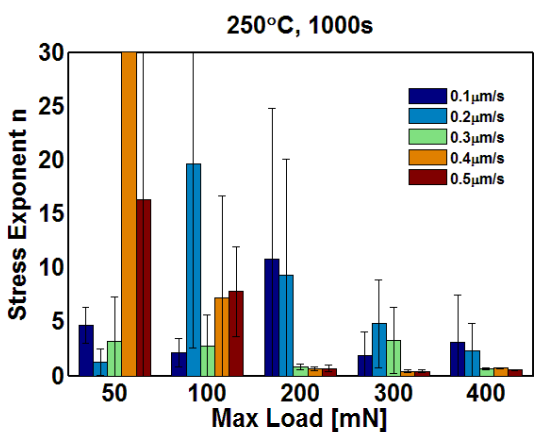
Room Temperature | 250°C | 500°C

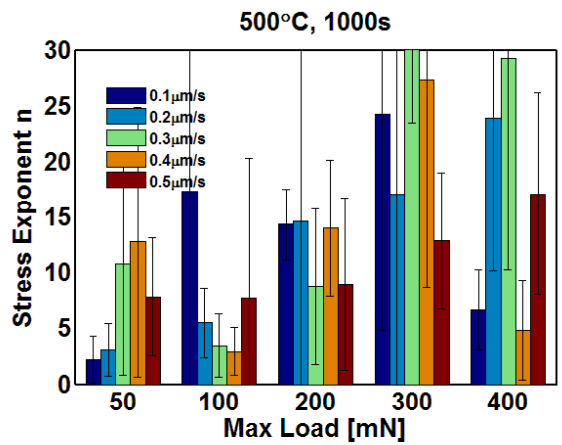












Creep Rate Changes as Maximum Load and Loading Rate

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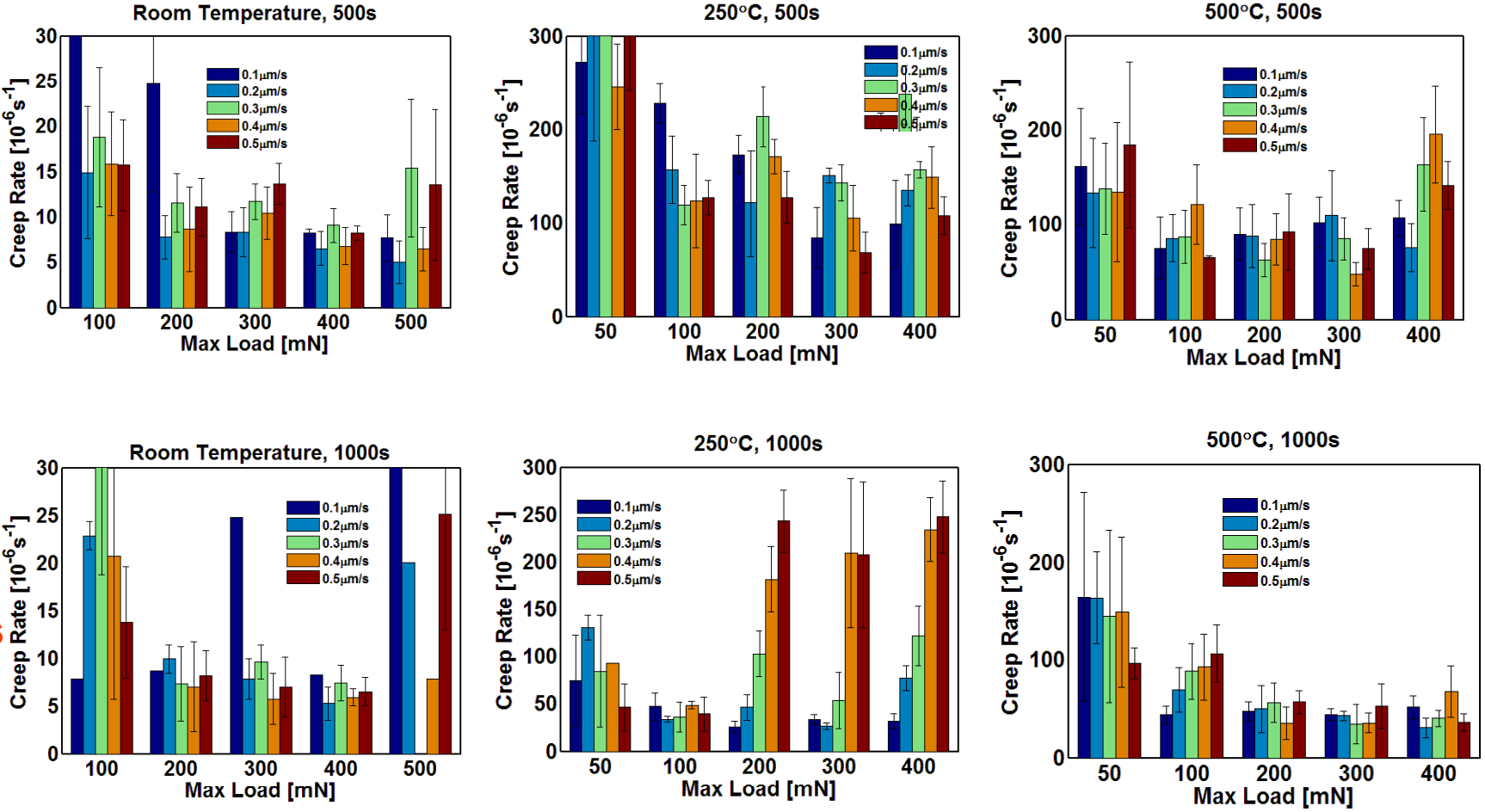
Room Temperature **250°C** **500°C**



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**hesive finite element based modeling of dynamic fracture failure in SiC-
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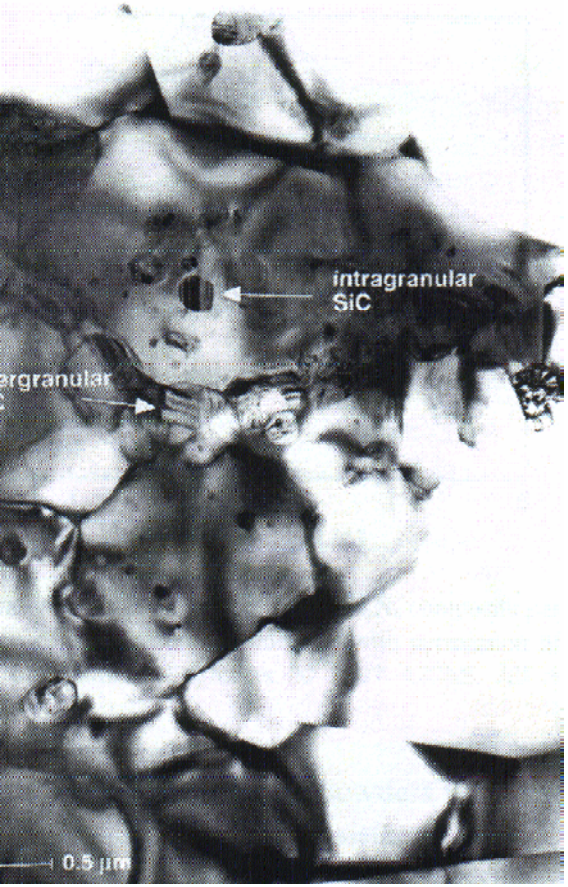
sights from molecular analyses

- Nanoscale fracture and strength at room temperature
- Role of temperature in strength upto 1500 K

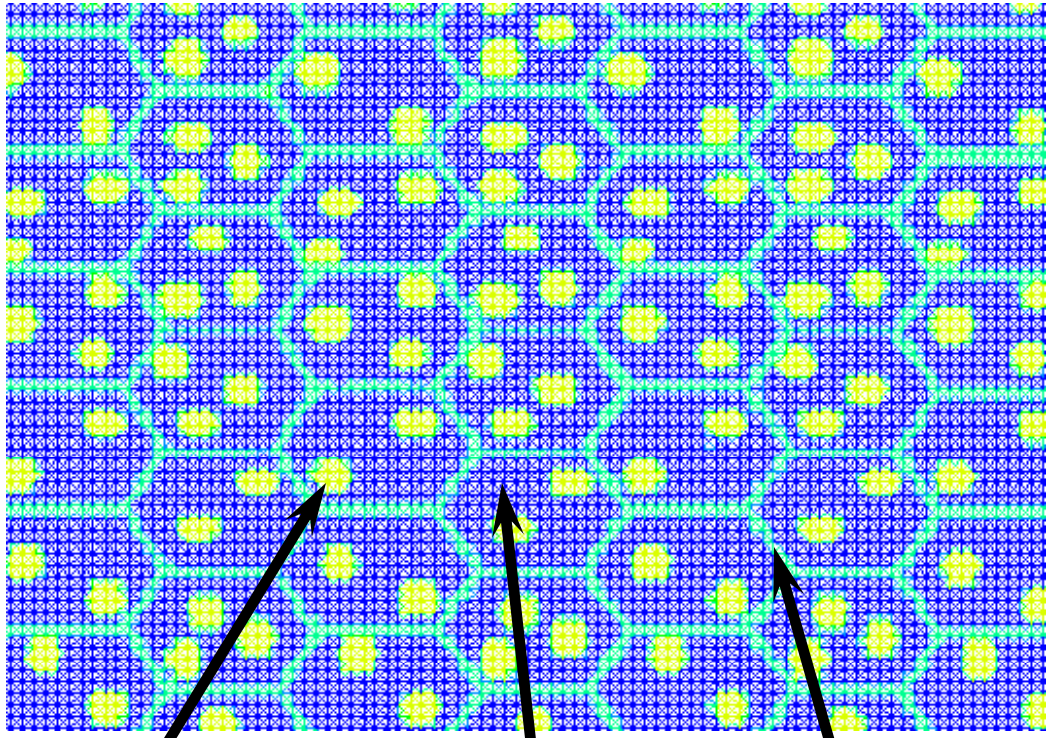
signing materials in an optimization framework

Idealized Microstructural Representation

Real Microstructure



Hypothetical Microstructure

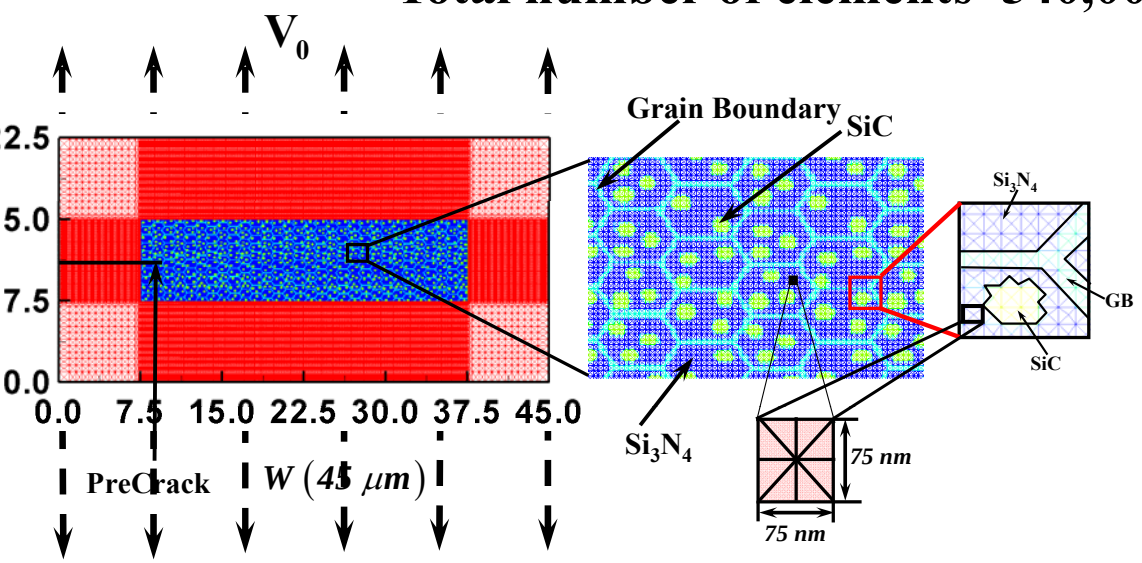


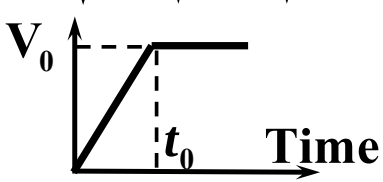
Silicon Carbide **Silicon Nitride** **GB**

AIAA, SDM-2007, Paper No. AIAA-2007-2345
Engg. Fract Mech., Mod. Sim. Mat. Sci. Engg.

Problem Setup

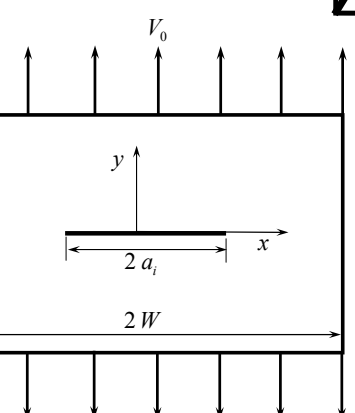
Total number of elements~540,000





$$0.1\times\frac{9\pi E\Phi_0}{32\left(1-\nu^2\right)T_{max}^2}<<h<<\left(\frac{\eta_0\Delta_{tc}}{T_{max}}\right)\frac{E'\left(\sqrt{2}+1\right)}{(1-\nu)}.$$

Tomar &Zhou, 2004

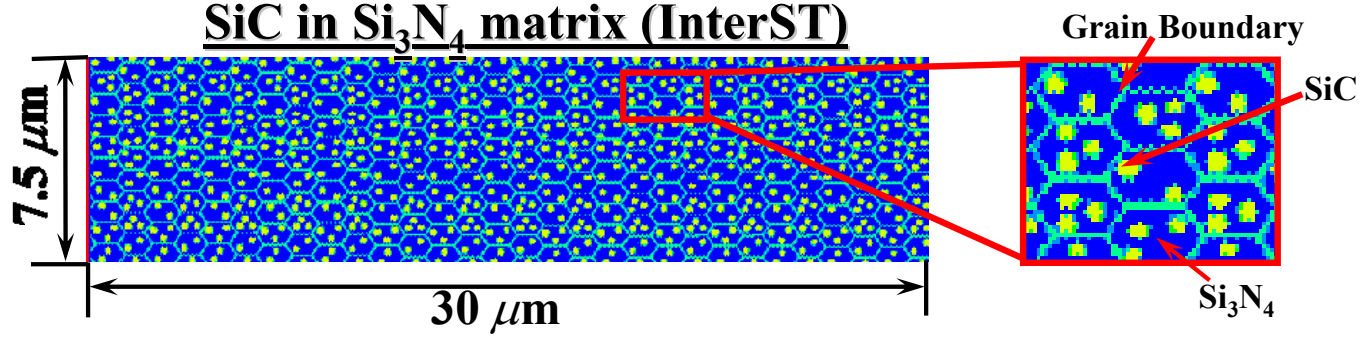


Its an image-analyses based procedure and can be easily extended to include real experimental microstructures

Microstructures of Focus-I

s-I

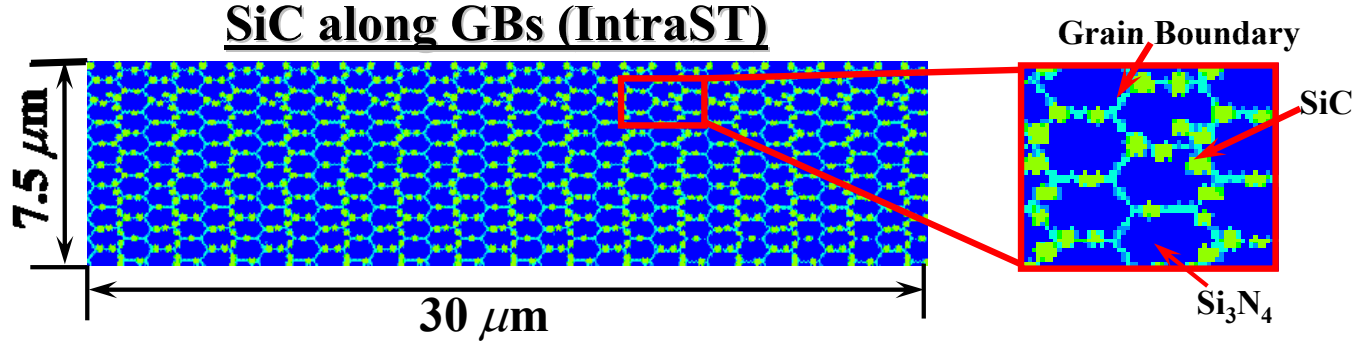
SiC in Si_3N_4 matrix (InterST)



(a)

s-II

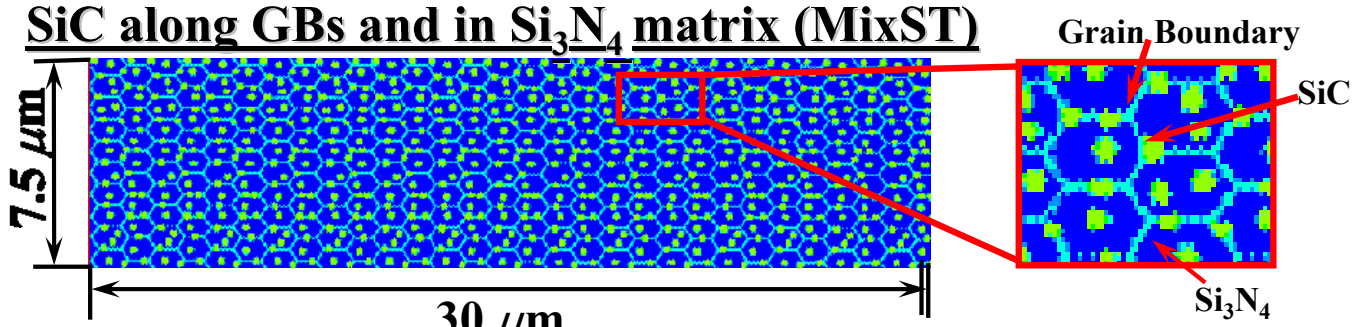
SiC along GBs (IntraST)



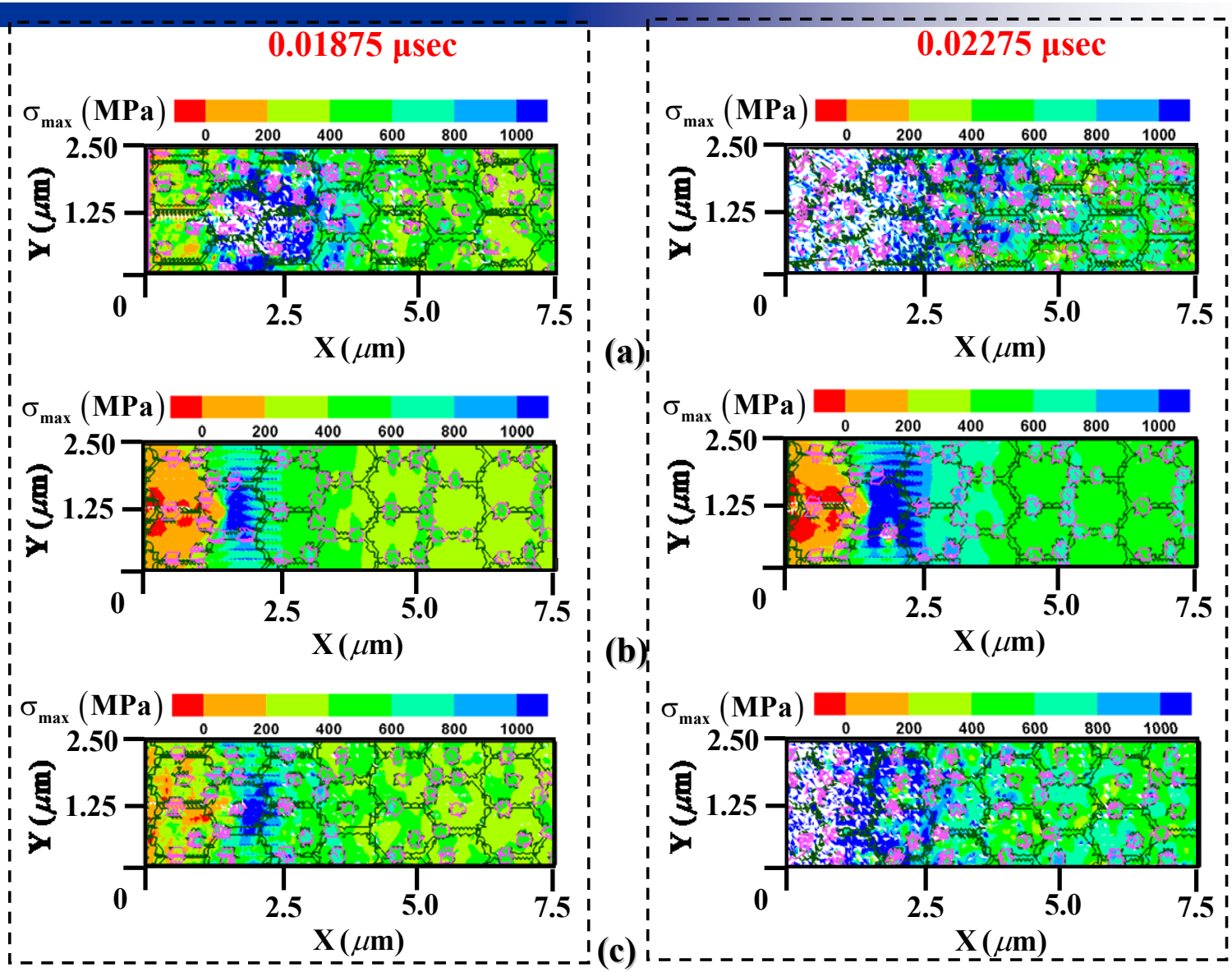
(b)

s-III

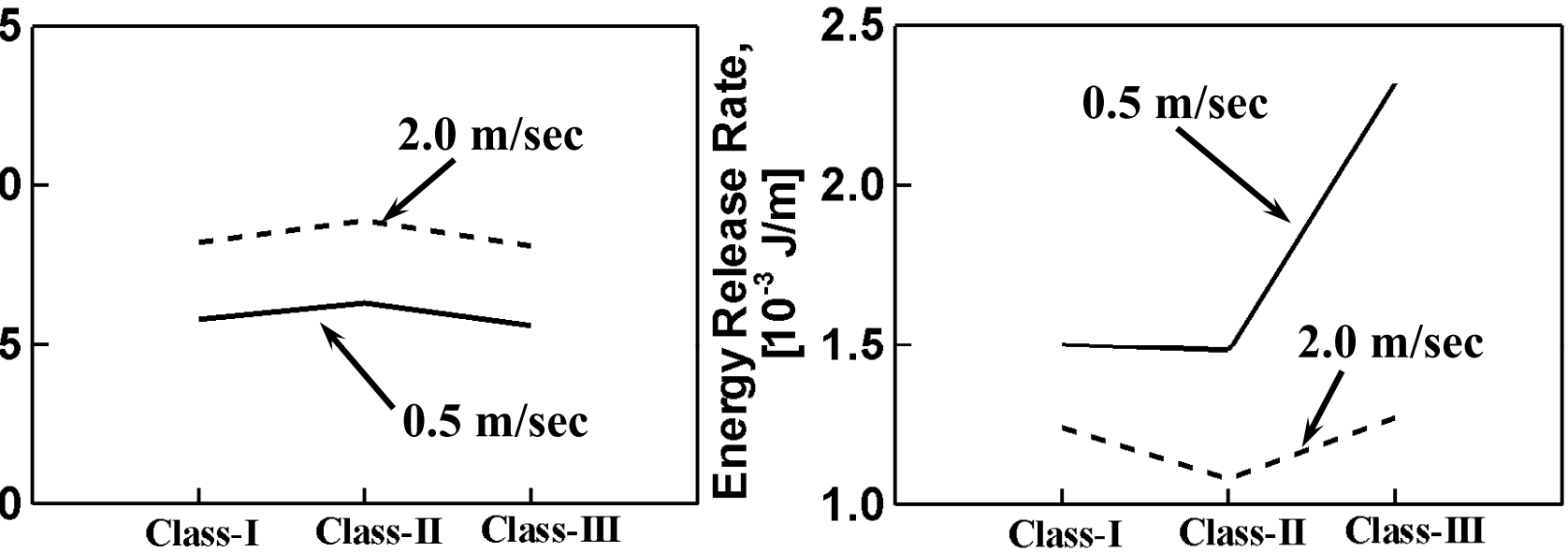
SiC along GBs and in Si_3N_4 matrix (MixST)



Damaging Effect of Second Phase Particles



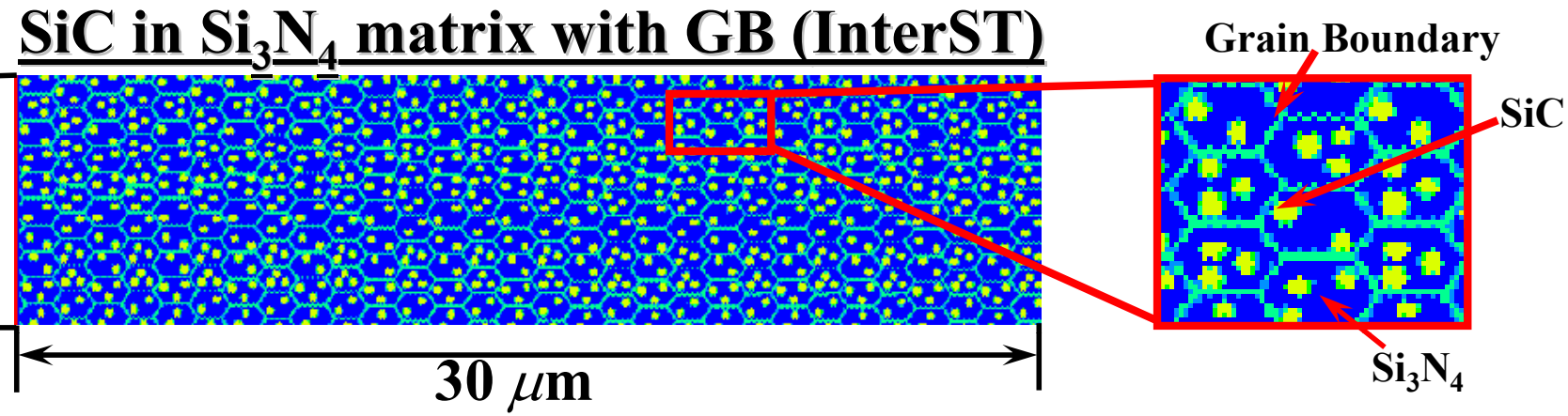
Second Phase Particles Effect on Crack Density



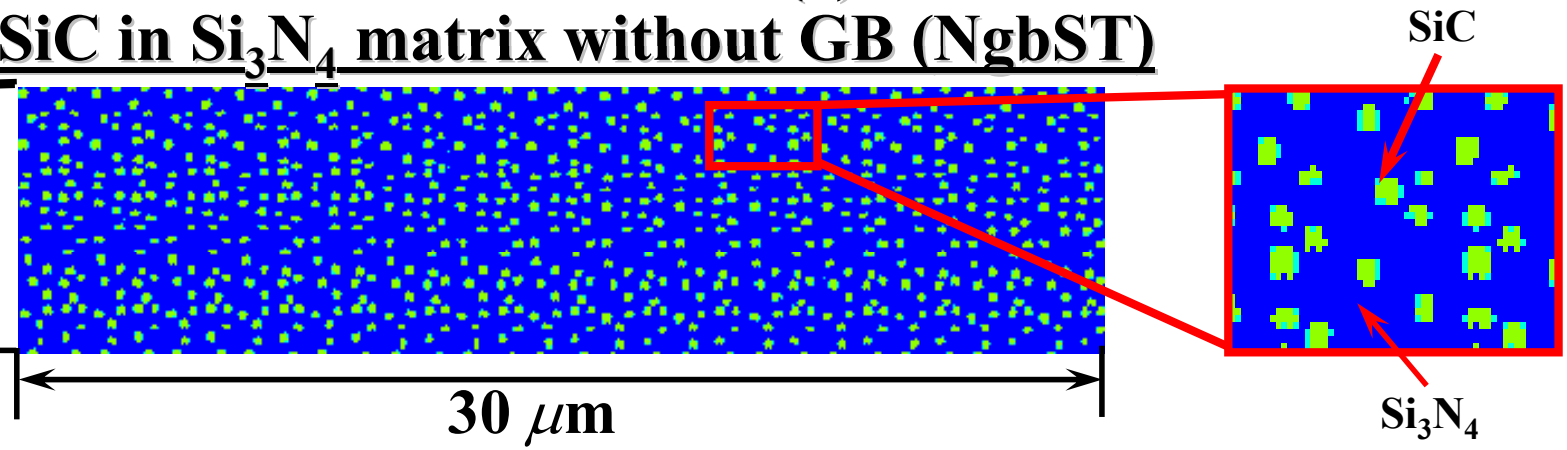
Higher second phase particle numbers in the matrix directly relate to higher crack density

In some cases second phase particles in matrix act to bridge the cracks leading to lowering of crack density

Microstructures of Focus-II

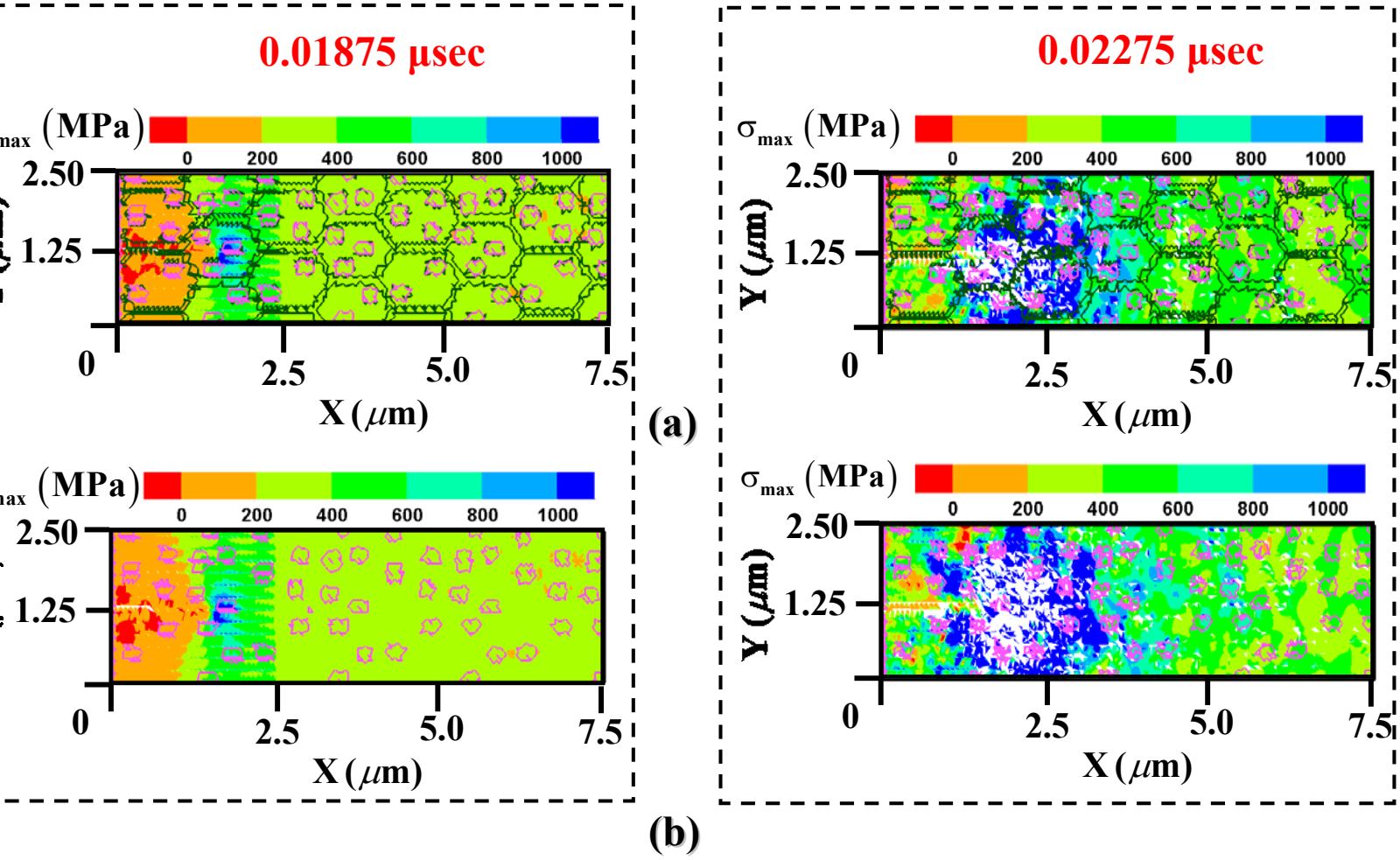


(a)



(b)

Damaging Effect of GBs



ield Function Model for Strength

$$\sigma = \sigma_0 \left[1 + \left(\frac{\varepsilon}{\varepsilon_0} \right)^n \right] \left[\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0} \cdot \exp\left(\frac{Q}{RT} \right) \right]^m$$

Lei et al., (2000)

Si_3N_4

= 28 MPa (yield stress at $\dot{\varepsilon}$ and $T = 1400°C$)

2× 10^{-9} s^{-1} (strain rate) *Wiederhorn S.M. et al. 1999*

740 KJ/mol (activation energy)

Krause et al. 1997, Wiederhorn and Luecke 1998

0.3 (reference strain) *Rouxel T. et al. 1997*

1 s^{-1} (reference strain rate)

0.34 (strain exponent)

0.09 (exponential coefficient)

Kusunose, T. et al 2004 and Park, H. et al 1998

SiC

$\sigma_0 = 400$ MPa (yield stress at $\dot{\varepsilon}$ and $T = 1400°C$)

$\dot{\varepsilon} = 1 \times 10^{-9}$ s^{-1} (strain rate)

$Q = 980$ KJ/mol (activation energy)

Wiederhorn S.M. et al. 1999 and Lane, J.E. et al 1998

$\varepsilon_0 = 0.17$ (reference strain)

$\dot{\varepsilon}_0 = 1$ s^{-1} (reference strain rate)

$n = 0.5$ (strain exponent)

$m = -0.001$ (exponential coefficient)

Munro R.G. et al. 1997

$R = 8.31447$ J/(mol °K) (universal gas constant)

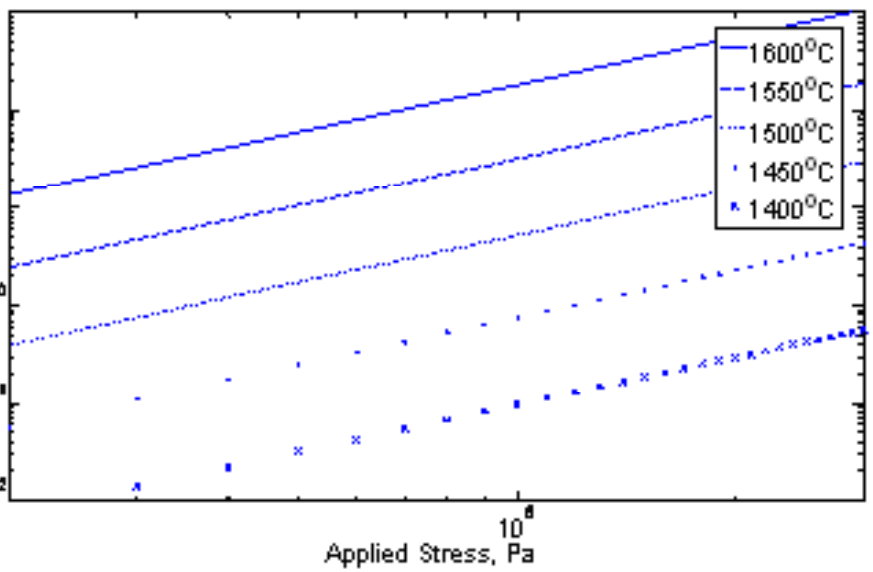
$\varepsilon = \varepsilon_e + \varepsilon_p$ (strain from FE)

$T \rightarrow$ absolute temperature in °K

Creep Model for SiC (Hexology)

By using the Norton relation for stress, the secondary creep elation for time and the temperature dependence, the creep strain ate expression for SiC used in this research takes the form:

$$\dot{\varepsilon}_c = A\sigma^n \exp\left(-\frac{Q}{RT}\right)$$



A fit to tensile data collected on Hexology

Wiederhorn 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 = 6.2e6 seg.$^{-1}$Pa^{-1}

n = 1.6

Q = 980 KJ/mol

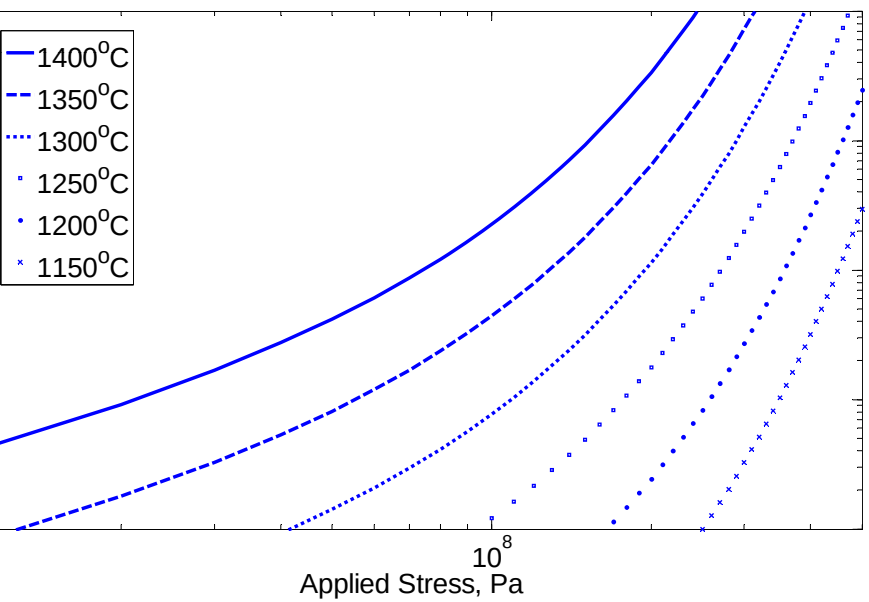
R = 8.31447 J/mol°K

Creep Model for Si_3N_4 (SN-88)

The creep strain rate the creep strain rate expression for Si3N4 used in this research takes the form:

Krause R.F. et al., 1999

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



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* = 3.89e6 seg^{-1}**

α = 0.0197±0.0008 MPa-1

***Q* = 715.3±22.9 KJ/mol**

***R* = 8.31447 J/mol°K (Boltzmann's constant)**

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

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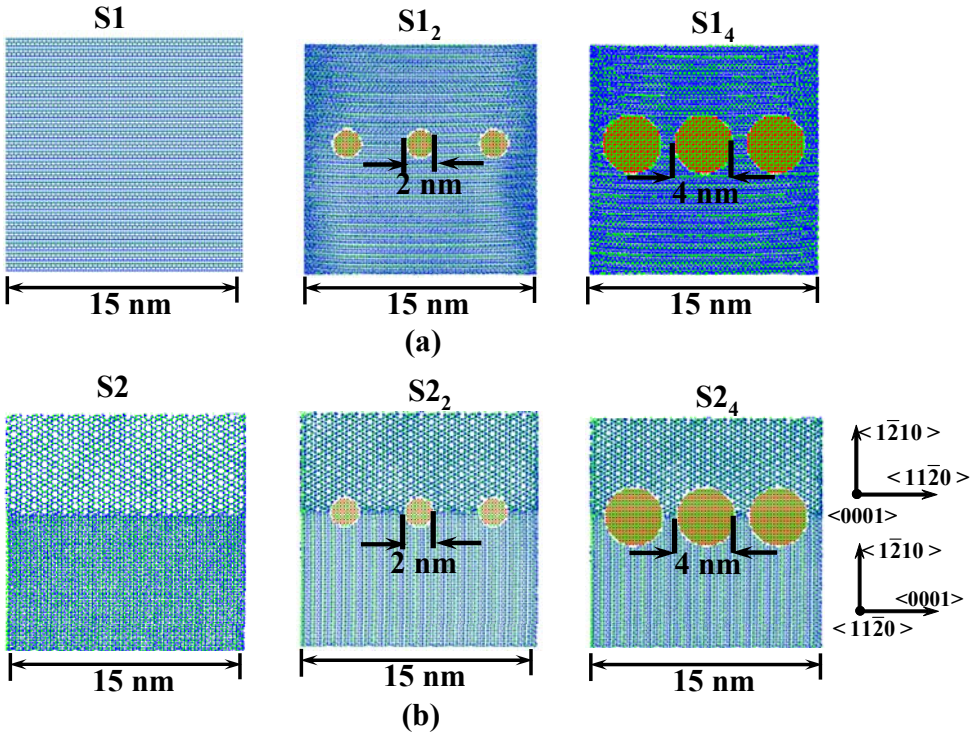
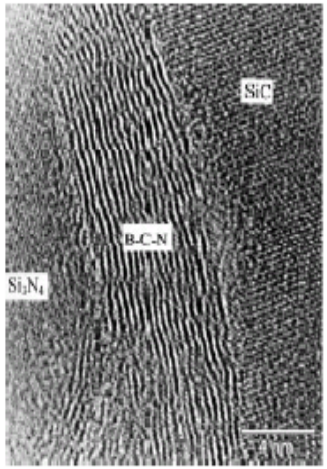
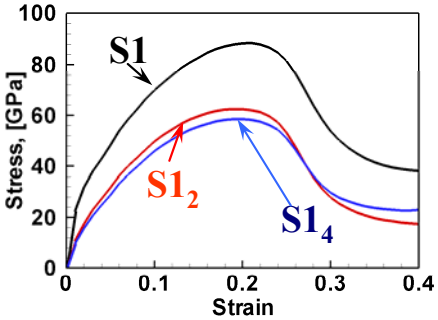
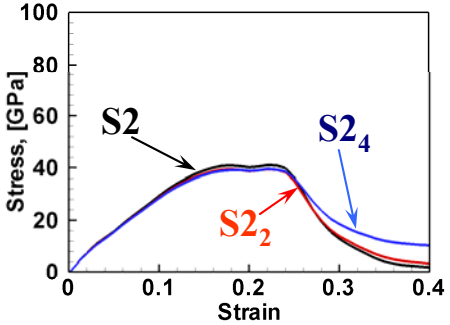
Microstructures of Focus

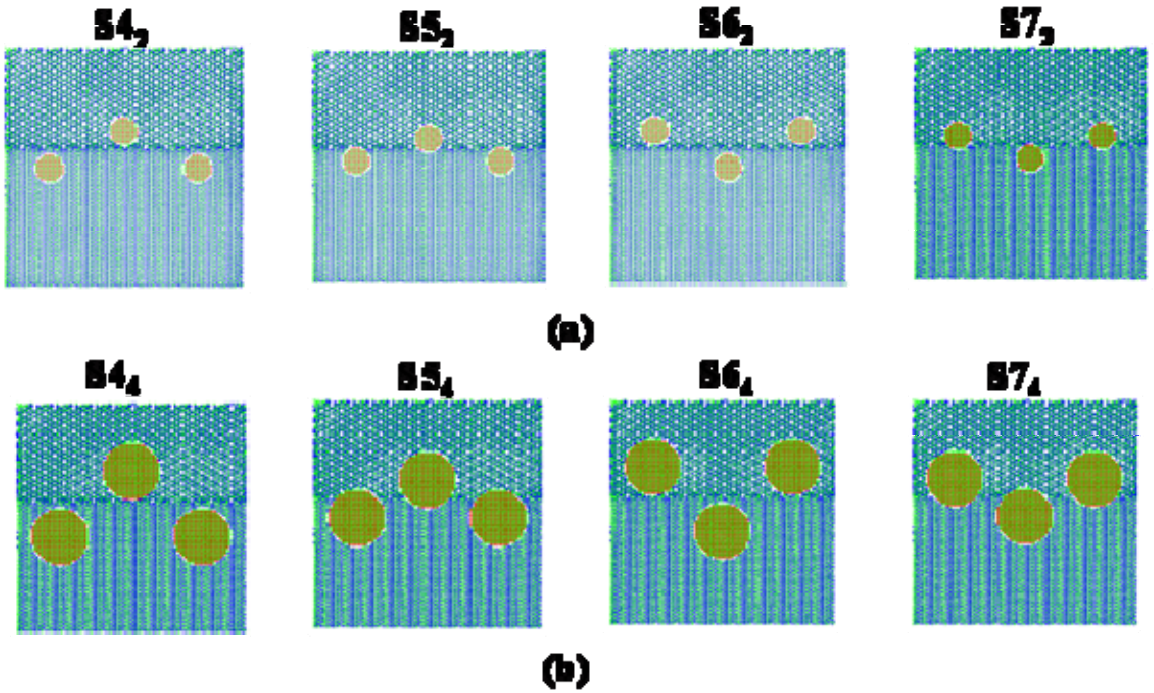


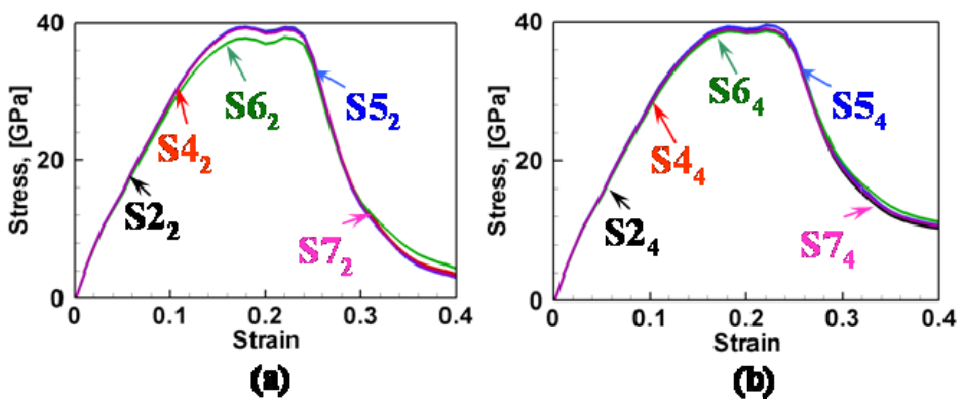




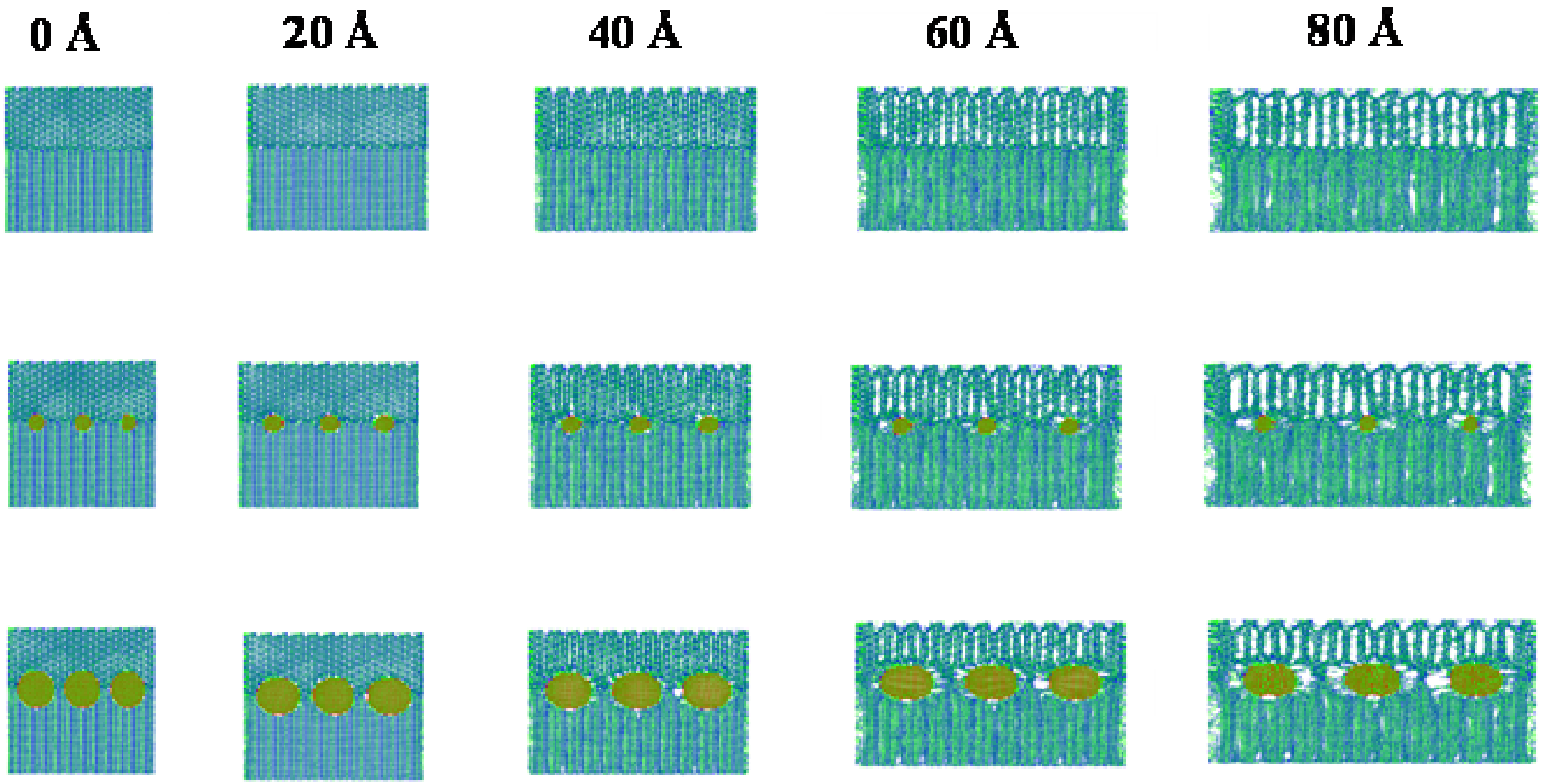


A Different Set of Microstructures



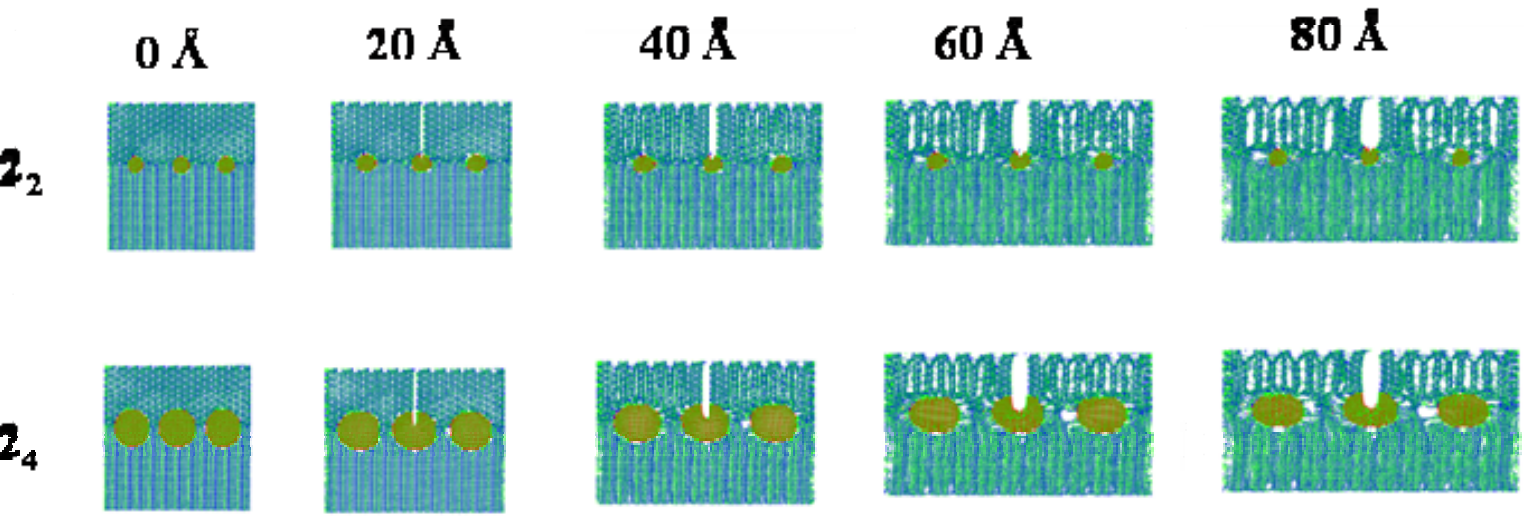


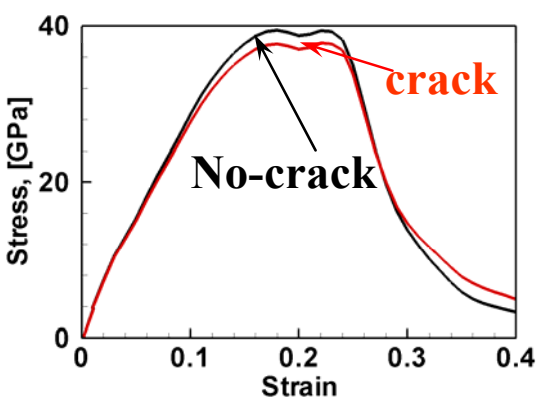
Deformation Mechanism



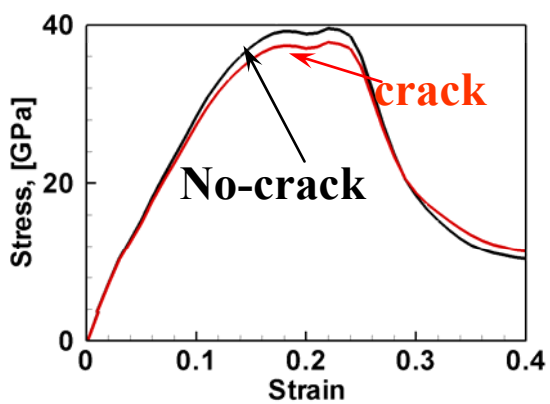
Tomar et al., Int. J. Multiscale Comp. Engg., Submitted

Crack Propagation





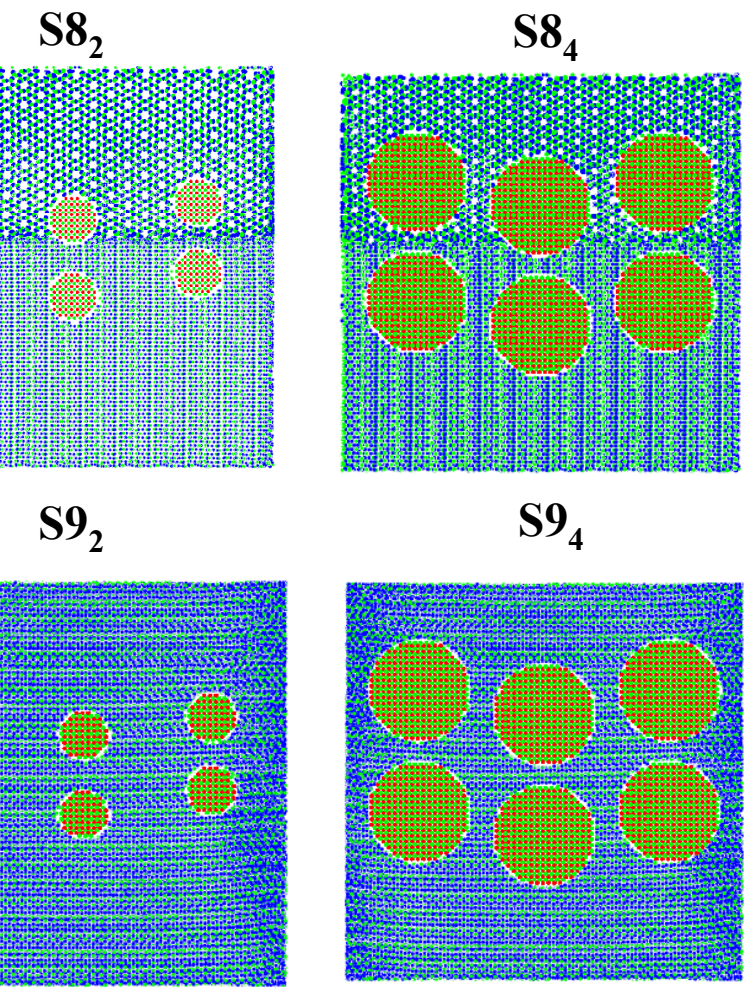
(a)

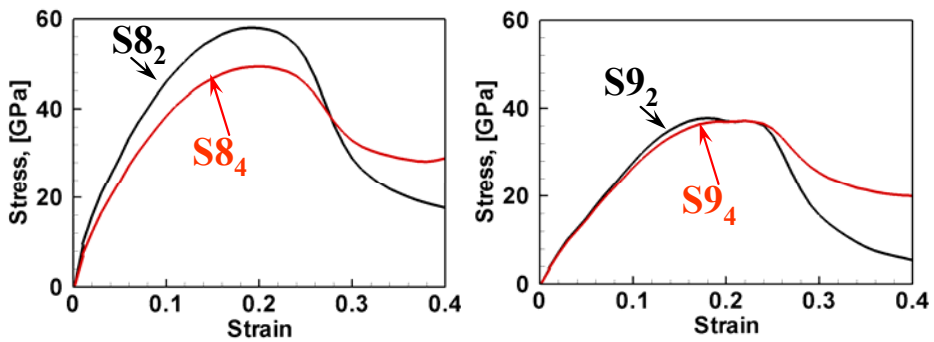


(b)

Flaw Tolerance!

Another Different Set of Microstructures

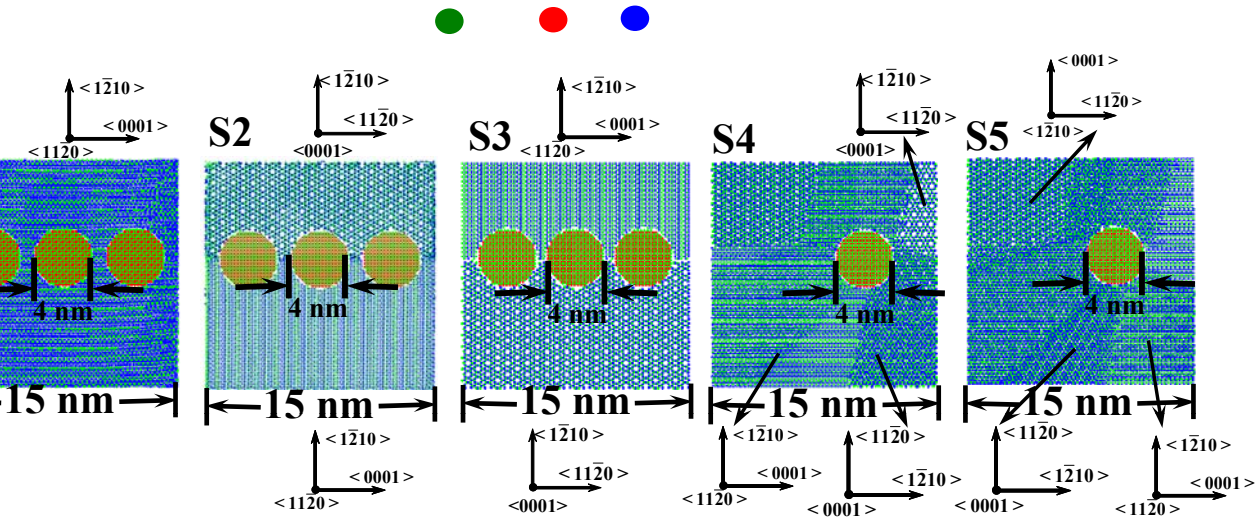




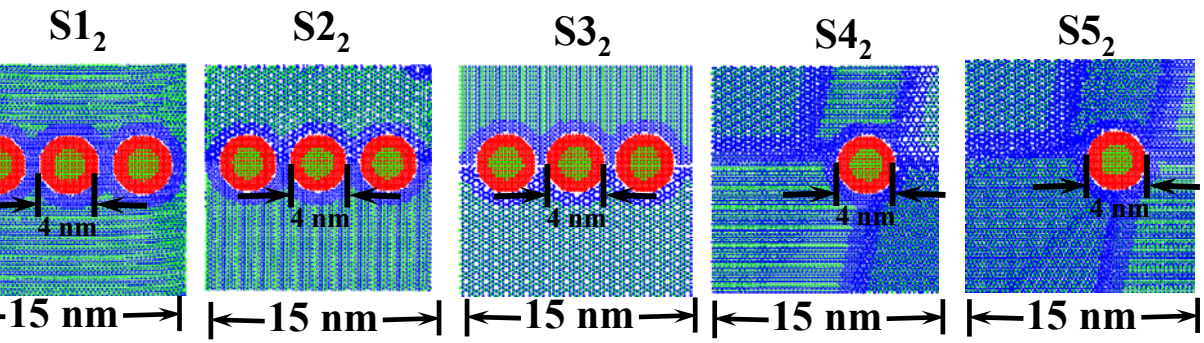
(a) (b)

Tomar et al., Int. J. Multiscale Comp. Engg., Submitted

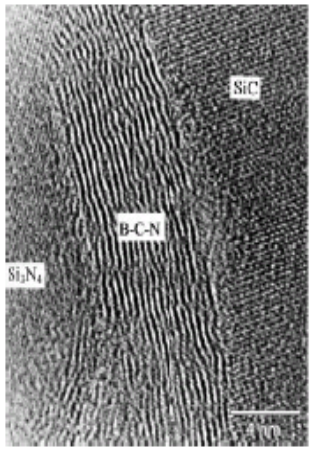
Another Set of Microstructures Analyzed



(a)

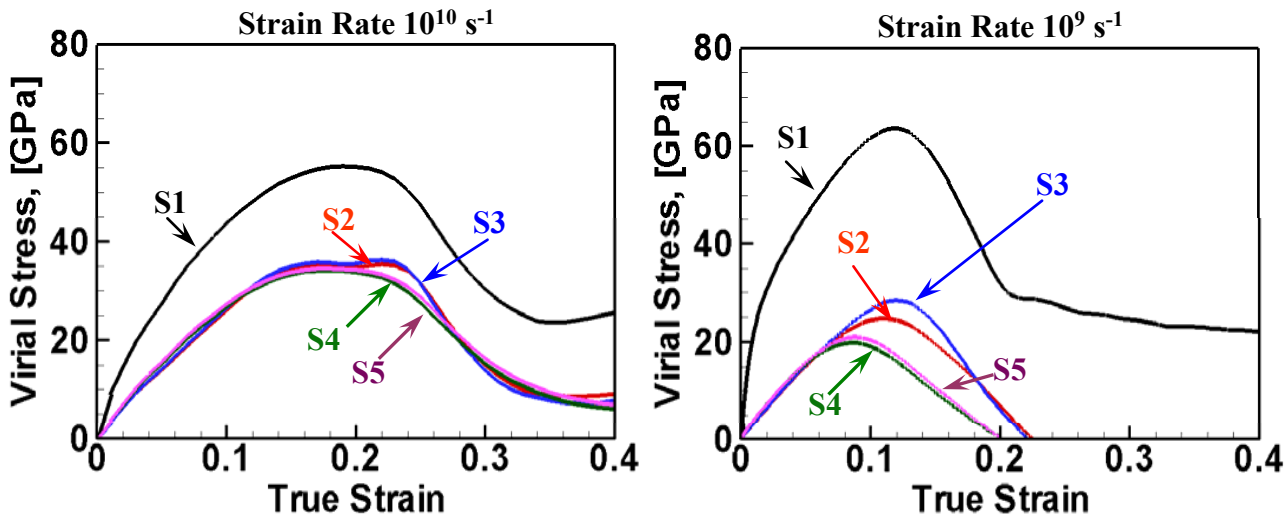


(b)

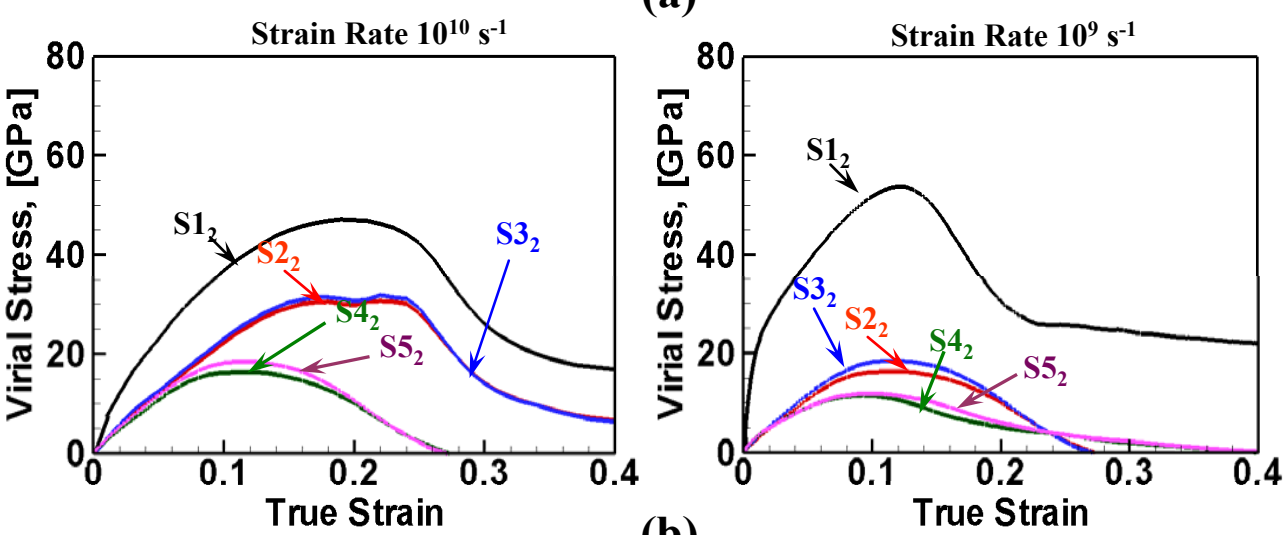


From R. Raj and co-workers

Rate Dependent Stress-Strain Behavior

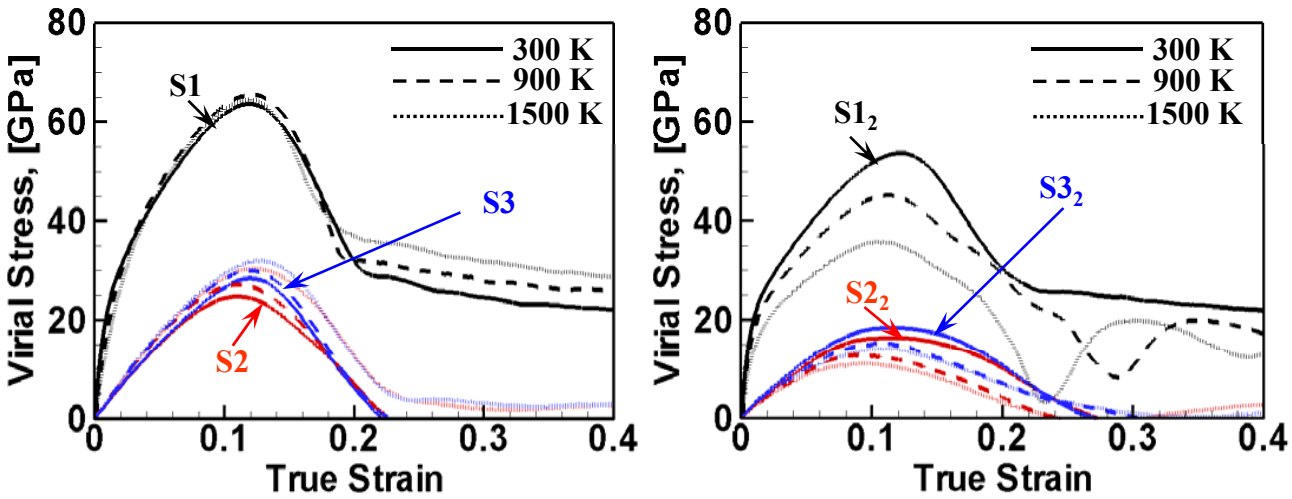


(a)

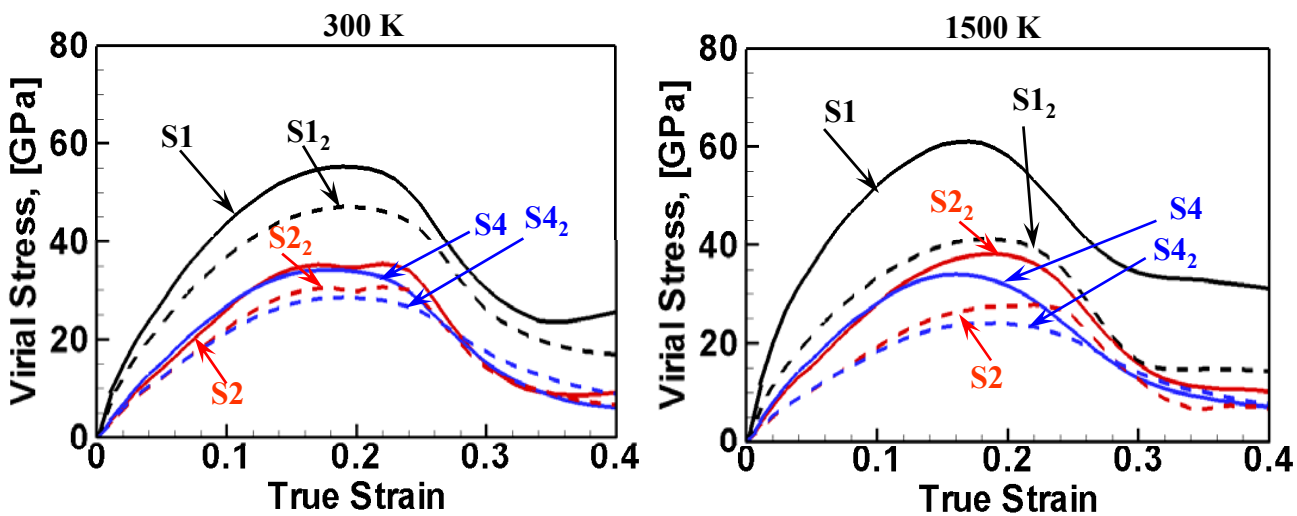


(b)

Temperature Dependent Strength



Temperature Dependent Strength-Effect of GB Thickness



Outline

gh temperature creep and nanoindentation experiments in high
nperature composites

- Scale dependent nanoindentation upto 773 K
- Nanoscale creep at different strain rates and maximum load

hesive finite element based modeling of dynamic fracture failure in SiC-
N_4 nanocomposites

- Cohesive analyses accounting for second phase particle distribution and grain boundary distribution effects
- High temperature constitutive models and trade-offs in designing these materials for high temperature performance

sights from molecular analyses

- Nanoscale fracture and strength at room temperature
- Role of temperature in strength upto 1500 K

signing materials in an optimization framework

Computer-aided design to investigate materials for use in future high temperature power plant applications

n
s

N_4

n
es

Fracture and creep resistance at about 1800 K

- Phase size
- Phase Volume Fraction

Variable Fidelity Optimization

Multiscale Material Simulation

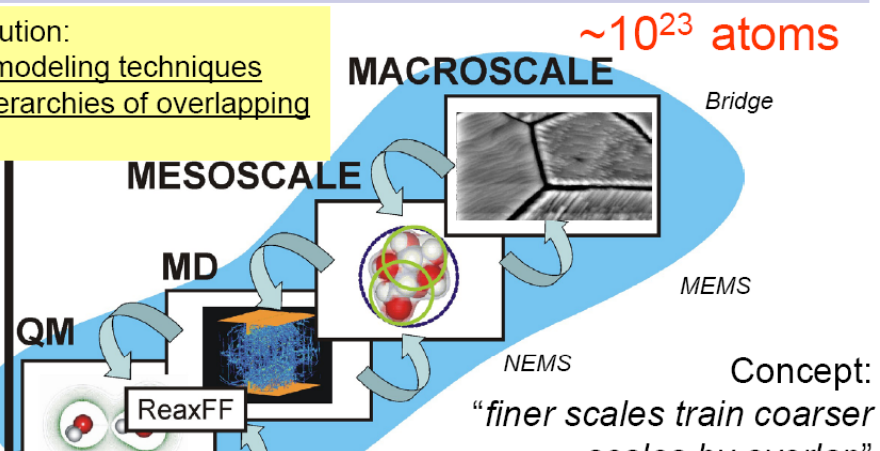
Optimal Morphologies

SiC-Si_3N_4 nanocomposite with desired properties

Accuracy of quantum mechanics (QM) in 10^{23} atom systems…

This is impossible (today and in the foreseeable future)

ution:
modeling techniques
erarchies of overlapping



Concept:
"finer scales train coarser scales by overlap"

Motivation

Materials for High Temperature Applications

Enhance efficiency and environmental performance.

Power plants

- **Gas turbine engine (rotors, nozzle guide vanes and the combustor liner)**
- **Fuel cells**

Desired Properties

- **Creep resistance.**
- **Fracture resistance.**
- **Oxidation Resistance.**

		α-SiC:	β-Si_3N_4:
Creep resistance (High Temp.)		✓	×
Strength	Room Temp.	×	✓
	High Temp.	✓	✓
Toughness (High Temp.)		×	✓

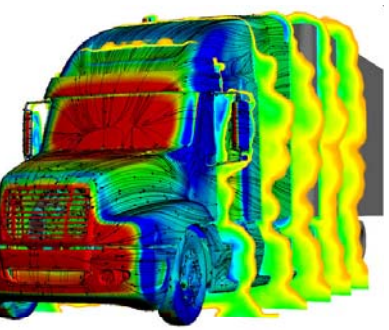
SiC-Si_3N_4 composites.

A trade-off must be made between creep resistance and material toughness to achieve an optimal high temperature microstructure.

Design Optimization

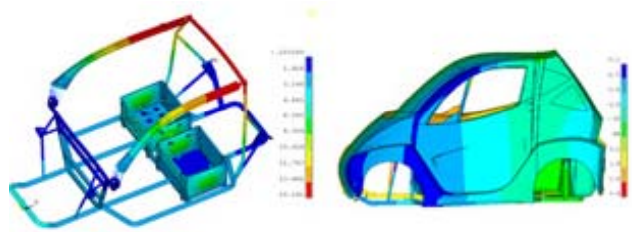
Computer simulation can be very expensive for optimization.

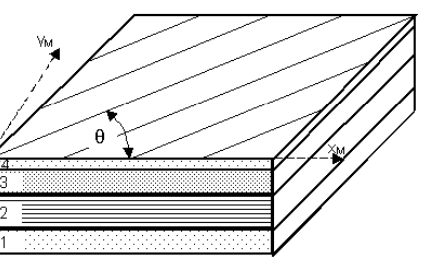
Need of optimization techniques for dealing with such simulations.



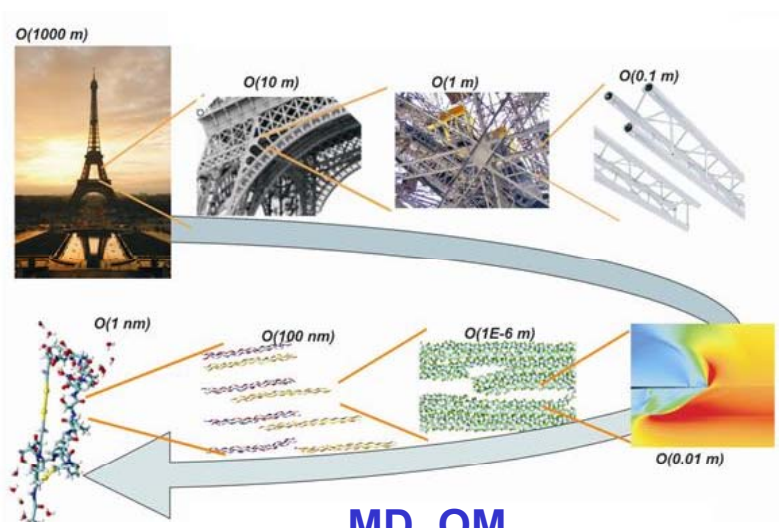
CFD

FEA





Layered Composites



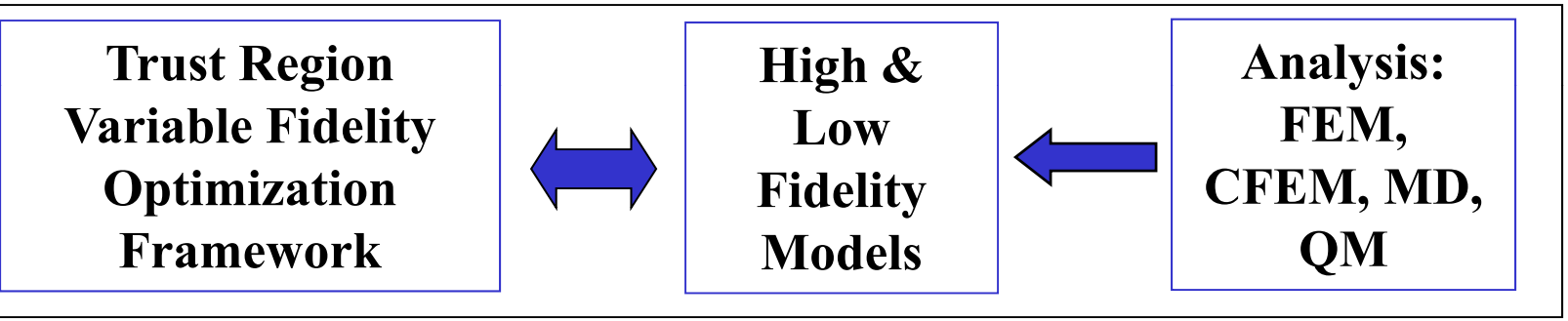
FEA

MD, QM

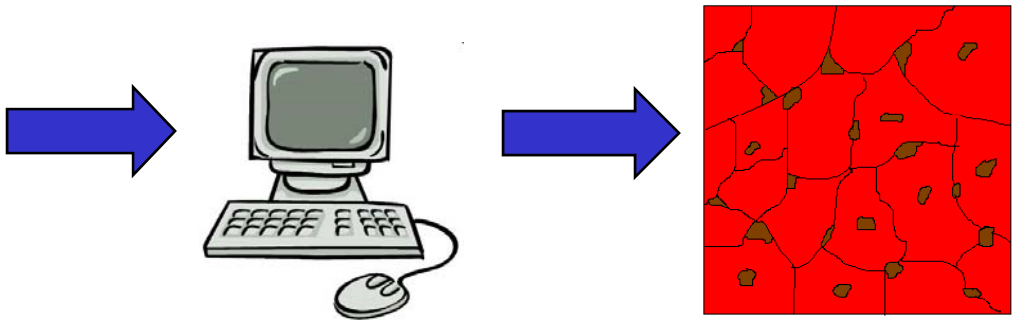
Material Design Methodology

Methods

- Address computational challenges.
- Address model management issues.

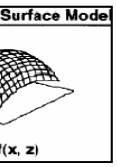
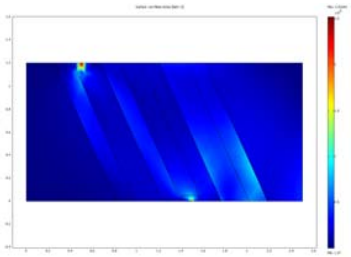
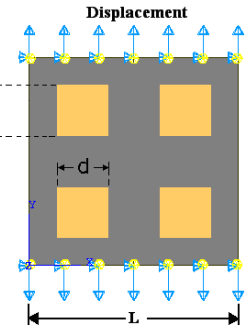
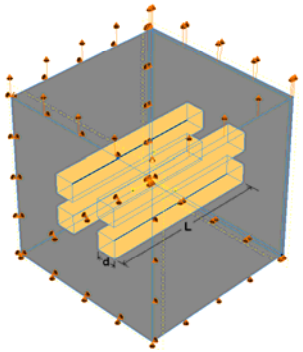
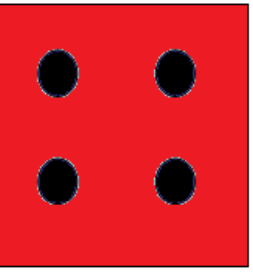
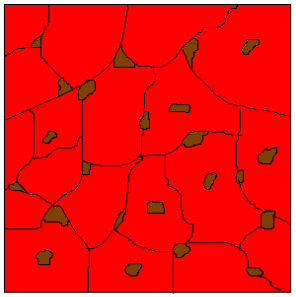


sign Targets
sign Variables

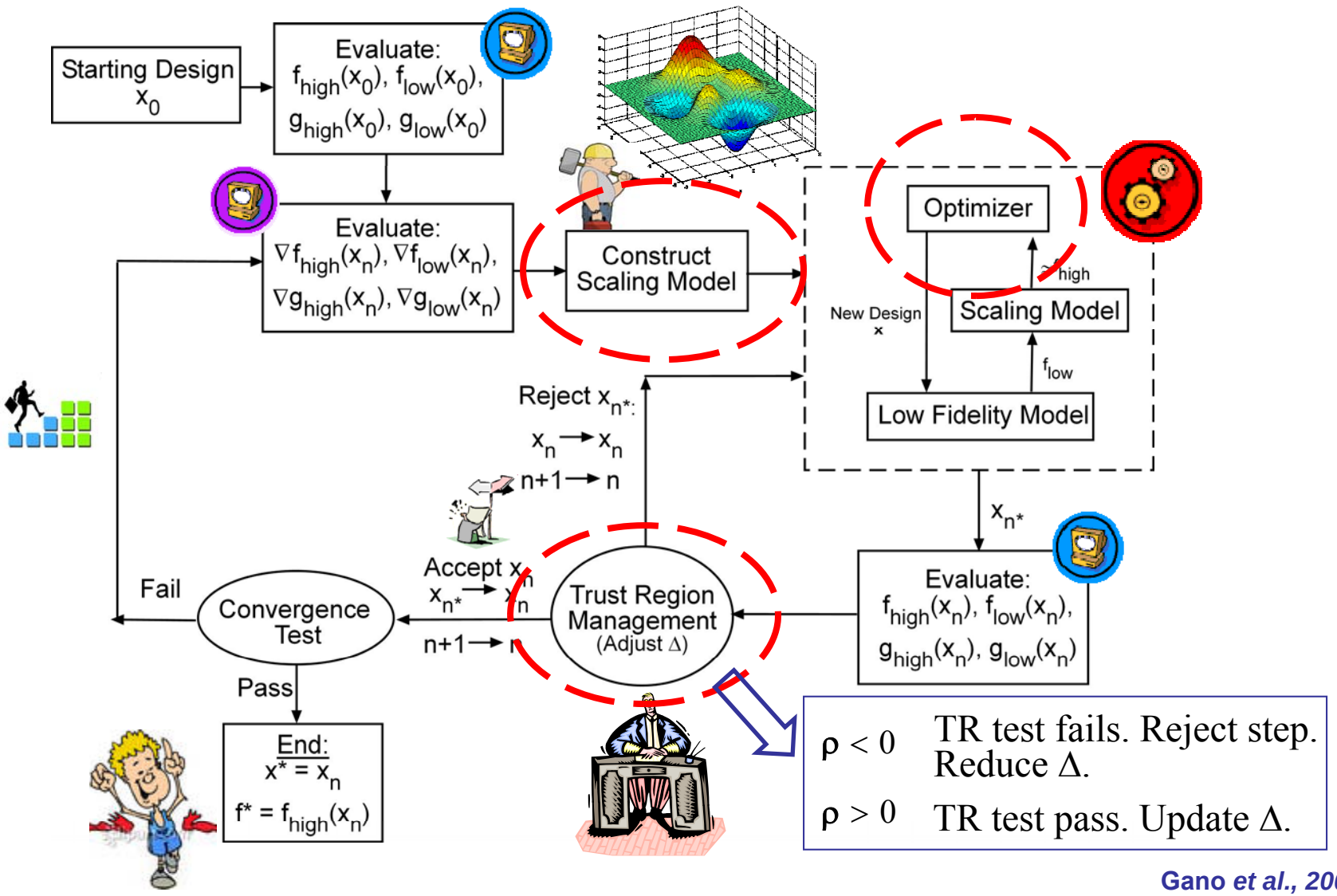


Optimal
Morphology

Objectives: Definition of Models

	LOW FM		HIGH FM
OBLEM 1  $K_I = \frac{PS}{BW^{3/2}} f\left(\frac{a}{W}\right)$	Analytical Fracture Mechanics & Surrogate Modeling	⟷ Fracture toughness	FEM (Comsol) 
OBLEM 2 	FEM (ABAQUS) 2D Model	⟷ Strength & Creep strain resistance	FEM (ABAQUS) 3D Model 
OBLEM 3 	FEM (ABAQUS) Ideal microstructure	⟷ Strength & Creep strain resistance	CFEM (ABAQUS) Real microstructure 

Variable Fidelity Optimization Algorithm



Gano *et al., 2004*

blem 1

Find optimal CFCC configuration for improving fracture toughness (K_{IC})

Construct scaling model

$$\beta(x) = \frac{f_{high}(x)}{f_{low}(x)} \Rightarrow f_{high}(x) \approx \beta(x) f_{low}(x)$$

Optimize scaling model

$$\begin{aligned} &\text{maximize} && \beta_n^1 K_{IC_{LOW}}(V_f, \theta), \\ &\text{subject to:} && \beta_n^2 K_{I_{LOW}} \le \beta_n^1 K_{IC_{LOW}}(V_f, \theta), \\ & && 0 \le V_f \le 0.2, \\ & && 0° \le \theta \le 90°, \\ & && -0.05 \le \Delta V_f \le 0.05, \text{ and} \\ & && -4° \le \Delta\theta \le 4° \end{aligned}$$

Fracture resistance

Variable Fidelity Optimization

Simulations

Optimal Morphology

s

- Second Phase orientation angle θ
- Second Phase Volume Fraction

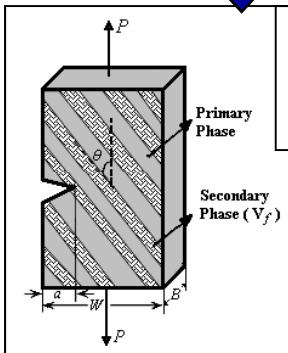
ze Mode-I $K_{IC}(V_f, \theta)$

to: $K_I \le K_{IC}(V_f, \theta)$

$0 \le V_f \le 0.2$

$0° \le \theta \le 90°$

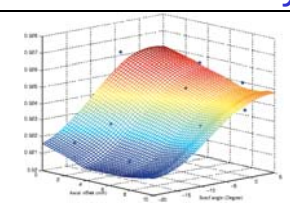
gn load
00 N → K_I



High fidelity (HF) model

K_{IC} and K_I obtained using FEM (COMSOL)

Low fidelity (LF) model



Response surface approximation for K_{IC}

$$K_I = \frac{PS}{BW^{3/2}} f\left(\frac{a}{W}\right)$$

Analytical fracture mechanics for K_I

Mejia-Rodriguez, G., J.E. Renaud, and V. Tomar, *A variable fidelity model management framework for designing multiphase materials.* Journal Of Mechanical Design, 2008. 130(9).

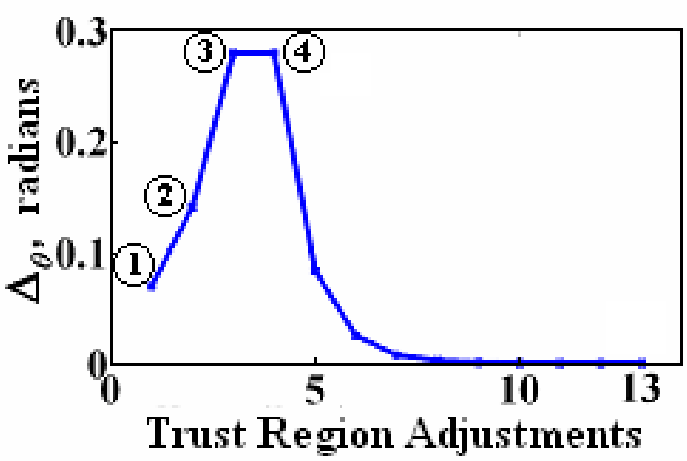
Results

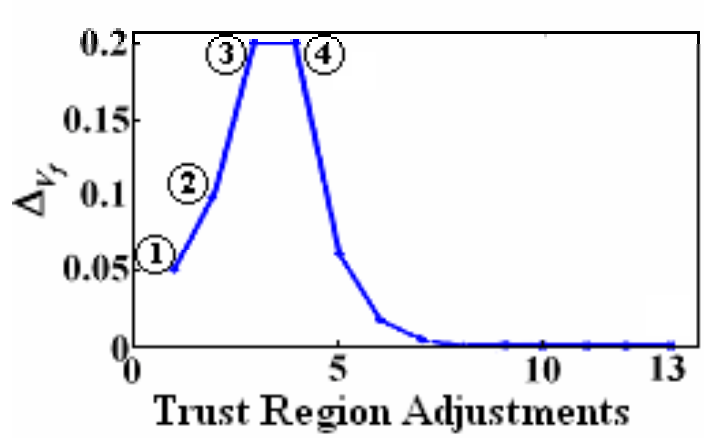
Problem 1

Initial design: $V_f^0 = 0.05$, $\theta^0 = 0.3$ rad
$K_{IC}^0 = 7013519.6$ MPa·m$^{1/2}$

80% reduction in HF simulations

Optimization Method	*Low Fidelity Simulations*	*High Fidelity Simulations*	*Iteration*	θ^*(rad.)	V^*_f	K_{IC}^*(MPa·m$^{1/2}$)
Conventional	0	109	10	0.08528	0.2	7239246.361
Variable Fidelity	187	24	4	0.08531	0.2	7239245.175





ration	*Trust Region Adjustments*	*Trust Region Size*		*Convergence*		*Design Varibles and Obj. Function*		
		$\Delta\theta$	ΔV_f	Δ_x	Δ_f	θ(rad.)	V_f	K_{IC} (MPa·m$^{1/2}$)
1	1	0.07	0.05	0.2828	0.0111	0.23	0.1	7091470.873
2	1	0.14	0.10	0.6859	0.0208	0.09	0.2	7239211.263
3	1	0.28	0.20	0.0214	4E-06	0.08530	0.2	7239241.029
4	10	2.1E-10	0.0000012	0.00002	6E-07	0.08531	0.2	7239245.175

blem 2

Find optimal CMC (ceramic matrix composite) configuration

- **Incorporate Multiobjective Optimization formulation**
- **Construct scaling model: It will allow to incorporate higher dimensionality that is not present in LF model**

Design Target

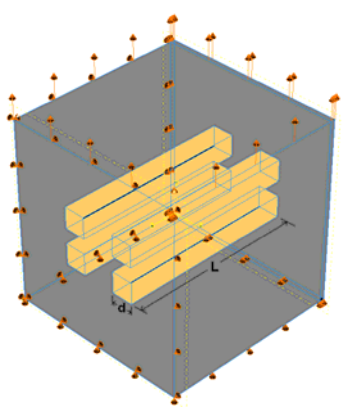
Strength & Creep resistance

Variable Fidelity Optimization

Simulations

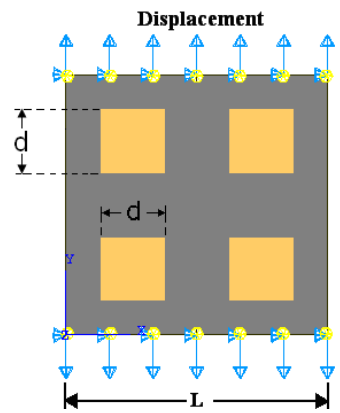
Pareto Optimal Morphology

Ideal microstructures



3-D HF FEA model

Design Variables
***d*-Grain width-height**
***l*- Grain length**
***T*-Temperature**



2-D LF FEA model

Design Variables
***d*-Grain width-height**
***T*-Temperature**

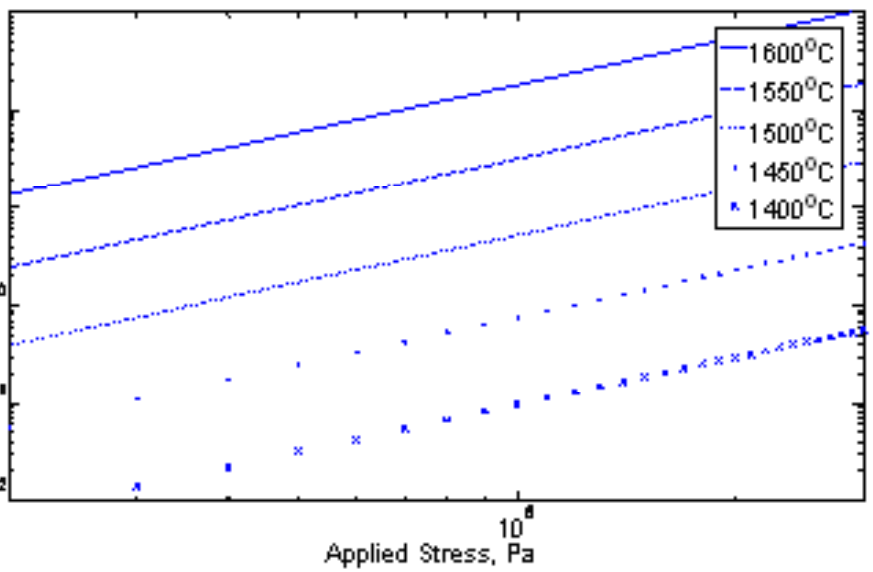
uum based models
ed to define:
Flow stress
Creep strain rate

- **Multiphysics models with different input variables**
- **2D LF captures physics of 3D HF**

Creep Model for SiC (Hexology)

By using the Norton relation for stress, the secondary creep elation for time and the temperature dependence, the creep strain ate expression for SiC used in this research takes the form:

$$\dot{\varepsilon}_c = A\sigma^n \exp\left(-\frac{Q}{RT}\right)$$



A fit to tensile data collected on Hexology

Wiederhorn 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 = 6.2e6 seg.$^{-1}$Pa^{-1}

n = 1.6

Q = 980 KJ/mol

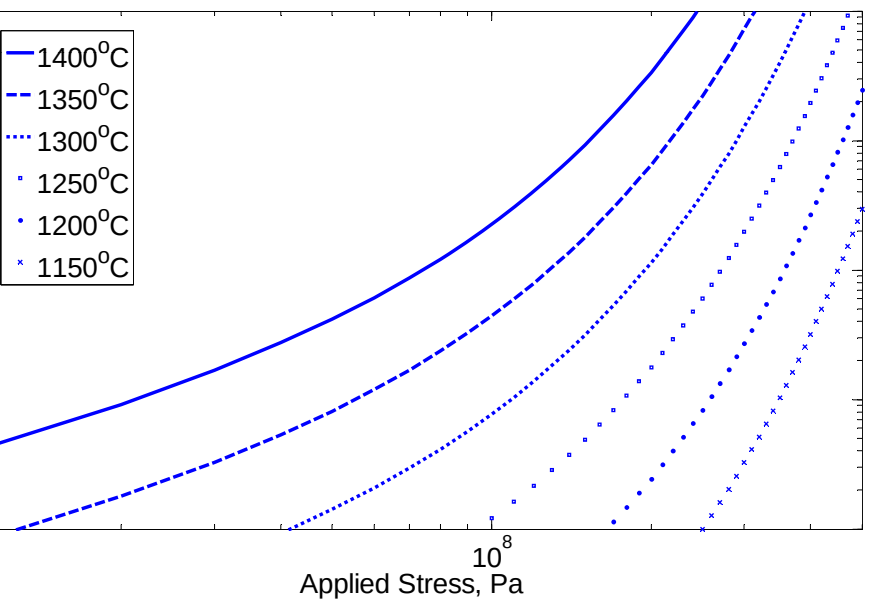
R = 8.31447 J/mol°K

Creep Model for Si_3N_4 (SN-88)

The creep strain rate the creep strain rate expression for Si3N4 used in this research takes the form:

Krause R.F. et al., 1999

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



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* = 3.89e6 seg^{-1}**

α = 0.0197±0.0008 MPa-1

***Q* = 715.3±22.9 KJ/mol**

***R* = 8.31447 J/mol°K (Boltzmann's constant)**

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

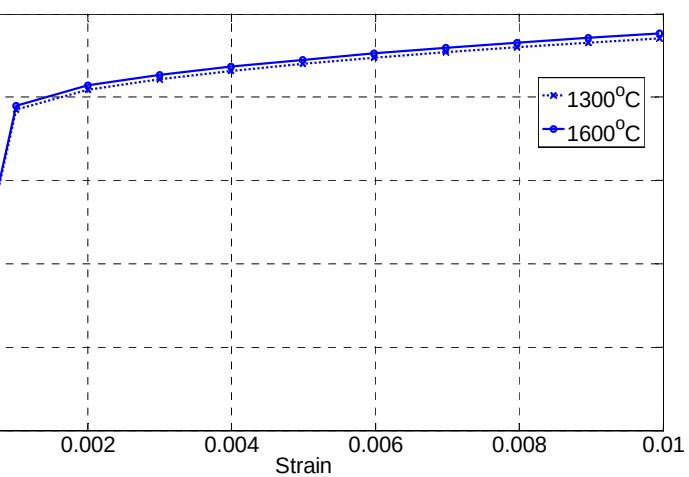
Stress-Strain curves

Problem 2

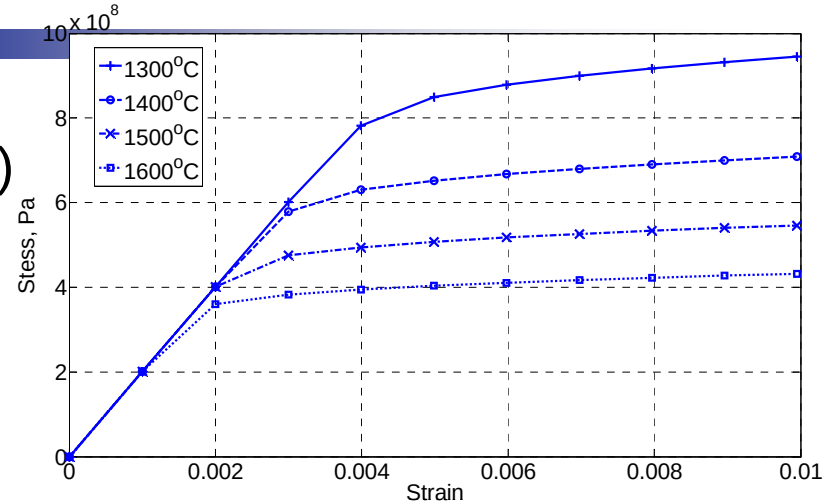
eld function for strength (Si_3N_4 & SiC)

$$= \sigma_0 \left[1 + \left(\frac{\varepsilon}{\varepsilon_0} \right)^n \right] \left[\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0} \cdot \exp\left(\frac{Q}{RT} \right) \right]^m$$

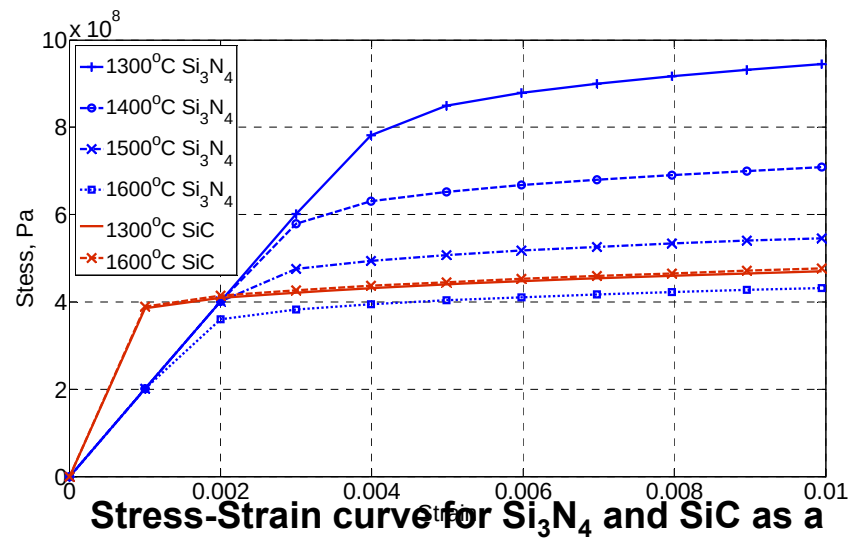
Lei et al., 2000



ss-Strain curve for SiC showing small ement in strength as temperature rises 1300°C to 1600°C.



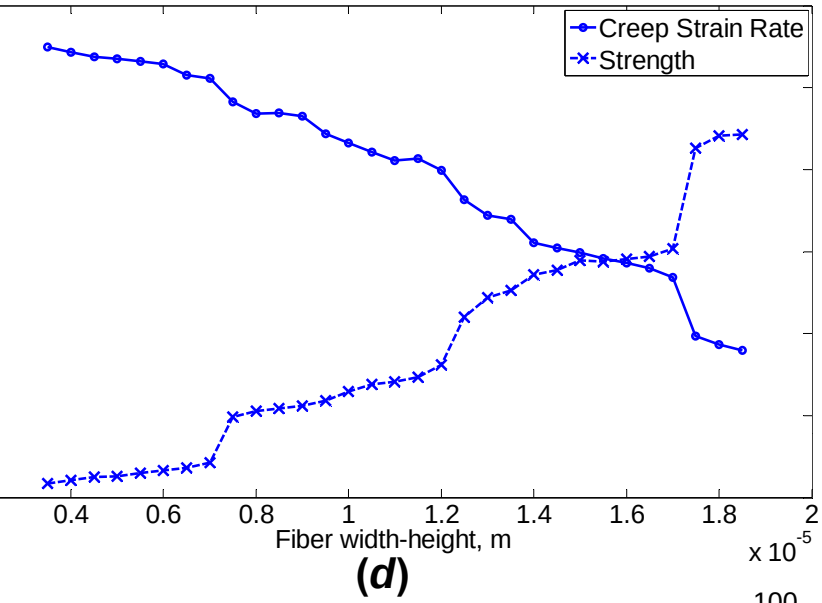
Stress-Strain curve for Si_3N_4 showing increment in strength as temperature goes down from 1600°C to 1300°C.



Stress-Strain curve for Si_3N_4 and SiC as a function of temperature.

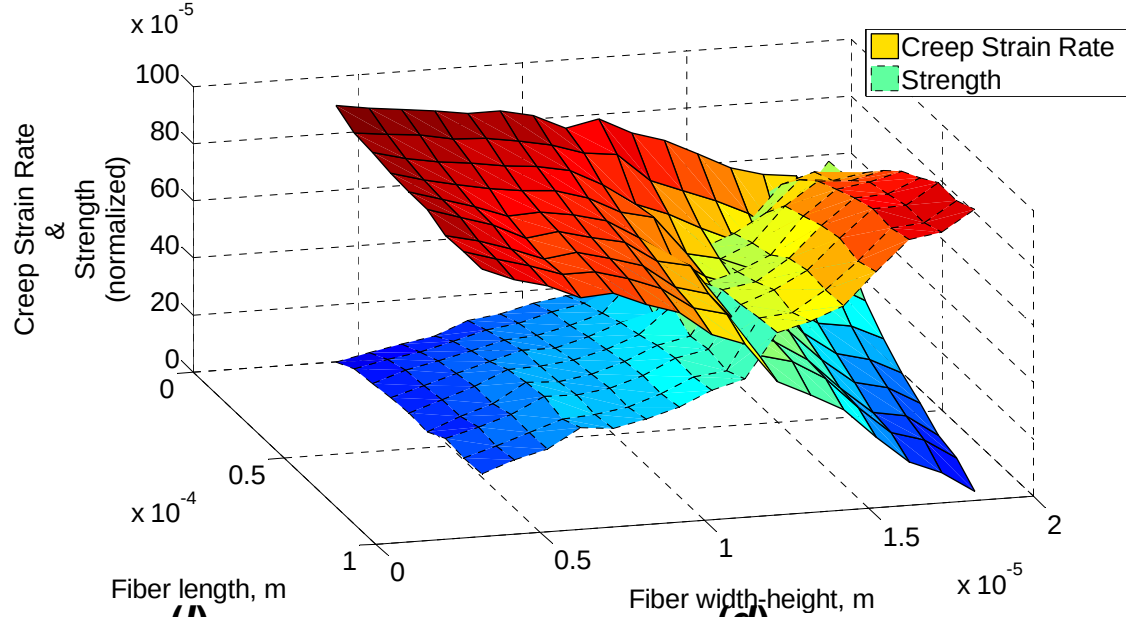
Strength and Creep behavior as a function of Design Variables

Problem 2



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.

gth and creep strain rate 00°C as a function of the n variables fiber width- t (*d*) and fiber length (*l*) the 3-D high fidelity l.



rent work

Develop nanocomposites whose performance variation is insensitive to processing (microstructure) variations

esign
rgets

- **Incorporate Multiobjective Optimization formulation**
- **Incorporate Robust Optimization approach.**
- **Construct scaling model**

- Fracture Toughness
- Strength
- Creep resistance

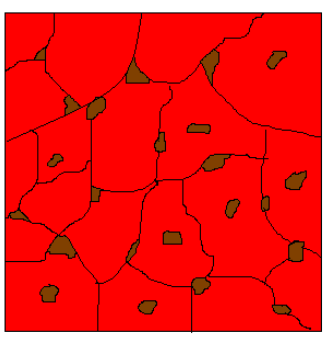
Variable Fidelity Optimization

Simulations

Pareto Optimal Morphology

hysics with t input es tures of HF

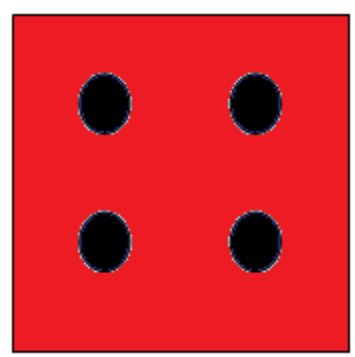
HF real microstructure



Design variables
- Grain size distribution
- Number of grains
- Volume fraction
- Grain orientation

CZM at grain boundaries & continuum models to define behavior of single phase

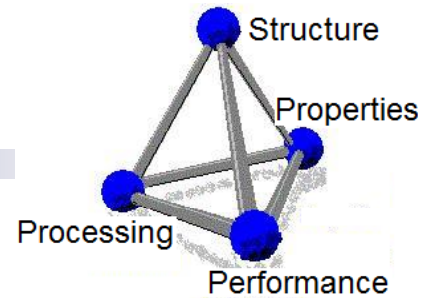
LF ideal microstructure



Design variables:
- Grain size
- Number of grains
- Volume fraction

Elasto-plastic continuum models to define behavior of single phases

Material Design

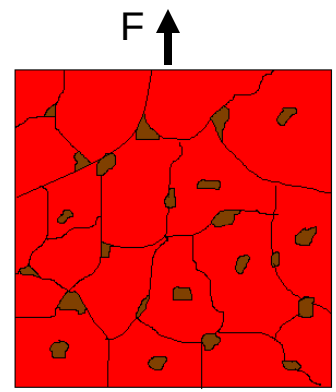


cessing | Transformation | Microstructure | Properties $\sigma^{u}, \dot{\varepsilon}_{c}$

V_f
Temp.
essure
ain size
⋮

X ?

Growth rate
Nucleation rate
GSD
grains
⋮



Processing variation → Microstructure variation → Properties variation

Robust Material Design

Find:
Optimal robust design variables

Maximum: σ^{u}

Minimum: $\dot{\varepsilon}_{c}$

Reduce variation
$\Delta\sigma^{u}, \Delta\dot{\varepsilon}_{c}$

Summary

Coal will become the energy and raw material resource of choice in the coming decades. The key is developing technologies for clean coal utilization. It is estimated that the use of bulk materials ($SiC-Si_3N_4$) with high temperature properties to operate at temperatures in excess of 1500 °C in power plants can result in enhancements in the power generation efficiency by 10% to 15%.

The Computer Aided Multiscale Design tool is expected to work efficiently on obtaining the most suitable sets of morphologies (design variables) to obtain a designated target set of properties by integrating multiscale simulations of the $SiC-Si_3N_4$ nanocomposite in a trust region variable fidelity framework.

The tool is based on a sound combination of atomistic simulations, continuum simulations, and experiments in a design optimization framework

The tool is efficient compared to existing approaches and has predicted many important insights about material behavior

Acknowledgements

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Refereed Archival Journal Publications

1. **Tomar, V. and Gan, M., 2009, Temperature dependent nanomechanics of si-c-n nanocomposites with an account of particle clustering and grain boundaries, *submitted to Int. J. Hydrogen Energy***

2. **Mejia-Rodriguez, G., Renaud, J.E., and Tomar V., 2009, Multiobjective composite material design using the variable fidelity model management optimization framework, *Submitted to the AIAA Journal***

3. **Samvedi, V, and Tomar, V., 2009, Role of interface thermal boundary resistance in overall thermal conductivity of Si-Ge multi-layered structures, *Submitted to Nanotechnology***

4. **Tomar, V., Samvedi, V., and Kim, H., 2009, Atomistic understanding of the particle clustering and particle size effect on the room temperature strength of SiC-Si_3N_4 nanocomposites *submitted to Int. J. Multiscale Comp. Engg. special issue on Advances In Computational Materials Science***

5. **Tomar, V., and Samvedi, V, 2009, Atomistic simulations based understanding of the mechanism behind the role of second phase SiC particles in fracture resistance of SiC-Si_3N_4 nanocomposites, *to Appear in International Journal of Multiscale Computational Engineering***

6. **Mejia-Rodriguez, G., Renaud, J.E., and Tomar V., 2008, A variable fidelity model management framework for designing multiphase materials, *ASME J. Mech. Design, vol. 130, 091702-1***

7. **Tomar, V., 2008, Analyses of the role of the second phase SiC particles in microstructure dependent fracture resistance variation of SiC-Si_3N_4 nanocomposites, *Modelling Simul. Mater. Sci. Eng. 16 (2008) 035001.***

8. **Tomar, V., 2007, Nanomechanical analyses of nanocrystalline Ni using accelerated molecular timesteps, Physica Status Solidi-a, vol. 204 (10), pp. 3340-3348.**

9. **Tomar, V., 2007, Accelerating the molecular timesteps for nanomechanical simulations: The hybrid Monte Carlo Method, Journal of Applied Physics, Vol. 101, pp 103512 (1-9)**