

**AFOSR FA9550-09-1-0633**  
Program Manager: Dr. Ali Sayir

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

**Part III**  
**Design optimization and uncertainty quantification**

**Andrés Tovar, Co-PI**

Kunal Khadke and An Weigang  
Indiana University-Purdue University Indianapolis, IN



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

Andrés Tovar

Indiana University-Purdue University Indianapolis, IN

Vikas Tomar

Purdue University, West Lafayette, IN

Rajendra Bordia

University of Washington, Seattle, WA

## Multiscale modeling, design optimization, and processing and experimental validation of SiC-Si<sub>3</sub>N<sub>4</sub> nanocomposites

### Multiscale Modeling and Experiments

- Molecular dynamics (MD) framework for complex nano-phase composites.
- Novel cohesive finite element method (CFEM) to analyze nanocomposite fracture.
- Prediction of SiC-Si<sub>3</sub>N<sub>4</sub> microstructure failure by numerical simulation.

### Design Optimization

- Robust SiC-Si<sub>3</sub>N<sub>4</sub> microstructure optimization for desired high temperature properties.
- Variable-fidelity model management for  $\mu$ -CFEM

### Processing and Experimental Validation

- New Processing strategies for SiC-Si<sub>3</sub>N<sub>4</sub> with engineered microstructure.
- Nanoscale to Macroscale Mechanical testing protocols for validation.

### Multiscale Modeling and Experiments

- MD simulations have provided fundamental insights in SiC-Si<sub>3</sub>N<sub>4</sub> composite failure mechanisms
- A universal fracture energy-material strength relation to predict microstructural fracture has been developed
- New nanoscale and microscale creep test protocols provide new insights into high temperature small scale material properties

### Stochastic Sampling

- An eigenvector sampling scheme (ETS) has been incorporated to reduce the number of samplings in the robust optimization method.

### Composite Processing

- Development of processing approaches to make a broad range of SiC-Si<sub>3</sub>N<sub>4</sub> composites.
- Effect of atmosphere and polymer composition of phase evolution

### MAIN ACHIEVEMENTS:

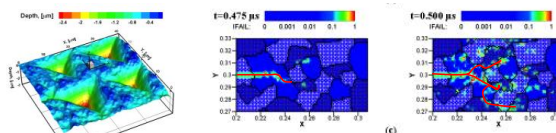


Fig. 1. New small scale creep experiments and microstructure dependent fracture models.

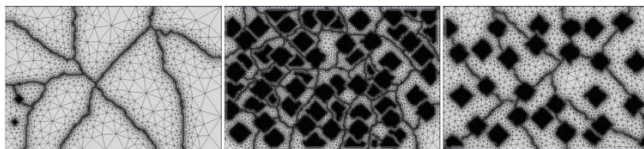


Fig. 2. Design Algorithms Have Predicted SiC-Si<sub>3</sub>N<sub>4</sub> Microstructures with Optimal Strength in Temperature Range 1500 to-1600 °C.

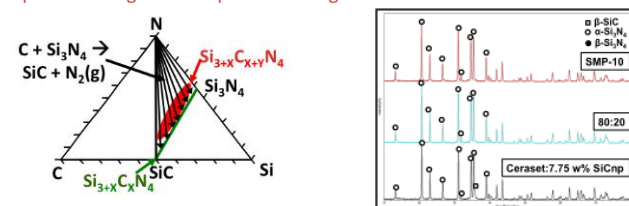


Fig. 3. Effect of polymer composition (let) and atmosphere (right) on the composition of the resultant composite. The ratio of C:N in the polymer and the pyrolysis atmosphere determines the phases in the ceramic.

### HOW IT WORKS:

- New protocols in creep experiments and microstructure based fracture models enable explicit microstructure dependent failure information to design models (Figs. 1 and 2).
- Ability to make a broad range of composition by controlling the polymer composition and pyrolysis atmosphere (Fig. 3).

### ASSUMPTIONS AND LIMITATIONS:

- Operating conditions are considered deterministic parameters in the optimization stage.
- The fixed assist sintering approach needs to be optimized to make high density composites

### Current Impact

- Simulation, analysis, and design method for optimal SiC-Si<sub>3</sub>N<sub>4</sub> microstructure established.

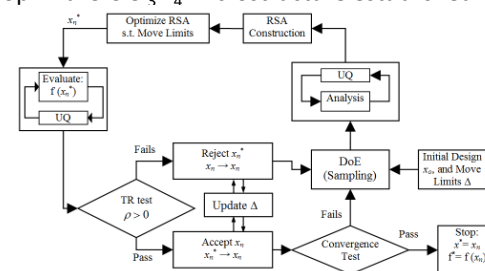


Fig. 4. Robust microstructure optimization algorithm.

- Processing and testing protocols have been established

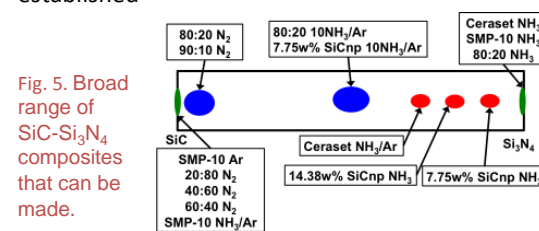


Fig. 5. Broad range of SiC-Si<sub>3</sub>N<sub>4</sub> composites that can be made.

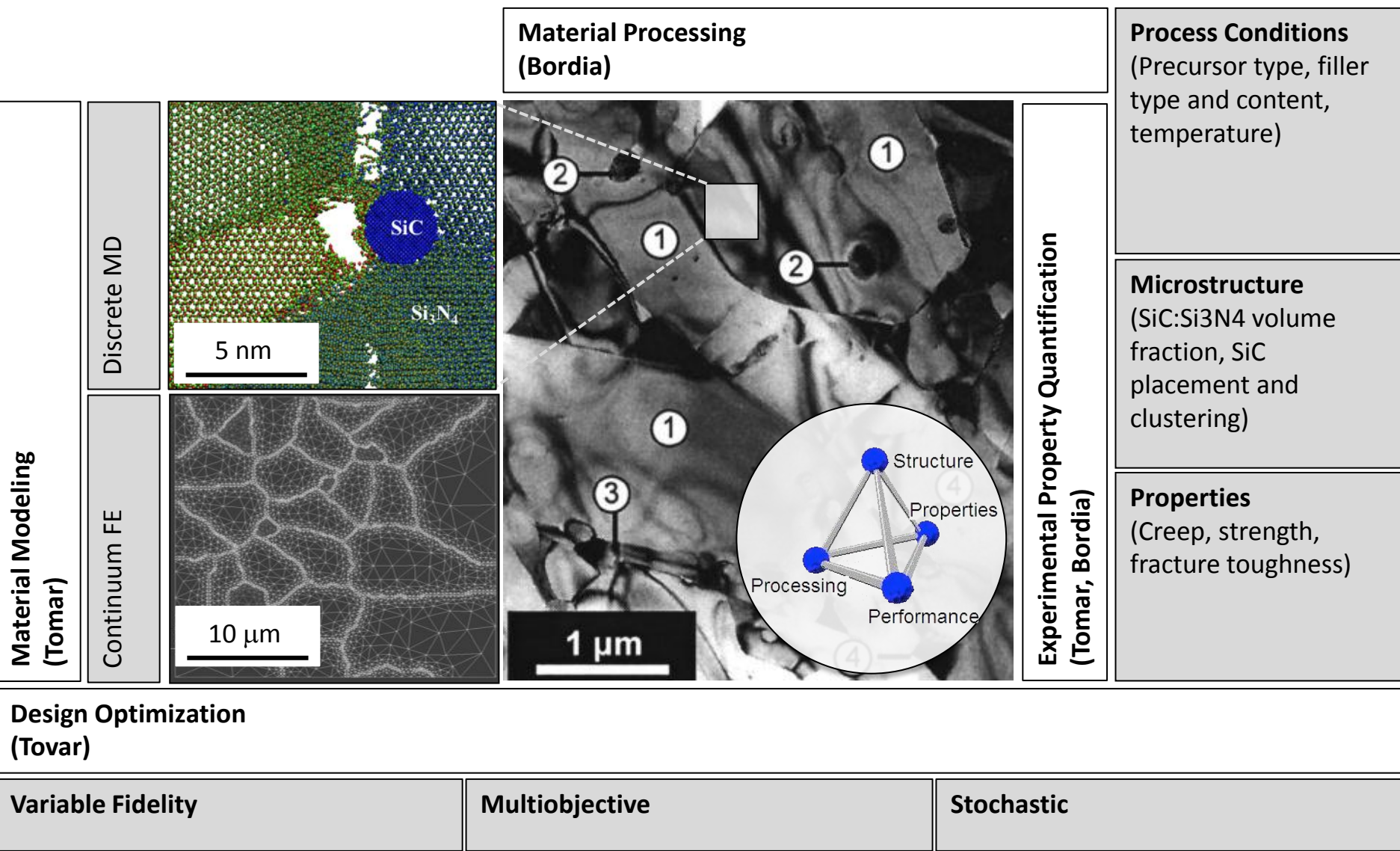
### Planned Impact

- A robust design optimization capable of predicting optimal SiC-Si<sub>3</sub>N<sub>4</sub> 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.

# Project Overview



**Collaborative Simulation and Experimental Development of SiC-Si<sub>3</sub>N<sub>4</sub> Microstructures**

# Outline of the research at IUPUI

## ❖ **Material design paradigm**

- Problem formulation
- Optimization methods and challenges

## ❖ **Microstructural simulation**

- Composite lattice model
- Finite element model

## ❖ **Variable fidelity optimization**

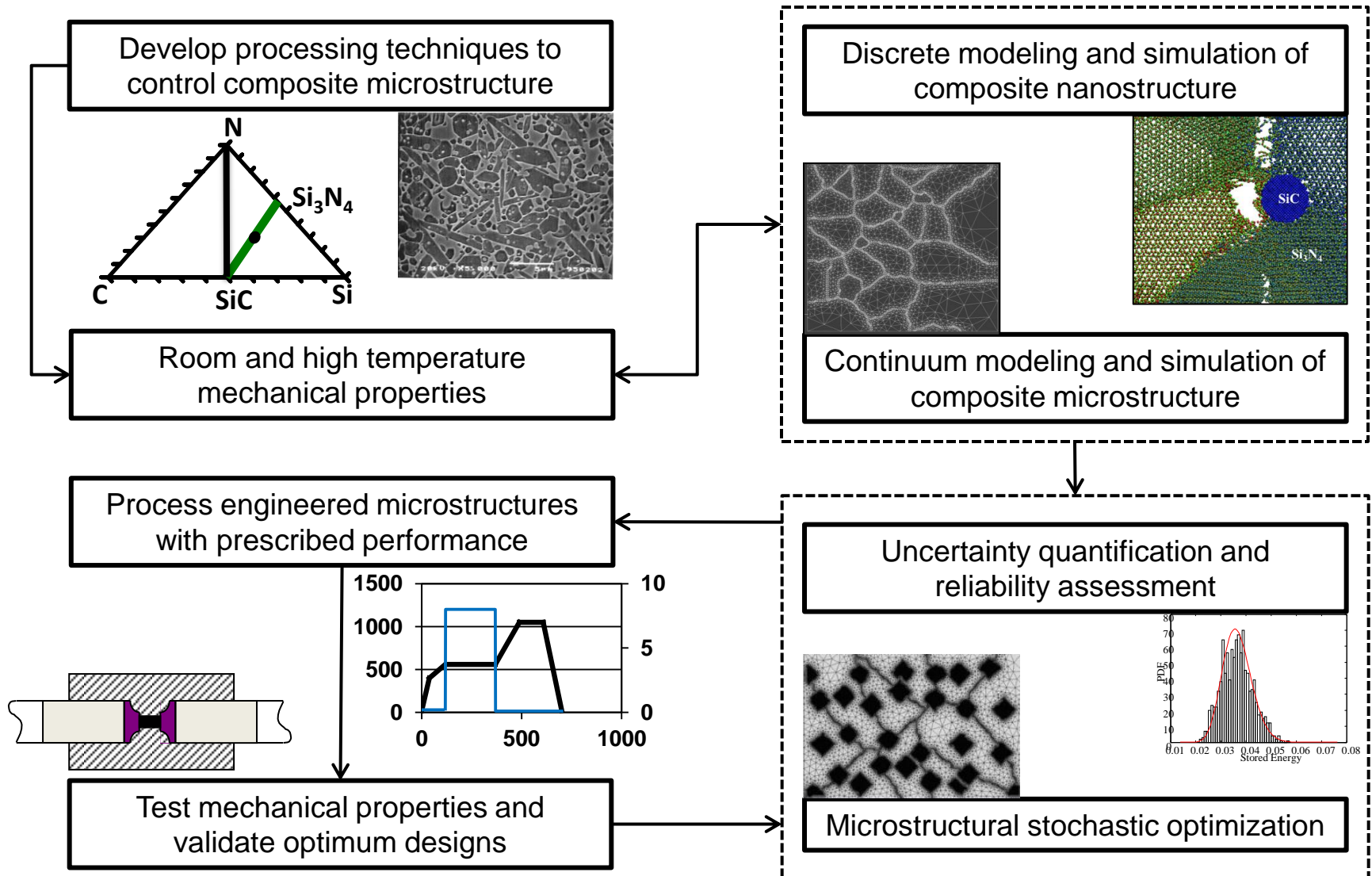
- Scaling and trust region management
- Application in composite design

## ❖ **Robust optimization**

- Uncertainty quantification
- Application in composite design

## ❖ **Final remarks**

# Simulation Guided Materials Development



Predictive tool to design composite microstructures with optimized properties

# 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
- ❖ SiC-Si<sub>3</sub>N<sub>4</sub> composites

		α-SiC:	β-Si <sub>3</sub> N <sub>4</sub> :
Creep resistance (High Temp.)		✓	✗
Strength	Room Temp.	✗	✓
	High Temp.	✓	✓
Toughness (High Temp.)		✗	✓

A trade-off of material properties must be made to achieve an optimal high temperature microstructure.

# Characteristics of SiC and Si<sub>3</sub>N<sub>4</sub>

## SiC

Creep resistance better than that of most materials. (low toughness)

Low density

High strength

Good high temperature strength (Reaction bonded)

Oxidation resistance (Reaction bonded)

Excellent thermal shock resistance

High hardness and wear resistance

Excellent chemical resistance

Low thermal expansion and high thermal conductivity

Electrical conductivity

### **Typical applications include:**

Fixed and moving turbine components

Seals, bearings, pump vanes

Ball valve parts

Wear plates

Kiln furniture

Heat exchangers

## Si<sub>3</sub>N<sub>4</sub>

Of all high-temperature ceramic materials with second phase at grain boundary, it is the toughest and strongest

Good thermal shock resistance

Good high temperature strength

Creep resistance

Low density

High fracture toughness

High hardness and wear resistance

Electrical resistivity

Good chemical resistance

Good oxidation resistance and not wetted by molten metals

### **Typical applications include:**

Bearing balls and rollers

Cutting tools

Valves, turbocharger rotors for engines

Turbine blades

Glow plugs

Molten metal handling

(Wiederhorn et al., 1999)

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

# Traditional optimization problem

**find** Composite  
architecture /  
Processing parameters

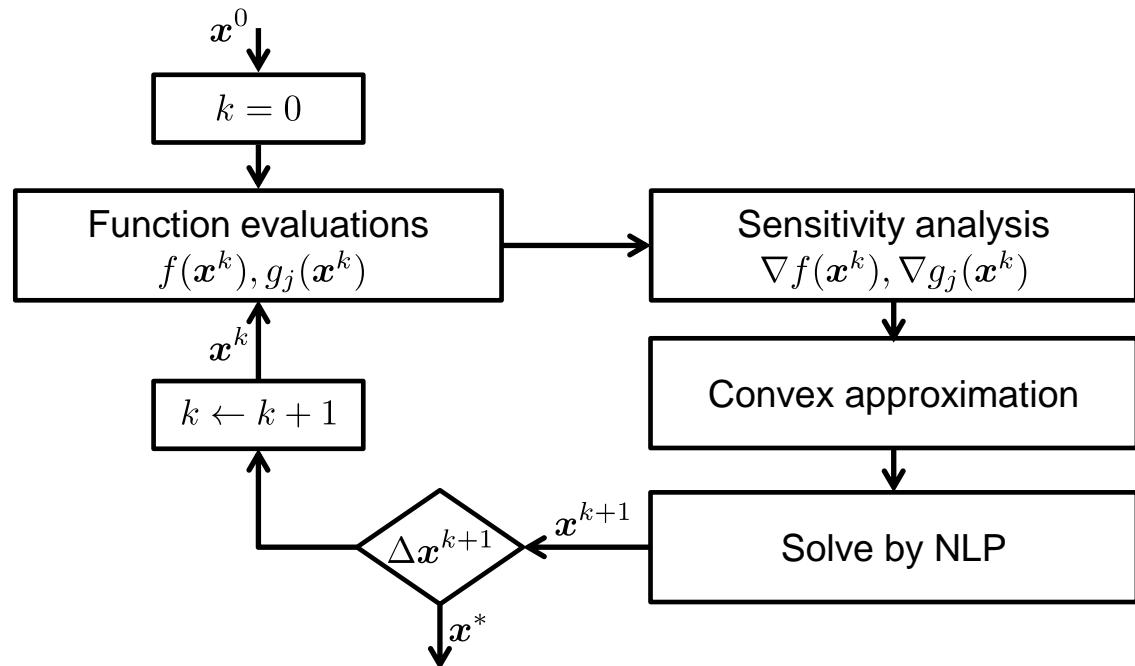
**maximize** Material  
performance

**subject to** Functional  
and geometric  
constraints

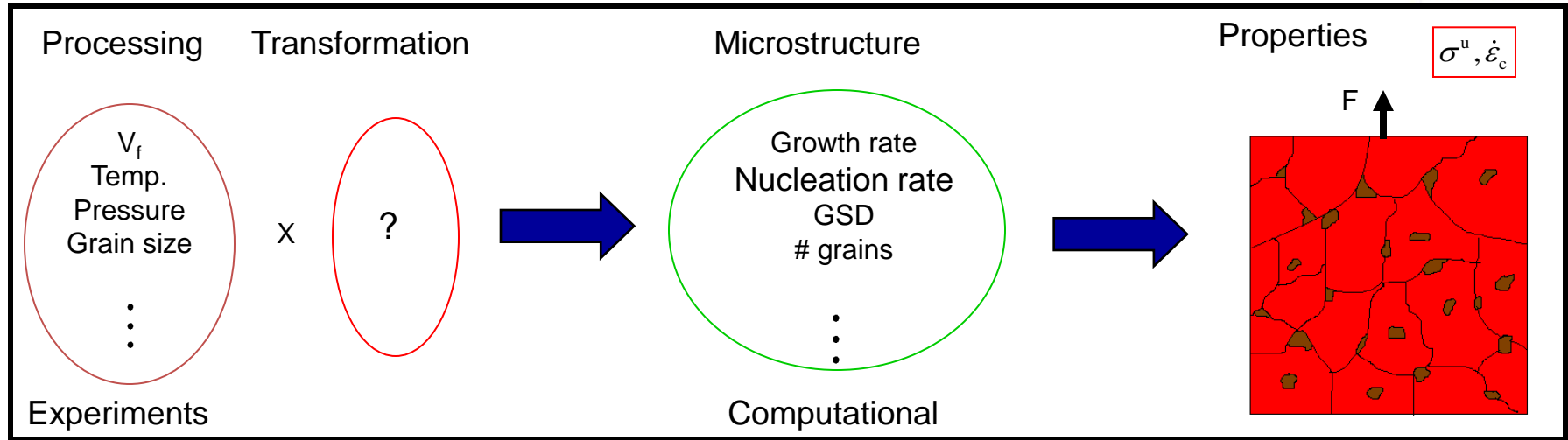
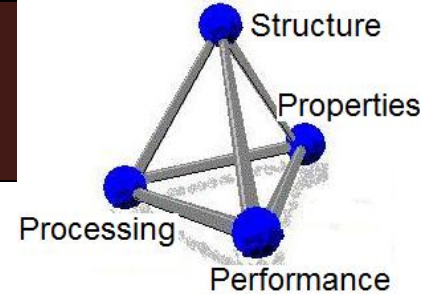
The optimization problem is

$$\begin{array}{ll}\text{find} & \mathbf{x} \in \mathbb{R}^n \\ \text{maximize} & f(\mathbf{x}) \\ \text{subject to} & \mathbf{g}(\mathbf{x}) \leq \mathbf{0} \\ & \mathbf{x}^l \leq \mathbf{x} \leq \mathbf{x}^u,\end{array}$$

The objective  $f$  and constraints  $\mathbf{g}$  are function of design variables  $\mathbf{x}$ .



# Material design paradigm



Processing  
variation



Microstructure  
variation



Properties &  
Microstructure  
variation

## Robust Material Design

Find  
Optimal robust  
design variables

Maximum:  $\sigma^u$

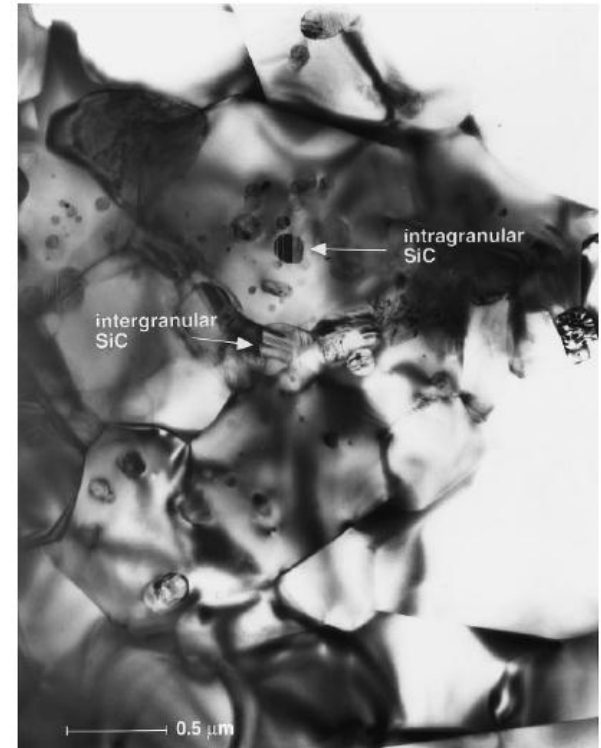
Minimum:  $\dot{\epsilon}_c$

Reduce variation

$\Delta\sigma^u, \Delta\dot{\epsilon}_c$

# Challenges in material design

- ❖ A wide **disagreement** has been observed in the effect of the nanosized reinforcements on the SiC-Si<sub>3</sub>N<sub>4</sub> nanocomposite properties.
- ❖ Large **properties variations** are due to:
  - Different testing techniques (3 pt. vs. 4 pt.).
  - Sample preparation methods (extent of polishing).
  - Sample size effect.
- ❖ **Systematic studies** are needed to investigate and understand the effect of reinforcement size and volume fraction on the microstructure, processing and properties of these nanocomposites.

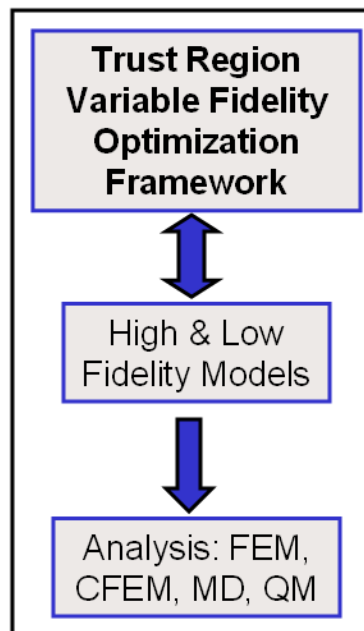


**SiC-Si<sub>3</sub>N<sub>4</sub> composite**  
(Weimer and Bordia, 1999)

# Material design methodology

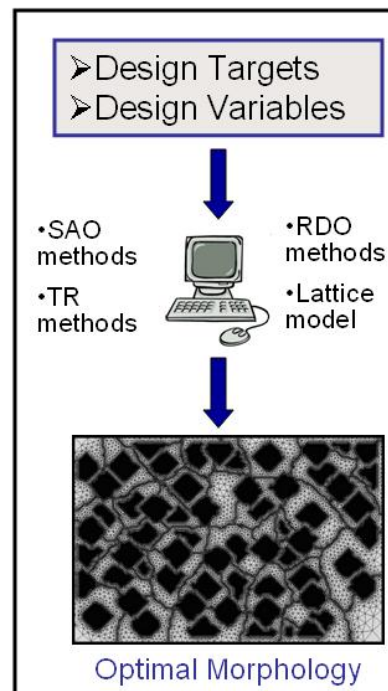
## Variable fidelity optimization (VFO)

- Address computational challenges.
- Address model management issues.



## Robust design optimization (RDO)

- Systematic method to synthesize optimal microstructures.
- Save time and cost.
- Composite materials tailoring.
- Simulation-based robust material design optimization methodology.



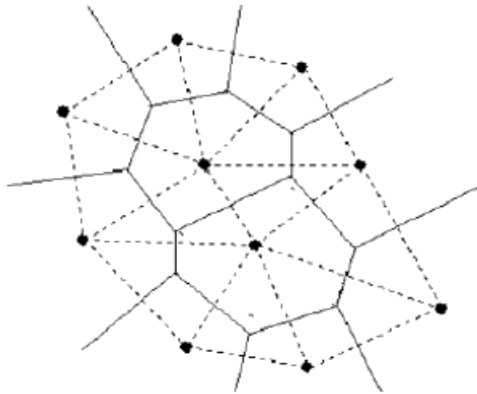
(Mejía-Rodríguez et al., 2008, 2010)

Composite materials can be tailored using a sequential approximate optimization framework

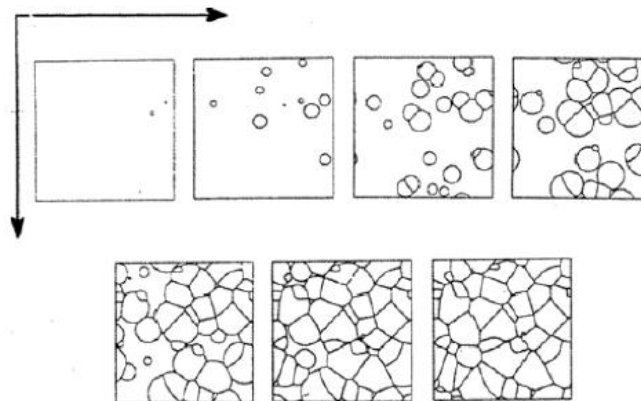
# Microstructural simulation techniques

- ❖ Predicting material behavior.
- ❖ Understanding the relationship between microstructure and properties.
- ❖ Simulate microstructure evolution:

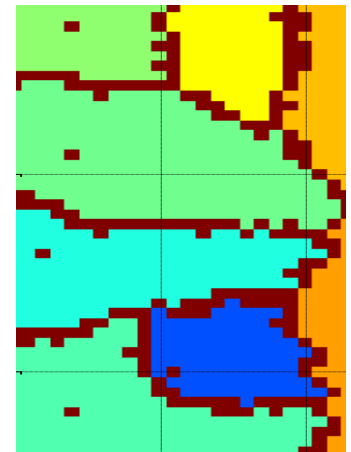
## ➤ Voronoi network



## ➤ Johnson-Mehl



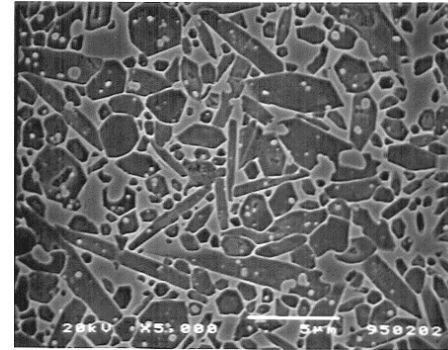
## ➤ Lattice model



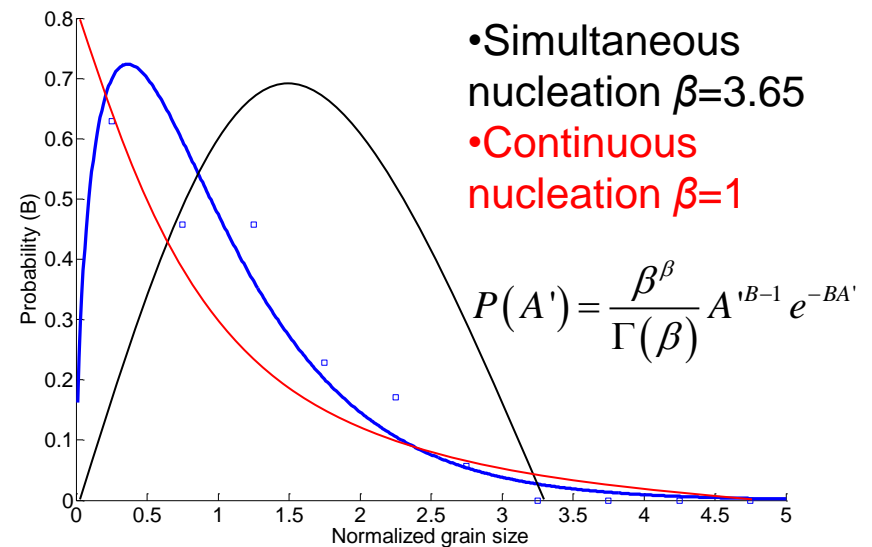
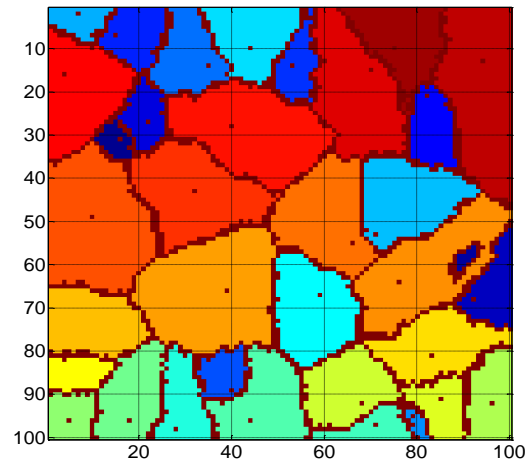
# Lattice model

## ❖ Nucleation and growth mechanisms

- Initial nucleation
- Nucleation rate



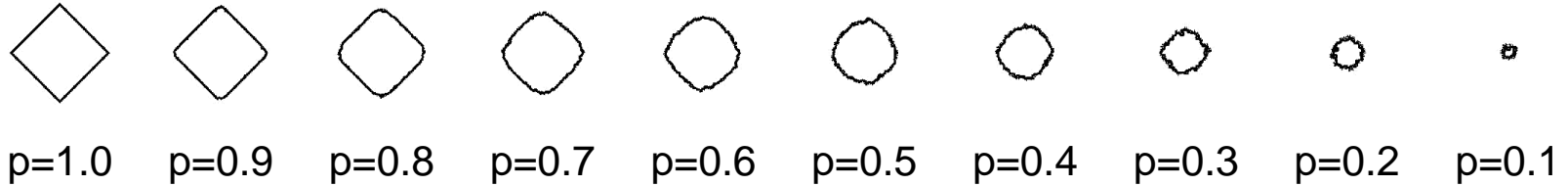
Target material:  
 $\text{SiC-Si}_3\text{N}_4$



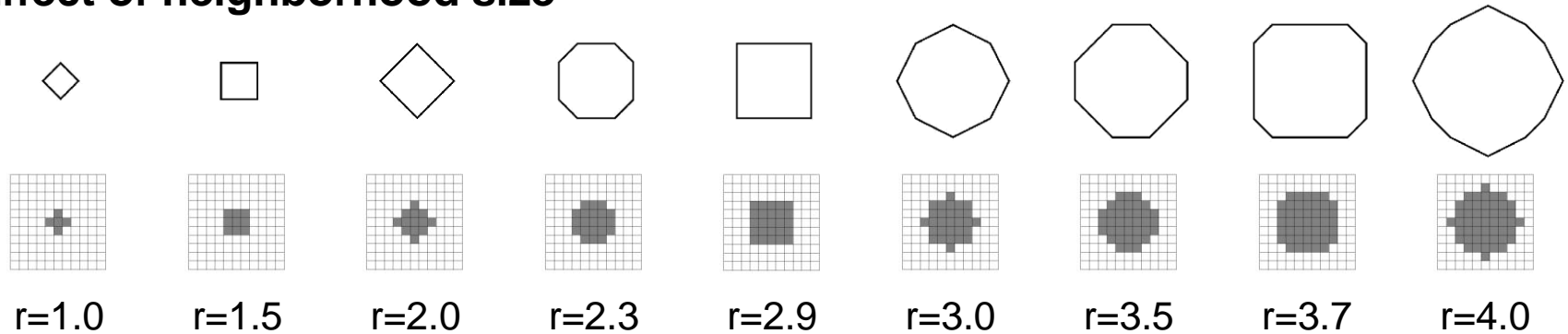
Probability density function (PDF) of Gamma distribution depending on the grain size distribution parameter  $\beta$

# Lattice model

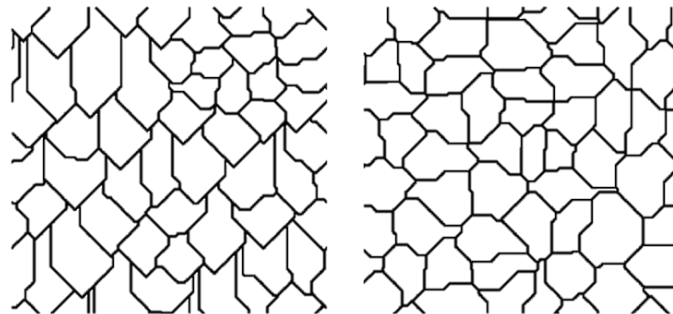
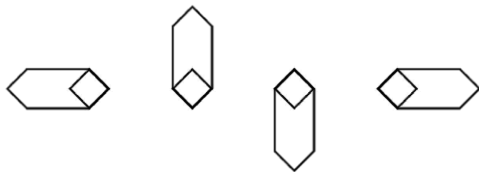
## Effect of the probability of growth



## Effect of neighborhood size

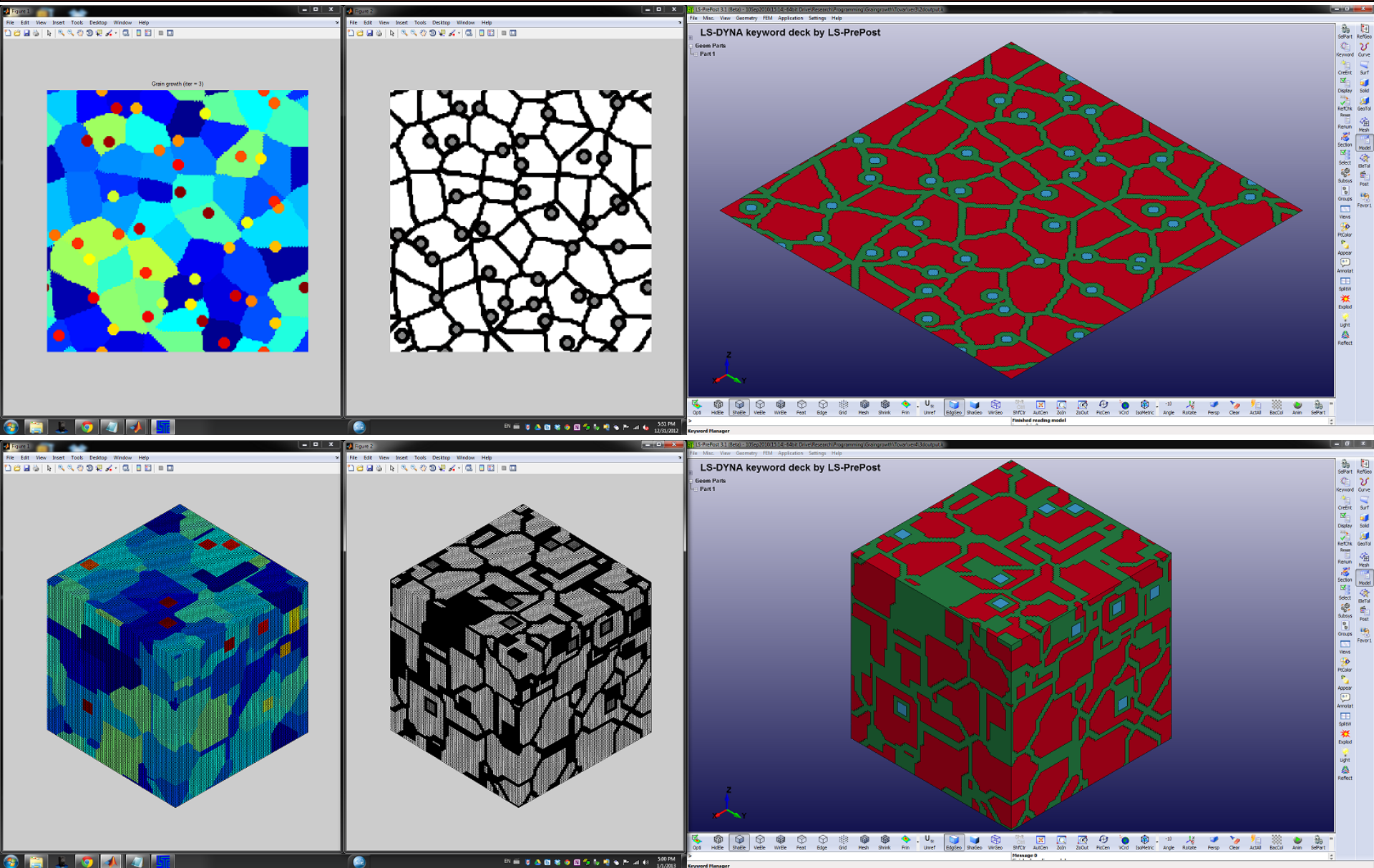


## Effect of bias



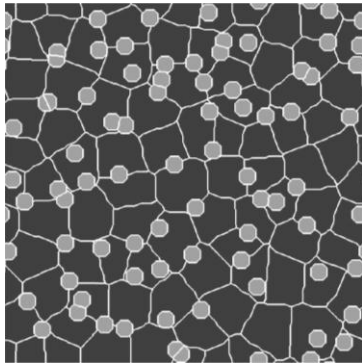
Mesh 500x500,  $p=1.0$ ,  $r=1.0$ , (left) with and (right) without bias.

# Simulation

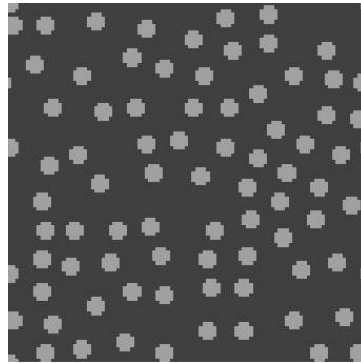


# Why does fidelity matter?

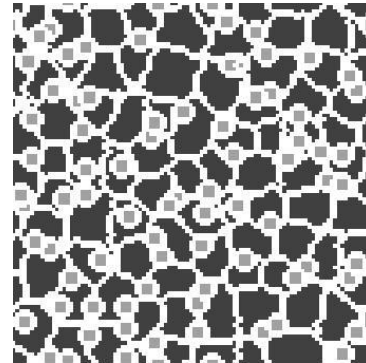
- ❖ What is the difference between high and low fidelity models?
  - Fewer assumptions in model
  - More physics
  - Higher resolution of computational meshes
- ❖ High fidelity simulations yield more accurate results
- ❖ High fidelity simulations are expensive to compute
  - Therefore, it is desired to reduce the number of these simulations



HF: 1kx1k



LF: 100x100

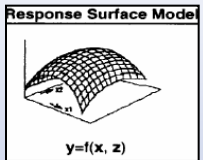


# Variable fidelity optimization

LOW FM

HIGH FM

## PROBLEM 1

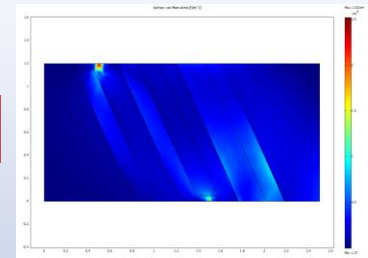


$$K_I = \frac{PS}{BW^{3/2}} f\left(\frac{a}{W}\right)$$

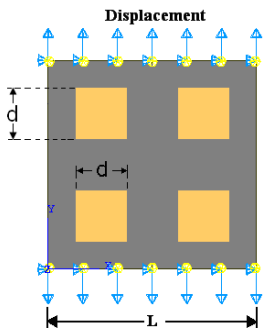
Analytical  
Fracture  
Mechanics &  
Surrogate  
Modeling

↔  
Fracture  
toughness

FEM (Comsol)



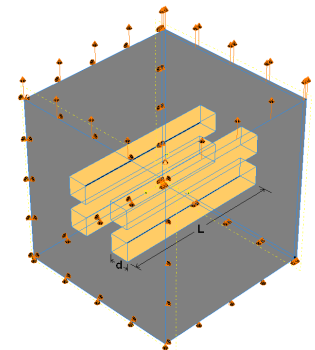
## PROBLEM 2



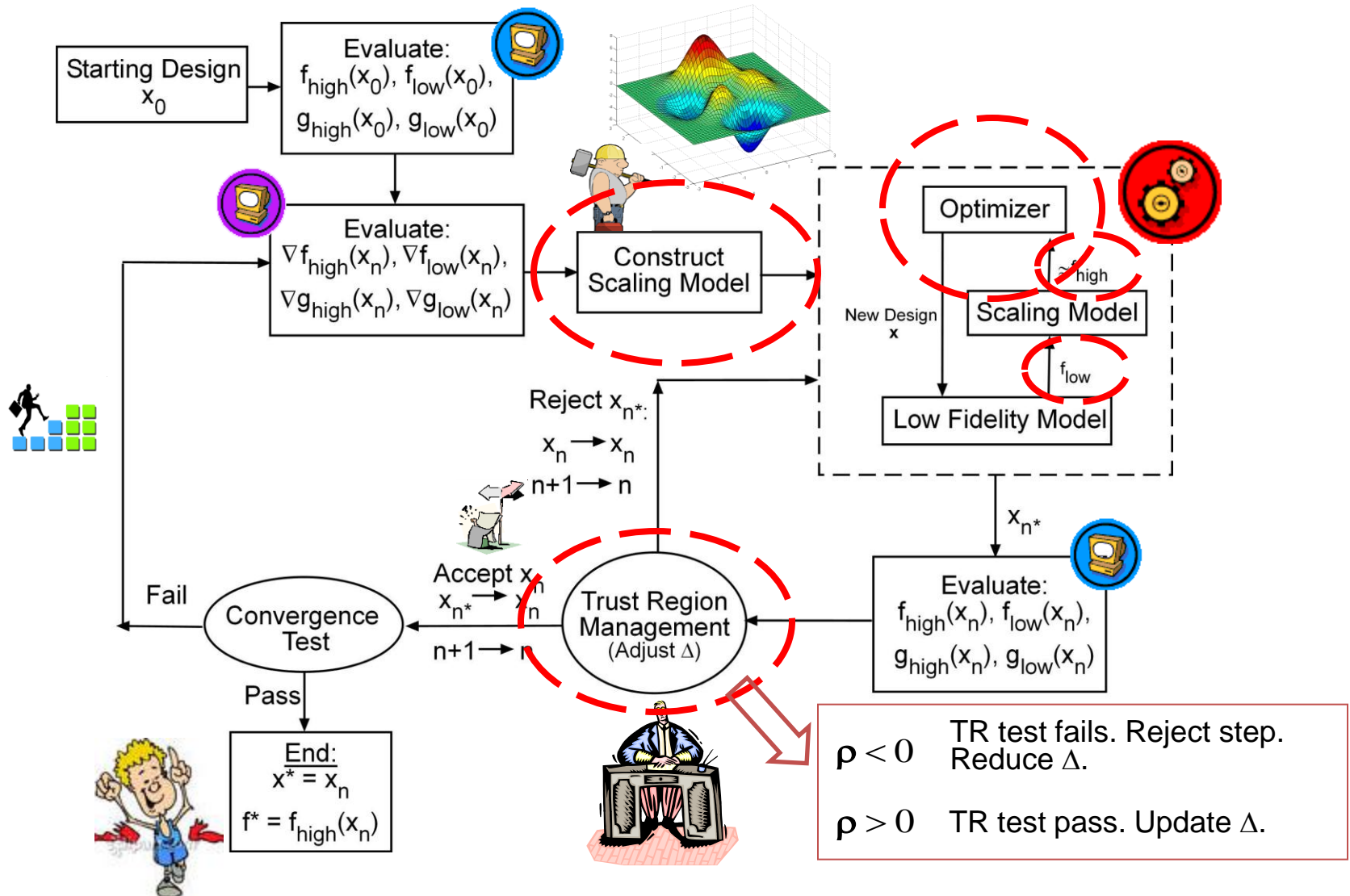
FEM (ABAQUS)  
2D Model

↔  
Strength &  
Creep  
strain  
resistance

FEM (ABAQUS)  
3D Model

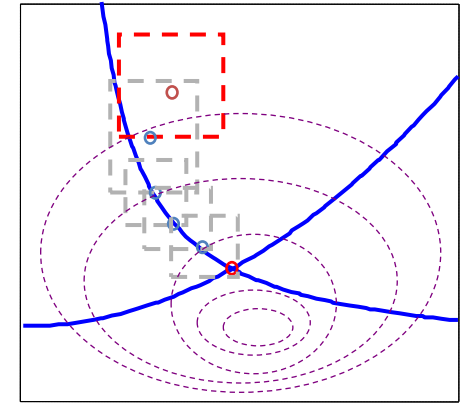


# Variable fidelity optimization



# Trust region model management

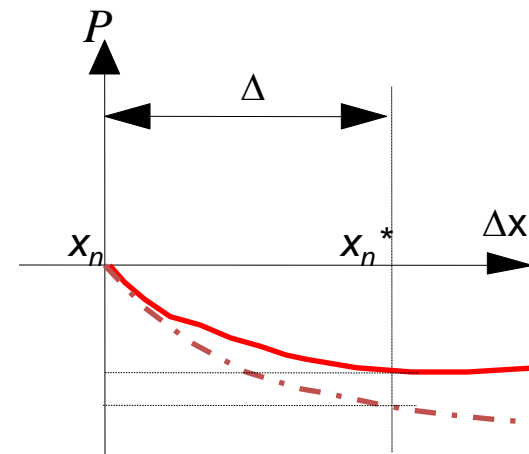
- ❖ Provides a mean for **adaptively managing** the allowable **move limits**.
- ❖ Each optimization finds a new candidate point  $x_n^*$  within the trust region radius  $\Delta$ .
- ❖ Trust Region ratio  $\rho$  : Physically represents **how good** of an **approximation** the **scaled low fidelity model** is **compared** to the **high fidelity model**.



$$\|x_n^* - x_n\|_{\infty} \leq \Delta_n$$

$$\rho_n = \frac{P(x_n)_{high} - P(x_n^*)_{high}}{P(x_n)_{scaled} - P(x_n^*)_{scaled}}$$

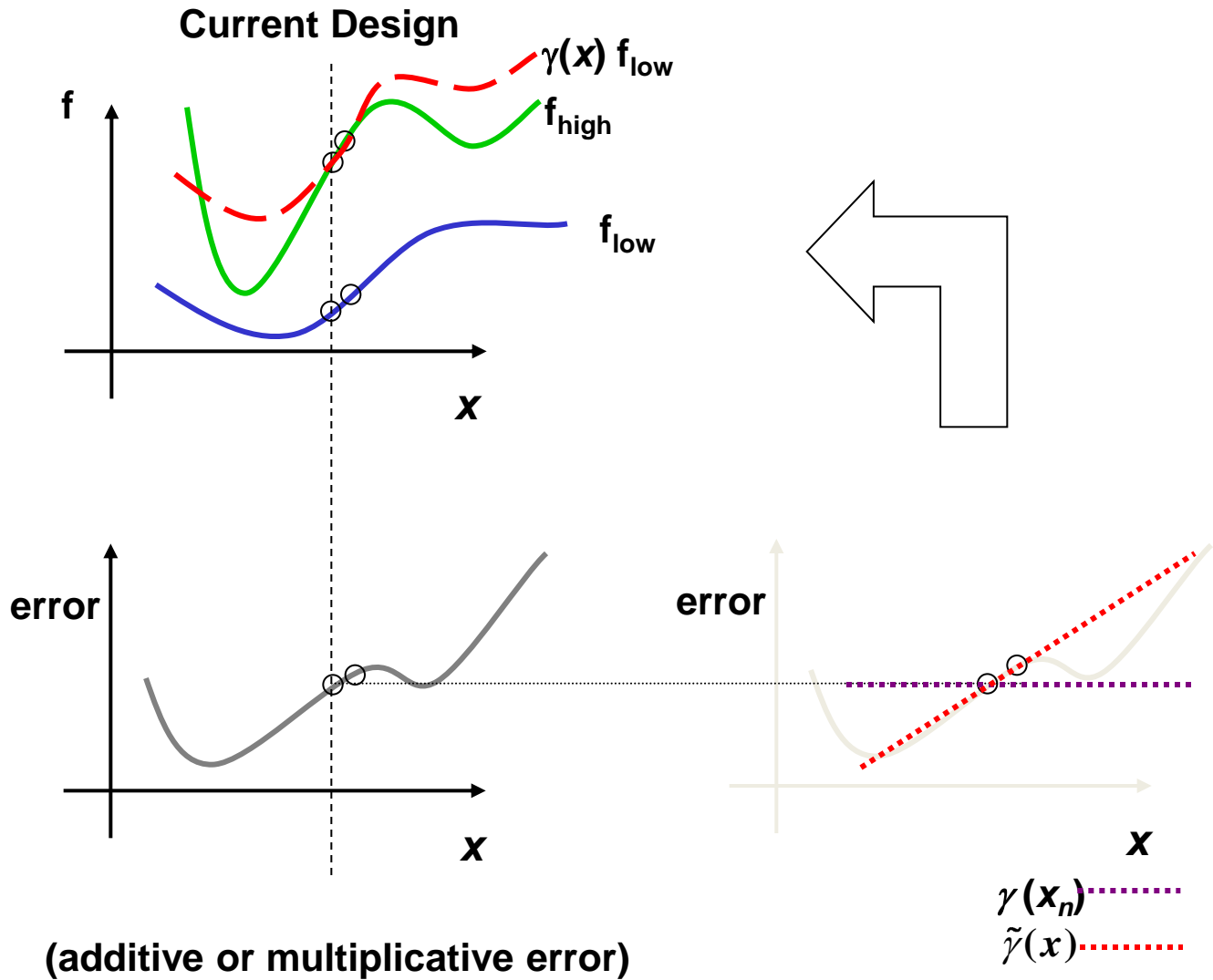
$$P = f_n + \frac{1}{\mu_n} \sum \max(0, g_i) + \frac{1}{\mu_n} \sum |h_j|$$



$$P(x_n^*)_{high}$$

$$P(x_n^*)_{scaled}$$

# First order scaling



# Variable fidelity optimization for fracture toughness

**find** optimal CFCC configuration

**max** 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

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

Design Target

Fracture resistance

Design Variables

- Second Phase orientation angle  $\theta$
- Second Phase Volume Fraction

Variable Fidelity Optimization

Simulations

Optimal Morphology

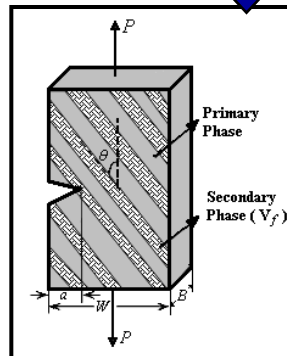
**maximize** Mode-I  $K_{IC}(V_f, \theta)$

**subject to:**  $K_I \leq K_{IC}(V_f, \theta)$

$$0 \leq V_f \leq 0.2$$

$$0^\circ \leq \theta \leq 90^\circ$$

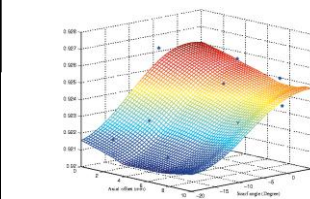
Design load  $\rightarrow K_I$   
93500 N



High fidelity (HF) model

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

Low fidelity (LF) model



Response surface approximation for  $K_{IC}$

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

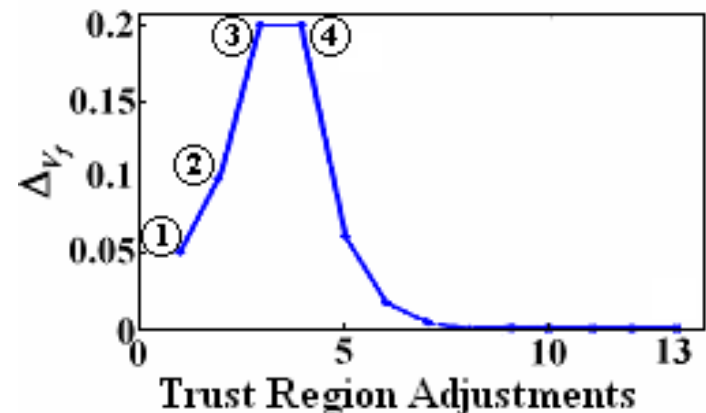
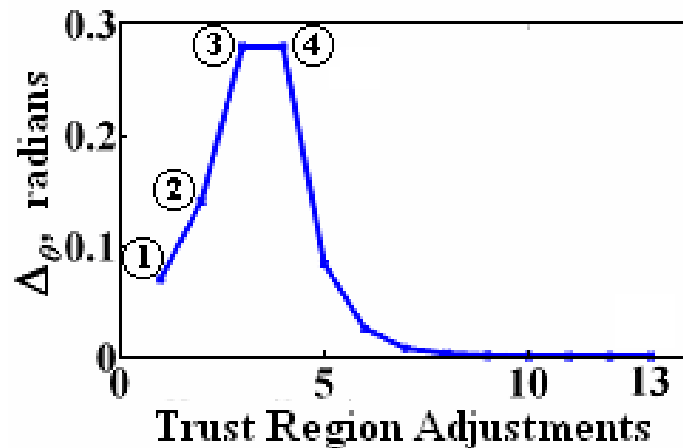
Analytical fracture mechanics for  $K_I$

# Variable fidelity optimization for fracture toughness

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

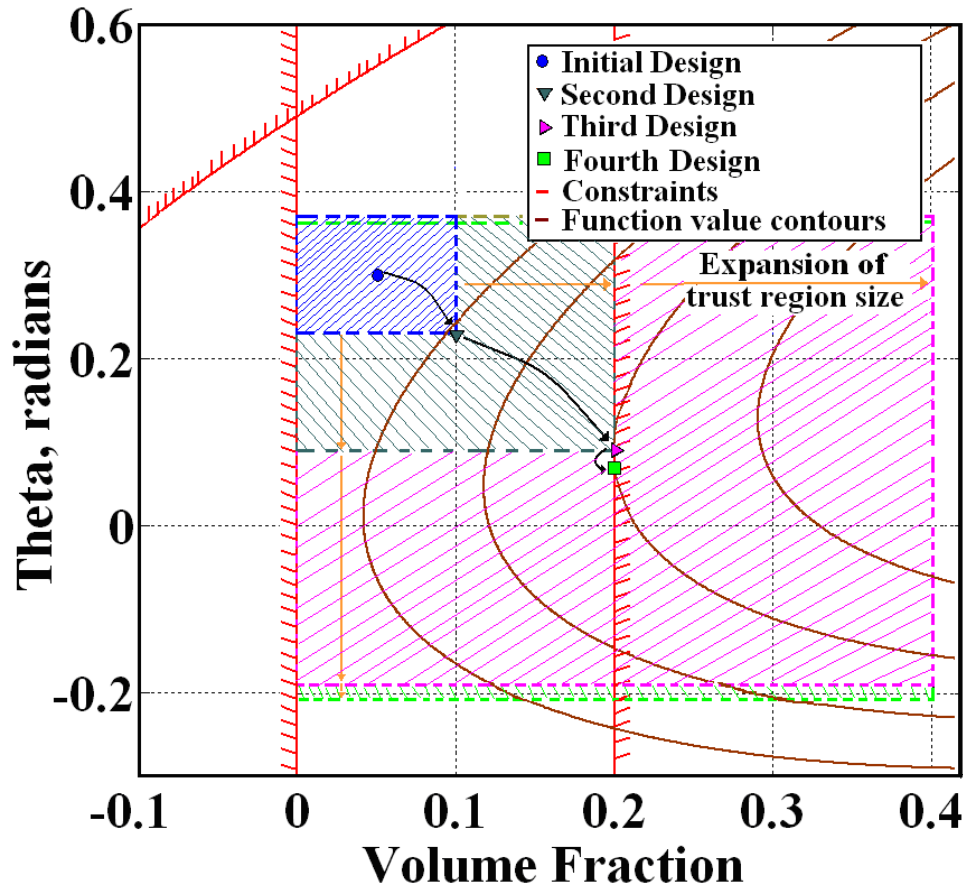
80% reduction in HF simulations

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



Iteration	Trust Region Adjustments	Trust Region Size		Convergence		Design Variables and Obj. Function		
		$\Delta\theta$	$\Delta V_f$	$\Delta_x$	$\Delta_f$	$\theta$ (rad.)	$V_f$	$K_{IC}$ (MPa·m <sup>1/2</sup> )
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

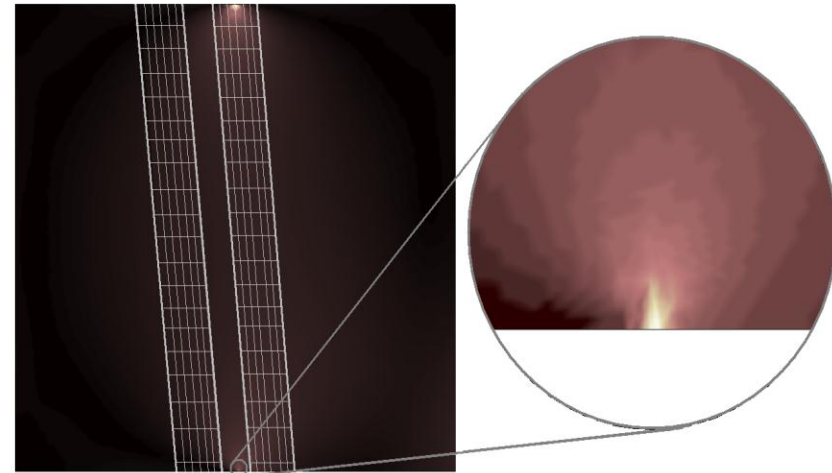
# Variable fidelity optimization for fracture toughness



First three iterations of variable fidelity framework

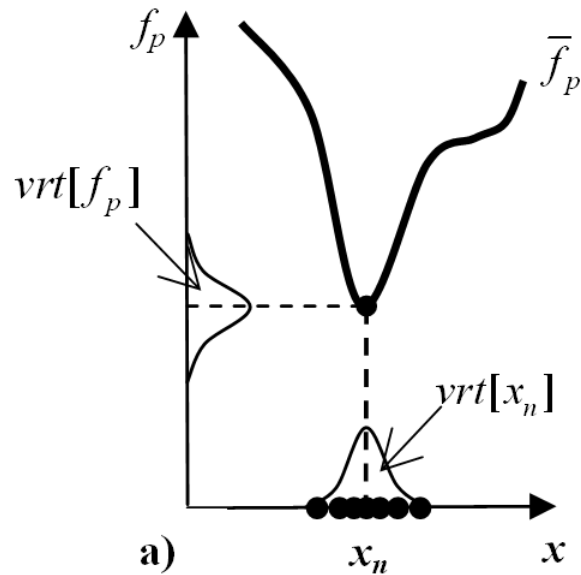
Von Mises Stress,  $\text{N/m}^2$

Min: 1372      1      2      3      4      5      6      7      Max:  $7.8 \times 10^8$



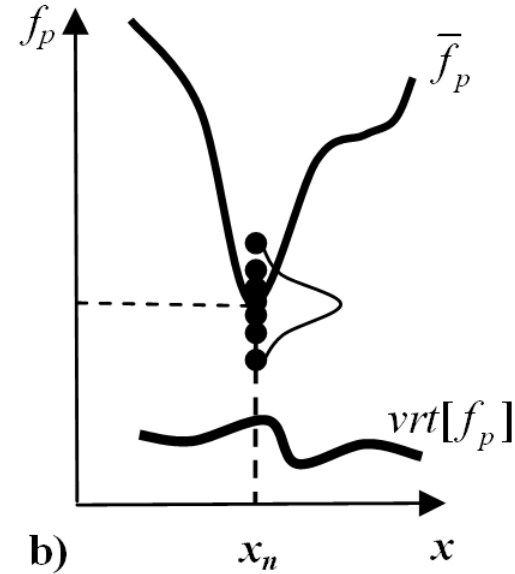
Von Mises stress distribution around the single edge crack tip

# Non-deterministic function



Non-deterministic function with uncertainty in design variables.

(Su and Renaud, 1997)



Non-deterministic function with no uncertainty in design variables.

# Robust optimization problem

**find** Composite  
architecture /  
Processing parameters

**maximize** Mean  
material performance  
and minimize variance

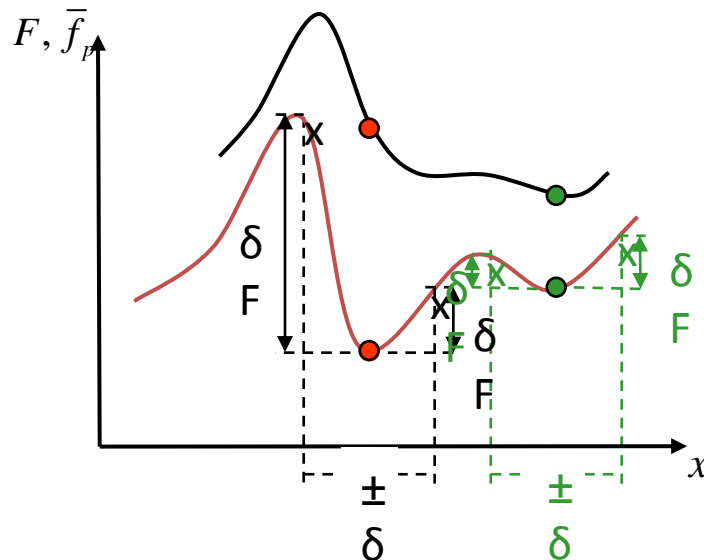
**subject to** Functional  
and geometric  
constraints

The optimization problem is

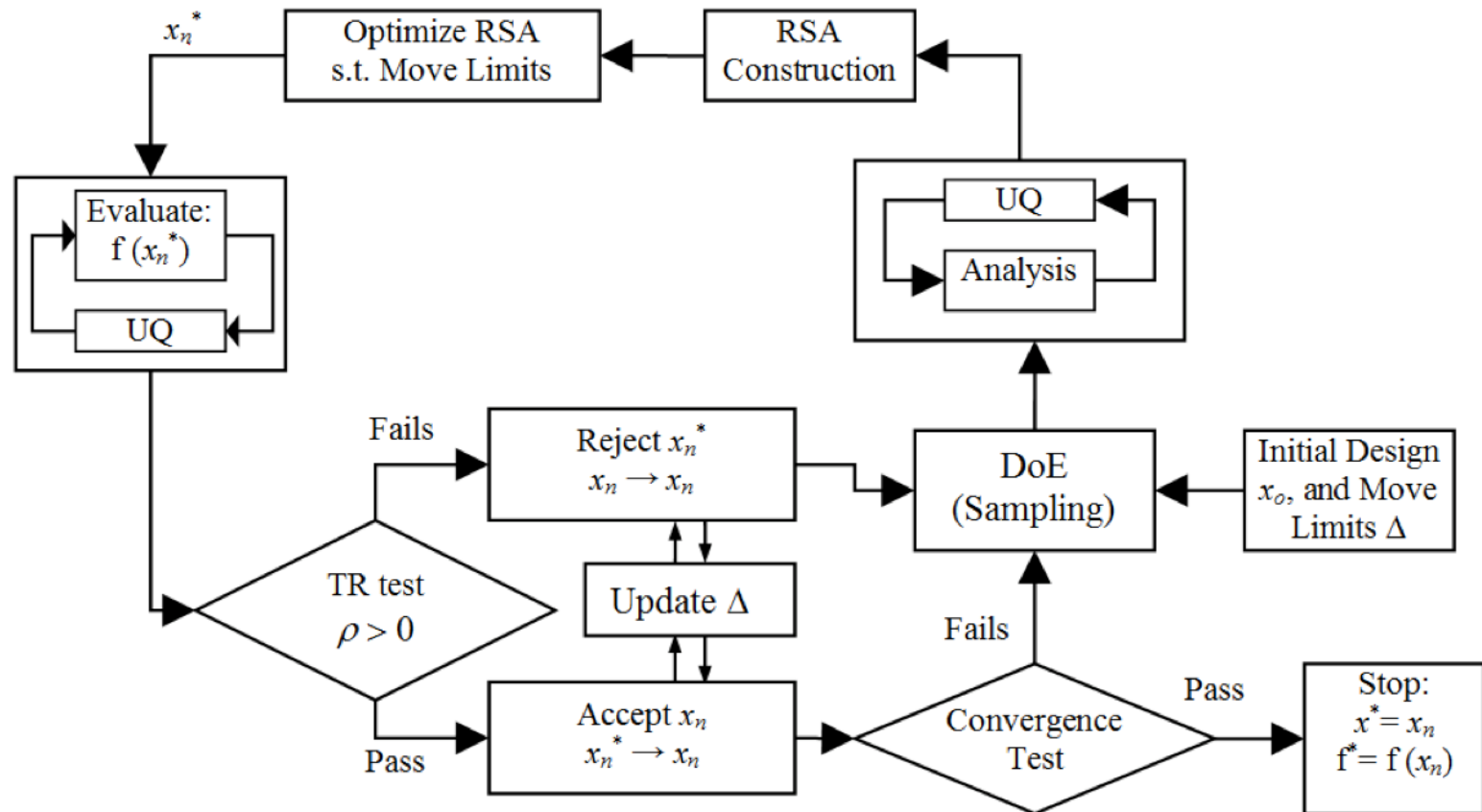
$$\begin{aligned} &\text{find} && \mathbf{x} \in \mathbb{R}^n \\ &\text{maximize} && F(\mathbf{x}) = \alpha \bar{f}_p(\mathbf{x}) - (1 - \alpha) \text{vrt} [f_p(\mathbf{x})] \\ &\text{subject to} && \mathbf{g}(\mathbf{x}) \leq \mathbf{0} \\ &&& \mathbf{x}^l \leq \mathbf{x} \leq \mathbf{x}^u, \end{aligned}$$

where

$$\text{vrt} [f_p(\mathbf{x})] = \sqrt{\frac{1}{ns} \sum_{i=1}^{ns} (f_{pi} - \bar{f}_p)^2}$$



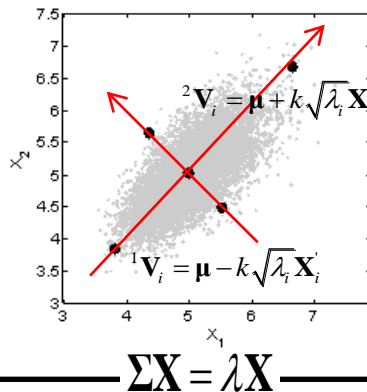
# Robust optimization problem



# Dimension reduction

## Eigenvector sampling

Least number of samples



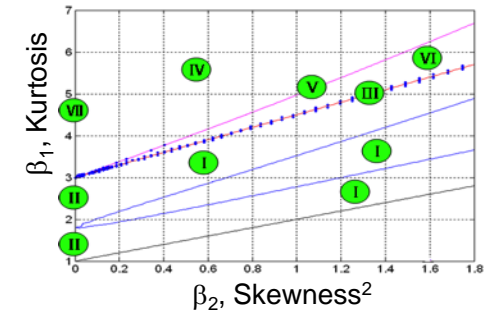
## Multi 1D integration

After additive decomposition

- Stepwise moving least squares (SMLS) method
- Adaptive numerical integration scheme

## Stabilized Pearson System

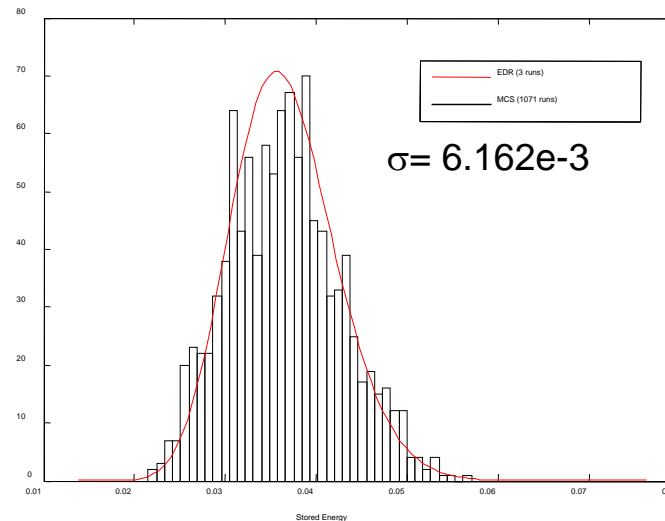
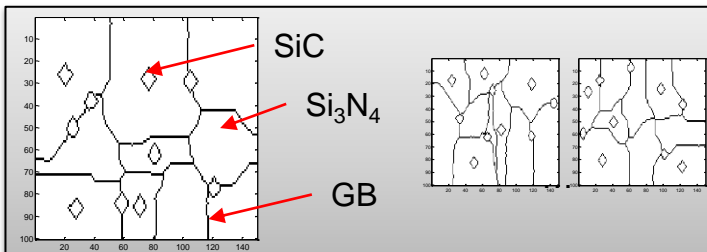
PDF generation from statistical moments



## UQ of Stored Energy for Temperature variation

$$S = S_0 \left( 1 + \frac{\epsilon}{\epsilon_0} \frac{\partial \ln \epsilon}{\partial \ln T} \right) \exp \left( \frac{Q}{RT} \right)$$

$T \sim \text{Normal}(1550, 100^2)$



MCS: >1000  
EDR; 3

# Robust material optimization

**find**

$x_1 \in \mathbb{N}$ : Total Si<sub>3</sub>N<sub>4</sub> grains

$x_2 \in \mathbb{N}$ : Inter-granular SiC grains

$x_3 \in \mathbb{N}$ : Intra-granular SiC grains

$x_4 \in \mathbb{R}$ : Initial nucleation of Si<sub>3</sub>N<sub>4</sub> grains

$x_5 \in \mathbb{R}$ : Nucleation rate of Si<sub>3</sub>N<sub>4</sub> grains

$x_6 \in \mathbb{R}$ : Growth of Inter-granular SiC grains

$x_7 \in \mathbb{R}$ : Growth of Intra-granular SiC grains

**maximize**

$$F_{MO}(\mathbf{x}) = \sum_i \omega_i F_{Ti}(\mathbf{x})$$

where

$$F_{Ti}(\mathbf{x}) = \alpha \bar{f}_p(\mathbf{x}) - (1 - \alpha) vrt[f_p(\mathbf{x})] \text{ at } T^\circ = T_i$$

and  $f_p$  is the deformation energy

**subject to**

$$10 \leq x_1 \leq 40$$

$$1 \leq x_2 \leq 25$$

$$1 \leq x_3 \leq 40$$

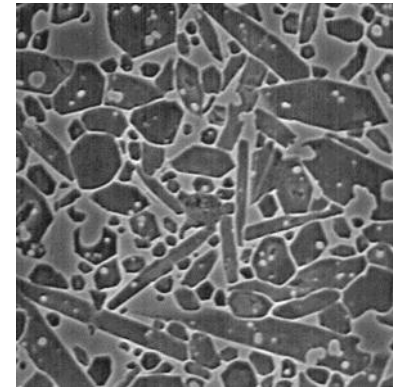
$$1 \leq x_4 \leq x_1$$

$$1 \leq x_5 \leq 10$$

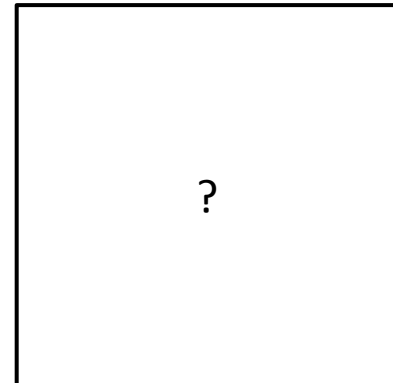
$$1 \leq x_6 \leq 8$$

$$1 \leq x_7 \leq 8$$

5  $\mu\text{m}$



Initial microstructure  
(Bordia's group)



Optimal microstructure

# Robust material optimization

maximize

$$F_{MO}(\mathbf{x}) = \omega F_{T_1}(\mathbf{x}) + (1 - \omega) \omega F_{T_2}(\mathbf{x})$$

where

$$T_1 = 1500^\circ\text{C}$$

$$T_2 = 1600^\circ\text{C}$$

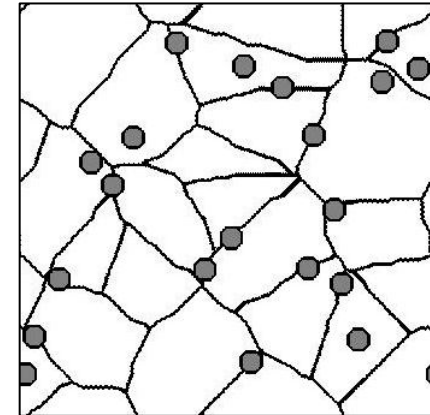
$$\omega = 1.0$$

Quadratic least square  
response surface

$$\tilde{T}(z) = a_0 + \sum_{i=1}^n a_i z_i + \sum_{i=1}^n \sum_{j=1}^n a_{ij} z_i z_j$$

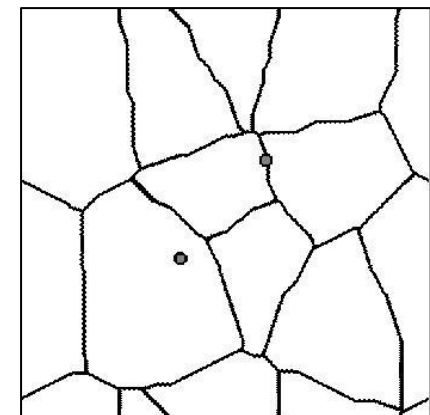
Fractional factorial design  
sampling

(get  $\mu$  and  $\sigma$  for each  
sample)



Initial microstructure

[20, 10, 10, 4, 4, 5, 5]



Optimal microstructure

[10, 1, 1, 1, 4, 3, 2]

# Robust material optimization

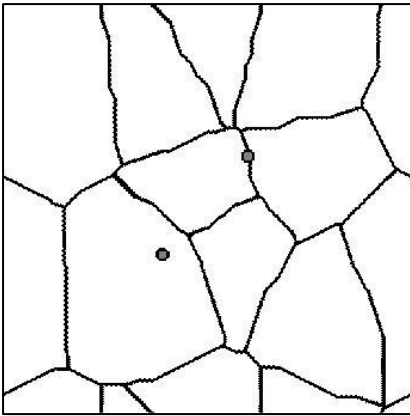
maximize

$$F_{MO}(x) = \omega F_{T1}(x) + (1 - \omega) \omega F_{T2}(x)$$

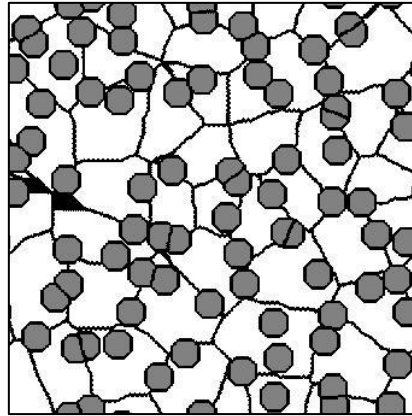
where

$$T_1 = 1500^{\circ}\text{C}$$

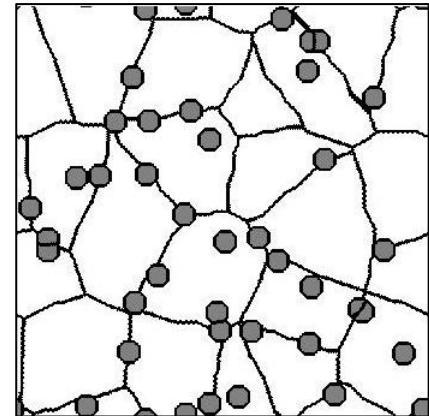
$$T_2 = 1600^{\circ}\text{C}$$



$$\omega = 1.0$$



$$\omega = 0.0$$



$$\omega = 0.5$$

# Final remarks

## Material modeling

- Computational fracture analyses by MD simulation to understand fracture mechanisms
- Continuum modeling and simulation of composite microstructure
- Numerical microstructure evolution (lattice) model
- Verification is undergoing

## Design Optimization

- Variable fidelity has been developed and tested using numerical and analytical models
- Stochastic optimization has been established using RSA
- Uncertainty quantification remains expensive
- Lower fidelity models may be used to approximate gradient of stochastic functions

Thank you