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HARD Solids

Micro-Scale Inverse Modeling

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Thomas A. Reichardt and Thomas J. Kulp, Sandia National Laboratories, Livermore, CA

National Nuclear Security Administration

Goals and Objectives

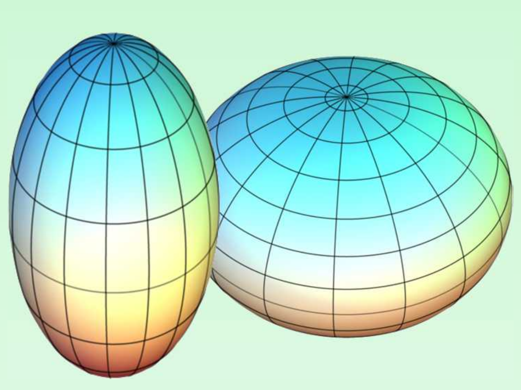
- Develop an invertible model for the reflectivity/emissivity of a surface chemical deposited on a substrate
- Model will incorporate fundamental physical properties (n,k) and account for morphological characteristics – particle size distribution (PSD), shape, and packing density
- Model should demonstrate agreement with reflectivity/emissivity measurements…
- …while the parameters extracted via model inversion (e.g., the PSD) should demonstrate agreement with independent measurements of related properties/characteristics

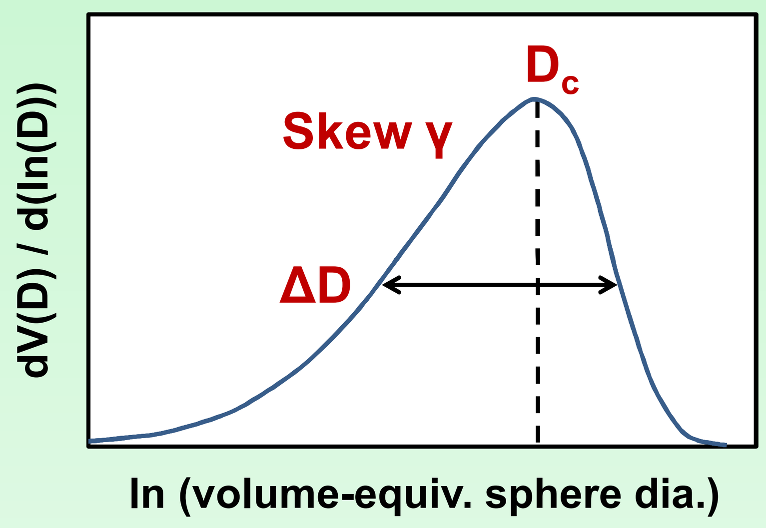
Introduction

- Model inversion addresses the question, “Given a model, what do the data say about it?”
- Optimizing the match between model and data sheds light on model shortcomings → Leads to improved model fidelity
- Framework is synergistic with SA/UQ

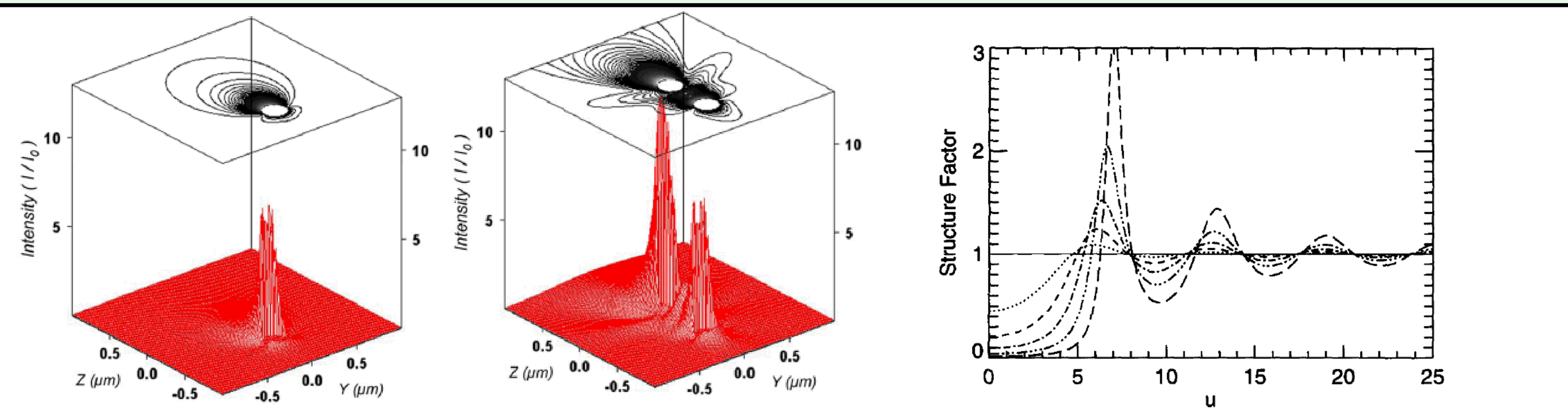
Methods

- Initially implement a solution for the reflectivity of an optically thick dilute particulate layer
  - Implement Ambartsumian invariant imbedding numerical solution (open-source code provided by NASA GISS)
- Allow for mixture of prolate/oblate spheroids with a skewed volume log-normal size distribution





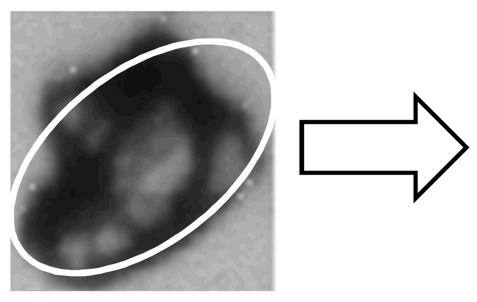
- Calculate scattering/extinction properties for size/shape mixture
  - For $D \sim \lambda$, calculate properties via T-matrix
  - For $D > \lambda$, calculate properties via ray tracing
- Invoke static structure factor (SSF), based on equivalent-volume spheres, to account for packing effects → fill factor f_{SSF}



J.-C. Auger and B. Stout, “Local field intensity in aggregates illuminated by diffuse light: T matrix approach,” Appl. Opt. **47**, 2897-2905 (2008).

M. I. Mishchenko, “Asymmetry parameters of the phase function for densely packed scattering grains,” JQSRT **52**, 95-110 (1994).

- Invoke effective media approximation (EMA) to account for “sub-spheroid” morphological features → fill factor $f_{EMA} = f_{air}$



$$\varepsilon_{eff} = \varepsilon_{mat}\left[1 + \frac{3 f_{air}\left(\dfrac{\varepsilon_{air} - \varepsilon_{mat}}{\varepsilon_{air} + 2\varepsilon_{mat}}\right)}{1 - f_{air}\left(\dfrac{\varepsilon_{air} - \varepsilon_{mat}}{\varepsilon_{air} + 2\varepsilon_{mat}}\right)}\right]$$

- 6 parameters: D_c, ΔD, γ, f_{SSF}, f_{EMA}, and angle of incidence θ (accounting for surface roughness)
- Vary these parameters to match model to data
  - Match optimized per forward-difference method in Sandia’s Dakota package
  - 125 λ-specific calculations distributed over 64 processors

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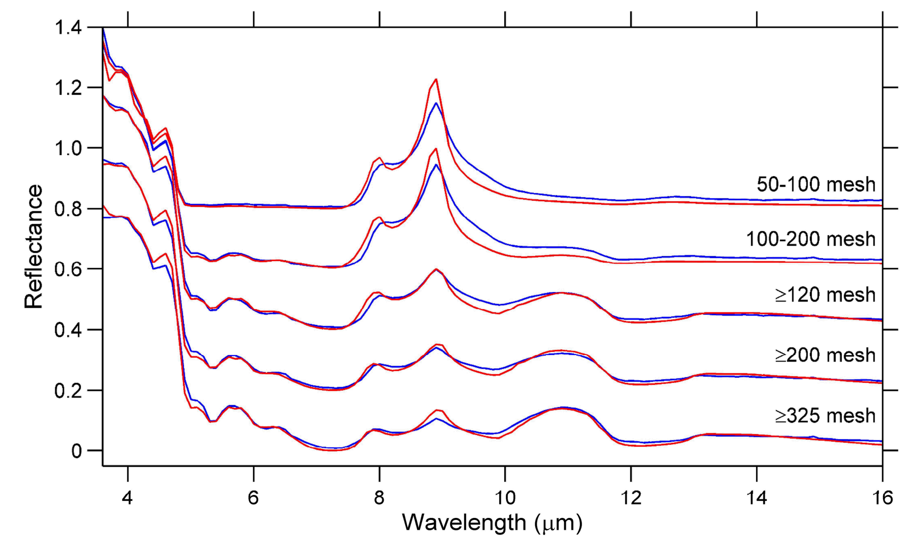
Results / Major Findings

Spectral fitting

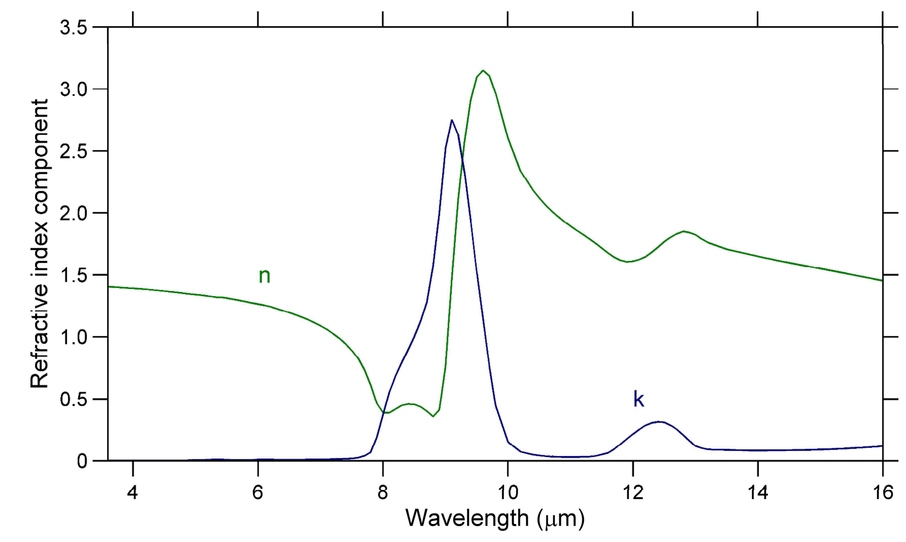
Size-dependent features, quite evident in the reflectance data of model systems (see “*Micro-Scale Measurements*” poster), are effectively captured by the reflectance model



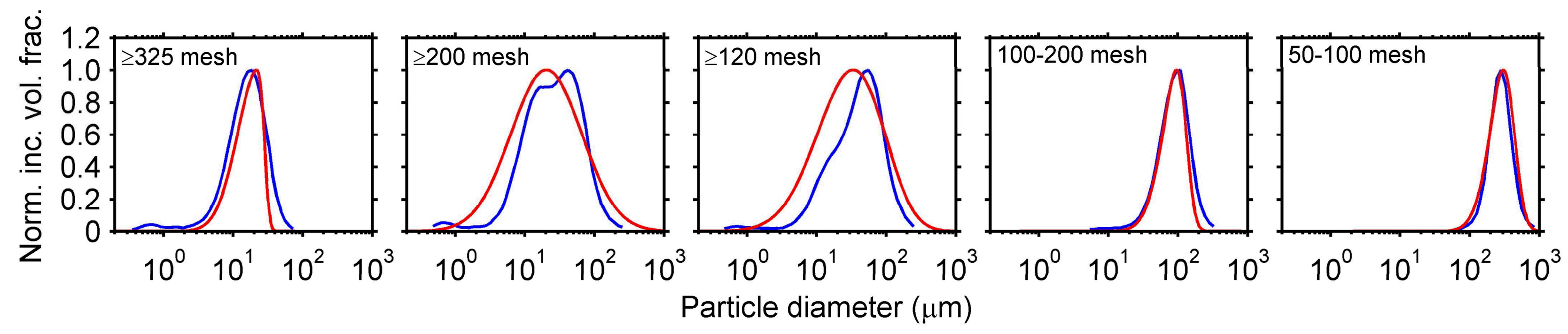
n/k spectra: Surface vs. volume scattering

k large: Reflectance dominated by field from particle surfaces

k small, n moderate: Reflectance dominated by field propagating into particulate medium



Particle size comparison



- PSDs measured by laser diffraction instrumentation
- Model-extracted PSDs exhibit good agreement with measurements

Packing density comparison

Extracted volume fractions deviate from those gravimetrically measured for finer particles.

- Significant approximations in applying the SSF and EMA

Volume Fraction

Mesh size	Gravimetrically measured	Model extracted
50-100	0.56	0.59
100-200	0.51	0.53
≥120	0.47	0.72
≥200	0.47	0.60
≥325	0.38	0.52

Discussion / Next Steps

- Given the approximations in the model, it nevertheless matches reflectance measurements of model systems
- Model developed on silica – extreme range of n (<1) and k (~3)
- As n,k measurements are performed on additional materials (see “*Lab Measurements of Optical Constants*” poster), the reflectance model can be applied to those materials as well
- Next: Perform SA/UQ on current model

Conclusion and Relevance to Program Objectives

- The numerically invertible model provides the means to account for particle size effects in the reflectance spectra of particulate media.
  - Based on fundamental physical properties (n,k)
  - Extracted parameters can be compared to auxiliary measurements → PSD agreement is especially encouraging
- Assessment via numerical inversion can continue to be applied as more advanced models become available (see “*Micro-Scale Forward Modeling*” poster)

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Pacific Northwest National Laboratory
Los Alamos National Laboratory
Lawrence Livermore National Laboratory
Sandia National Laboratories

CONTACT
Thomas Reichardt
Phone: 925.294.4776
tareich@sandia.gov