Complex Hierarchical Optimization Algorithms for the Design of Nanoporous Materials

Paul T. Boggs, David M. Gay, Stewart Griffiths and Bob Nilson
Sandia National Laboratories
Robert Michael Lewis, College of William and Mary
Kevin R. Long, Texas Tech University
Stephen G. Nash, George Mason University

Abstract: Our goal is the optimal design of nanoporous materials for the efficient storage of gas and/or electric charge. The physics at the nano-scale allows for significantly denser storage than at the macro-scale. Nanoporous materials have been proposed for use as hydrogen gas storage vessels and as ultra-capacitors. One form of the optimization problem is to construct the material in a manner that provides the maximum possible energy discharge from a storage system of fixed volume within a specified discharge period. We seek the optimal balance between rapid discharge and storage capacity. We developed a novel mathematical description of the problem, where the geometry depends on the mesh. For computational efficiency, we also identified surrogate steady-flow problems having nearly the same optima. We developed a multilevel optimization algorithm framework for the solution of such hierarchical problems where the physics changes fundamentally from the nano- to the macro-scale.

The Promise of Nanoporous Materials

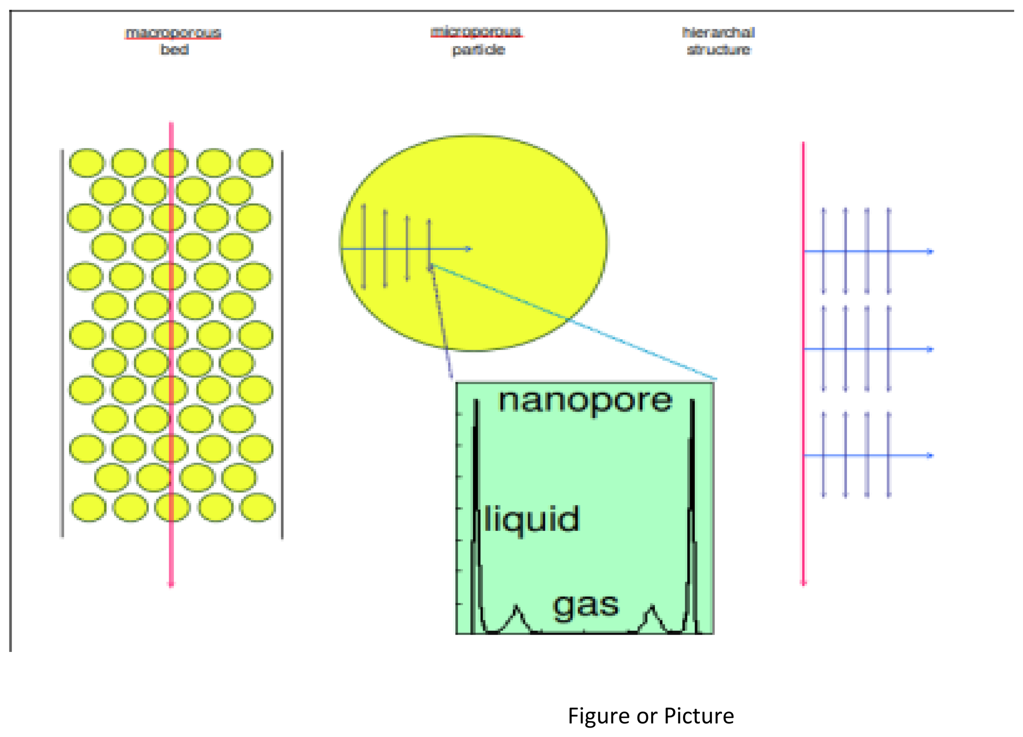
Objectives

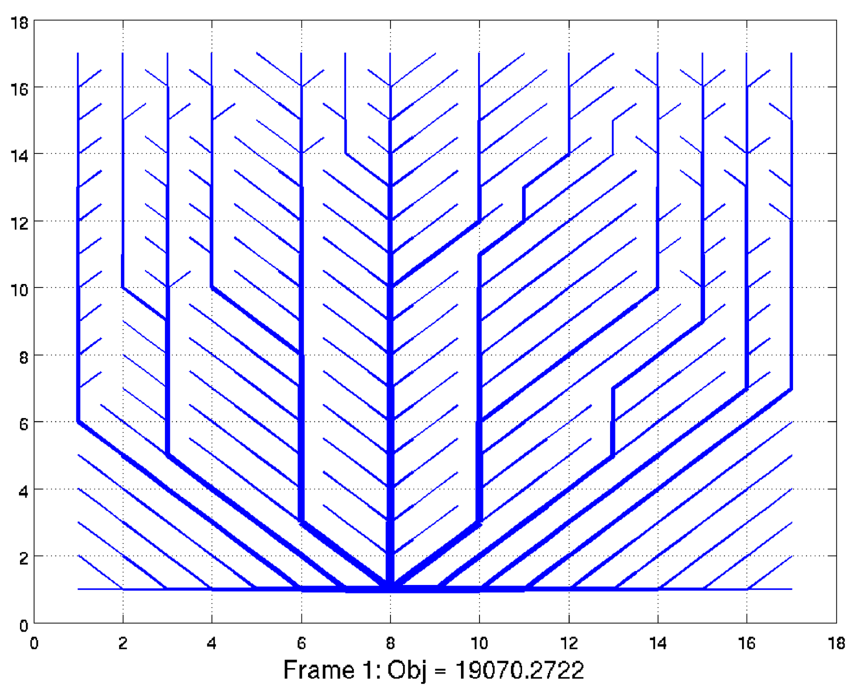
- Formulate and solve multi-scale optimization problems for energy storage applications
- Specifically, develop multi-scale models for optimizing the internal structure of nanoporous materials
  - Allow different physics at different scales
  - Derive methods to communicate between scales
- Create a general multi-grid algorithm to solve such problems on high-performance computers

Impact

- Applications include super capacitors, hydrogen storage, catalytic beds, and filters
- Algorithm can be applied to general hierarchical design problems arising from complex systems
- Nanoporous materials are potentially important energy storage systems

The Challenge of Nanoporous Materials



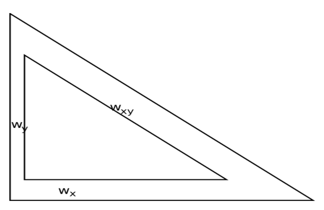


Left: Channels of different widths in the material

Right: Channels in model problem

Transport Model

- Multi-level model as a function of channel widths
- This is a nonstandard problem since the channel locations are a function of the mesh



The mean viscous flow speed through a channel of width w_i oriented along an edge's tangential unit vector $\mathbf{e}_i$ will be

$$v = (\mathbf{e}_i \cdot \nabla p)\,\frac{w_i^2}{4(d^2-1)\eta}$$

where d is the dimension of the problem and η the dynamic viscosity. The velocity is

$$\begin{aligned}\mathbf{v} &= -\mathbf{e}_i(\mathbf{e}_i \cdot \nabla p)\,\frac{w_i^2}{4(d^2-1)\eta} \\ &= -B\nabla p\end{aligned}$$

where

$$B = -\frac{w_i^2}{4\,(d^2-1)\,\eta}\,(\mathbf{e}_i \otimes \mathbf{e}_i)\,,$$

Let $\chi_e(\mathbf{x})$ be 1 in the channels, 0 in the bulk medium. Assuming isotropic permeation with permeability κ in the bulk, the net mass transfer current will be

$$\mathbf{j} = -\chi_e(\mathbf{x})\rho B\nabla p - (1-\chi_e(\mathbf{x}))\frac{\rho\alpha}{\eta}\nabla p.$$

The capacity is

$$c(\mathbf{x}) = \chi_e(\mathbf{x}) + \beta\,(1-\chi_e(\mathbf{x}))$$

and the transport equation is

$$c\frac{\partial\rho}{\partial t} + \nabla\cdot\mathbf{j} = 0$$

Surrogate steady-flow models that approximate this equation have also been developed.

Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

Sample Optimization Model

Minimize <average pressure in the material>
Subject to <transport model of flow>
<porosity constraint: how much volume can be used for channels>
<bounds on channel widths>

Could also minimize extraction time

Hierarchical Optimization: χOpt

χOpt: Complex Hierarchical OPTimization

Prototype high-performance software for hierarchical optimization

- MG/Opt: optimization-based multigrid framework
  - The optimization problem can be better suited to a multilevel algorithm than the underlying PDE
  - General framework for developing and analyzing multilevel optimization algorithms for general equality and inequality constraints
    - Convergence theory
      - Handles unconstrained and constrained problems
      - Proves convergence based on an underlying optimization algorithm under standard conditions
- Sundance
  - Powerful tool for handling PDEs, based on finite-element method
  - Transforms high-level description of PDE in weak form into numerical operators
  - Automates computation of objective function, constraints, and their gradients
  - Extended to handle the mixed channel-medium transport model

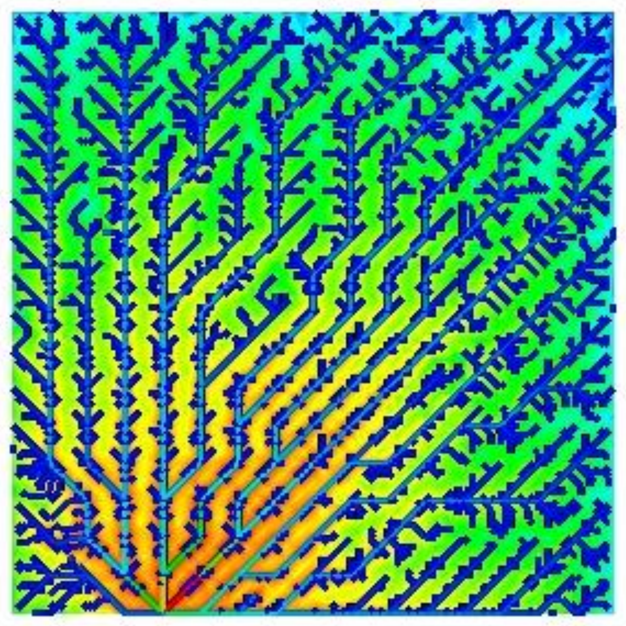
Results

Algorithm

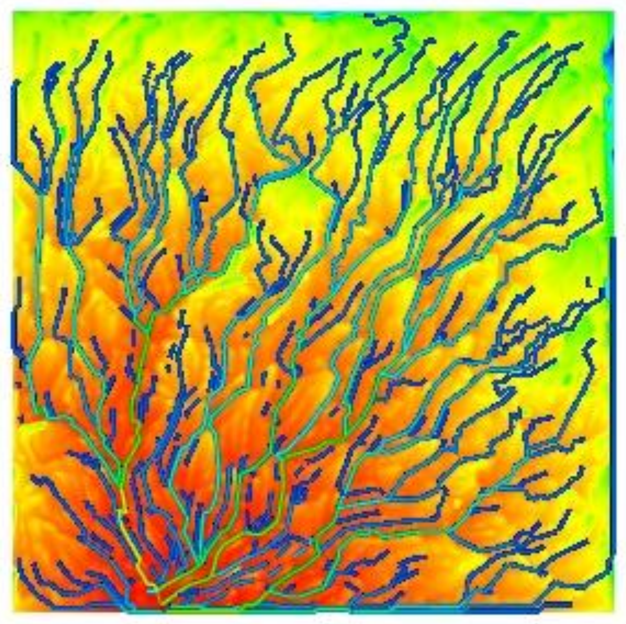
- Based on flexible software framework
- Meshing
  - Automatic mesh refinement
  - General mesh allowed
- Can have outlets from multiple nodes or complete edges
- Updates/downdates exploit physics
- Initialization
  - Use greedy algorithm on network approximation for initial guess

Material Design Problem

- Designs with tree structure are better than those with more general network structure
  - We can enforce the tree structure in the network in the optimization model
- Solutions are not unique (regularization is needed)
- Incorporation of physics into the algorithm design improves the algorithm



3-Level Model [restricted channels]



3-Level Model [general channels]

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- Stephen G. Nash, "Convergence and descent properties for a class of multilevel optimization algorithms," www.optimization-online.org/DB_HTML/2010/04/2598.html (2010).