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**

![Image showing channels of different widths in the material](image)

**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 $e_i$ will be

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

where $d$ is the dimension of the problem and $\eta$ the dynamic viscosity. The velocity is

$$v = -e_i (e_i \cdot \nabla p) \frac{w_i^2}{4(d^2 - 1)\eta} = -B \nabla p$$

where

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

Let $\chi_s(x)$ be 1 in the channels, 0 in the bulk medium. Assuming isotropic permeation with permeability $k$ in the bulk, the net mass transfer current will be

$$j = -\chi_s(x)pB \nabla p - (1 - \chi_s(x)) \frac{\rho g}{\eta} \nabla p.$$

The capacity is

$$c(x) = \chi_s(x) + \beta (1 - \chi_s(x))$$

and the transport equation is

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

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

**Sample Optimization Model**

<table>
<thead>
<tr>
<th>Objective</th>
<th>Subject to</th>
</tr>
</thead>
<tbody>
<tr>
<td>Minimize</td>
<td>&lt;average pressure in the material&gt;</td>
</tr>
<tr>
<td>Subject to</td>
<td>&lt;transport model of flow&gt;</td>
</tr>
<tr>
<td></td>
<td>&lt;porosity constraint: how much volume can be used for channels&gt;</td>
</tr>
<tr>
<td></td>
<td>&lt;bounds on channel widths&gt;</td>
</tr>
</tbody>
</table>

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
- Sandia: 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
- Update/set among 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

**Selected References**