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Sandia National Laboratories

Additive Manufacturing of Porous Material Lattices with Spatially Optimized Permeability

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PRESENTED BY

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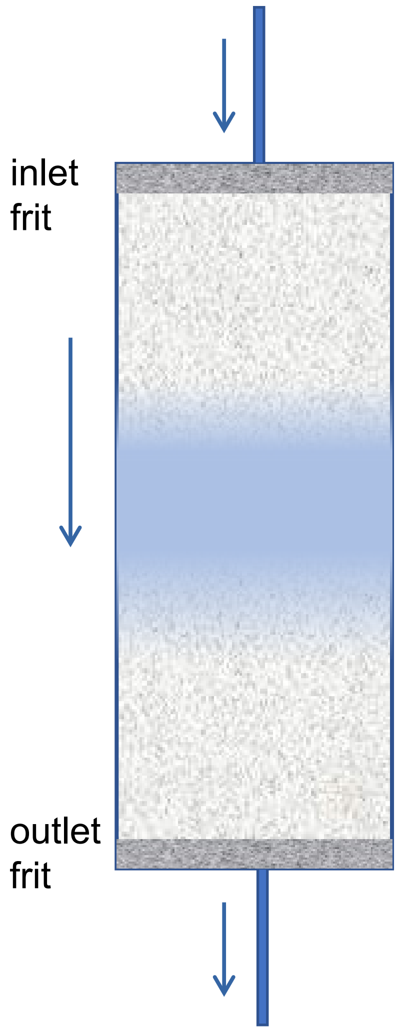
Background and Motivation



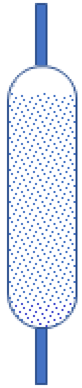
Energy conversion and storage systems often rely on porous media.

Microscopic pore geometry defines macroscopic permeability, defining flow rate versus pressure.

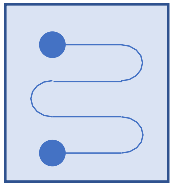
Fluid phase can travel over varying cross sections or around corners, where flow can be nonuniform.

Examples:

Catalytic converters
Catalytic columns for fuel processing
Battery and fuel cell electrodes



Challenge: Adjust permeability to maintain spatially uniform fluid velocity despite nonuniform geometries.



3D Printing of Porous Material

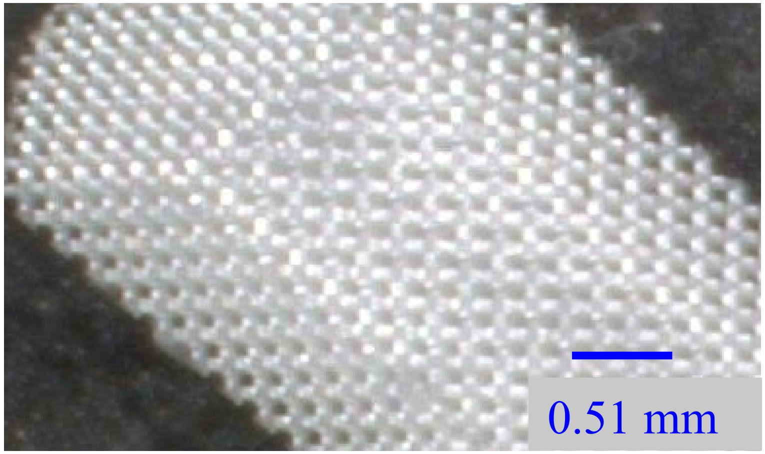
Additive manufacturing (AM) techniques raise the possibilities that porous media can be fabricated in which the permeability can be arbitrarily specified in three dimensions

By varying laser power and speed, and tuning particle size distribution, Mott Corporation has claimed the ability to spatially vary permeability using metal laser sintering AM methods.[1]

We have previously studied flow through additively manufactured polymer lattices with precisely defined pores.[2,3]

How can we use the tunability of AM to tailor permeability and control flow?

Photopolymer lattice from Autodesk Ember 3D printer with 150 μm pores



1. V.P. Palumbo et al. “Porous Devices Made by Laser Additive Manufacturing.” US Patent Application 2017/0239726 A1, Mott Corporation, 2017.
2. M. Salloum and D.B. Robinson “A Numerical model of exchange chromatography through 3-D lattice structures”, *AIChe J.* vol 64(5), pp. 1874-1884, 2018
3. D.B. Robinson. 3D-Printed Apparatus for Efficient Fluid-Solid Contact. US Patent 10493693 B1 (2019).

Outline

1. Theory: Flow properties of lattices and porous media
2. Numerical modeling of lattice permeability
3. Additive manufacturing of lattices
4. Characterization of lattices
5. Optimization of flow by tuning permeability

Flow in Porous Media: Permeability and Porosity

We focus on Darcy's Law:
- Assumes slow flow
- Applicable scale: greater than pore length scale

$$v = \frac{\kappa}{\mu L} \Delta p$$

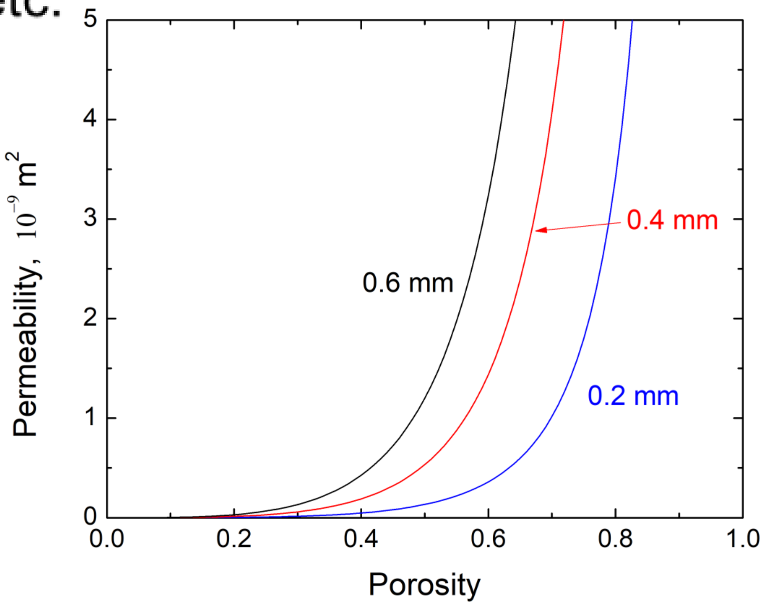
v = fluid velocity $\left(\frac{\mathrm{m}}{\mathrm{s}}\right)$
κ = permeability $\left(\mathrm{m}^2\right)$
μ = viscosity (Pa s)
L = length (m)
Δp = pressure drop (Pa)

Permeability is related to porosity ε (pore volume fraction)

- Depends on pore geometry and how it is changed
  - Compaction, sintering, 3D printing, etc.
- Packed spheres, diameter D: Kozeny-Carman equation

$$\kappa = \frac{D^2 \varepsilon^3}{150(1-\varepsilon)^2}$$

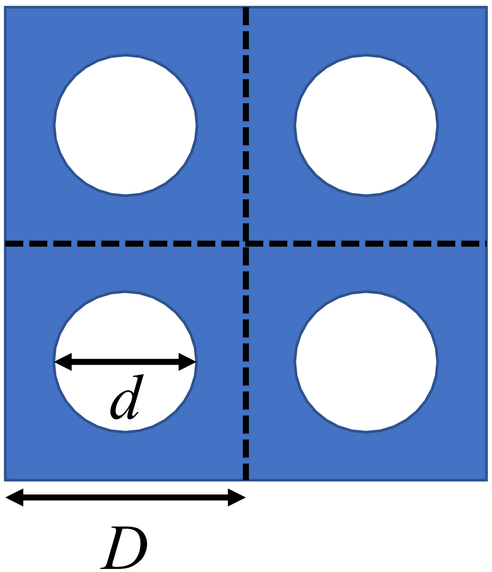
- Sharp nonlinearity could make control of permeability difficult.



Lattices as porous media: parallel pipes

Lattices have more precise geometries than randomly packed spheres. What is the permeability of a lattice?

Simple example: parallel pipes, flow perpendicular to screen



Poiseuille pipe flow:

$$v_{pipe} = \frac{d^2}{32\mu L}\Delta P$$

Darcy's Law:

$$v_{area} = \frac{\kappa}{\mu L}\Delta P$$

Geometry defines porosity:

$$\varepsilon = \frac{\pi d^2}{4D^2}$$

where

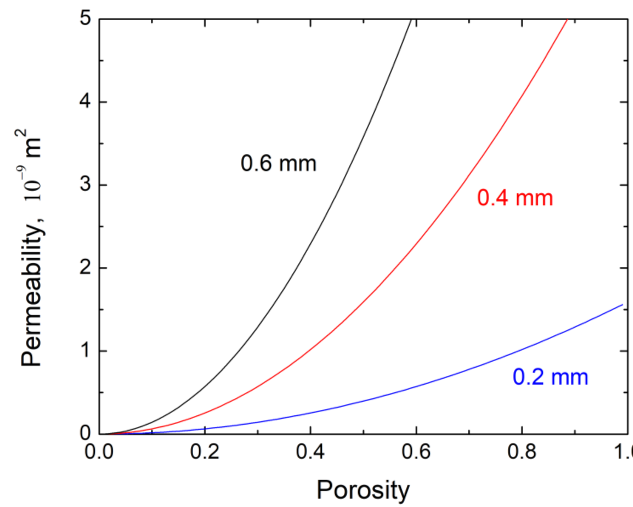
$$v_{area} = v_{pipe}\,\varepsilon$$

Combining these gives

$$\kappa = \frac{D^2\varepsilon^2}{8\pi}$$

$$0 < \varepsilon < \pi/4$$

The softer ε dependence may allow easier tuning of permeability than for sphere packing.



Additively manufactured 3D lattices

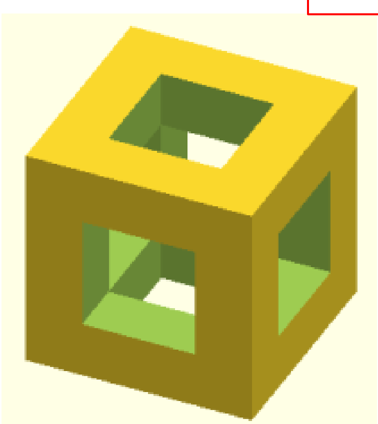
For chemical engineering applications, we often desire µm-scale pores.

- Only photopolymer AM techniques can achieve this.
- These methods typically print cubic voxels.
- We propose unit cells requiring a minimum number of voxels.

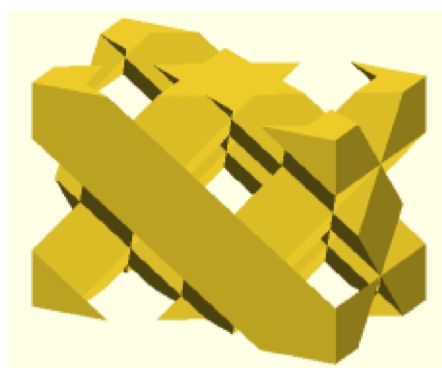
3D lattices are less sensitive to pore diameter variations than parallel tubes.

- The cube-edge unit cell allows flow in 3 dimensions.
- A column aligned with the cube diagonal allows lateral mixing in 3D.

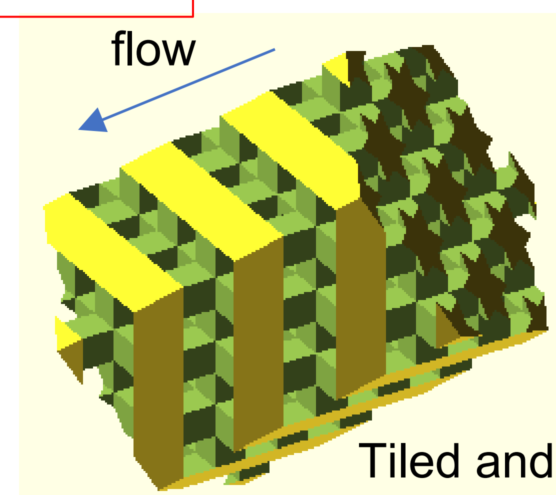
We will explore how to design and build high-resolution, diagonally oriented cube-edge lattices.



Cube-edge unit cell



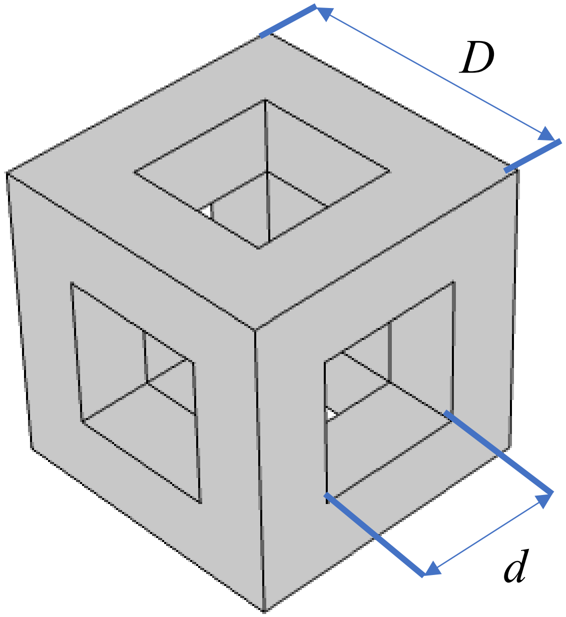
Derived rectangular prism unit cell



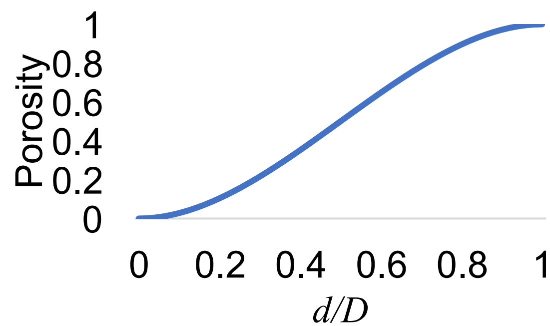
Tiled and cropped unit cells

Porosity of 3D lattices

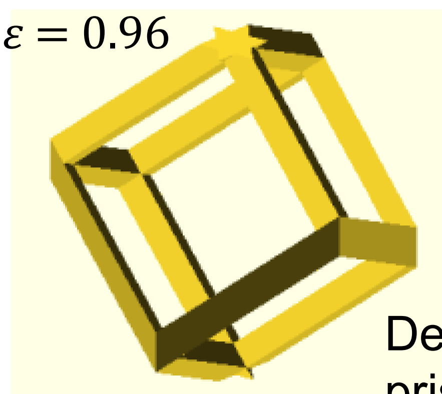
Porosity is a smooth function of unit cell geometry.



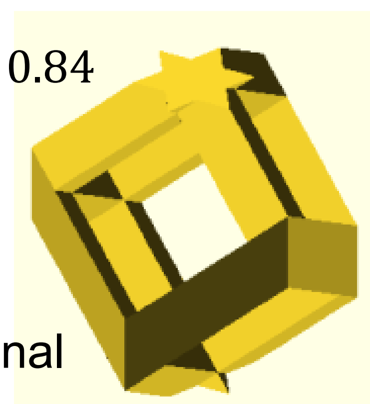
$$\varepsilon = 3\left(\frac{d}{D}\right)^2 - 2\left(\frac{d}{D}\right)^3$$



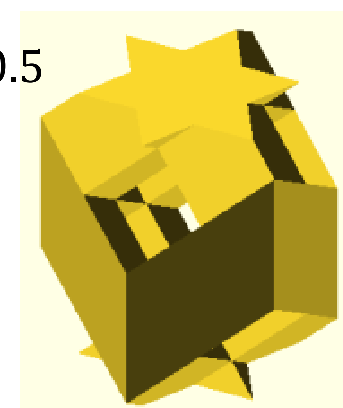
$\varepsilon = 0.96$



$\varepsilon = 0.84$



$\varepsilon = 0.5$



Derived hexagonal prism unit cells

1. M. Salloum and D.B. Robinson “A Numerical model of exchange chromatography through 3-D lattice structures”, *AIChe J.* vol 64(5), pp. 1874-1884, 2018
2. D.B. Robinson. 3D-Printed Apparatus for Efficient Fluid-Solid Contact. US Patent 10493693 B1 (2019).

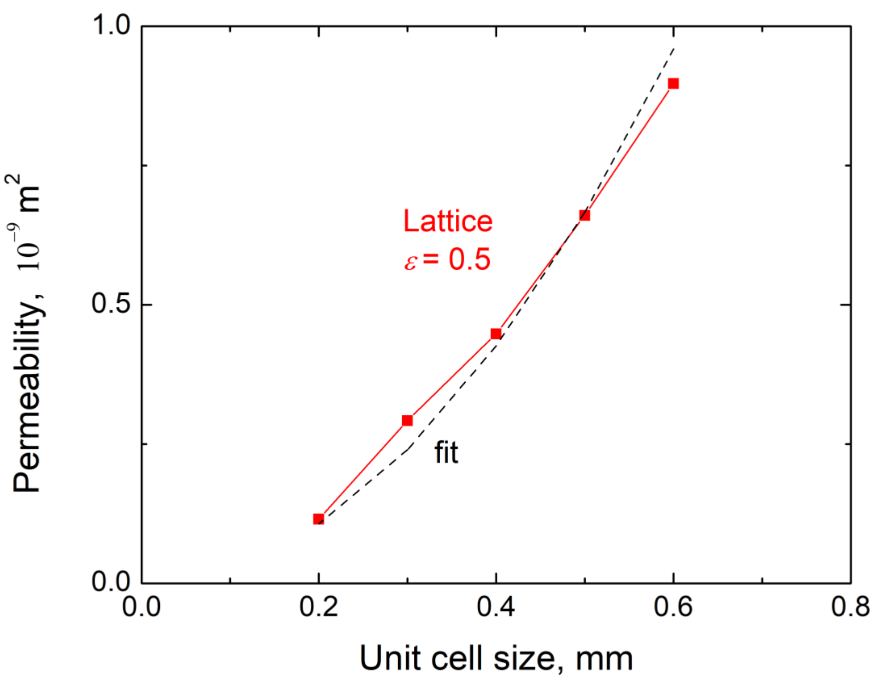
Cube edge lattice permeability

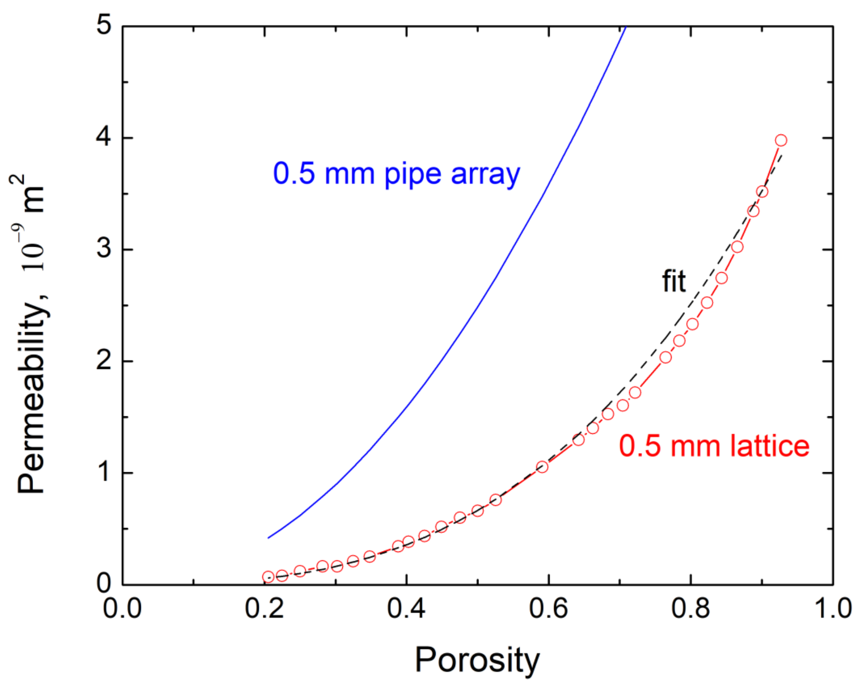
We have computed the $\kappa(D, \varepsilon)$ relationships for the cube-edge lattice by solving the Navier-Stokes equations using COMSOL numerical modeling software.

We tiled 4x4x10 rectangular unit cells (10 cells in the flow direction).

The data can be fit as $\kappa = \frac{D^2\varepsilon^2}{8\pi}(0.056 + 0.42\varepsilon)$.

Lattice permeability is lower than a tube array with the same D and ε, presumably due to narrower channels, sharp corners, and zigzag flow paths.





Printed lattice columns

At the Spring 2021 ACS meeting, we met Acrea3D, a company that prints high-resolution photopolymer structures using a specialized resin.

Voxels are 7.6 μm.

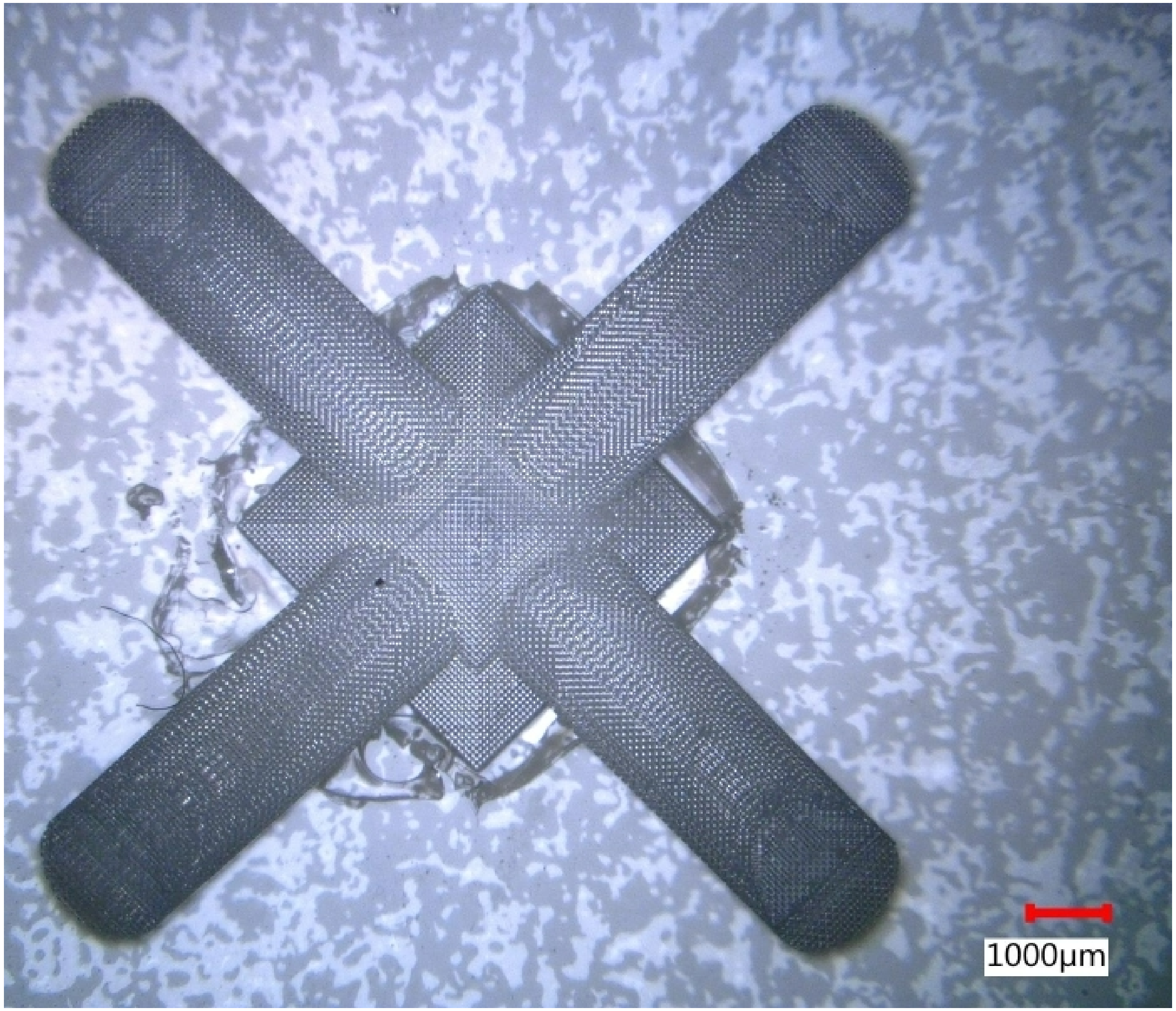
This lattice is designed with 92 μm unit cells, $\varepsilon = 0.5$.

Cubic unit cells are parallel to the build plate.

Cylinders are oriented in cube-diagonal direction and can be trimmed from pyramidal base.

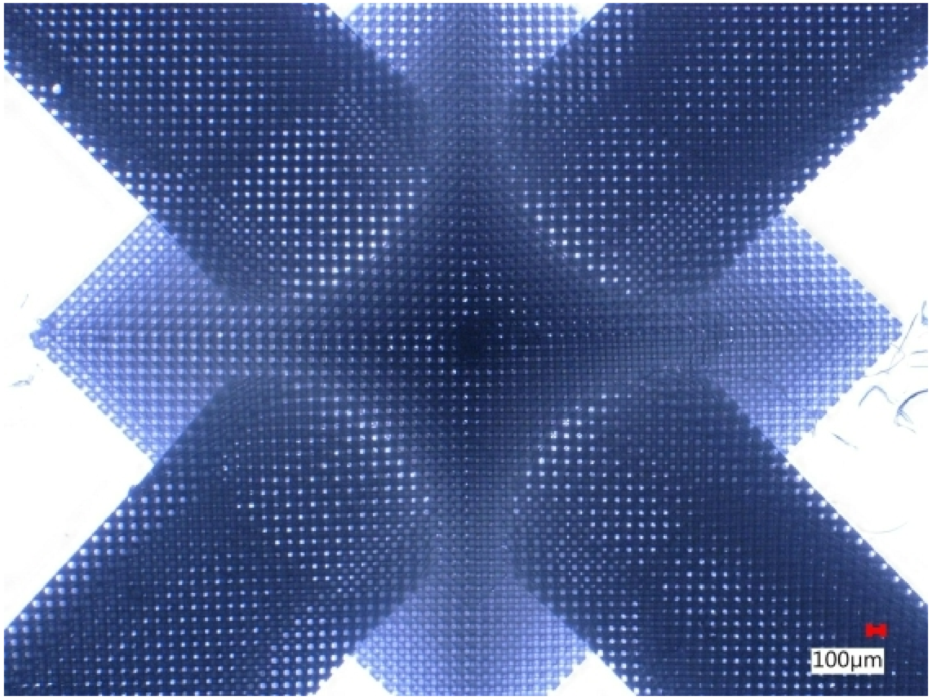
Bending of cylinders during printing led to some defects at the ends.

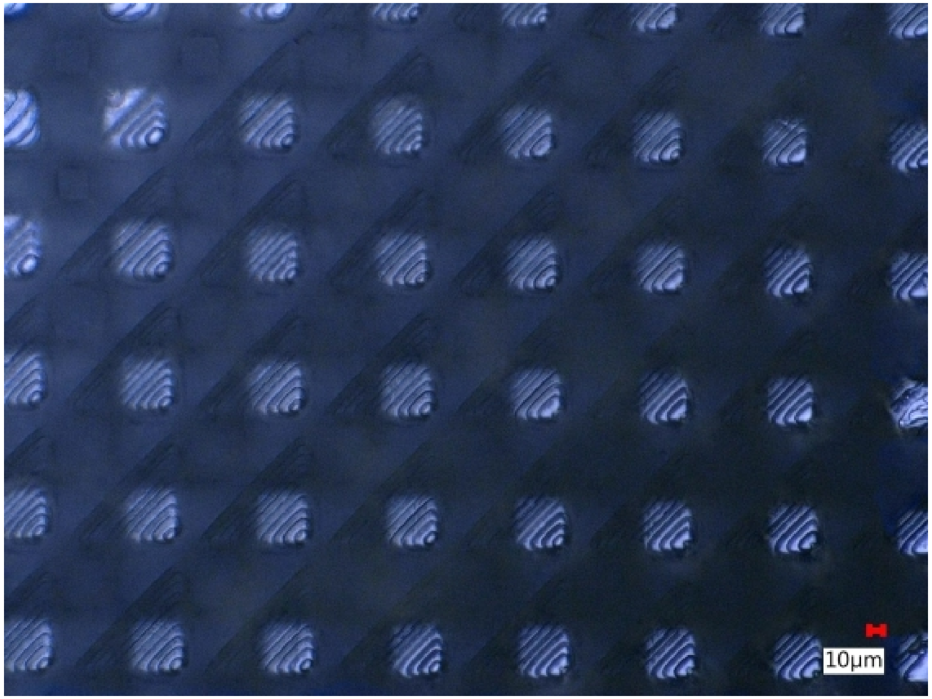
Overall, part geometry is accurate.



Optical microscopy of 92 µm lattice

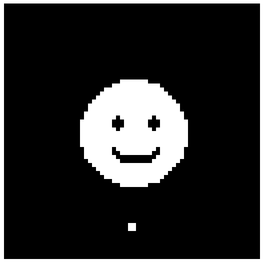
- Pore dimensions in this part appear quite uniform.
- Polymer is overexposed; pore width appears to be about 31 µm, $\varepsilon = 0.26$.
- By changing exposure conditions or input file porosity, we expect we could obtain the target porosity.
- To evaluate the lattice in 3 dimensions, we can use x-ray tomography.

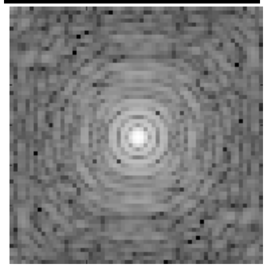




Tomography: 2D example

Original 2D image





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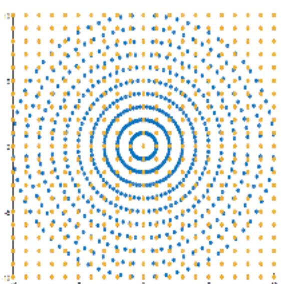
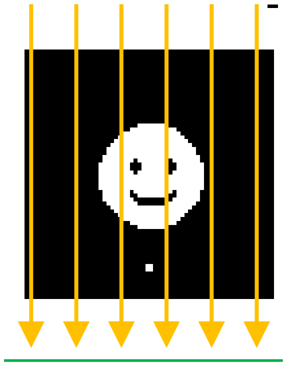
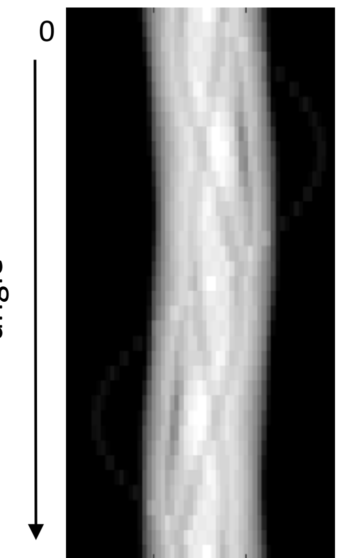


Sampled point density in Fourier domain



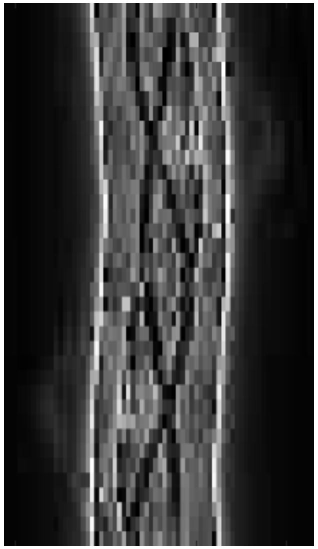
Project to 1D detector - Integrate intensity along line

Repeat at many angles



1D projections

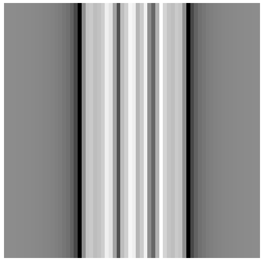
Apply |ω| filter

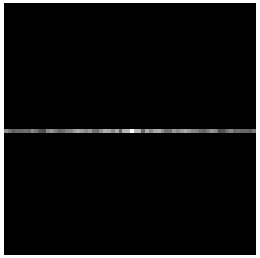


Filtered projections

Place 1D fft as angled line in 2D fft

Invert fft





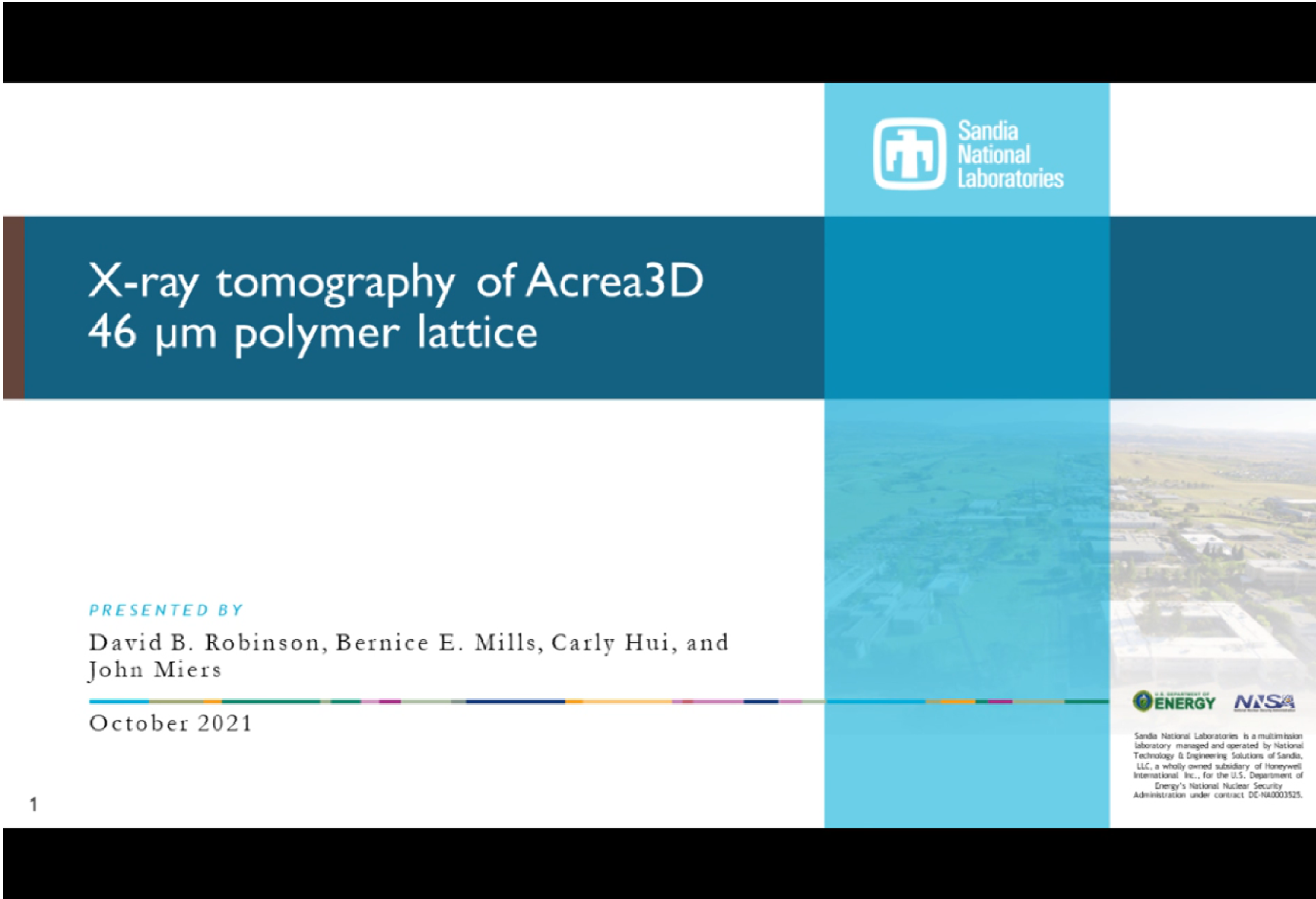
Reconstruction

- Method assumes projected intensity is proportional to amount of material in beam path.
- High frequencies are undersampled and noisy.
- Polar grid must be converted to Cartesian grid.
- Projections must be precisely aligned by cross-correlation or marker tracking.
- Grayscale reconstruction must be converted to binary solid/void map by filtering and thresholding.

- 3D reconstruction of well aligned projections on a single tilt axis is just many parallel 2D reconstructions.

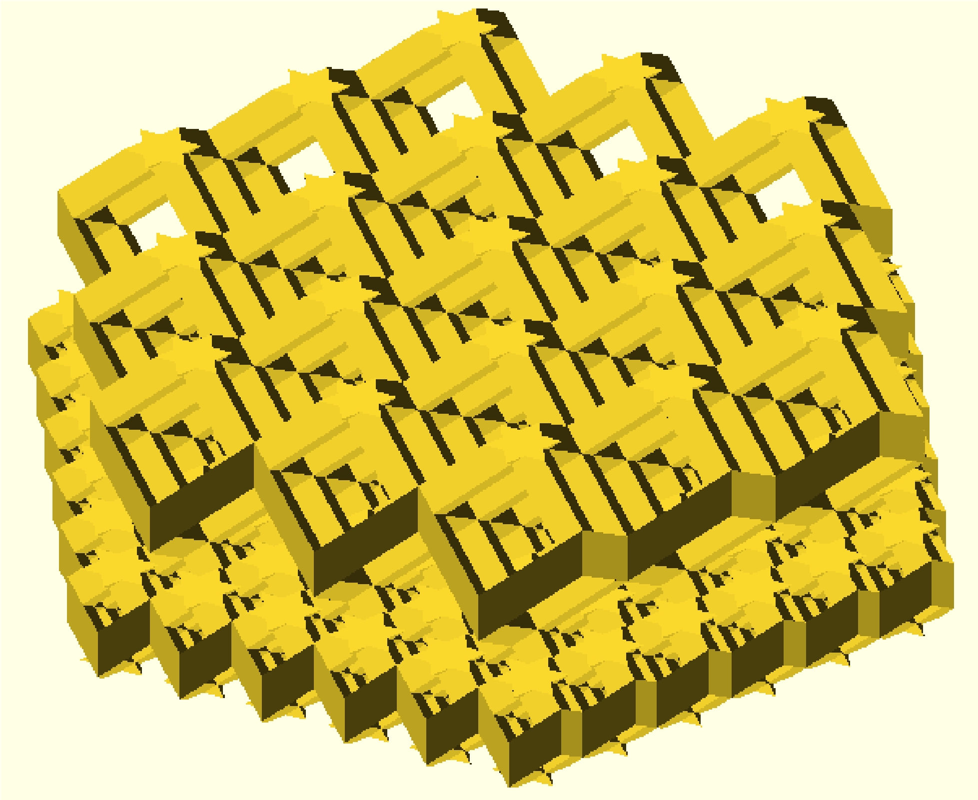
Lab x-rays are not parallel beams. This can be accounted for.

X-ray tomography of printed lattice



Spatially tuning permeability

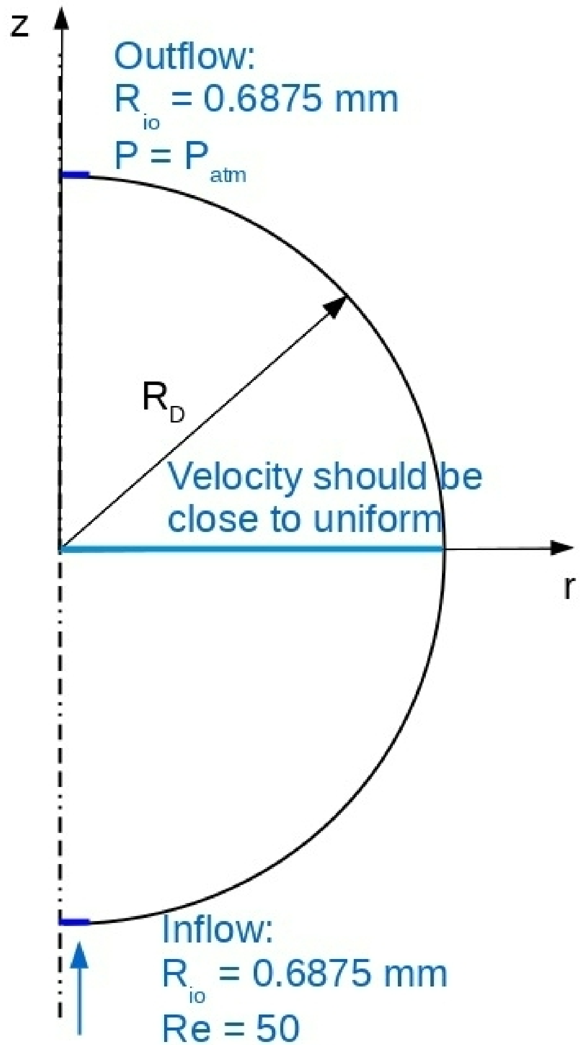
- For packed, sintered spheres, we can tune sizes of particles added to a part.
- Porosity of packed spheres can be adjusted through more/less local sintering.
- For lattices, we can change lattice parameter and porosity.
- Lattice must stay in registry, so it changes in big jumps (integer factors).
- Porosity changes in steps defined by the 3D printer's voxel size.
- These two factors allow us to access a range of permeabilities.



We can use this approach to design lattices that optimize flow properties.

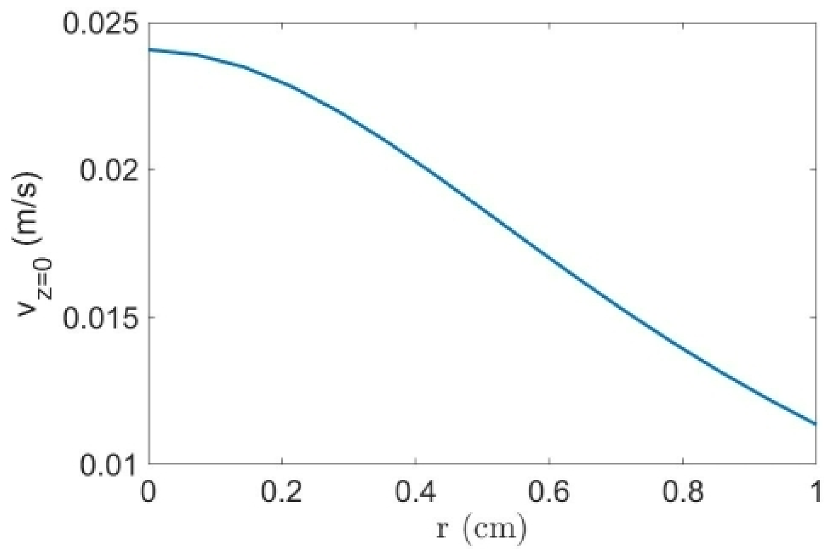
Spherical Column Optimization

As a test optimization problem, we consider a spherical column is a simple model of a column with hemispherical ends.



We start by studying a sphere of axi-symmetric model geometry.

For a constant permeability of 10^{-12} m^2, the resulting midplane velocity profile is:



We seek a graded permeability that meets criteria such as a uniform flow velocity mid-way between the inlet and outlet, or uniform transit times along streamlines.

Spherical Column with Graded Permeability

Iterative Optimization

In our optimization, we do not impose any functional form of the permeability field.

We obtain the optimal solution iteratively by adjusting the permeability field model input according to the midplane velocity and transit time results at each iteration.

We start with a constant permeability $K = K_0$ and repeat the following until convergence:

$$K(r,z)_i = K(r,z)_{i-1} \cdot \sqrt{\frac{max[v(r)_{i-1}]}{v(r)_{i-1}} \cdot \frac{\tau(r,z)_{i-1}}{max[\tau(r,z)_{i-1}]}}$$

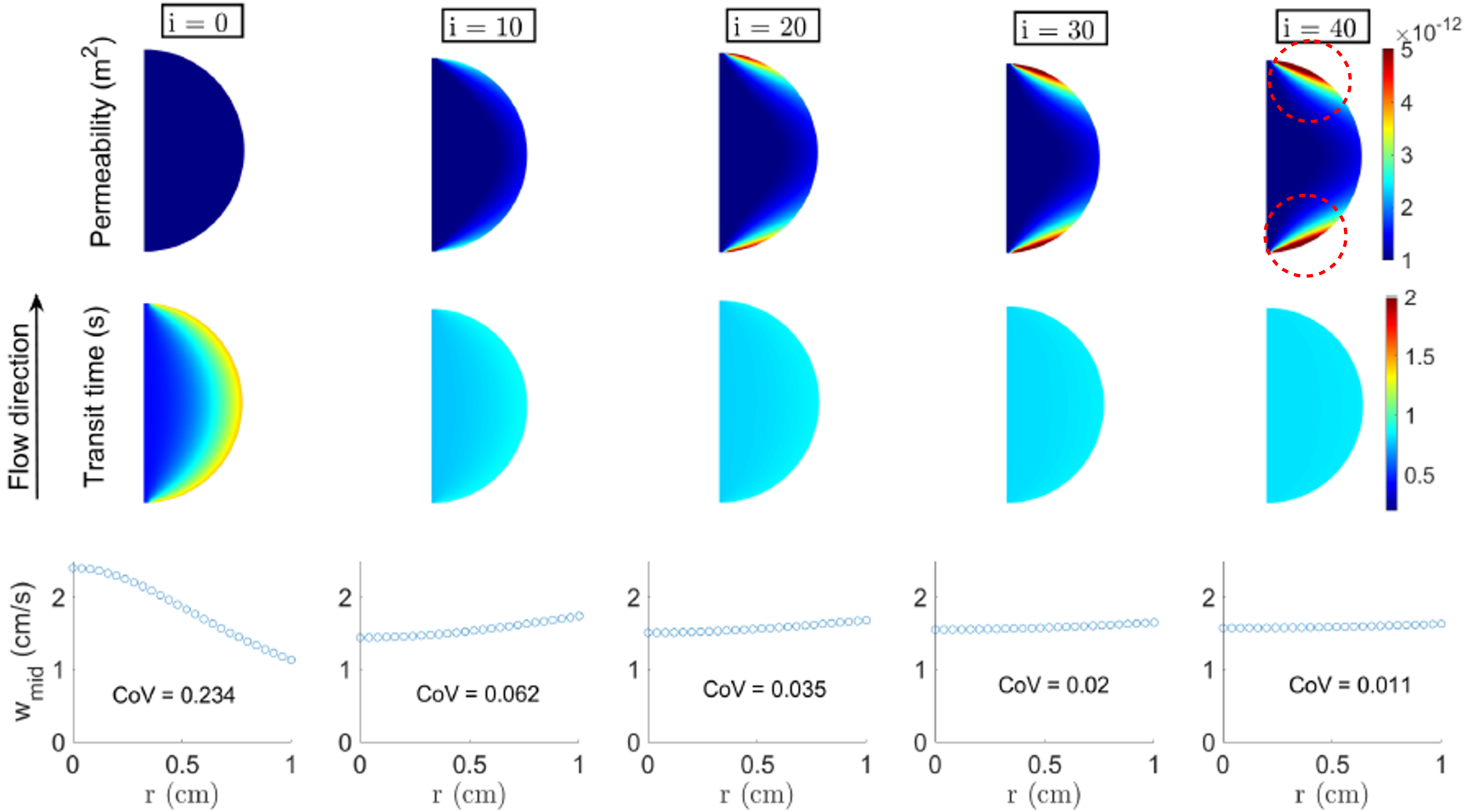
This algorithm adjusts the local value of the permeability according to the midplane velocity at that radius and transit time on that streamline. The square root damps large changes and aids convergence.

Spherical Column with Graded Permeability

Optimization Results

An optimal solution is obtained within 40 iterations with uniform velocity and transit time.

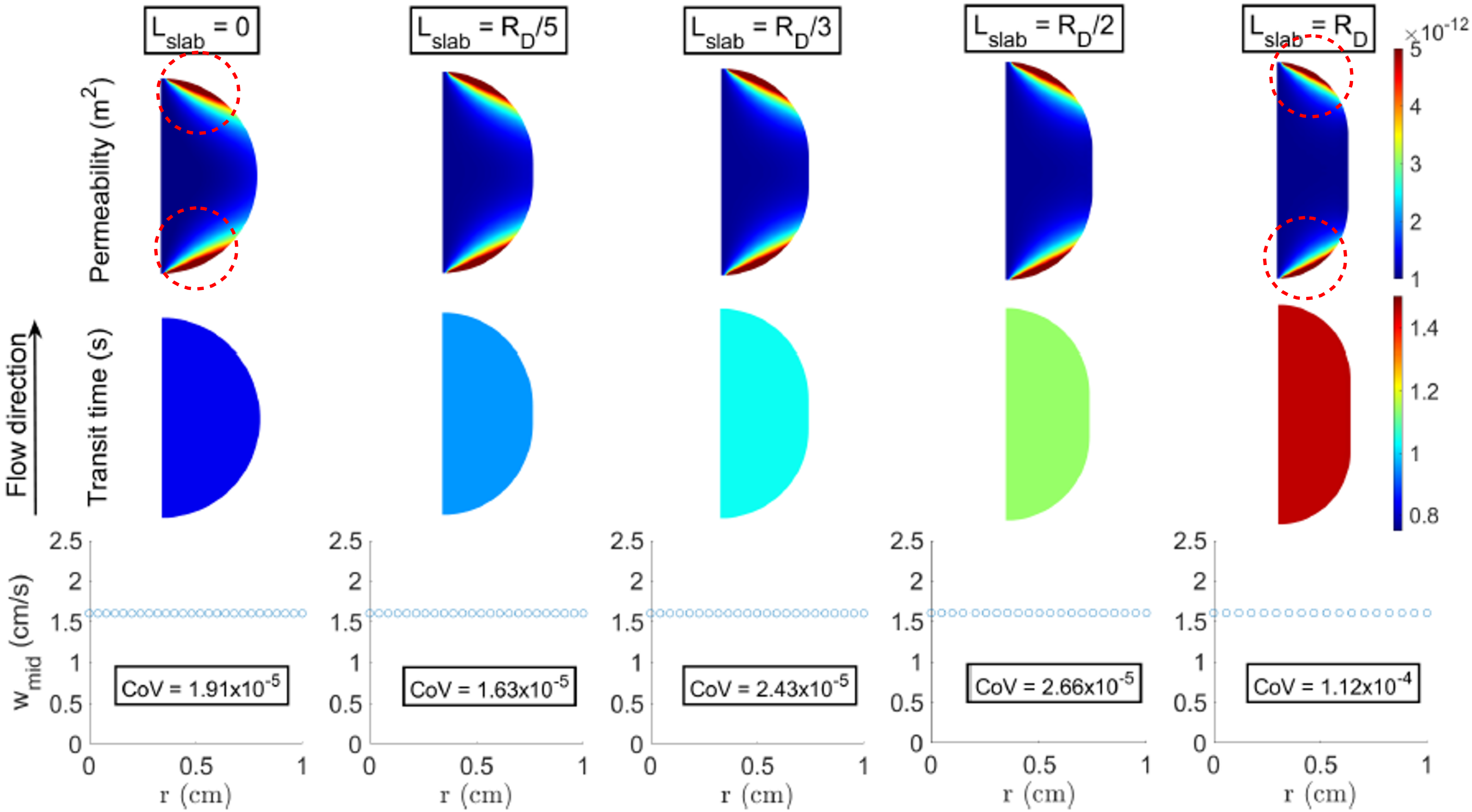
By grading the permeability only near the sphere inlet and outlet over a 10x range, we obtain uniformity in midplane velocity and transit time within a few percent.



Spherical Column with Graded Permeability

Effect of an Inserted Slab

Similar permeability field trend is obtained for a slab inside the sphere which simulates a more cylindrical geometry (e.g., glass chromatography column)



Conclusions

Additive manufacturing techniques enable novel architectures for porous media using sintered powders or lattices.

Lattices approximately follow $\kappa \propto D^2\varepsilon^2$ and likely allow more precise tuning of permeability than sintered powders obeying the Kozeny-Carman equation.

Optimized graded permeability can achieve desired fluid velocity fields and transit times in flow columns.

Columns with hemispherical ends can have uniform transit time by simply grading permeability in regions near the inlet and outlet.

We aspire to apply these methods to practical applications in the handling of renewable fuels such as hydrogen, and electrochemical energy devices.

Acknowledgements and Contact Info

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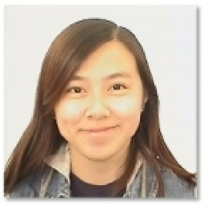

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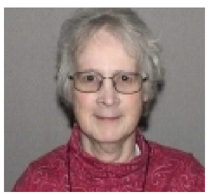

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John Miers aided
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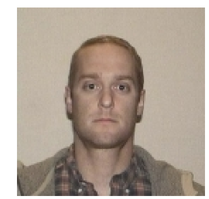

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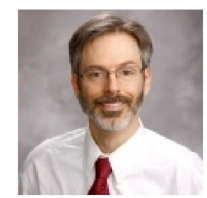

Acrea3D printed the polymer lattice. Acrea3D.com

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