Impacts of biomass feedstock pre-processing on heat and mass transfer during pyrolysis using X-ray computed tomography and multiscale modeling

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Abstract

Knowledge of the transport properties of biomass particles such as porosity, tortuosity, and permeability is paramount for high-fidelity modeling of biomass pyrolysis due to the heat and mass transfer limitations imposed by particle microstructure. X-ray computed tomography (XCT) is a non-destructive imaging method that enables full 3D reconstructions of the biomass particle microstructure with high resolution, permitting direct calculation of porosity, tortuosity, and permeability from real particle geometries. In this study, XCT imaging revealed the 3D microstructures of particles and chars from pyrolytic conversion of cylindrically cut or milled/pelletized loblolly pine samples. The porosity, tortuosity, and permeability were calculated directly from the XCT geometries via open-source microstructural analysis tool MATBOX (https://github.com/NREL/MATBOX_Microstructure_analysis_toolbox) and computational fluid dynamics (CFD) simulations using our solver, Mesoflow (https://github.com/NREL/mesoflow). These properties were used in a reactor scale model developed in COMSOL of the single particle reactor at NREL to investigate the impact of feedstock pre-processing on biomass conversion during pyrolysis with rigorous experimental validation.

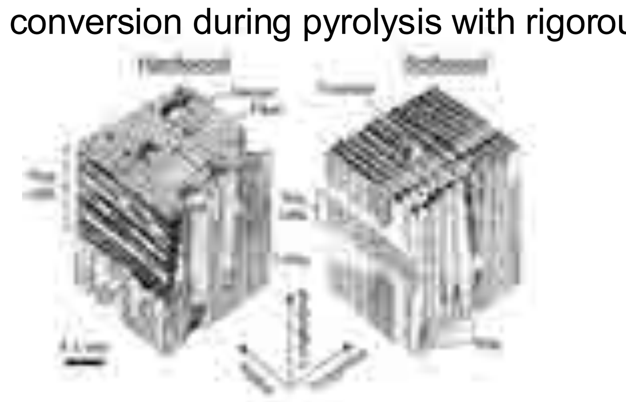


Figure 1. The natural anisotropy of wood microstructure. Heat and mass transfer is most facilitated in the longitudinal direction. Reproduced from Jakes et al., Forests, 2019 . DOI: 10.3390/f10121084

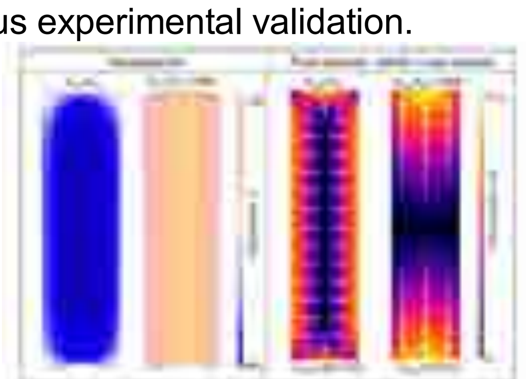


Figure 2. Anisotropic transport properties lead to anisotropic heat and mass transfer and significant temperature gradients during pyrolysis Reproduced from Pecha et al., Energy and Fuels, 2021 DOI: 10.1021/acs.energyfuels.1c02679

Figure 3. Photograph of cut and pelletized pine and char samples characterized in this study

Methods

- Cylindrically cut or pelletized pine particles sourced from Idaho National Laboratory were converted to char via pyrolysis in the single particle reactor at the National Renewable Energy Laboratory
- The 3D microstructures of cut or pelletized pine samples along with chars after pyrolytic conversion were imaged using X-ray computed tomography (XCT) at Colorado School of Mines
- Rectangular regions were cropped from the original 3D XCT reconstructions, filtered, and segmented using *Dragonfly* v. 2022.2 (Comet Technologies Canada Inc., Montreal, Canada) and *MATBOX* for microstructural analysis
- The porosity and tortuosity for each sample were calculated directly from the 3D particle geometries using *MATBOX* microstructural analysis toolbox
- The permeability tensor was computed from computational fluid dynamics (CFD) simulations using *Mesoflow* by applying a unidirectional pressure gradient in each direction and solving for the velocity via the compressible Navier-Stokes equations.
- The calculated properties were used in reactor and particle scale models of the single particle reactor in *COMSOL Multiphysics*® v. 6.1 (COMSOL AB, Stockholm, Sweden) to investigate the impact of particle microstructure on pyrolytic conversion using the CRECK pyrolysis kinetic scheme (Debiagi, P. *et al. J. Anal. Appl. Pyro.* 2018 DOI:10.1016/j.jaap.2018.06.022)
- Experimental measurements of mass loss of the particle over time from the single particle reactor were used to validate model predictions.

Porosity: *The volume fraction of void space*

$$\varepsilon_k = \frac{1}{N}\sum_{i=1}^{N} v(i), \text{with } v(i) = \begin{cases} 1 \text{ if } v(i) \in \text{phase } k \\ 0 \text{ if } v(i) \notin \text{phase } k \end{cases}$$

Tortuosity: *The ratio between free diffusion and diffusion in pore network*

$$\tau_i = \frac{\varepsilon D_{bulk}}{D_{eff,i}}$$

Compressible Navier-Stokes equations:

$$\frac{\partial \rho}{\partial t} + \frac{\partial(\rho U_i)}{\partial x_i} = 0$$

$$\frac{\partial(\rho U_i)}{\partial t} + \frac{\partial}{\partial x_j}(\rho U_i U_j) = -\frac{\partial P}{\partial x_i} + \frac{\partial \tau_{ij}}{\partial x_j}$$

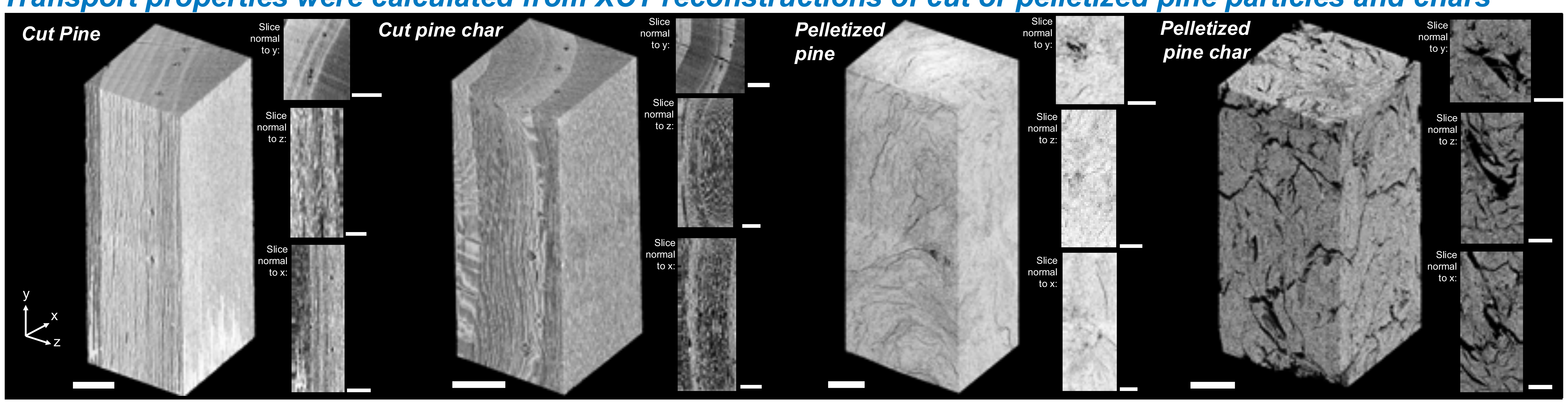
$$\frac{\partial(\rho e)}{\partial t} + \frac{\partial}{\partial x_i}(\rho e + P) = \frac{\partial}{\partial x_i}\left(k\frac{\partial T}{\partial x_i}\right) + \frac{\partial(\tau_{ij} v_j)}{\partial x_i}$$

Permeability: *Ease of fluid flow through pores*

$$K_i = \frac{U_i \mu}{\Delta P}$$

Results & Discussion

Transport properties were calculated from XCT reconstructions of cut or pelletized pine particles and chars



** All scalebars indicate 1 mm*

XCT geometries were used in CFD simulations to calculate permeability

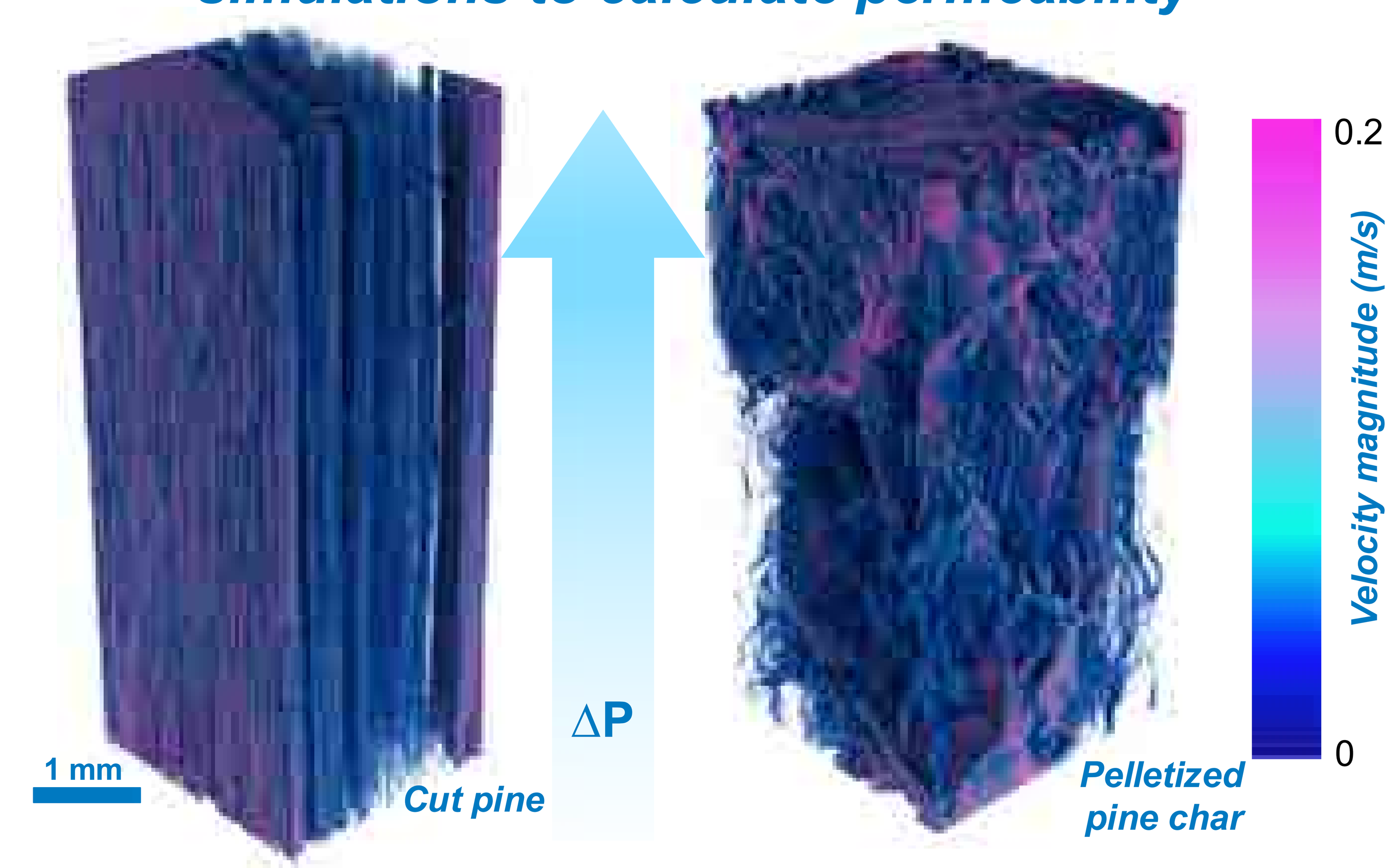


Table 1. Porosity and tortuosity of cut pine or pelletized pine particles and chars

	Porosity	Tortuosity, x	Tortuosity, y	Tortuosity, z
Cut Pine	0.44	40.74	2.34	116.55
Cut Pine char	0.60	8.03	1.19	9.91
Pelletized Pine	0.16	-	330.95	-
Pelletized Pine char	0.62	2.76	2.44	2.74

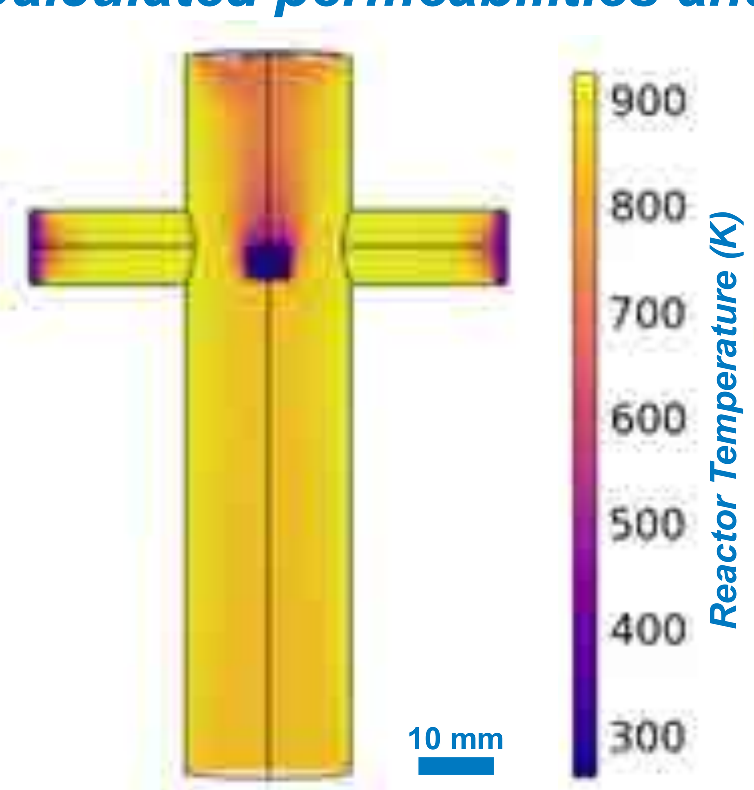
** Dashes indicate lack of pore connectivity required to calculate tortuosity*

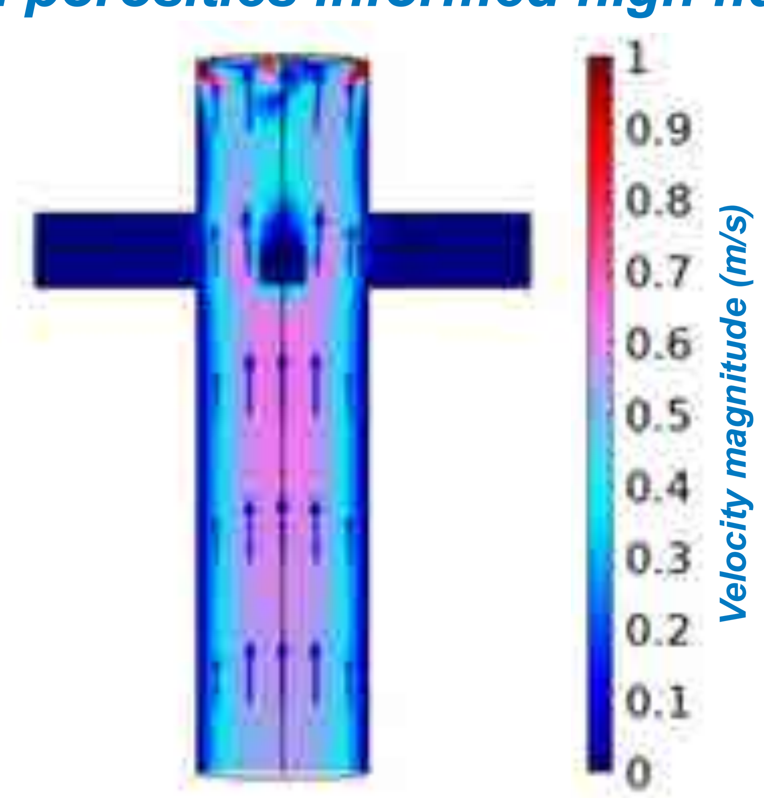
Table 2. Permeabilities cut pine and pelletized pine particles and chars in m^2

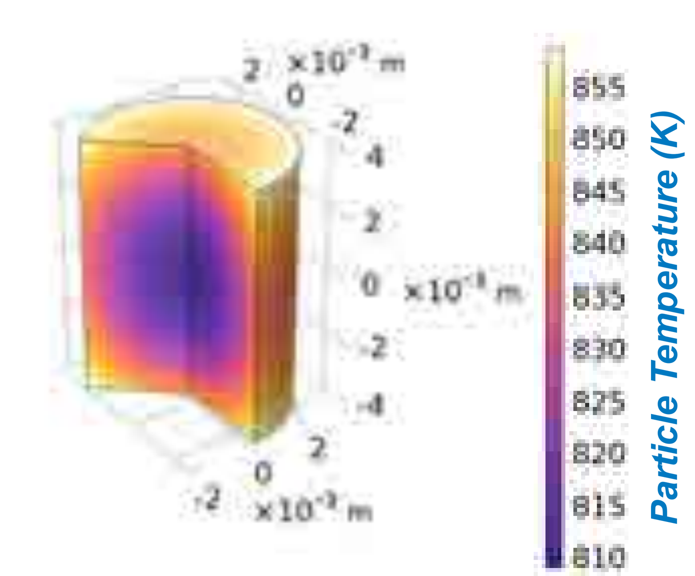
	x	y	z
Cut Pine	1.60E-14	5.64E-13	1.99E-27
Cut Pine char	7.98E-14	9.86E-13	5.76E-14
Pelletized Pine	6.23E-27	1.28E-15	2.22E-16
Pelletized Pine char	1.48E-12	1.26E-12	1.45E-12

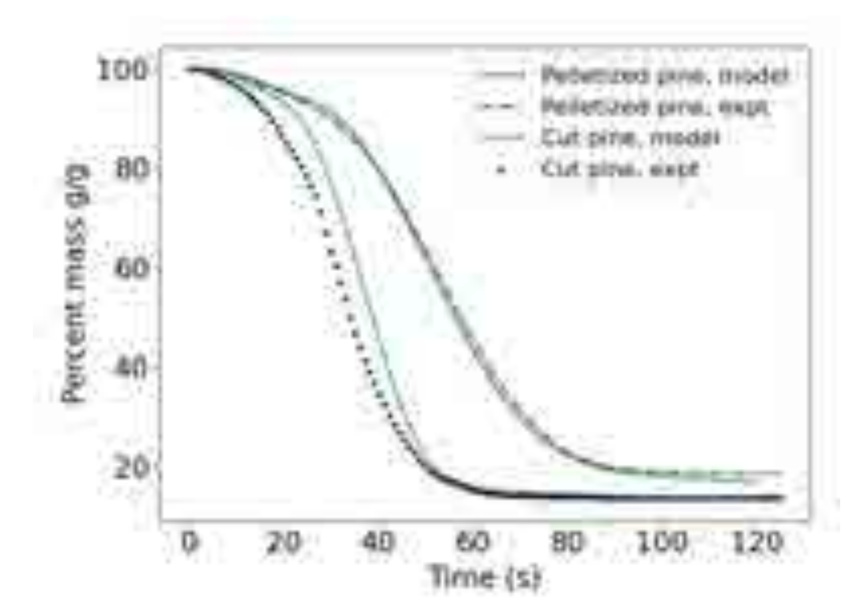
The anatomical features and natural anisotropy in tortuosity and permeability observed in the cut pine particle and char were erased in the pelletized pine particle which had lower porosity and permeability. The pelletized pine char had the highest, most isotropic tortuosity and permeability and of all samples with comparable porosity to cut pine char.

Calculated permeabilities and porosities informed high fidelity reactor and particle scale pyrolysis models









Higher permeability and porosity in the cut pine particles resulted in faster conversion and less residual char compared to the pelletized pine. Discrepancies in model and experimental mass loss may be remedied by adjustment of thermal conductivity in models.

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