Title: Stereolithography 3D Printing as a Versatile Tool to Manufacture Porous Electrodes for Redox Flow Batteries

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Redox flow batteries show potential for large-scale energy storage; yet their current costs limit widespread implementation. As the porous electrode microstructure defines the available surface area for electrochemical reactions, electrolyte transport, and fluid pressure drop [1], enhancing the electrode performance is a promising strategy to increase power density and thus reduce system costs. Conventional fibrous porous electrodes are repurposed from fuel cell gas diffusion electrodes and have not been tailored to sustain the requirements of liquid-phase electrode microstructure and resulting properties need to be developed [2]. In this context, 3D printing techniques are uniquely suited to manufacture controlled and deterministic architectures, enabling the tuning of electrochemical performance and hydraulic resistance [3].

In this work, we employ stereolithography 3D printing using a commercial desktop printer, followed by carbonization, to manufacture model grid structures and investigate their potential as redox flow battery electrodes. We use microscopy, tomography, spectroscopy, fluid dynamics, and electrochemical diagnostics, to evaluate the thermal behavior, manufacturing fidelity, and fluid and mass transport performance of ordered lattice structures in non-aqueous redox flow cells. We find that the flow field geometry (flow through versus interdigitated), printing orientation with respect to the printing platform, and pillar geometry (Figure 1a) influence the flow cell performance. The printing orientation impacts the surface roughness and resin spreading, which determines the electrode morphology, influencing the internal surface area, shrinking direction after carbonization, and thus the charge transfer, mass transfer, and hydraulic resistances. Moreover, mass transfer rates within the electrode are increased by altering the pillar shape to a helical or triangular design, or by using an interdigitated flow field design, providing directions to improve electrolyte mixing through the electrode. Although the commercial electrodes feature a greater internal surface area and therefore a better electrochemical performance, the area-normalized mass transfer coefficients and pressure drop can be reduced by utilizing 3D printed electrodes (Figure 1b-c). Going forward, 3D printing technologies that enable finer features combined with carbonization at elevated temperatures can be used to manufacture multi-scale electrode structures simultaneously providing excellent electrochemical performance and low hydraulic resistance. We anticipate that combining 3D printing with emerging computational topology optimization approaches could enable the bottom-up design of advanced electrode materials for electrochemical devices [4].



Figure 1: (a) The process workflow to obtain a conductive electrode from a nonconductive 3D print to be used in a flow cell, including a subsection of the tomographic image after carbonization together with the evaluated pillar geometries. **(b)** The pressure drop over a range of velocities for the 3D printed electrode and two commercial electrodes (Freudenberg H23 and ELAT Cloth). **(c)** The mass transfer for different superficial velocities represented by the Sherwood over the Peclet number plotted on a double-log scale for the 3D printed electrode and the two commercial electrodes.

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