

Title: Investigating mass transfer relationships in stereolithography-based 3D printed electrodes for redox flow batteries

Authors: Maxime van der Heijden, Marit Kroese, Zandrie Borneman, and Antoni Forner-Cuenca

Redox flow batteries are a promising option for large-scale energy storage but remain too costly for widespread deployment. One approach to increase cost competitiveness is to improve the power density and efficiency of the electrochemical cell. Porous electrodes determine the performance of the cell, and their three-dimensional structure determines the electrolyte transport, fluid pressure drop, and the available surface area for electrochemical reactions [1]. Conventional porous electrodes are fibrous mats assembled in coherent structures [2]; however, they are repurposed from fuel cell gas diffusion electrodes and have not been tailored for redox flow batteries. Hence, an effective means to improve the performance of redox flow batteries is by engineering porous electrode structures to sustain the requirements of liquid-phase electrochemistry. To this purpose, there is a need to develop new manufacturing techniques affording a higher degree of control over the electrode microstructure and resulting properties. Additive manufacturing, or 3D printing, can be employed to manufacture customized, controlled, and deterministic architectures, enabling the fine-tuning of the electrical and hydraulic performance of porous electrodes [3].

In this study, we manufacture model grid structures using stereolithography 3D printing followed by carbonization (**Figure 1a**) to tune the physiochemical properties of electrodes to be used in redox flow batteries. We employ a suite of microscopy, tomography, spectroscopy, fluid dynamics, and electrochemical diagnostics to understand the impact of the electrode structure on the mass transport and hydraulic performance of ordered lattice structures in flow cells. Here, the influence of the printing direction, pillar geometry (**Figure 1b**), and flow field type on mass transport is investigated using an organic electrolyte. We elucidate correlations between the electrode structure and performance metrics, namely surface area, pressure drop, and mass transfer correlations. We find that the printing orientation impacts the electrode performance through a change in electrode morphology caused by resin spreading and surface roughness, affecting the shrinking direction upon carbonization, internal surface area, and thus the charge transfer, mass transfer, and hydraulic resistances. Furthermore, we find that mass transfer rates within the electrode are enhanced by using an interdigitated flow field or by altering the pillar shape to a helical or triangular design, which we hypothesize improves mixing. Compared to commercial carbon-fiber electrodes, the pressure drop is significantly reduced (**Figure 1c**) as expected due to larger pore sizes ($\sim 500\ \mu\text{m}$ for the grids vs. $2\text{-}100\ \mu\text{m}$ for the Freudenberg H23 paper electrode and $2\text{-}300\ \mu\text{m}$ for the ELAT Cloth electrode). Even though the commercial electrodes feature a superior internal surface area compared to the 3D printed electrodes, their area normalized mass transfer coefficients are lower (**Figure 1d**). Going forward, the use of printing technologies enabling finer features in combination with carbonization at elevated temperatures can be used to manufacture multiscale electrodes simultaneously providing low hydraulic resistance and excellent electrochemical performance. Additive manufacturing in combination with emerging computational approaches in topology optimization [4] might enable the bottom-up design and manufacturing of advanced electrode materials.

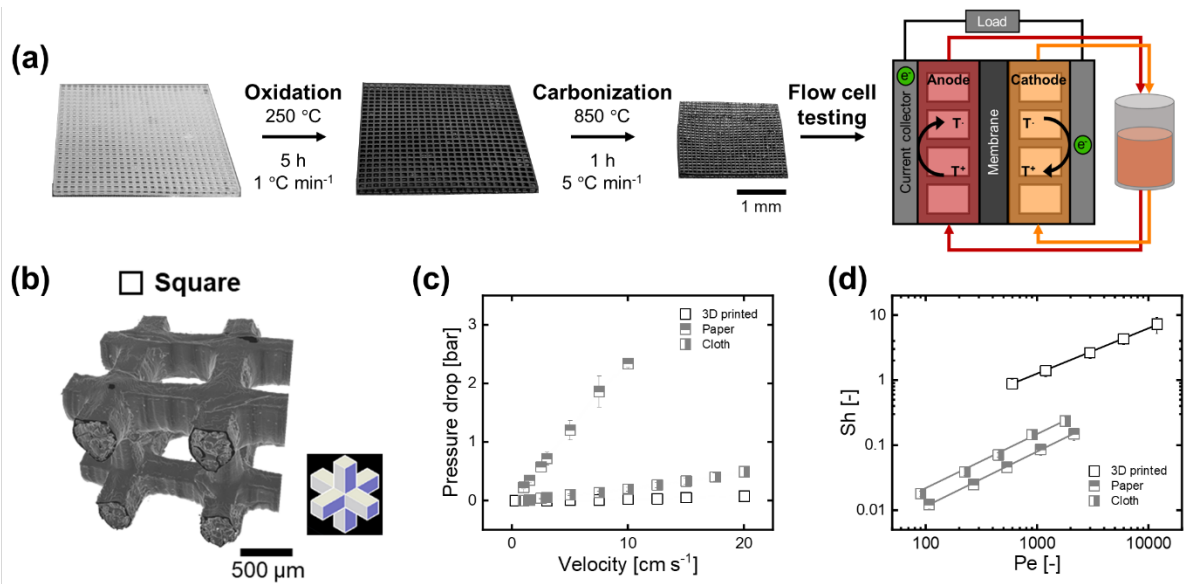


Figure 1: (a) Representation of the process workflow to obtain a conductive electrode from a nonconductive 3D print to be tested in a flow cell. (b) Subsection of the tomographic image of the 3D print after carbonization. (c) The pressure drop over a range of velocities for the 3D printed electrode and two commercial electrodes (Freudenberg H23 and ELAT Cloth). (d) 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.

References

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