Research group Membrane Materials and Processes

Towards bottom-up design of porous electrode microstructures for redox flow batteries

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Redox Flow Batteries

Redox flow batteries (RFBs) (Figure 1) are a promising technological platform for grid-level storage of intermittent renewable electricity. RFBs are rechargeable electrochemical systems that enable decoupling of the energy (i.e., tank volume) and power (i.e., reactor size), facilitating their large-scale deployment[1]. Porous electrodes need to fulfil several performance-relevant functions related to kinetics and transport which determine the overall efficiency and costs of RFBs.

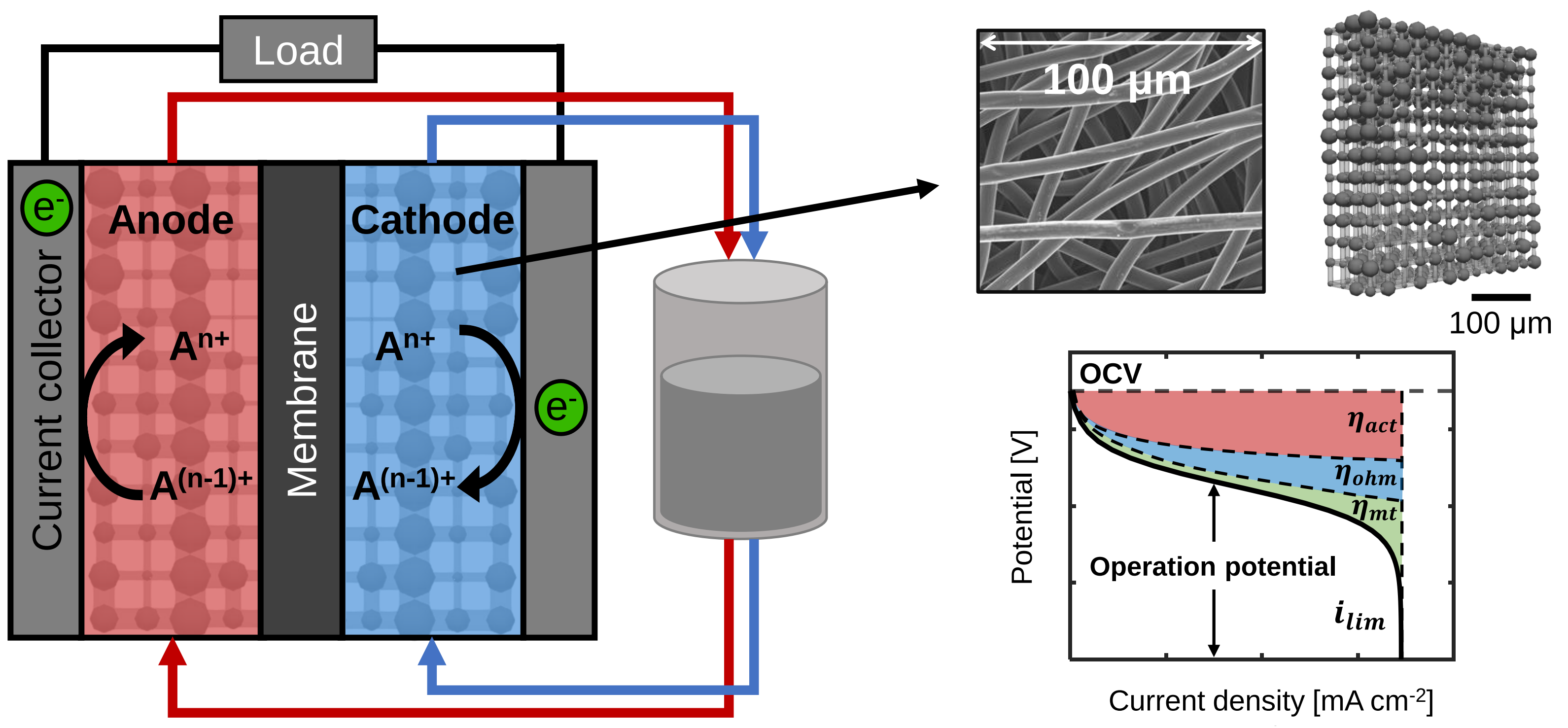

Figure 1: Schematic diagram of a symmetric flow cell, the porous electrode, and the discharge polarization curve showing the activation, ohmic, and mass transfer losses[1,2].

Bottom-up design of porous electrodes

We aim to predict and fabricate optimal electrode geometries for RFBs from the bottom-up, using a combination of experimental characterization, pore network modeling, genetic algorithms, and manufacturing techniques (Figure 2).

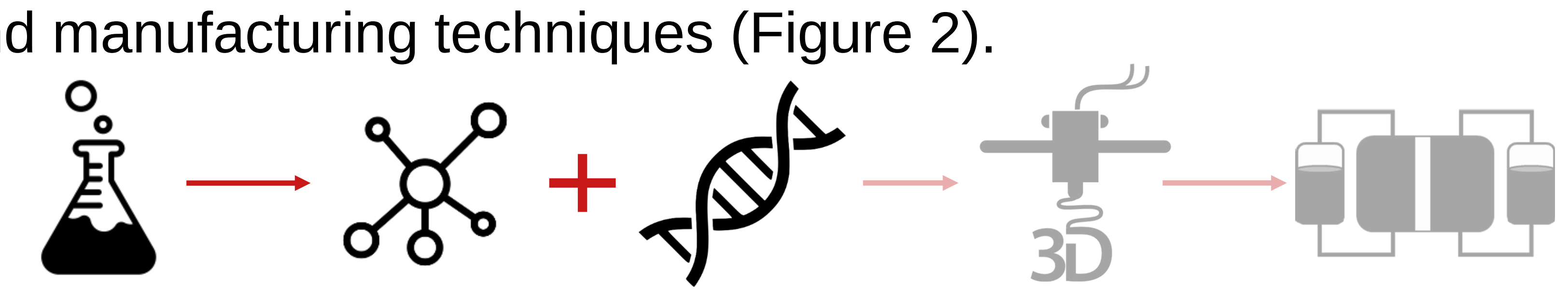

Figure 2: Illustration of the research pipeline.

Iterative pore network model

A three-dimensional electrochemical modelling toolkit integrated in an open access platform (OpenPNM) was developed. The simulation domain is electrolyte-agnostic and computes the electrolyte fluid transport (1), charge transport (2), and species transport (3) (Figure 3) in an iterative fashion at low computational cost. The pore networks of two commercial electrodes were used in the pore network model, extracted using X-ray tomography and the SNOW algorithm[3] (Figure 3).

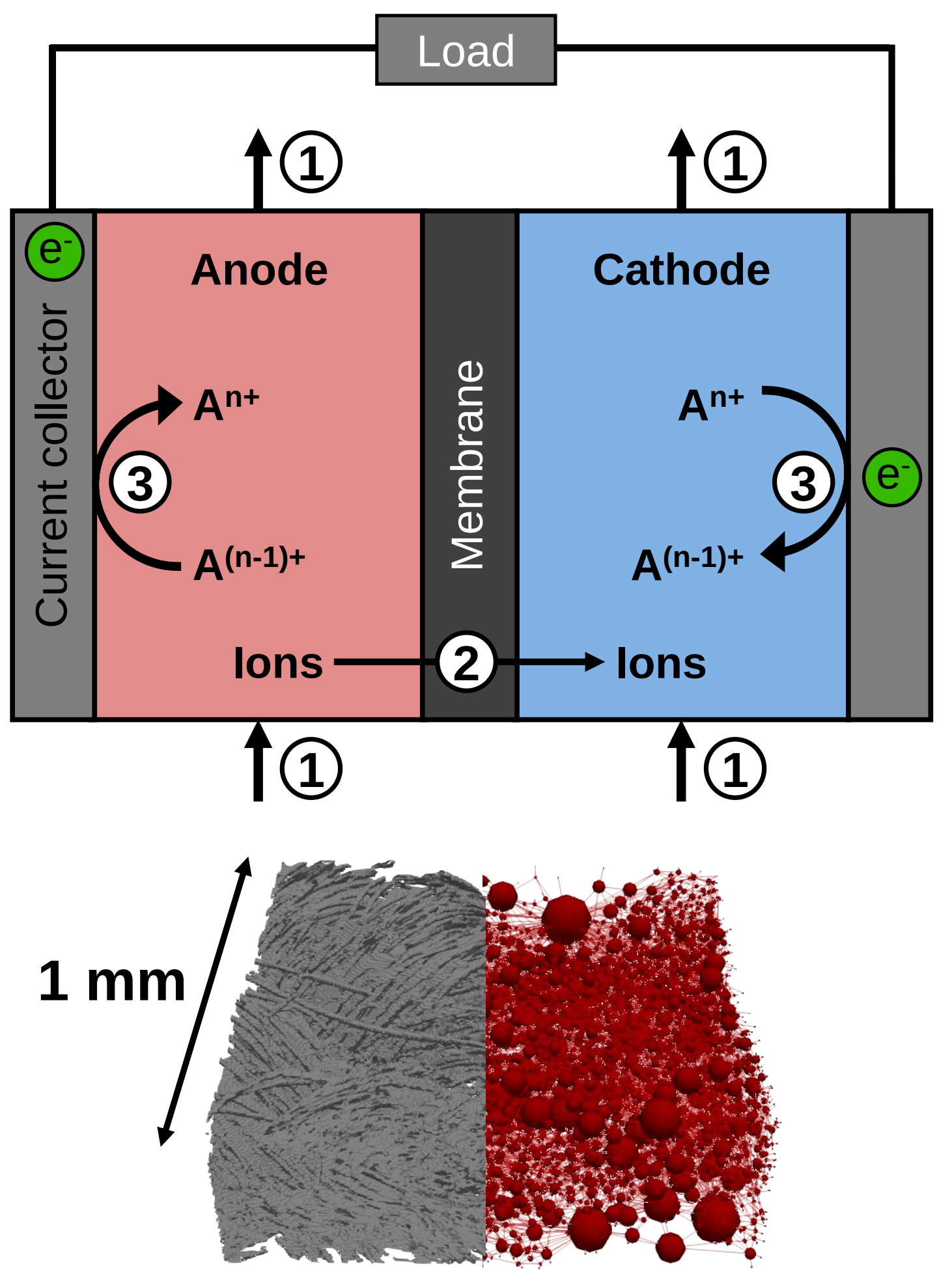

Figure 3: Schematic representation of the pore network modelling domain (top) with a dry tomograph and pore network of a carbon cloth (bottom).

The model was validated for an organic electrolyte using a symmetric cell (Figure 4) for two commercial electrodes without the use of fitting parameters, besides the experimental determination of the exchange current density.

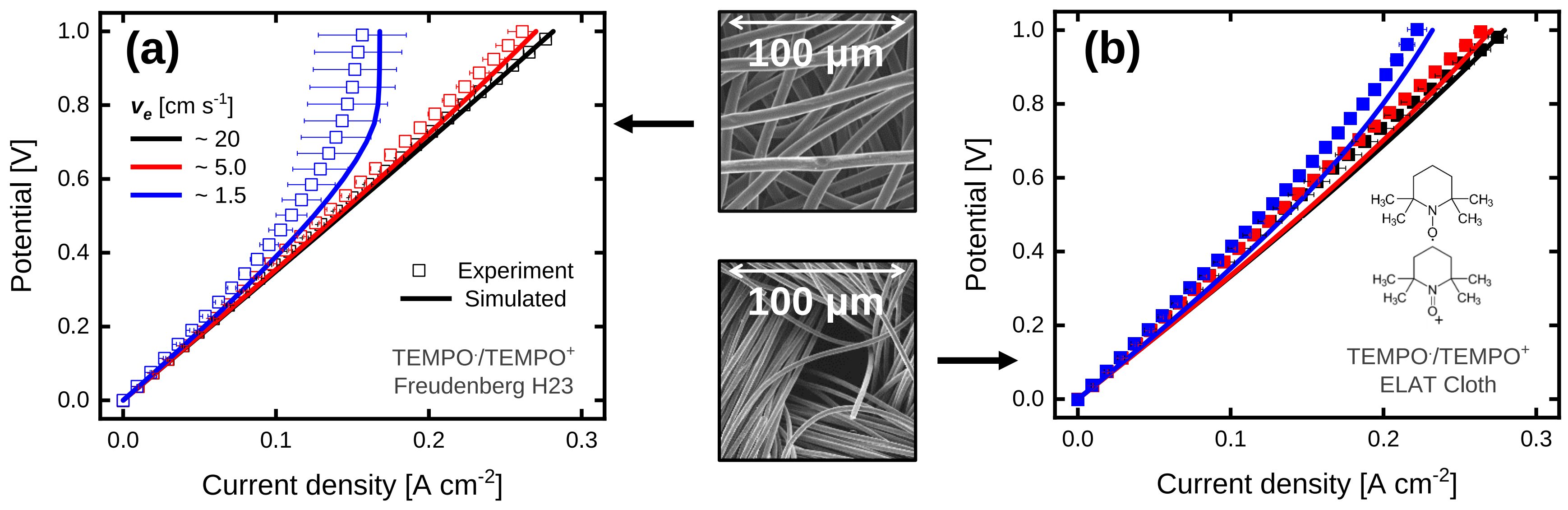

Figure 4: Polarization curves of: **(a)** a carbon paper (Freudenberg H23), and **(b)** a carbon cloth (ELAT Cloth) electrode. Where the experiments are shown with symbols and the model with a line. A single electrolyte cell with an organic electrolyte (0.1 M TEMPO·/TEMPO$^+$ in 1 M TBAPF$_6$/MeCN) was used[3].

Genetic Algorithm

A genetic algorithm was coupled with the PNM for the bottom-up design of porous electrode microstructures, improving the fitness over generations (Figure 5a). The resulting pore network (Figure 6) features: 1) the generation of fluid highways, decreasing the pumping losses (P_{pump}) by 73%, and 2) hierarchical organization of the pores, resulting in a bimodal pore size distribution (Figure 5b) and an improved electrical performance (P_{max} - P_{el}) of 42%.

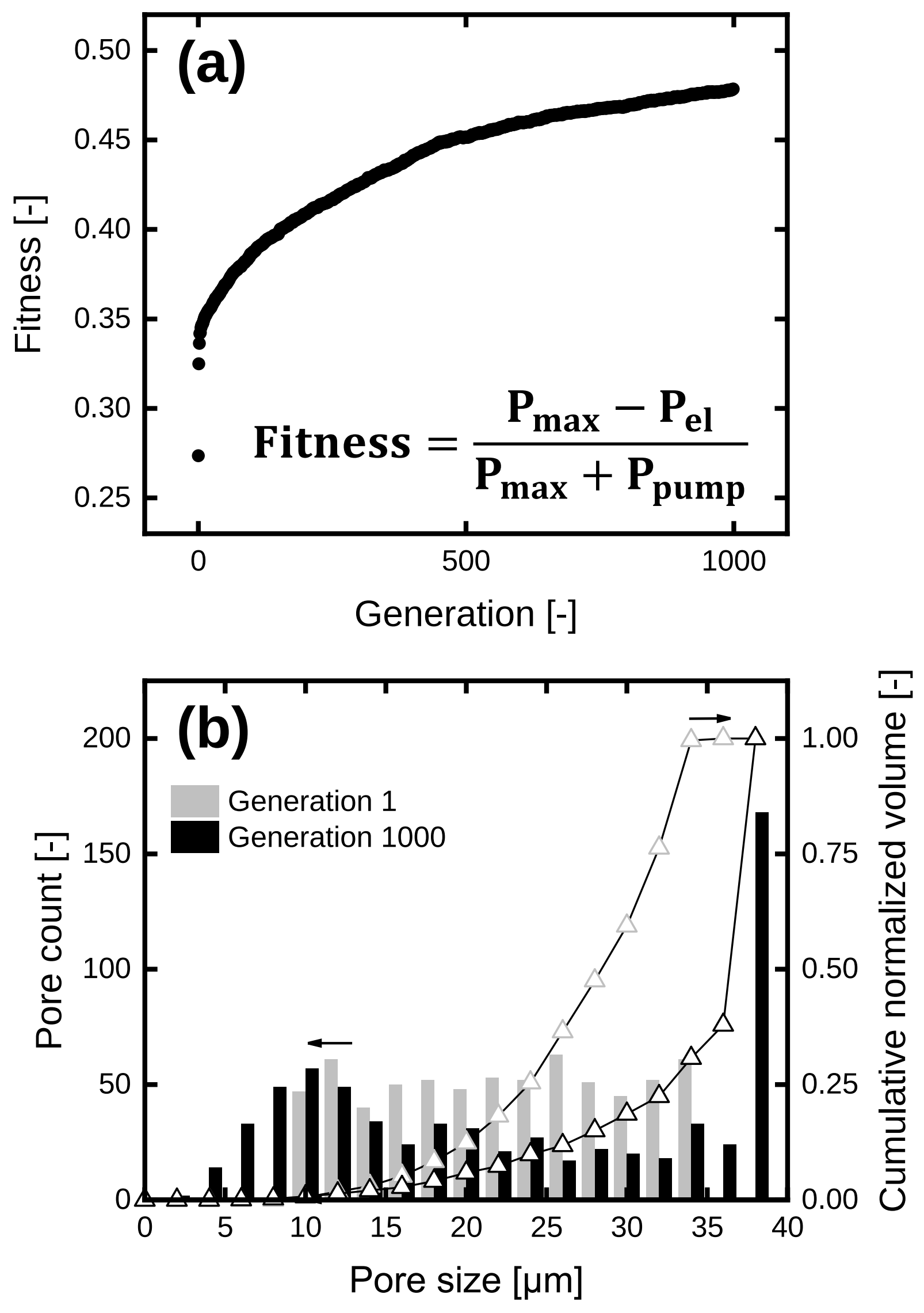

Figure 5: **(a)** The fitness function evolution over the number of generations, and **(b)** the pore size distribution for the first and last generation[2].

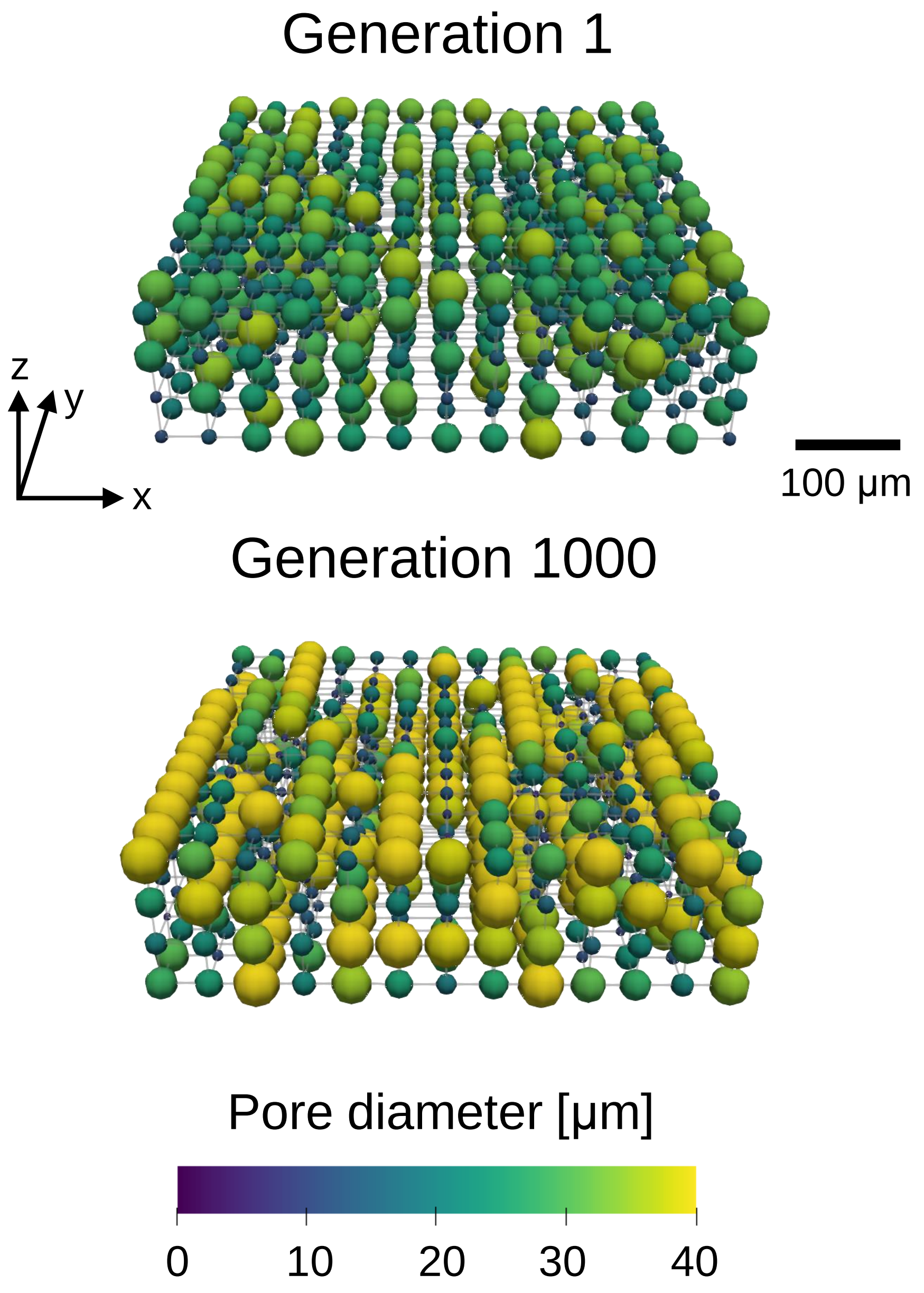

Figure 6: The first and final pore networks showing the generation of fluid highways along the flow direction[2].

Acknowledgements

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References

[1] van der Heijden *et al.*, *Encyclopedia of Energy Storage*, 480-499 (2022)
[2] van der Heijden *et al, J. Electrochem. Soc.*, **169** 040505 (2022)
[3] van Gorp *et al, Chem. Eng. J,* 139947 (2022)

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