



Contribution ID: 565

Type: Oral Presentation

Non-solvent Induced Phase Separation: A Synthetic Approach Towards High Performance Redox Flow Battery Electrodes

Monday, 31 May 2021 16:10 (15 minutes)

Transitioning to a sustainable energy economy is one of the greatest challenges of this century. Integrating renewables (e.g., wind and solar power) into the grid must be accelerated to limit the devastating effects of climate change. Due to their intrinsic intermittency, large-scale energy storage must be deployed to balance the mismatch between supply and demand[1]. Redox flow batteries (RFBs) stand out as a promising candidate due to their ability to independently scale power and energy and projected lower costs[2,3]. The performance of RFBs largely depends on the porous electrodes microstructure and chemical composition as they must simultaneously provide high surface area for electrochemical reactions, low pressure drop, high electrical conductivity, and facile mass transport[4]. State-of-the-art electrodes are composed of carbon fibers which are arranged together using mechanical methods forming idiosyncratic structures such as papers, cloths and felts[5]. Their fabrication involves multiple complex subprocessing steps[6] impacting the final manufacturing cost and offering limited versatility to control the three-dimensional structure of the material, which ultimately hampers widespread commercialization of the technology.

Here, we introduce the non-solvent induced phase separation (NIPS) as a simple and versatile fabrication method for carbonaceous porous electrodes[7]. Drawing inspiration from membrane science and technology[8], the NIPS method has been leveraged to synthesize morphologically-diverse microstructures (e.g., isoporous, macrovoids, porosity gradient) which are appealing to electrode manufacturing. A polymer solution, containing polyacrylonitrile (PAN, carbon-containing) and polyvinylpyrrolidone (PVP, pore-forming agent) dissolved in N,N-dimethylformamide (solvent) was casted in a mold and subsequently immersed in water (non-solvent). Finally, the polymeric scaffold is carbonized under inert conditions to form a conductive network. Easily adjustable parameters, such as solvent type, polymer concentration and temperature enable control of the final electrode microstructure. In this work, we study the influence of the PAN:PVP ratio on the electrode microstructure and its resulting effect on RFBs performance.

Microstructural characterization revealed a multimodal pore size distribution composed of fine, interconnected microvoids (pore diameter \approx 2-15 μ m) coupled with through plane, finger-like macrovoid channels (throat diameter > 50 μ m) forming honeycomb networks. The unique microstructure, not attainable with traditional carbon-fiber manufacturing techniques, enables large surface area at the membrane-electrode interface and fast electrolyte replenishing which reduces mass transfer resistance within the electrode. Flow battery tests with Fe²⁺/Fe³⁺ electrolyte revealed a considerable reduction of the charge transfer (RCT \approx 0.016 Ω) and mass transfer (RMT \approx 0.025 Ω) overpotentials of the novel electrodes compared to the commercial baseline (SGL29AA, RCT \approx 0.326 Ω and RMT \approx 0.151 Ω at a linear velocity of 5 cm s⁻¹) at the expense of a slight increase in pressure drop. In the final part, we demonstrate the use of NIPS-electrodes in a full all-vanadium RFB. The polarization analysis revealed a ca. 70% improvement in power density compared to the baseline material, which can be attributed to reductions in the charge transfer and mass transport overpotentials. Although nascent, NIPS emerges as a promising platform to engineer porous electrodes for RFBs and other convection-enhanced electrochemical systems.

Time Block Preference

Time Block B (14:00-17:00 CET)

References

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Session Classification: MS19

Track Classification: (MS19) Electrochemical processes in porous media