

Electronic Supplementary Information

## **Gel Composite Electrolyte - An Effective Way to Utilize Ceramic Fillers in Lithium Batteries**

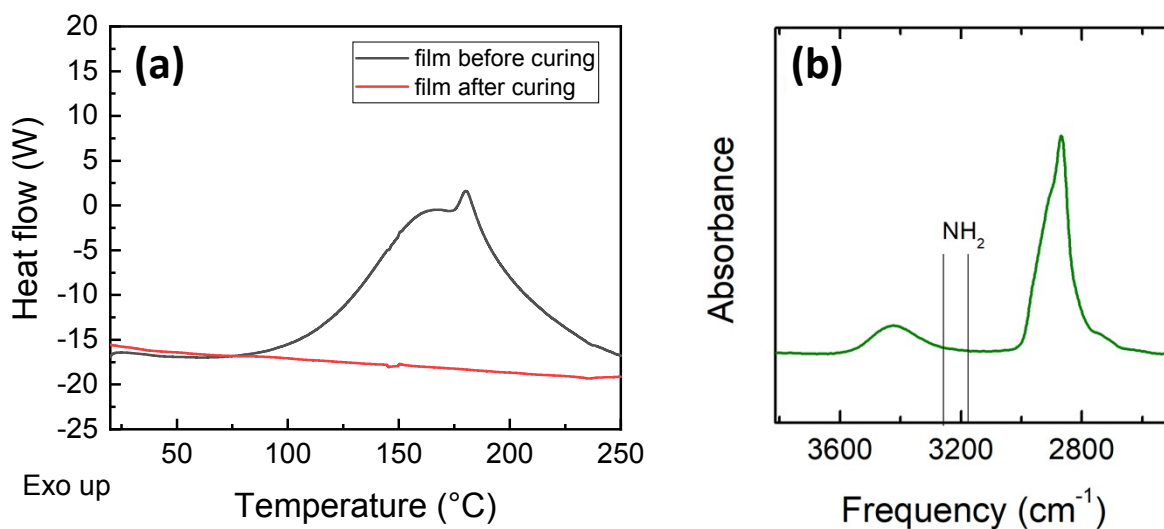
X. Chelsea Chen<sup>a, \*</sup>, Yiman Zhang<sup>a</sup>, Laura C. Merrill<sup>b</sup>, Charles Soulen<sup>a</sup>, Michelle L. Lehmann<sup>c</sup>,  
Jennifer L. Schaefer<sup>b</sup>, Zhijia Du<sup>d</sup>, Tomonori Saito<sup>a, c</sup>, and Nancy J. Dudney<sup>a</sup>

<sup>a</sup>Chemical Sciences Division and <sup>d</sup>Energy and Transportation Science Division, Oak Ridge National  
Laboratory, Oak Ridge, TN 37830, USA.

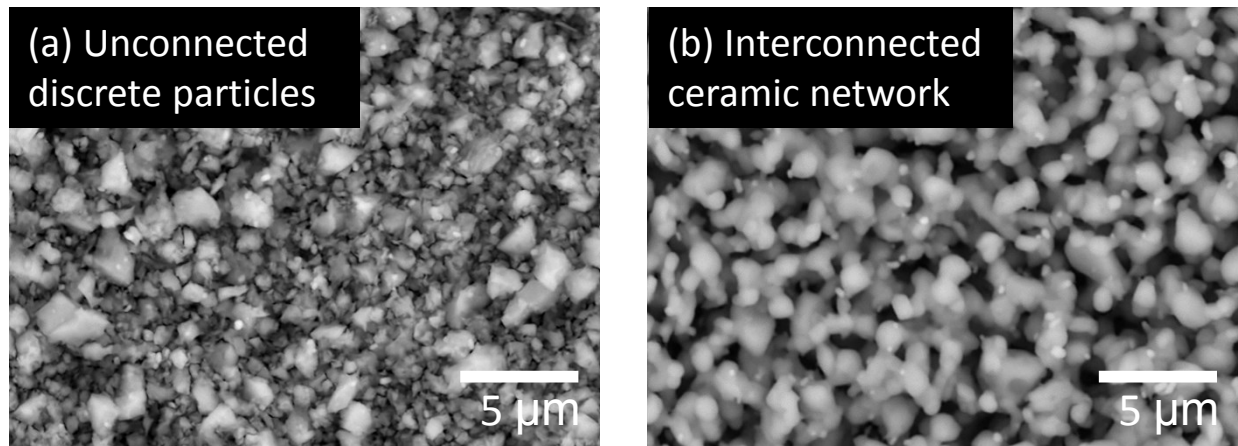
<sup>b</sup>Department of Chemical and Biomolecular Engineering, University of Notre Dame, Notre Dame, IN  
46556, USA.

<sup>c</sup>The Bredesen Center for Interdisciplinary Research and Graduate Education, University of Tennessee,  
Knoxville, TN 37996, USA.

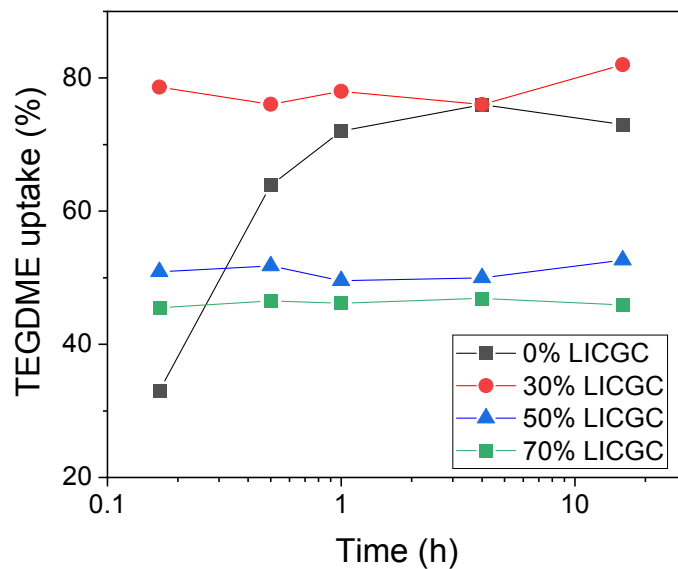
\*corresponding author email: [chenx@ornl.gov](mailto:chenx@ornl.gov)



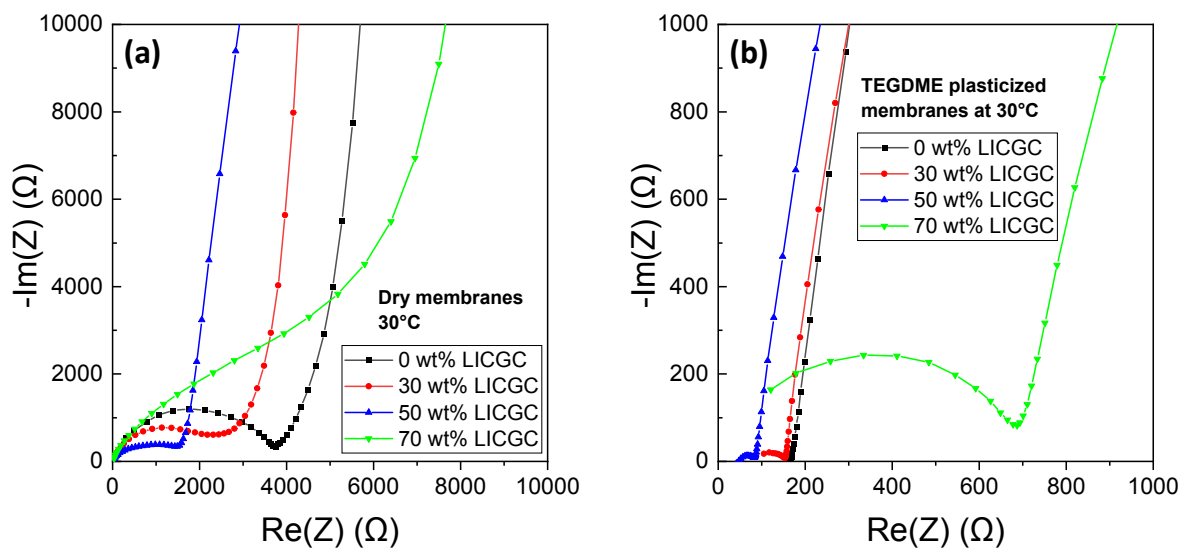
**Fig. S1** (a) Differential scanning calorimetry (DSC) thermograms of polymer and composite electrolyte films before and after curing. A large exothermic peak was observed for the uncured film. After curing, the exothermic peak disappeared, indicating completion of the crosslinking reaction. (b) Infrared spectrum of cured polymer electrolyte film, indicating that all NH<sub>2</sub> groups have reacted, and likely all NH groups. The broad stretching vibrations of OH and NH begin to overlap in the 3370 cm<sup>-1</sup> to 3400 cm<sup>-1</sup> regions.



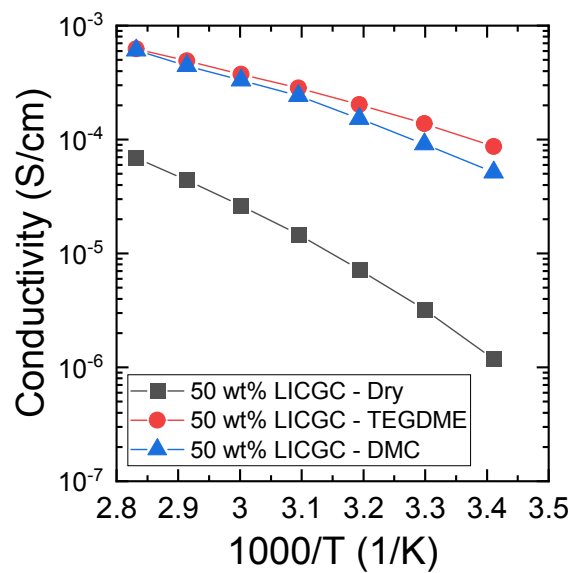
**Fig. S2** SEM images showing the morphology differences between (a) densely packed but unconnected discrete LICGC<sup>TM</sup> particles and (b) partially sintered and interconnected LICGC<sup>TM</sup> ceramic network.



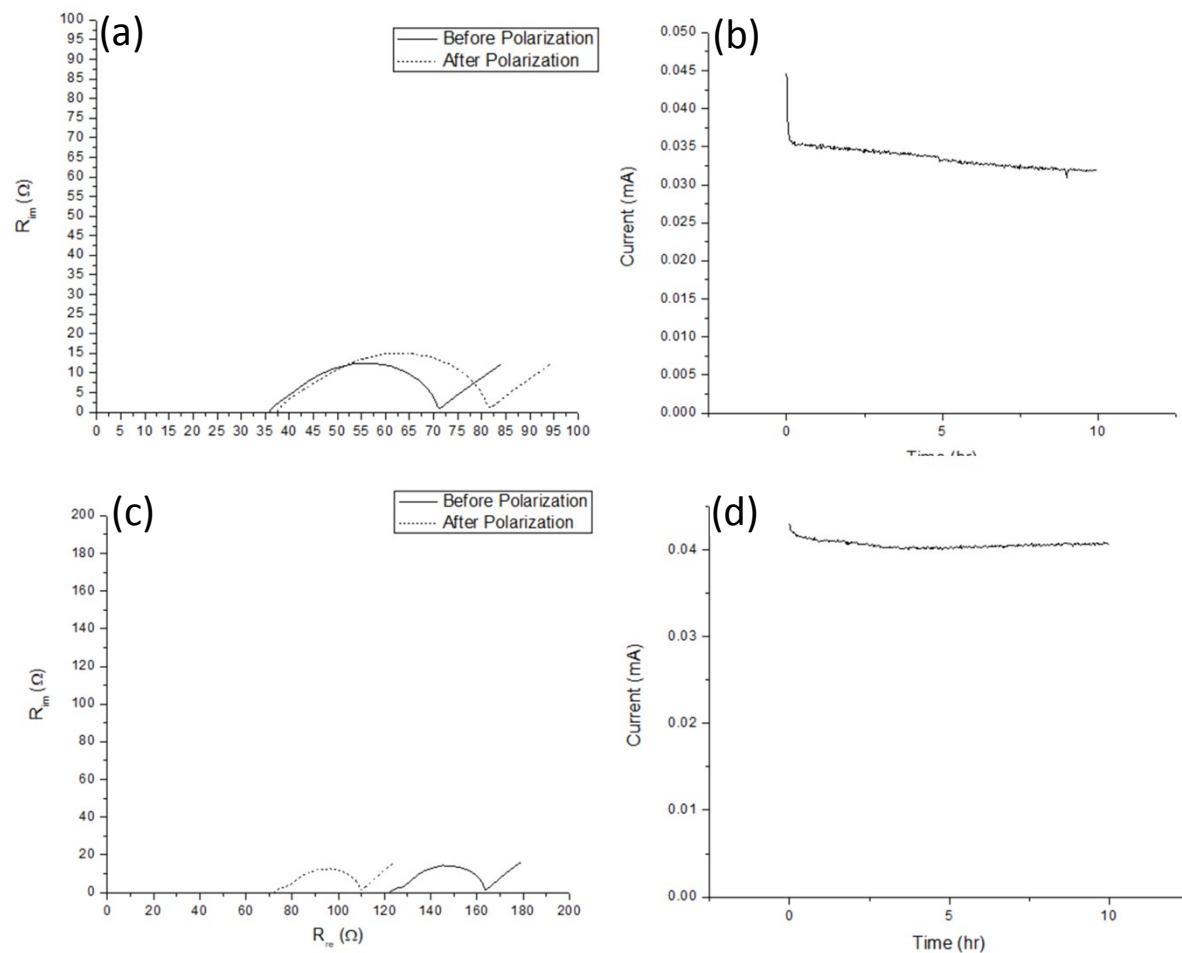
**Fig. S3** Tetraethylene glycol dimethyl ether (TEGDME) uptake of the polymer and composite electrolyte membranes as a function of time. The thinner composite membranes with less than 100  $\mu\text{m}$  thickness reached saturated uptake within 10 min. The thicker pristine polymer membrane reached saturated uptake in 1 hour.



**Fig. S4** Representative Nyquist plot of dry (a) and TEGDME plasticized (b) polymer and composite electrolyte membranes at 30 °C, with stainless steel blocking electrodes.



**Fig. S5** Ionic conductivity of composite electrolyte membrane containing 50 wt% LICGC (dry weight) as a function of inverse temperature. Black curve, dry membrane; red curve, plasticized with TEGDME; and blue curve, plasticized with dimethyl carbonate (DMC). Similar conductivity was observed in the two plasticizers.



**Fig. S6** Details of  $\text{Li}^+$  transference number measurements. (a,b), EIS (a) and polarization (b) of film with 0 wt% ceramic loading. (c, d), EIS (c) and polarization (d) of film with 50 wt% ceramic loading.