

Highly microporous carbon aerogel as sulfur host in lithium-sulfur batteries

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Lithium-sulfur (Li-S) batteries are currently one of the promising systems among next generation batteries due to their high theoretical specific capacity and high specific energy density. However, the commercialization of Li-S batteries is left challenged by the capacity loss induced by the polysulfide shuttle effect. Encapsulation of active material in the cathode matrix is one of the many strategies inhibiting the polysulfide shuttle effect^[1].

Carbon Aerogels (CA), first introduced by Richard Pekala in 1989^[2], are highly promising materials to be used as cathode matrix encapsulating sulfur. Starting from organic resorcinol-formaldehyde (RF) aerogels, CA exhibits highly porous structure with huge porosity up to 97 %, high surface area about 500-2000 m²/g, large micropore volume about 0.1-0.6 cm³/g and sufficient electrical conductivity^[3-4]. In addition, the crucial advantage of CA is its adjustable porous structure and pore size distribution. The microstructure can be tuned during synthesis and carbonization of organic aerogels. Furthermore, the flexibility of CA enables the elimination of the crack formation during volume change of sulfur.

In the present study, we synthesized and investigated highly microporous CA as conductive matrix embedding sulfur for cathodes in Li-S batteries^[5]. The improved carbonization process leads to an increase in micropore volume. Thus, the amount of active material sulfur in micropores is maximized. The innovative gas phase sulfur infiltration of the CA traps short sulfur chains in the micropores (< 2 nm). Complementary characterization techniques such as TGA and XPS are used for demonstration. It is shown that sulfur infiltrated microporous CA cathodes are able to suppress the polysulfide shuttle effect, leading to a higher cycle stability of the cell in both ether and carbonate based electrolytes.

Moreover, the influence of the structural and physical properties of different CAs, including density and pore size, on the electrochemical performance of the cell is discussed.

[1] L. Ma, K. Hendrickson, S. Wei, L. Archer, *Nano Today* **2015**, *10*.

[2] R. W. Pekala, *Journal of Materials Science* **1989**, *24*, 3221-3227.

[3] R. W. Pekala, J. C. Farmer, C. T. Alviso, T. D. Tran, S. T. Mayer, J. M. Miller, B. Dunn, *Journal of Non-Crystalline Solids* **1998**, *225*, 74-80.

[4] R. W. Pekala, S. T. Mayer, J. L. Kaschmitter, F. M. Kong, in *Sol-Gel Processing and Applications* (Ed.: Y. Attia), Springer US, **1994**, pp. 369-377.

[5] M. Schwan, L. Ratke, *Journal of Carbon Research* **2016**, *2*, 22.