Supplementary Information

Sol-gel encapsulated lithium polysulfide catholyte and its application in lithium-sulfur batteries

Leland C. Smith[a], Peter Malati[ab], Jonathan Fang[ac], Wade Richardson[ad], David Ashby[a], Chun Han Lai[a] and Bruce S. Dunn[a]

[a] Department of Materials Science and Engineering, University of California at Los Angeles, Los Angeles, CA 90095, USA
[b] Teledyne Scientific Company, 1049 Camino Dos Rios, Thousand Oaks, CA, USA
[c] TLC Biopharmaceuticals, Inc., 432 North Canal Street, Suite #20, South San Francisco, CA 94080
[d] StemBios Technologies, Inc., 2530 Corporate Place, STE A112 Monterey Park, CA 91754

E-mail: lcsmith50@g.ucla.edu

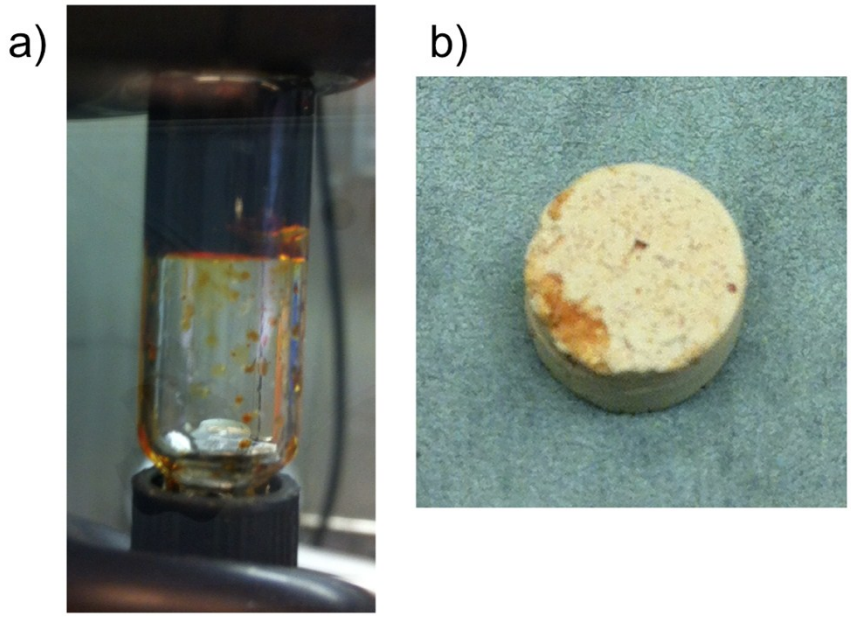

Fig. S1 (a) Image of the dark red PG in an overturned vial, showing that it is completely solid (b) monolith of silica obtained after the lithium polysulfide was removed from the PG by solvent exchange.

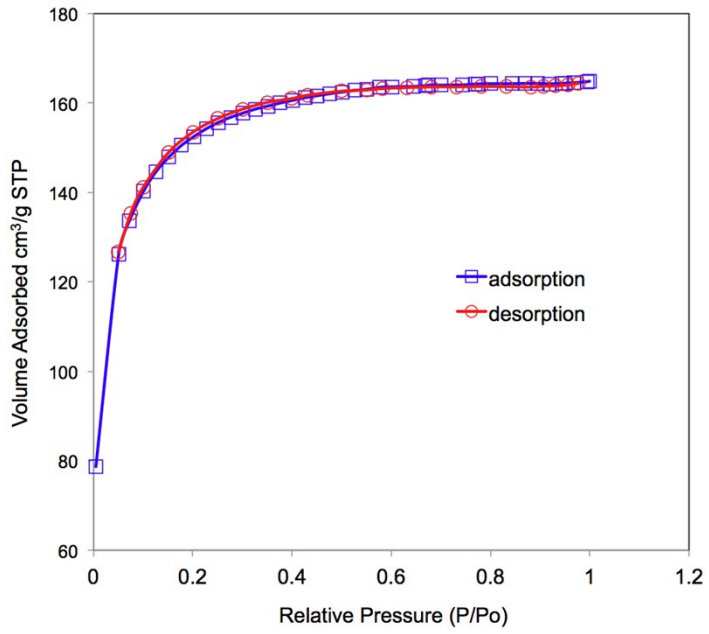

Fig. S2 Nitrogen adsorption and desorption plot obtained for the silica matrix of the PG.

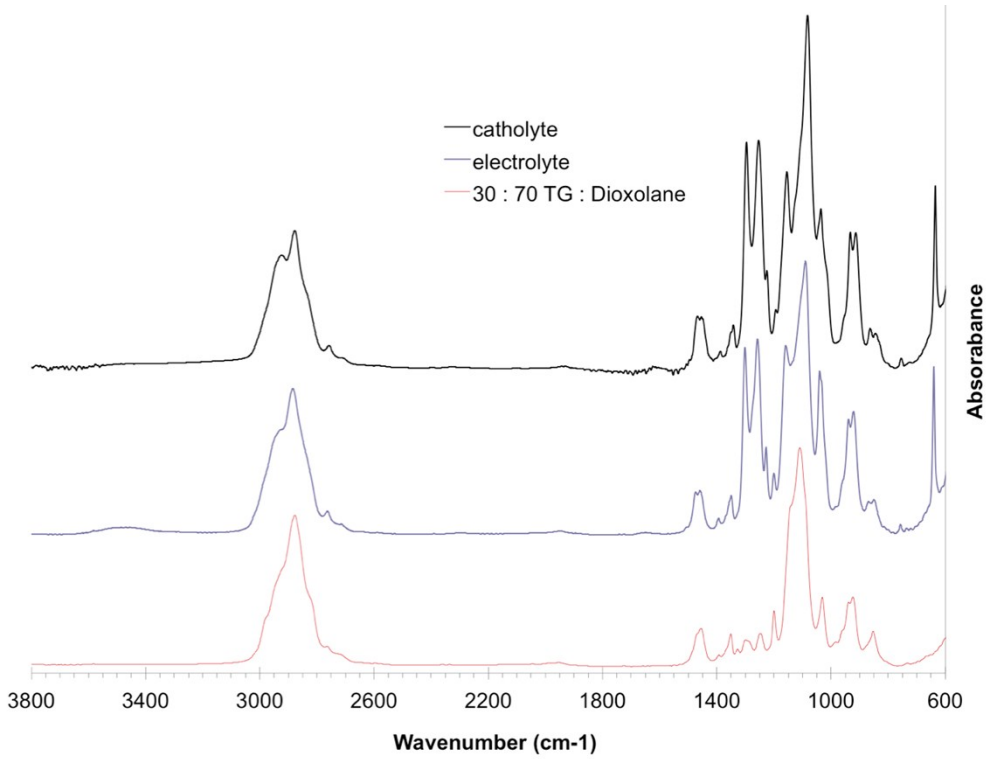

Fig. S3 The peak identities for the catholyte (Table 3) were determined by measuring the FTIR spectra of its various components. The FTIR spectrum of the catholyte (top) is compared to that of the electrolyte (middle) and the 30 : 70 :: TG : DXL solvent mixture (bottom). The electrolyte contains 0.8 M $LiSO_3CF_3$ dissolved in 30 : 70 :: TG : DXL. In addition to the electrolyte, the catholyte contains 0.5 M Li_2S_6.

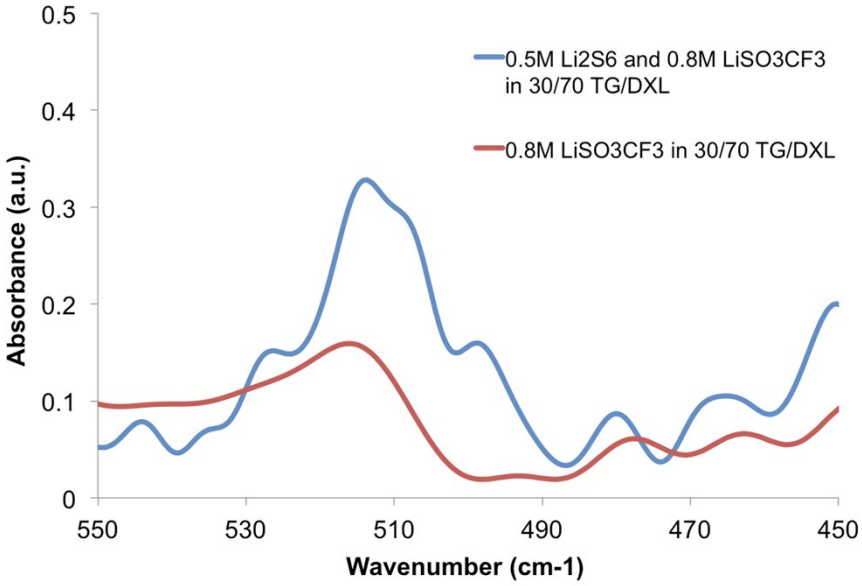

Fig. S4 The only evidence for the presence of lithium polysulfides in the FTIR data is a small peak at 517 cm^{-1}.

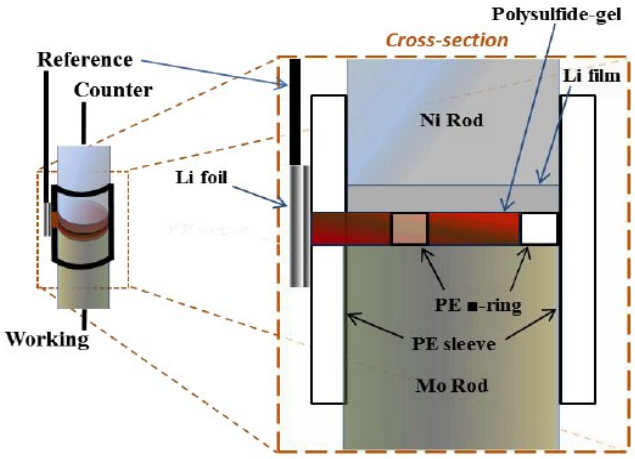

Fig. S5 Schematic of three-electrode cell used for electrochemical testing of PG. The anode is lithium metal applied to a polished Ni rod. The cathode current collector is a polished Mo rod. The rods are pushed together in a polyethylene sleeve and separated by a 1 mm spacer. Before gelation PG is cast into the opening and an external Li reference electrode is applied.

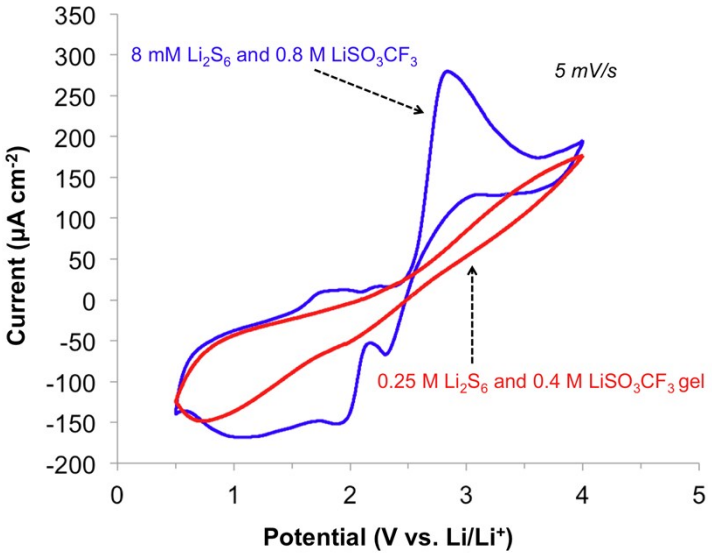

Fig. S6 Cyclic voltammograms of: 1. A battery prepared from 1:1 volume mixture of dehydrated sol and catholyte (0.5 M Li_2S_6 and 0.8 M $LiSO_3CF_3$ in 30/70 TG/DXL) and 2. A battery prepared with polysulfide catholyte (8 mM Li_2S_6 and 0.8 M $LiSO_3CF_3$). Electrodes: Mo, working; Li, counter; Li, reference. The PG displays a similar shape, but with wider voltage separation between peaks.

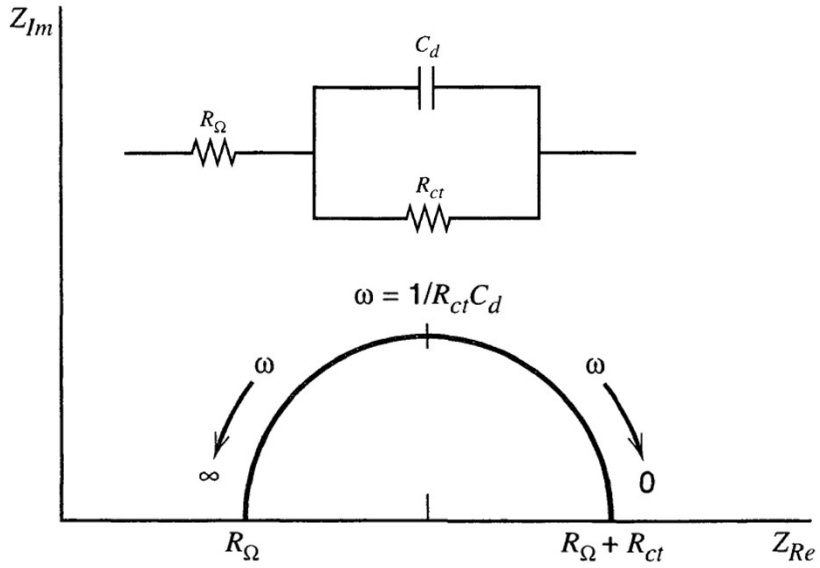

Fig. S7 An equivalent circuit consisting of a capacitor (C_d) with one resistor in parallel (*Rct*) and one resistor in series (R_Ω). R_Ω is attributed to the electronic contact resistance and electrolyte resistance and *Rct* is attributed to charge transfer in the PG. Adapted from Ref [32].

Table S1 – Summary of Galvanostatic Cycling Data for Polysulfide Gel

cycle number	current density ($\mu A/cm^2$)	charge capacity (mAh/cm^2 // mAh/g S)	discharge capacity (mAh/cm^2 // mAh/g S)
1 - 5	100	0.032 // 6.7	0.055 // 12
6 - 9	50	1.65 // 345	0.69 // 144
10	10	1.19 // 249	1.00 // 209