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Investigation of the capacity retention mechanisms in novel composite sulfur copolymer-base cathodes for high-energy density Li-S batteries[1] VLADIMIR OLESHKO, JENNY KIM, KEVIN MASSER, STEVEN HUDSON, CHRISTOPHER SOLES, Material Measurement Laboratory, National Institute of Standards and Technology, 100 Bureau Drive, Gaithersburg, MD 20899, JARED GRIEBEL, WOO JIN CHUNG, ADAM SIMMONDS, JEFFREY PYUN, Department of Chemistry, University of Arizona, 1306 E University Blvd, Tucson, AZ 85721 — Utilization of the active cathode material in high-energy density Li-S batteries limited by the insulating nature of sulfur and losses in the form of insoluble polysulfides was improved by the use of 1,3-diisopropenylbenzene (DIB) copolymerized with molten sulfur. This approach termed, inverse vulcanization, transforms elemental sulfur into chemically stable processable copolymer forms with tunable thermomechanical properties. According to dielectric spectroscopy and dc conductivity measurements, composite sulfur-DIB copolymer cathodes exhibit a glassy-state beta relaxation related to short sulfur segments or to the DIB cross-linker. High-resolution AEM and FESEM studies down to the atomic scale reveal multiscale 3D-architectures created within the pristine and cycled composite cathodes with various contents of the electroactive copolymers. The morphology, structures, bonding and local compositional distributions of the constituents (sulfur, copolymers, aggregated conductive carbon nanoparticles) as well as extended pore structures and their transformations under cycling have been examined to provide insights into mechanisms of the enhanced capacity retention in the modified Li-S cells.

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