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Structured Battery Materials by Polymer Self-Assembly and Sol-Gel Chemistry

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Lithium batteries are currently one of the most promising energy storage devices. Improving and understanding these systems will help to advance several key technologies such as renewable energies and electric mobility. Lithium batteries consist of two electrodes that store lithium (ions) and an electrolyte that facilitates ion transport between these electrodes while forcing electrons to pass through an external circuit. Optimizing the performance of these batteries requires the choice of appropriate materials and a morphology that allows for efficient transport of both lithium ions and electrons. The synthesis of electrode materials on various length scales has therefore received tremendous attention.

Nevertheless, the fabrication of well-defined structures on both the micro- and nanoscale has remained a challenge. This impedes a more detailed study of the relationship between morphological parameters and electrochemical performance.

This work addresses this problem by combining the structure formation properties of polymer self-assembly with the sol-gel synthesis of inorganic compounds. The presented approach achieves defined (nano)structures that can be applied to various battery materials.

Using both block copolymer microphase separation and macrophase separation, mesoporous microspheres with adjustable pore sizes have been produced. This structure has been realized for titania and lithium titanate, two potential anode materials. Important electrochemical parameters, including cyclability and rate capability, have been correlated to the pore size.

A network of hollow lithium iron phosphate nanospheres was further synthesized by conserving the morphology of block copolymers in solution by a sol-gel process. The porosity of this cathode material was retained during crystallization, resulting in excellent electrochemical properties.

Besides electrode materials, these concepts were used to fabricate mesoporous silica interlayers. These interlayers were able to improve the cyclability of lithium-sulfur batteries.

This thesis thus discusses the importance of structural features for the performance of lithium batteries, providing insights for the development of the next generation of battery materials.

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