Towards High-Capacity Silicon Anodes for Lithium-Ion Batteries: Comparative Performance Study of Commercially Available Polymer Binders and Silicon Nanopowders <u>Torsten Brezesinski</u>,¹ Christoph Erk,^{1,2} Heino Sommer,^{1,2} Christoph Weidmann¹ and Juergen Janek^{1,3} ¹Institute of Nanotechnology, Battery and Electrochemistry Laboratory (BELLA), Karlsruhe Institute of Technology, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany ²BASF SE, 67056 Ludwigshafen, Germany ³Institute of Physical Chemistry, Justus-Liebig-University Giessen, Heinrich-Buff-Ring 58, 35392 Giessen, Germany

Silicon is a promising anode material for nextgeneration lithium-ion batteries because of its natural abundance and high theoretical specific capacity of either 4008 mAh/g for $\text{Li}_{21}\text{Si}_5$ or 3340 mAh/g for metastable $\text{Li}_{15}\text{Si}_4$.^[1] However, major issues arise from large volume changes (up to 300 %) during alloying with lithium. As a result, electrodes containing silicon typically suffer from severe mechanical stress and often show signs of capacity fading after few charge/discharge cycles. This is due in part to pulverization which results in both loss of electrical contact of the silicon particles and increase in electrical resistance.

In recent years, much effort has been spent on preparing nanostructured silicon materials and composites thereof with the goal of preserving the electrode integrity upon lithiation/delithiation.^[1-3] Nevertheless, the overall performance depends on a range of different "key" parameters, including binder, electrolyte, loading and so forth, and, thus, the definition of a so-called "standard" is virtually impossible.

Here, we present results from a comparative performance study of electrodes made from commercially available silicon nanopowders and polymer binders. The data establish that the binder itself has a profound effect on the electrochemical performance. Several binders have been tested and it turned out that electrodes made from poly (acrylic acid) exhibit the best cycling stability. Overall, the results indicate that polymer binders with a large amount of polar functional groups allow for facile electrode processing and, further, stabilize the active components to some extent. The latter helps improve the cycle life.

We also show that the use of silicon nanopowders with narrow size distribution and thin native oxide layer is beneficial to achieving high reversible capacities and low first cycle irreversibilities and to keeping the overpotential relative to lithium low. Lastly, we demonstrate that independent of polymer binder and silicon starting material, monofluoroethylene carbonatebased electrolyte systems have the potential to significantly improve the overall cell performance.

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