How Nanoporous Silicon-Polypyrrole Hybrids Flex Their Muscles in Aqueous Electrolytes: In Operando High-Resolution X-Ray Diffraction and Electron Tomography-Based Micromechanical Computer Simulations

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The datasets presented here contain different data on an in situ, combined electrochemical and x-ray diffraction investigation on the electrochemical actuation of a polypyrrole-filled porous silicon hybrid material. The pores of nanoporous silicon are filled by the electroactive polymer polypyrrole. An applied potential to the hybrid material immersed in a perchloric acid electrolyte solution leads to an incorporation of ions into the polymer and an expansion of the whole hybrid material. The samples used in this study are bi-layers. A polypyrrole-filled porous silicon layer with a thickness of approximately 25µm is attached to an underlying 500µm thick bulk silicon layer. Thus, the porous silicon layer cannot freely expand since it is clamped by the bulk silicon.

In the range of 0.2-0.6V the applied potential leads to a capacitive incorporation of the anions. They are stored in the polypyrrole and do not partake in any chemical or electrochemical reactions. Therefore, the ions can be reversibly extracted from the polypyrrole. The potential is applied in a step-wise fashion with single potential steps at 0.2V, 0.4V and 0.6V. The current and the charge are recorded during these potential steps. Once the current settles at a constant level, an x-ray measurement is performed.

In this study the x-ray measurements are conducted by θ-2θ diffraction measurements. Two different measurement geometries are used. In one, the out-of-plane type, the x-ray hits the front side of the porous silicon layer and is scattered off it. The second type is the in-plane geometry, in which the x-ray is hitting the back side of the sample and the scattered beam is measured in transmission coming through the front side. In both cases, the mono-crystalline porous as well as the bulk silicon layers produce a diffraction signal, which can be seen in the x-ray datasets. Through the difference of these two diffraction peaks it is possible to extract the strains occurring in the porous silicon layer.

The file ‘Experimental_data’ contains two folders ‘out of plane’ and ‘in plane’. In both a folder labelled ‘steps’ contains the electrochemical (‘Echem_’) and x-ray diffraction (‘Xray_’) files measured at different potential steps. The information is contained in the file names. In the ‘out of plane’ folder an additional ‘timeseries’ folder exists. These time series measurements are conducted to probe the dynamical actuation properties of the hybrid sample. Here, the potential is not applied step-wise but continuously – linearly or in a square-like fashion changing from 0.2V to 0.6V, see the sub-folders. For the square potential, the x-ray data consists of single 2D-detector pictures, which contain the diffraction peaks of the porous silicon and the bulk silicon layer. With the knowledge of the detector distance of 955.6mm and a pixel size of 172 x 172 μm^2 an evaluation is possible and a diffractogram can be obtained.

Moreover, a transmission electron microscopy tomography is performed on the polypyrrole-porous silicon hybrid material to obtain a three-dimensional model. This tomography reconstruction is included in the dataset. It can be transformed into an FEM model so that the actuation-induced strains and stresses in the complex porous silicon host structure can be studied and compared to the experimentally gained insights.

The hybrid material is installed in a scanning electron micrograph with a focused ion-beam so that an approximately 200nm thin needle can be prepared from the material. The needle is then installed in a transmission electron microscope (TEM), in which a series of transmission-micrographs are recorded. The TEM (FEI Talos F200X) is equipped with a high brightness Schottky-field emission gun (X-FEG) and acquires a tilt series of the needle. The microscope parameters are 200 kV acceleration voltage, 50 pA beam current and a camera length of 98 mm for the scanning transmission electron microscopy (STEM) high angle annular dark-field (HAADF) detector. The resolution of the images is 2048 × 2048 pixels with a pixel size of 384 pm. The tilt range was +80 to -80 degrees with an increment of 2 degrees resulting in 81 images. The images can then be reconstructed to form a three-dimensional model. This model is also included in the upload. Its voxel size is 0.36 x 0.36 x 0.36 nm^3.