1. Supplementary Information

Influence of Pillar Size

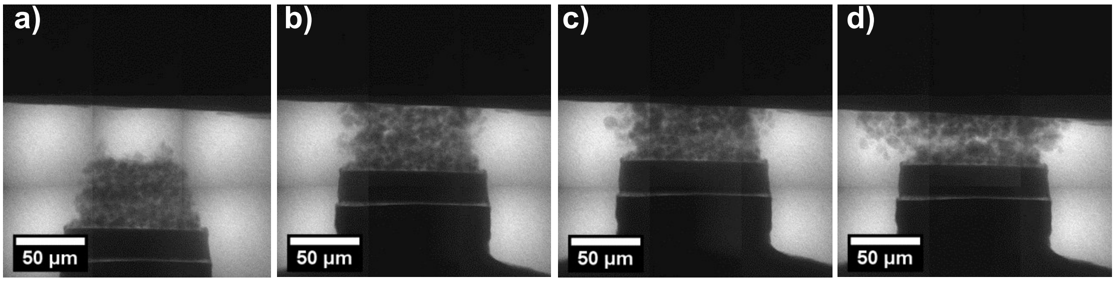


Figure S1 a-d) A pillar of ca. 65 µm diameter during uniaxial compression. The pillar does not maintain its structural integrity and as compression proceeds particles spill out laterally.

A smaller pillar, with a diameter of ca. 65 µm was compressed to study how the sample size affects the structural integrity of the sample during compression. As presented in the radiographs in Figure S1, the sample breaks apart after the first compression step. During the successive compression steps, the particles start spilling laterally until the sample is crushed. The loss of particles from the field-of-view renders this dataset unusable for further analysis, as the digital volume correlation (DVC) algorithm relies on correlating sub-volumes of pixels between datasets. However this analysis is useful to conclude that this technique is more suited for larger samples.

Digital Volume Correlation

The DVC algorithm is used to measure the strain within an electrode and operates by cross-correlating grey-level intensity vales of the datasets provided. The reference and deformed datasets are split into cubical or rectangular sub-volumes. The maximum cross-correlation between the squares of the grey-scale intensity values within each sub-volume for the reference and deformed datasets is then used to identify the 3D translation vector. The correlation coefficient (CC) is a measure of the similarity between the distinctive intensity patterns contained within different sub-volumes for the two datasets and is measured between the 0-1 range [1]. The sub-volume size is key in obtaining adequate correlation coefficients that can be related to the material and phenomena that is being observed: if the sub-volume window is too small, the measurement will be susceptible to noise, if the sub-volume window is too large, the resolution of the measurement will not be adequate [2]. In the context of the work presented in the main manuscript, smaller particle fragments bellow the sub-volume size generate lower correlation as their intensity patterns are more susceptible to smaller variations in their grayscale value due to noise or minor morphological changes.

Sub-volume Edge Length Determination

To determine the most suitable cuboid sub-volume edge length for the subset-based DVC approach, its influence on the correlation coefficient (CC) was studied as follows. Its influence on the result was gauged by increasing the edge length from 4 to 16 µm in 2 µm increments with the main aim of obtaining the best compromise between a high CC and fine sub-volume size to grant a correct identification of features. The variation in CC is presented in Figure S2.

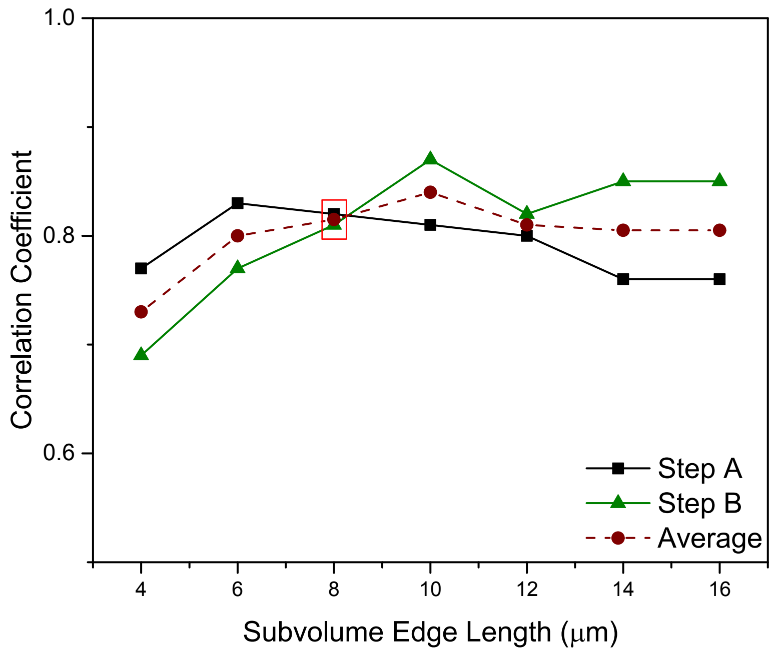
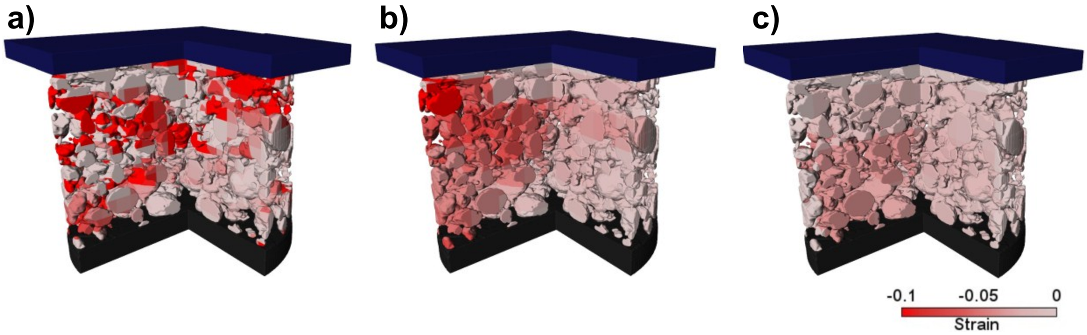


Figure S2: The CC for the DVC calculation for Step A, B along with their averaged value.

From the average CC, it is possible to note that the highest combined CC is obtained for a sub-volume edge length of 10 µm with a value of 0.84, however as this is too coarse, it leads to a low-resolution highly averaged strain map. An example of a coarse strain map can be viewed in Figure S3 c). This is not deemed an accurate representation because averaging the strain over large sub-volume sizes does not result in a map that is locally representative. As the sub-volume size decreases, the CC also decreases and the resulting strain map is more susceptible to the noise present in the image as viewable in Figure S3 a). A sub-volume edge length of 8 μm is hence selected as this offers the best compromise between resolution and high CC. This can be viewed in Figure S3 b).



4 μm **8 μm** **12 μm**

Figure S3: Strain maps obtained with sub-volume sizes of a) 4, b) 8 and c) 12 μm.

Load Profile

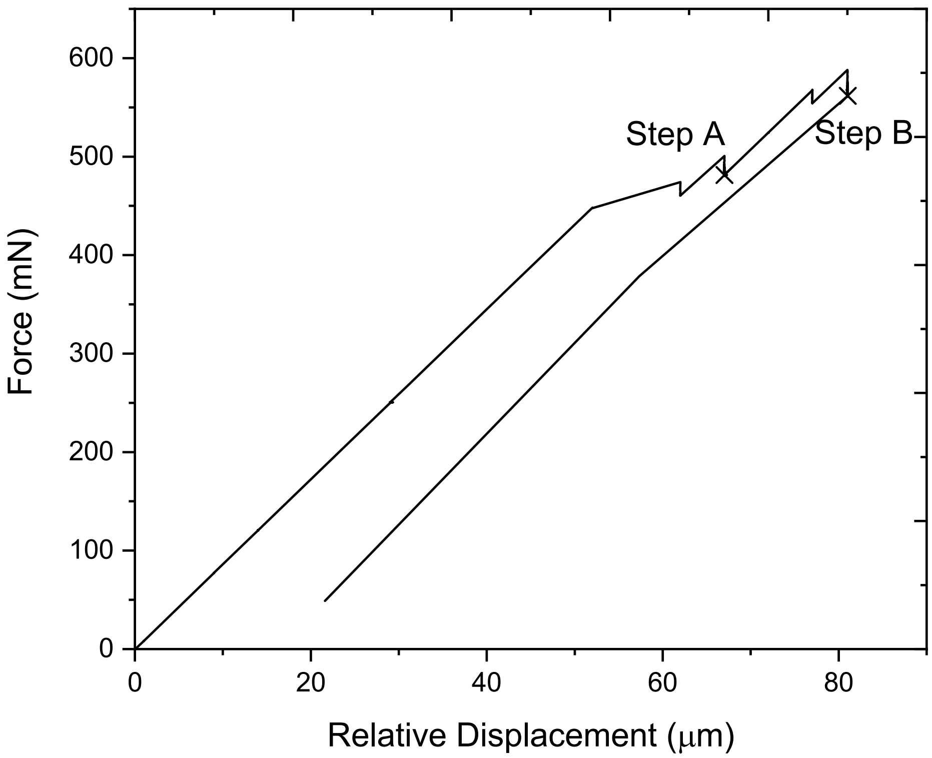


Figure S 4: Force vs relative stage displacement for the two compression steps.

The load profile obtained during compression is presented in Figure S4. The relative displacement indicates the displacement of the stage from the point of first contact between the compression tip and the sample. The two scans were taken at Step A (65 um, 500 mN) and Step B (80 um 560 mN). After the scan, the sample was left to rest leading to a characteristic relaxation that can be observed from Figure S4 with a decrease in force prior to the scan. Other relaxation steps occurring before and after the scans are due to short pauses taken to evaluate whether to continue compression or interrupt and scan. For a further understanding of the load response, compression needs to be repeated on a pillar composed solely of carbon and binder in order to decouple its behaviour from the electrode as a whole.

1 M. Peña Fernandez, A. H. Barber, G. W. Blunn and G. Tozzi, *J. Microsc.*, 2018, **00**, 1–16.

2 E. Dall'Ara, D. Barber and M. Viceconti, *J. Biomech.*, 2014, **47**, 2956—2963.