Electronic Supplementary Material (ESI) for Materials Horizons.

Striping Modulations and Strain Gradients within Individual Particles of a Cathode Material upon Lithiation

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Supporting information

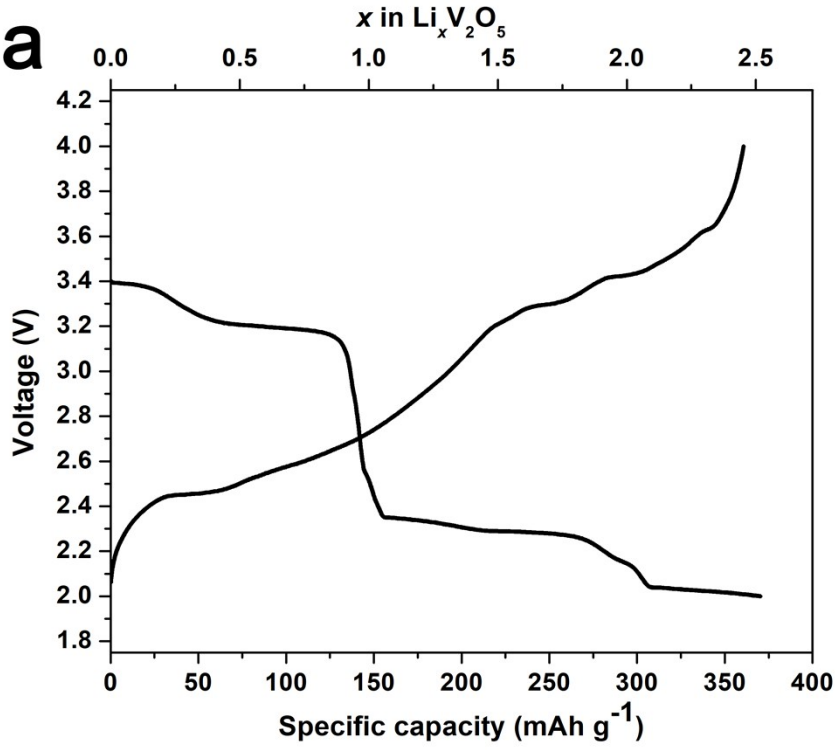

Figure S1. Galvanostatic charge–discharge curves measured for V_2O_5 in a coin-cell configuration at a constant 0.2 C rate. The plateaus correspond to distinctive intercalation-induced phase transitions in V_2O_5: α→ε at 3.4 V, ε→δ at 3.2 V, and δ→γ at 2.3 V.

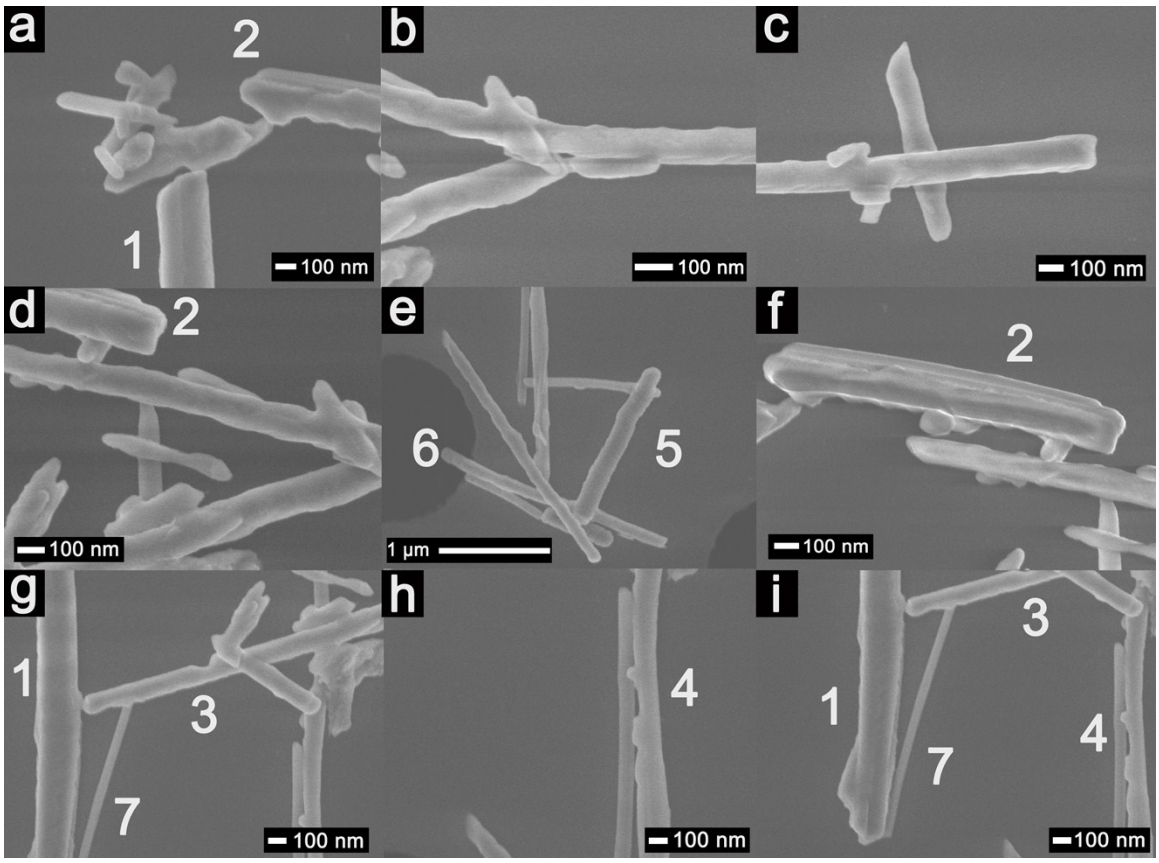

Figure S2. High-resolution SEM images of the cluster of wires imaged by STXM . High-resolution images of segments of the interlaced wires depicted in Figure 2a. The segments are numbered as in Figures 2 and 3.

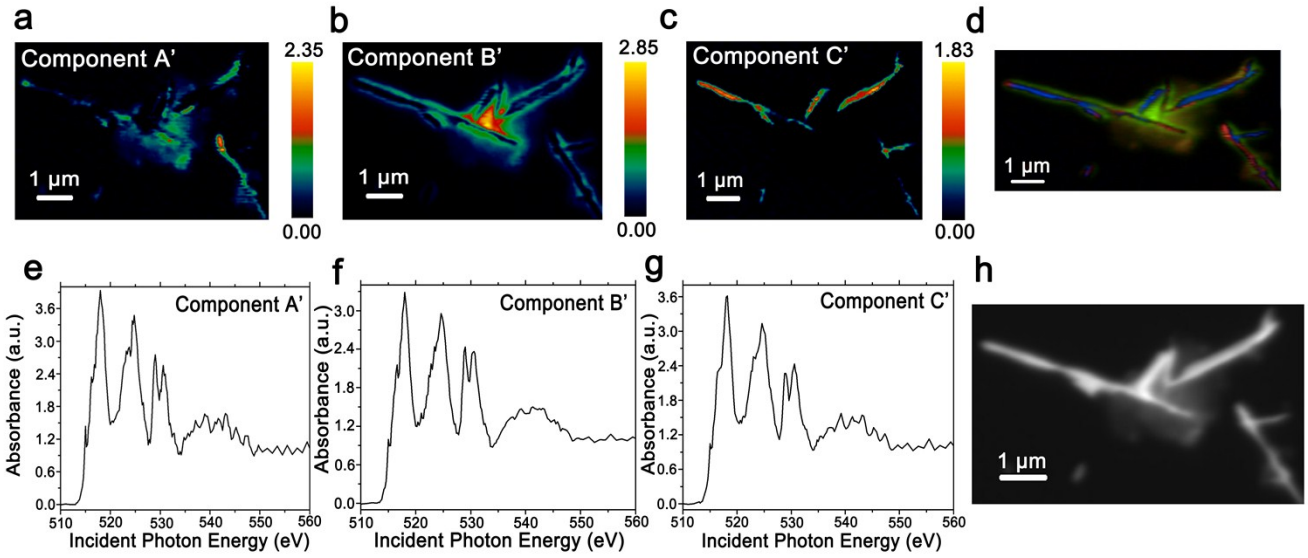

Figure S3. Intensity maps derived from region-of-interest singular value decomposition of electrochemically lithiated samples at a discharge of 2.75 V. a–c) Intensity maps depict the location of the spectral signatures, Components **A**'—**C**' plotted in (e)-(g). (d) An overlay of the three spectral components illustrating the compositional and phase heterogeneity across the interconnected network. Red, green, and blue regions demarcate majority contributions at the specific pixel from spectral components in (e), (f), and (g), respectively. h) Integrated STXM image of the cluster of wires.

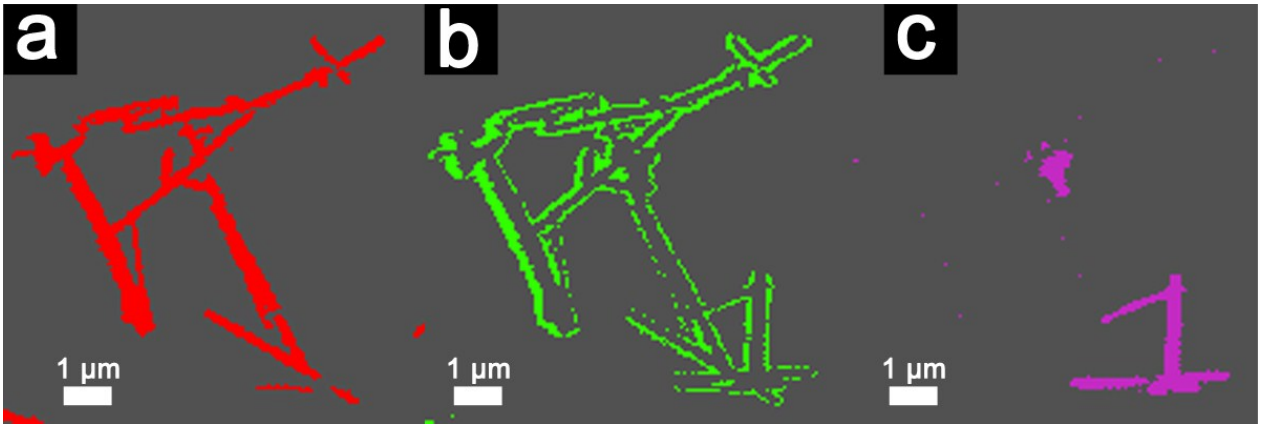

Figure S4. **Spatial Distribution of Components III—V as Deduced from PCA analysis of Hyperspectral STXM Data Acquired for an Ensemble of Lithiated V_2O_5 Nanowires.** (a)–(c) show the spatial location of the separated components **III—V** generated by PCA of the hyperspectral stack. The corresponding eigenspectra are plotted in Fig. 2e.

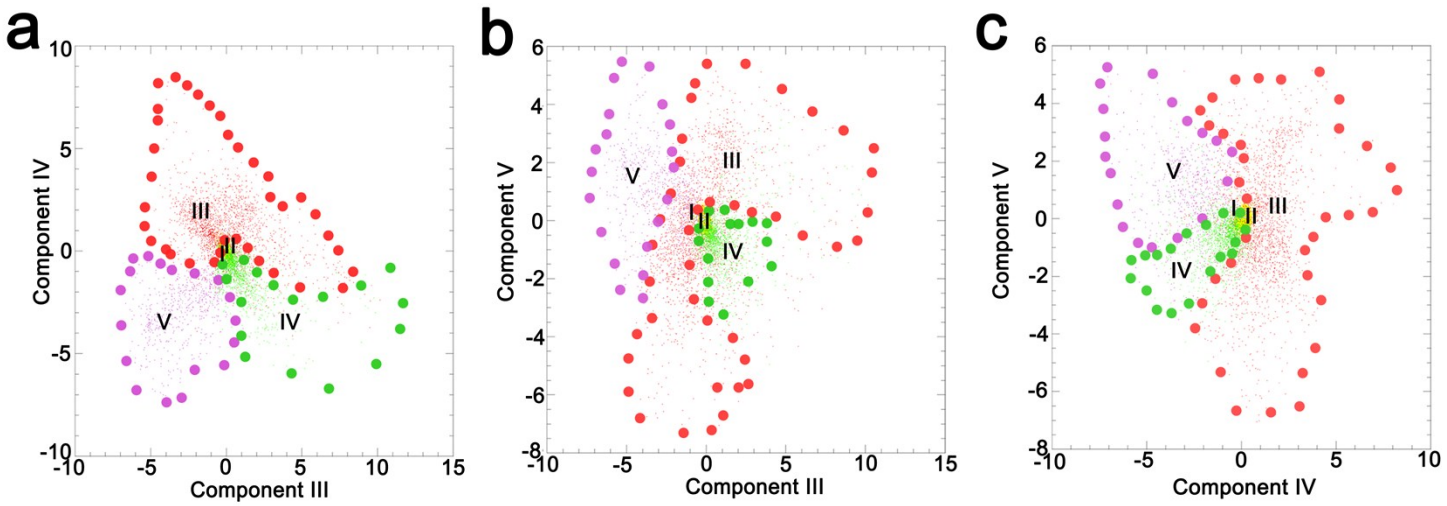

Figure S5. **Clustering of pixels in PCA analysis and spatial profiles of individual principal components.** (a)–(c) show the comparison between components in terms of separation of generated pixels by PCA of the hyperspectral stack. Blue and yellow represent background components, whereas **III**, **IV**, and **V** correspond to the components with eigenspectra plotted in Figure 2e. Dotted lines establishing boundaries are guides to the eye.

Components	Cluster weights 1	Cluster weights 2	Cluster weights 3	Cluster weights 4	Cluster weights 5
I	0.104 (31.84%)	-0.0704 (21.62%)	0.0398 (12.21%)	-0.0858 (26.34%)	-0.0260 (7.99%)
II	1.75 (47.31%)	-1.62 (43.82)	-0.125 (3.38%)	0.166 (4.49%)	0.0372 (1.00%)
III	9.17 (75.36%)	1.74 (14.33%)	-0.0822 (0.68%)	0.948 (7.79%)	-0.225 (1.85%)
IV	4.19 (54.27%)	-1.10 (14.26%)	0.794 (10.28%)	-1.10 (13.09%)	-0.627 (8.11%)
V	7.51 (49.88%)	1.08 (7.20%)	-2.86 (18.99%)	-2.76 (18.31%)	0.846 (5.62%)

Table S1. Summary of the cluster weights of each component deduced from the PCA analysis performed in Figure 2.

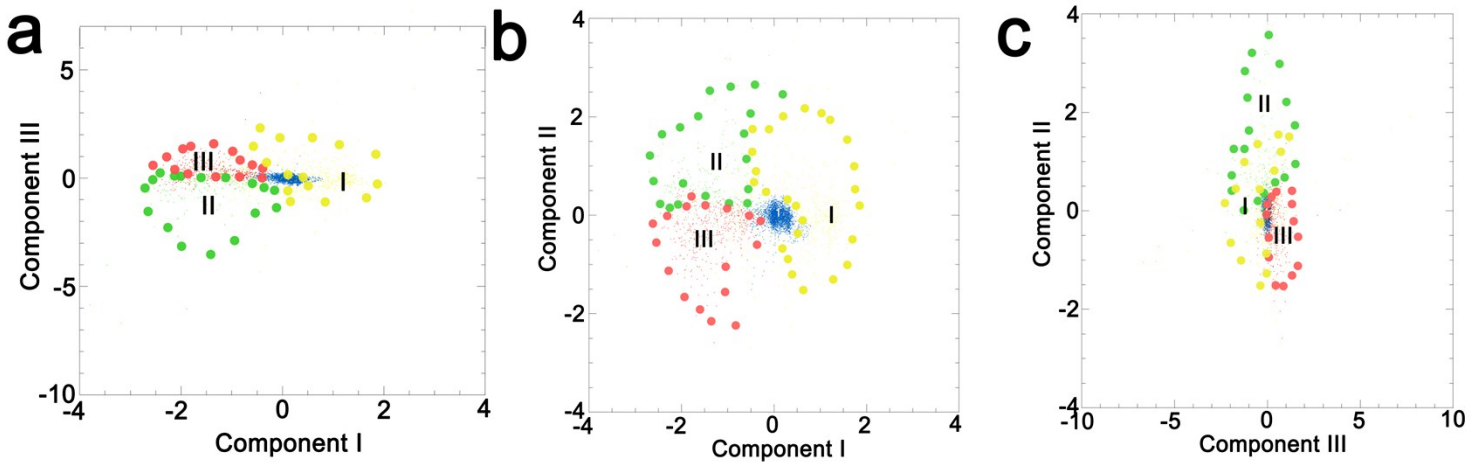

Figure S6. **PCA Analysis of STXM data acquired for an individual V_2O_5 nanowire after 1 min of chemical lithiation** (an average composition of $Li_{0.28\pm0.07}V_2O_5$ is deduced based on ICP-MS analysis)**.** ((a–c) Scatter plots depicting the scores at each pixel for each of the three components. The analysis allows for effective classification of each of the pixel to three components. Dotted lines serve as a guide to the eyes.

Components	Cluster weights 1	Cluster weights 2	Cluster weights 3	Cluster weights 4
1	0.00578 (2.92%)	0.121 (61.28%)	0.00463 (2.34%)	0.0661 (33.45%)
2	5.05 (91.89%)	0.421 (7.67%)	0.00355 (0.06%)	-0.0208 (0.38%)
3	1.18 (43.40%)	-1.14 (42.15%)	0.348 (12.86%)	0.0429 (1.58%)
4	1.33 (46.58%)	-1.14 (39.94%)	-0.377 (13.22%)	-0.00770 (0.27%)

Table S2. Summary of the cluster weights of each component deduced from the PCA analysis performed in **Figure 4**.

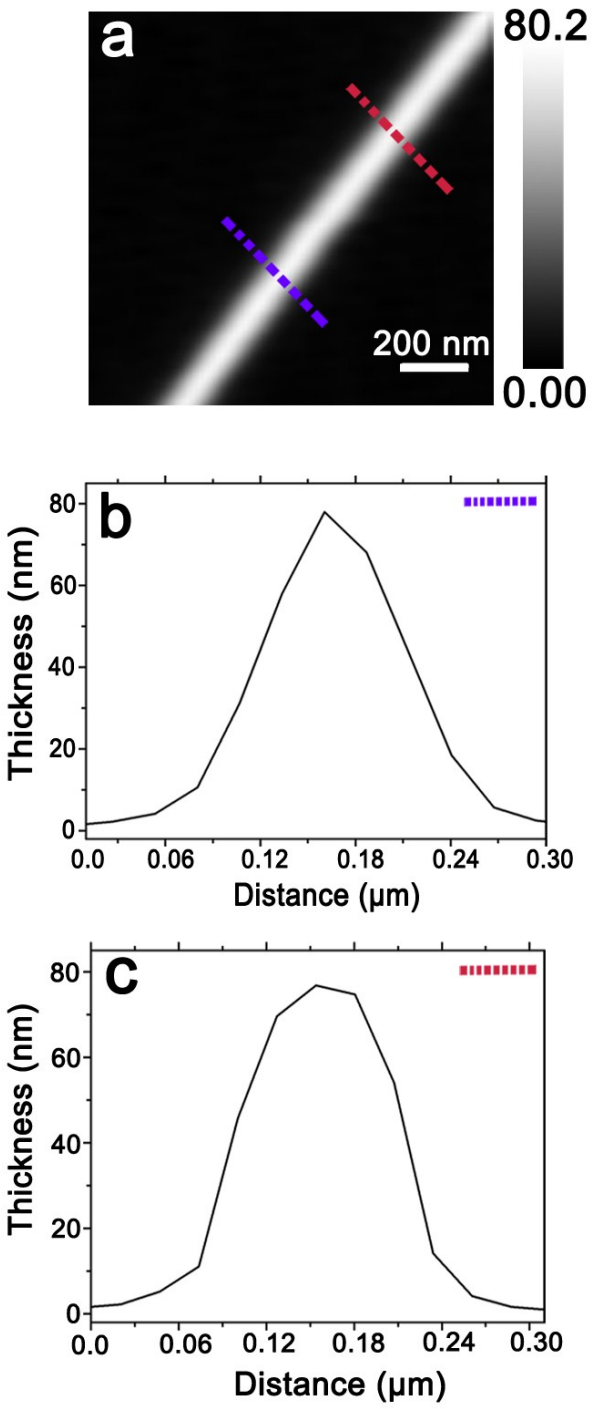

Figure S7. **Thickness map of an individual chemically lithiated V_2O_5 nanowire that is 245 nm in diameter.** (a) Thickness map of the chemically lithiated V_2O_5 nanowire depicted in **Figure 4,** where the gray scale bar depicts the thickness in nm. The colored dashed lines represent the cross-sections measured in (b) and (c).

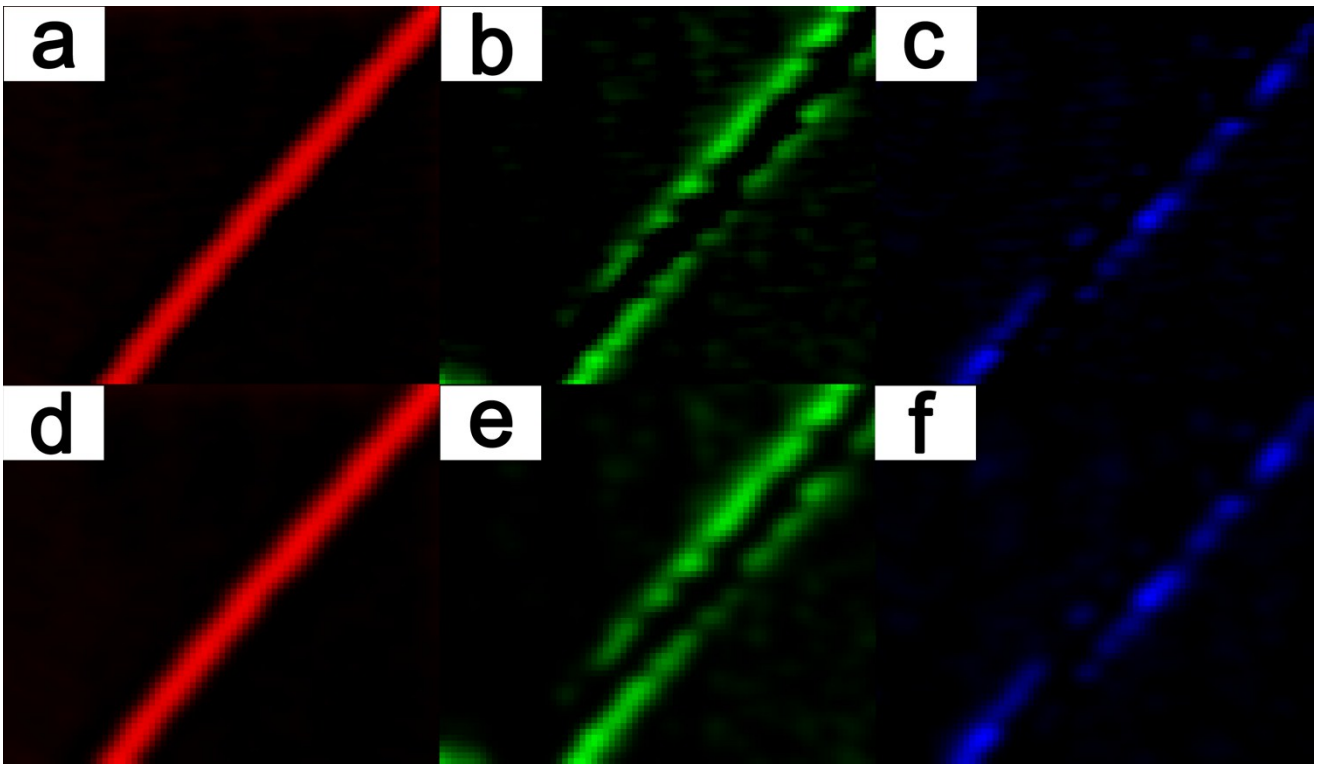

Figure S8. **Conversion of STXM scan data into a composition map.** The normalized intensities in (a)–(c) were subjected to Gaussian filtering in order to remove noise (d)–(f).

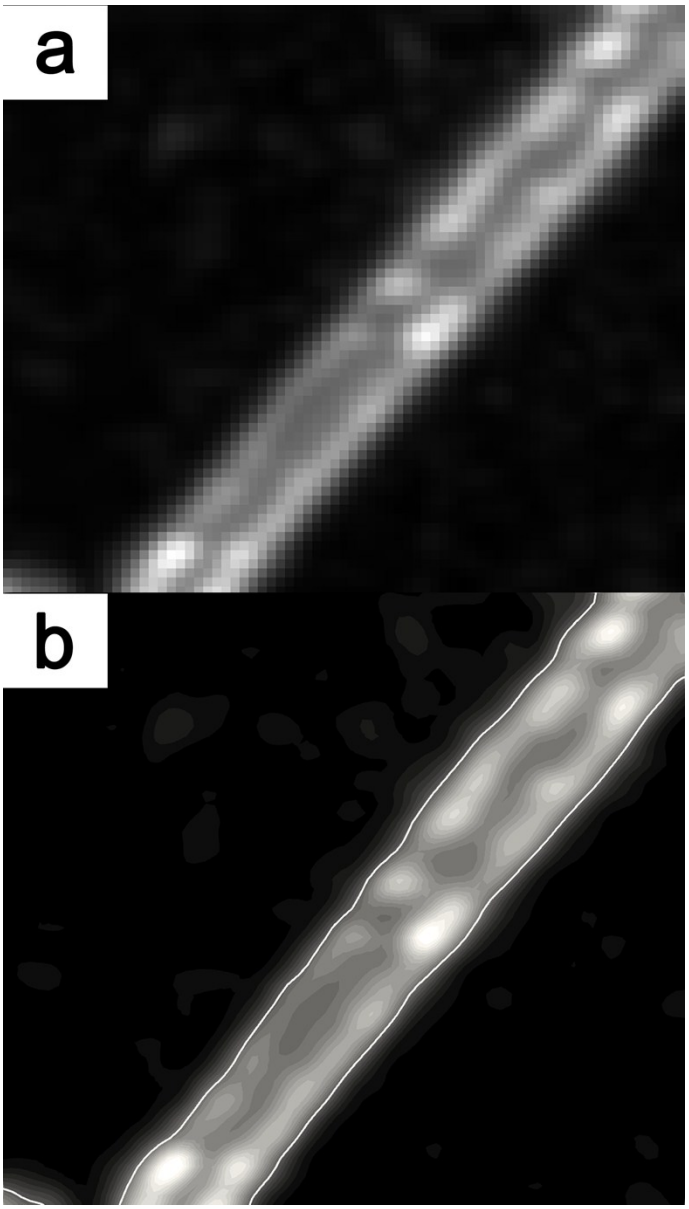

Figure S9. **Superposition of the filtered intensities.** Using the stoichiometry fractions x_i as weighting factors, yields the distribution of x over the pixel space (a). Bilinear interpolation of the pixel data yields a smooth composition map in pixel space, from which the outlines of the nanowire geometry have been extracted using a Marching Cubes algorithm (b).