

Three-Dimensional Visualization of Conductive Domains in Battery Electrodes with Contrast-Enhancing Nanoparticles

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Abstract

Replacing conductive carbon black with commercial carbon-coated iron nanoparticles yields an effective contrast-enhancing agent to differentiate between active material, conductive additive, and binder in lithium-ion battery electrodes. Nano XCT resolved the carbon-binder-domain with 126 nm voxel resolution, showing partial coatings around the active material particles and inter-particle bridges. In a complementary analysis, SEM/EDS determined individual distributions of conductive additives and binder. Surprisingly, the contrast-enhancing agents showed that the effect of preparation parameters on the heterogeneity of conductive additives was weaker than on the binder. Incorporation of such contrast-enhancing additives can improve understanding of processing-structure-function relationships in a multitude of devices for energy conversion and storage.

The ever-increasing demands of power and energy for automotive and consumer product applications require optimal design of battery electrodes. Of major concern in battery design is the carbon-binder-domain (CBD) of conductive carbon additives and polymer binder. Although the CBD makes up only a small fraction of electrode mass and volume (typically 3-6 wt% in commercial formulations, but much higher in academic settings), its microstructure has a critical influence on battery performance. Insufficient connectivity of the CBD leads to poor electron transport and insufficient mechanical strength, while excessive CBD adds dead weight and volume at the cell level and may even slow ion transport. Continuum-scale battery performance models do not explicitly consider the CBD, and instead predict discharge curves based on effective parameters such as long-range electron conductivity (solid phase), long-range ion tortuosity (liquid phase), and short-range contact resistances (solid-phase). The fundamental relationships between CBD microstructure and these transport parameters are currently very poorly understood, as evidenced by the trial-and-error approach to electrode processing optimization.

A major cause for poor understanding of the relationships between processing, microstructure, and performance is the inability to adequately visualize the location and distribution of carbon and binder inside battery electrodes. Scanning electron microscopy coupled with energy dispersive x-ray spectroscopy (SEM/EDS) is currently the most reliable method for visualizing carbon in electrodes¹⁻⁶. Infiltrating electrode voids with a silicon-based resin provides the needed contrast between porosity and CBD regions¹. Combining SEM with focused ion beam (FIB) milling can obtain 3D reconstructions, but the reconstructions are valid only if the resin does not expand or contract upon hardening, which would alter the microstructure in unknown ways. Furthermore, the cost and time of FIB/SEM is considerable,

and the field of view is generally very small. Accordingly, many studies have been devoted to higher-throughput methods of electrode imaging based on x-ray computed tomography (XCT)⁷⁻¹². High-resolution nano-XCT can image electrodes with resolution as small as 50 nm, while lab-scale tomography can commonly resolve images of entire electrodes with resolution of 100-600 nm. However, XCT is fundamentally limited in distinguishing between active material and CBD. If high-energy x-rays are chosen for optimal imaging of a transition metal oxide, the CBD and porosity (voids) appear equally transparent. If low-energy x-rays are chosen for a carbonaceous active material, then low x-ray penetration levels prohibit the study of realistically-sized specimens. Continuing efforts to avoid this problem combine XCT with FIB/SEM and image-matching¹⁰, statistical pore-filling models of the CBD¹¹, or separate tomograms in Zernike phase-contrast mode¹².

In addition to the described limitations, neither XCT nor SEM/EDS is able to separately resolve carbonaceous active materials from conductive carbon. In this work, we present an innovative solution to these problems by replacing conductive carbon with C-coated Fe nanoparticles (Fe-C). Fe absorbs x-rays hundreds of times more strongly than C or F, thereby enhancing the contrast of the CBD with XCT. In SEM/EDS, the Fe provides a strong signal that differentiates conductive carbon from active materials to determine the homogeneity of the CBD.

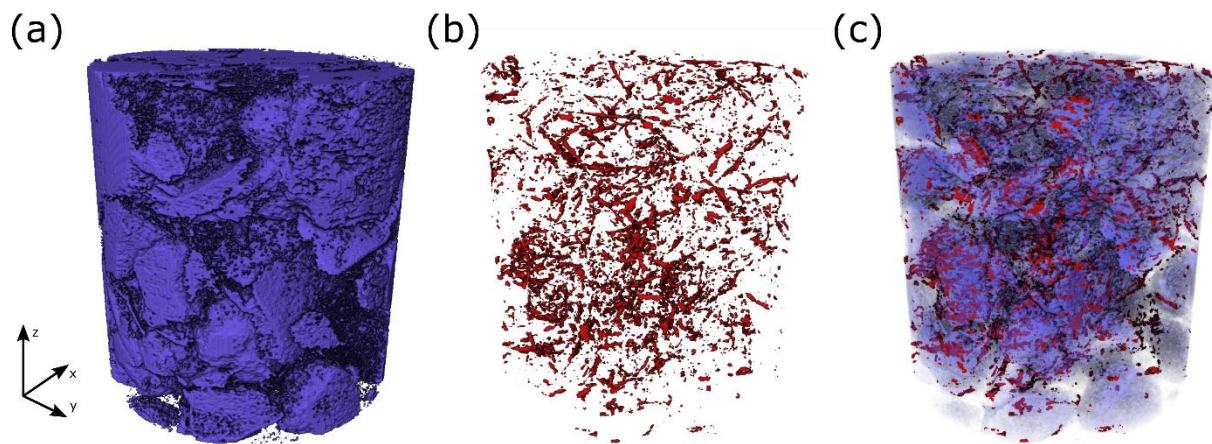
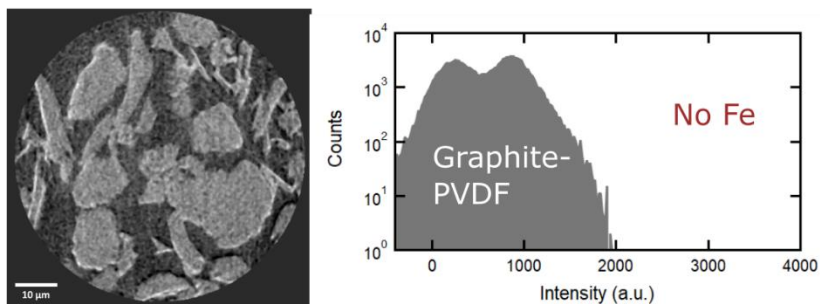


Figure 1: 3D renderings of (a) graphite phase (b) Fe-labeled CBD phase and (c) both phases with graphite transparency of 1%. The voxel size is 126 nm and the imaged cylinder is 65 μm in diameter.

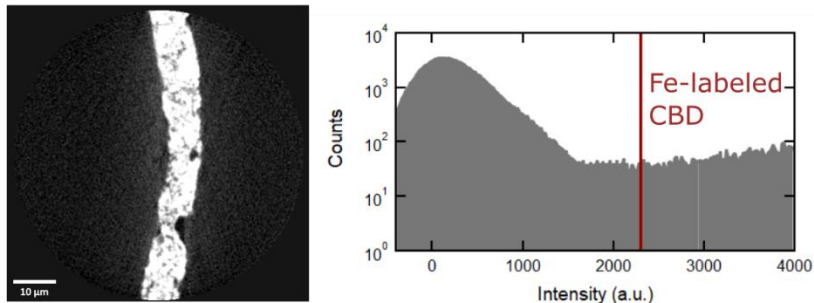
As shown in Figure 1, commercial Fe-C nanoparticles were demonstrated as contrast-enhancing agents for XCT by preparing and imaging thin electrodes of Fe-PVDF, graphite-PVDF, and graphite-Fe-PVDF according to literature methods¹³⁻¹⁶. With a diameter of 25 nm, the particle size is very similar to conventional conductive carbon nanoparticles. The carbon coating around the Fe additionally provides a surface chemically similar to conventional conductive carbon. Figure S1 demonstrates the ability of the Fe-C nanoparticles to perform in a functional graphite electrode. All three electrodes were imaged using identical exposure parameters and reconstructed using consistent bounds, which allowed a direct comparison of CT numbers between datasets. Virtual slices of the 3D XCT volume for each of the three electrode samples are accompanied by histograms of voxel intensity in Figure 2. Each dataset was reconstructed using the full 32-bit computational dynamic range available in the reconstruction software, in order to avoid CT number (or radiodensity) scaling issues imposed by

downsampling to lower bit depths. As such, all reconstruction values were accepted, including the negative values resulting from small amounts of noise in the x-ray radiographs.

(a) Graphite-PVDF



(b) Fe-PVDF



(c) Graphite-Fe-PVDF

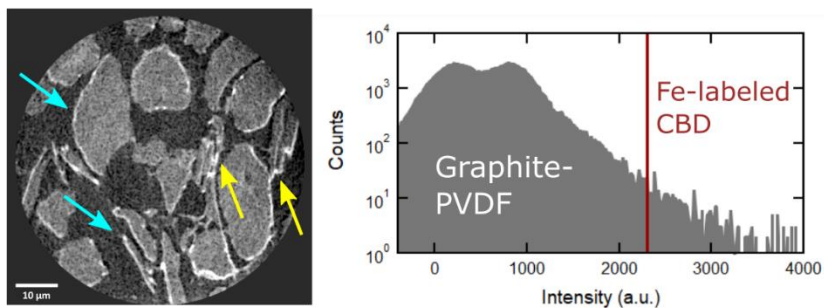


Figure 2: Virtual slices of 3D XCT volume with accompanying histograms for (a) graphite-PVDF (b) Fe-PVDF (c) graphite-Fe-PVDF. Red line indicates the assigned CBD threshold of 2300 (for details, see Supporting Information). Blue and yellow arrows in (c) indicate CBD coatings and bridges between particles, respectively.

Comparison of Figure 2a and 2b shows that, as expected, the graphite-PVDF (a) is much less bright than Fe-PVDF (b). A visual binarization of the Fe-PVDF dataset provided a clear means of determining the cut-off threshold value between Fe and PVDF of ~ 2300 , as shown in Figure 2b. This cut-off was then verified in the graphite-PVDF case (Figure 2a), which registered a negligible number of CT values above 2300 and, thus, confirmed that 2300 was an appropriate Fe threshold. A similar procedure was performed for the graphite-PVDF case in order to set a threshold value for graphite vs. PVDF. The resulting final threshold values for PVDF, graphite, and Fe were applied to the graphite-Fe-PVDF case (Figure 2c). These images therefore indicate that the bright spots in (c) surrounding the larger graphite particles represent Fe-C nanoparticles coating the active material surface. In contrast, some brightness can be observed on the edges of the graphite particles in (a), but the quantitative greyscale values are far below the Fe thresholds indicated by the Fe-PVDF (b) or graphite-Fe-PVDF (c) intensities.

The virtual slices of the graphite-Fe-PVDF electrode were rendered into 3-D images as shown in Figure 1. The 3-D rendering of the graphite-PVDF film is shown in Figure S1. Figure 1 (a) illustrates the microstructure of the graphite phase, (b) illustrates the microstructure of Fe-labeled CBD, and (c) overlays the graphite and CBD phases. The calculated volume percent of Fe-C is 1.2%, while the volume percent of Fe-C nanoparticles based on the electrode composition is 0.7%. However, the expected volume percent of CBD is 3.9%, assuming homogeneous distribution of Fe-C within the CBD. The missing CBD is attributed to the resolution of the XCT. With a voxel size of 126 nm, and a particle diameter of 25 nm, voxels will be detected as pure Fe only if particles are densely packed in clusters of more than 100 particles. Fe-C particles that have been diluted by binder or void will absorb with lower intensity. At sufficiently high dilution, the Fe-PVDF phase will be detected indistinguishably from

graphite. Higher resolution imaging would therefore increase the fraction of Fe registered and yield unambiguous structures of the CBD, but at the cost of a smaller field of view and less statistical power.

From Figures 1 and 2, contrast-enhanced XCT clearly affords the specific spatial variations of active material, conductive additive, and void space. From this information, the interconnectivity of conductive additive, void, and graphite can be calculated to provide critical inputs for detailed electrochemical models that predict effective ion and electron transport properties¹⁷ as well as mechanical and thermal behavior¹⁸. For example, finding the fraction of voxels that share common vertices yields the lower-bound percolation ratios, or connectivities, of graphite, void, and conductive additive. Performing this calculation on a selected subvolume (Figure S3) yields a percolation ratio of 100% for graphite, 85% for the CBD, and 99% for voids. The complete connectivity of graphite is predictable, as particles cannot be standing in free space, while the 99% connectivity between voids (which are filled with electrolyte during battery operation) suggests the absence of isolated pockets, which would be unable to transport ions.

The methods presented here can be used to relate processing conditions to electrode microstructure, which in turn influences performance. For example, some recent efforts to model charge transport and mechanical deformation in battery electrodes have considered the effects of CBD present as conformal coatings around active material particles versus ‘binder bridges’ between particles¹⁹. Figures 1 and 3 show the clear presence of such bridges between particles (indicated by yellow arrows), and the subvolume in Figure S3 shows that they are generally connected. For the entire sample volume imaged, the computed surface area of graphite is 0.19 mm², while the surface area of CBD is 0.025 mm². The surface area of contacts between graphite and CBD is 0.012 mm², showing that CBD bridges between particles cover approximately 6% of

the graphite surface. The fraction of surface area covered by CBD bridges will certainly affect particle-particle resistances, which are critical to electrode performance for a variety of systems^{14,20–22}.

While bridges between particles are clearly visible from Figures 1 and S3, Figure 2c also appears to show conformal CBD coatings around some of the graphite particles (blue arrows). However, for 20 μm graphite particles, conformal coating of 3.9 vol% CBD around each particle would yield a thickness of approximately 140 nm. With a voxel size of 126 nm, 140 nm features are smaller than the spatial resolution of the XCT images, as convention suggests that a minimum of two voxels are needed in order to clearly resolve a feature²³. Considering that much of the CBD is already present as large clusters in the bridges, the actual volume fraction of CBD present as a conformal coating is even less than 3.9 vol%. Thus, thin coatings cannot be visualized without higher resolution XCT.

A major advantage of the contrast-enhancing nanoparticles is the ability to determine the influences of electrode heterogeneity²⁴ on the distribution of CBD. Table 1 shows that the CBD volume is slightly higher in the subvolume of Figure S3 than in the overall volume, demonstrating mesoscale heterogeneities. The mesoscale heterogeneity is confirmed by completing a similar analysis on varying sub-volumes in the electrode, the results of which are shown in Table 1. Because the CBD is the most dilute component, variation in volume fraction leads to the greatest variation in connectivity.

Table 1: Volume fractions of graphite, CBD, and void on entire sample volume as well as selected sub-volumes with z-direction connectivity.

	Entire image	Sub-volume A	Sub-volume B	Sub-volume C
Volume (voxels)		60 x 90 x 35	60 x 90 x 35	160 x 160 x 505
Volume fractions				
Vol % graphite	44.9%	63.6%	48.9%	54.7%
Vol % CBD	1.7 %	1.7%	2.9%	0.6%
Vol % void	53.7%	34.7%	48.2%	44.7%
Connectivity				
Graphite		~100%	99%	~100%
CBD		86%	85%	0%
Void		99%	94%	99%

While the results of Table 1 demonstrate the local heterogeneities of CBD distribution, Figure 1b shows that the electrode preparation technique yields Fe-C that is distributed uniformly throughout the macroscopic structure, as evidenced by the homogeneous distribution of bright voxels (>2300) throughout the specimen volume. A homogeneous distribution of Fe-C within the CBD is also supported by the EDS of Fig. S4. Thus, contrast-enhanced XCT can be used to determine both microscopic and macroscopic distributions of CBD and active material, if it is demonstrated that the nanoparticles are uniformly distributed in the CBD.

If electrode processing parameters are not chosen correctly, the CBD itself can be inhomogeneous. The contrast-enhancing nanoparticles can also be used to determine this homogeneity, by separately determining binder and conductive additive distributions through SEM/EDS. Thick electrodes (100 μm dry) were chosen to amplify gradients formed during coating and drying²⁵. Images are shown in Figure 3. In both images, the SEM shows graphite

particles with particle sizes ranging from 20 to 50 μm , consistent with manufacturer specifications. Both the Fe and F maps show the presence of CBD. For the homogeneous electrode, Fe and F show considerable overlap, while the heterogeneous electrode shows enhanced F content close to the electrode surface. Such binder migration to the electrode surface has been well-documented and may be caused when concentration gradients formed by capillary forces during drying have insufficient time to relax²⁶⁻²⁹. The Fe gradient appears less obviously than the strong F gradient, indicating that *homogeneity of the contrast-enhancing agent does not ensure homogeneity of the CBD*.

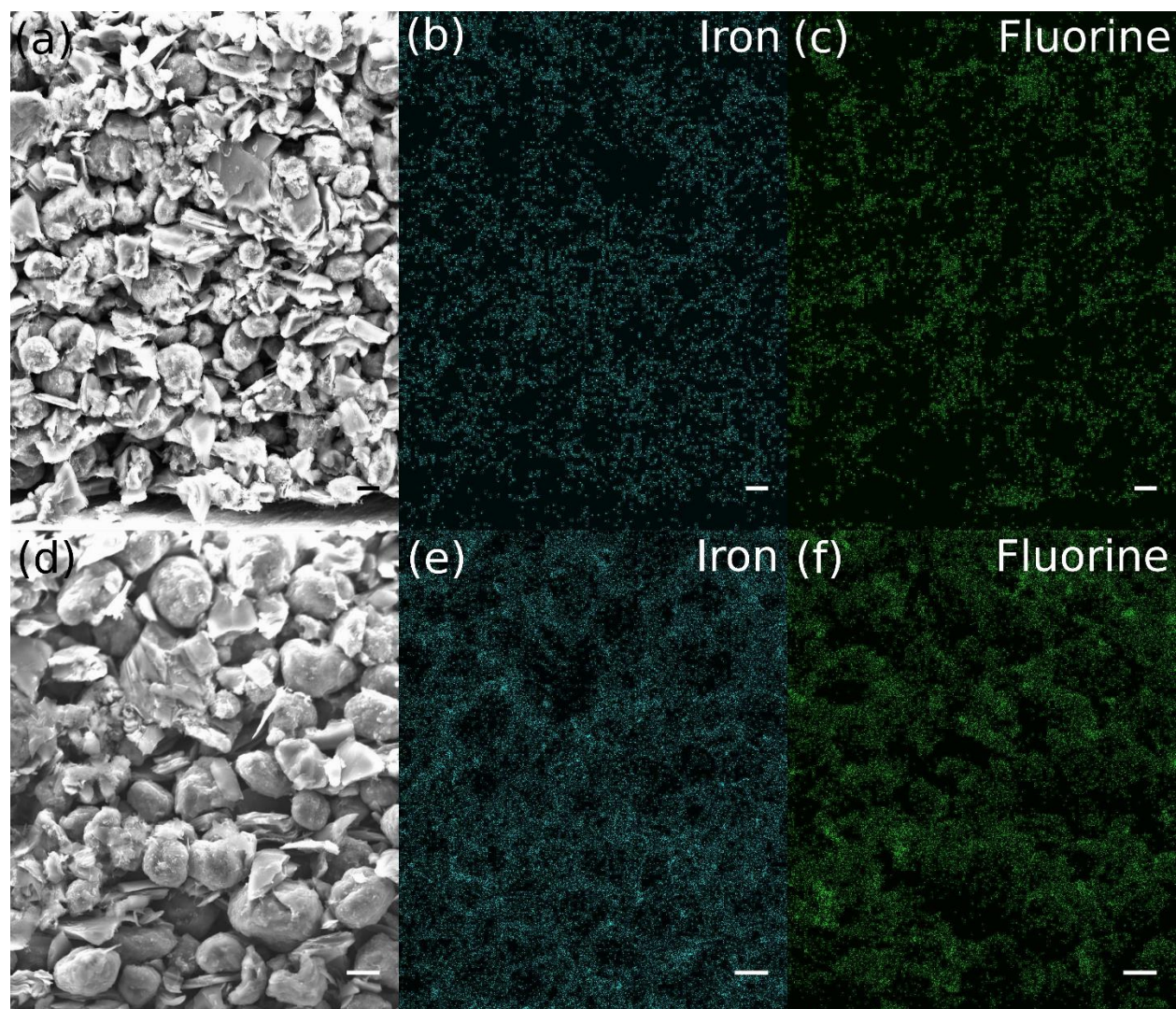


Figure 3: SEM and EDS maps for homogeneously (a-c) and heterogeneously (d-f) prepared graphite electrodes with Fe contrast-enhancing agent. For each image, the current collector appears at the bottom, and the scale bar corresponds to 10 microns. (a, d) SEM images (b, e) Fe EDS map in cyan (c, f) F EDS map in green. For the homogeneously prepared electrode, the Fe and F maps are almost indistinguishable and show a well-dispersed CBD. For the heterogeneously prepared electrode, Fe and F maps show different regions of deficiency, with stronger F enrichment at the electrode surface.

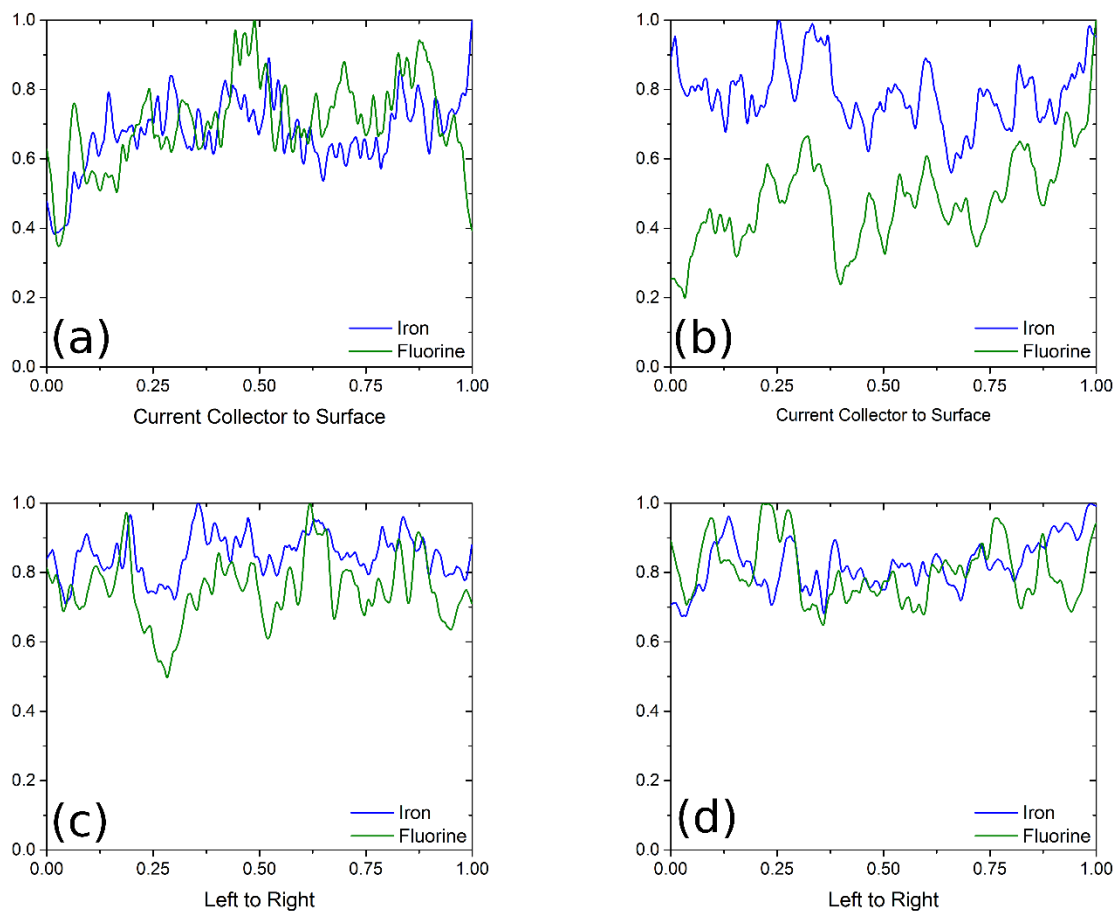


Figure 4: Normalized integrated intensity vs position for a) homogeneously prepared, bottom-top b) heterogeneously prepared, bottom-top c) homogeneously prepared, left-right d) heterogeneously prepared, left-right. Intensities were normalized by the maximum line-integral value for each elemental map.

The heterogeneity of the electrodes in Figure 3 can be analyzed via line-integrals of EDS intensity, as shown in Figure 4. Integration of the EDS signal from left to right as shown in Figure 4a shows that for the homogeneous electrode, neither the F nor the Fe show an obvious trend from current collector to surface. Figure 4b shows that for the heterogeneous electrode, the surface F concentration is approximately four times greater than that at the current collector, but

the Fe concentration is approximately the same at the current collector and the electrode surface. Integrating the SEMs from top to bottom as shown in Figure 4c and d shows that the oscillations in Fe and F roughly track each other both electrodes, likely because of variation in local porosity. The combined F/Fe integral scans show that, depending on how the electrode is processed, the CBD may be either homogeneous or heterogeneous on a macroscopic length scale, and that conductive additive and binder are independently influenced by processing conditions.

In summary, we have developed a simple, low-cost method to incorporate Fe nanoparticles into battery electrodes as contrast-enhancing agents and have demonstrated their ability to improve visualization of the CBD in both XCT and SEM/EDS. XCT rapidly yields detailed 3-D microstructures of the phases, while SEM/EDS provides higher spatial resolution and chemical specificity with widely available instrumentation. Neither method requires synchrotron access, and the two techniques are in fact strongly complementary if SEM/EDS is used to verify homogeneous mixing of the CBD before imaging samples in XCT. These techniques are not limited to Li-ion chemistries; carbonaceous or other low-Z materials are also relevant to beyond-Li systems like Na-ion, Li- and Na-S, pseudocapacitors, and multivalent intercalation chemistries. Even in fuel cells, researchers must consider how the mesoscale structure of Nafion binder and carbon nanoparticles affect oxygen transport in the catalyst layer. This work presents a simple method to determine the microstructure of active material, conductive additive, and binder that is easily generalizable to a great many of these systems. Efforts to quantify specific microstructures and relate them to electrode lifetime and performance are ongoing.

ASSOCIATED CONTENT

Supporting Information. Materials and methods, electrode formulations, battery discharge profiles, additional XCT and SEM/EDS images.

ACKNOWLEDGMENT

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TOC Figure

