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Learning From Scanning Transmission Electron Microscopy to Enhance Transmission X-ray Microscopy: How We Can Merge STEM and TXM Datasets?

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Lithium-ion batteries are the important part of various electronic devices, such as mobile phones, laptops and electric vehicles. The chemical and physical processes occurring between electrodes and electrolyte are the fundamental problems to understand the battery performance. Studying the three-dimensional (3D) structures of cathode particle agglomerates in the batteries is one of these concerns, which is essential in designing new and improved battery material [1].

The imaging modalities, such as transmission X-ray microscopy (TXM) and scanning transmission electron microscopy (STEM) have been individually used to quantify the agglomerate structure. Each technique has unique advantages and limitations. TXM is capable of capturing the structural details of larger volume of particles but yields relatively poor resolution and signal-to-noise-ratio. STEM offers superior spatial resolution but only for a thin section of the sample. A correlative analysis combining these two techniques will promise improved image quality for structural determination of the whole agglomerate.

In this paper, we present a dictionary learning approach that enhances TXM images from high-resolution STEM images. We first perform TXM to obtain tomographic dataset for a cathode particle agglomerate of a Lithium-ion battery. Then we use focused-ion beam to slice a thin section from it and obtain another tomographic dataset using STEM. With patches that extracted from the STEM images, we train a dictionary containing the structure information of the agglomerate. We use the learned dictionary for enhancing the TXM reconstruction images at any slice without a registration process.

The dictionary learning is one of the most active fields for image processing in recent years. The basic idea is that signals can be represented as a linear combination of few columns (called atoms) from a redundant dictionary, $\boldsymbol{D}$. In the image processing, we extract patches from an image and consider they are a set of signals $\boldsymbol{X} = [x_1, x_2, , , , x_n]$ in $\mathbb{R}^{m\times n}$ and solving the following optimization function [2-4]:

$$\min_{D\in C, \alpha\in\mathbb{R}^{k\times n}} \frac{1}{n}\sum_{i=1}^{n}\left(\frac{1}{2}\|x_i\text{-}\boldsymbol{D}\alpha_i\|_2^2 + \lambda\|\alpha_i\|_1\right) \tag{1}$$

where α_i is a sparse vector that contains a small number of significant coefficients, while the rest of the coefficients are close or equal to zero; λ is a regularization parameter; *n* is the number of sample, here it is the number of patches; *m* is the signal dimension; *k* is the number of atoms. The patch is typically considered as a square matrix with size p ($p\times p$, and $m = p^2$) and it should contain the smallest structure we expect to evaluate from the objective image. For the typical image process, *n* is suggested to be a large number ($n \geq 100{,}000$), and also $k \ll n$.

With the basic principle above, we apply the following process for reconstructed TXM images:

1. Extract patches $\boldsymbol{X}$ from the reconstructed STEM images and learn dictionary $\boldsymbol{D}$ from these patches, which can be denoted as $\boldsymbol{D\alpha}_{stem} \approx \boldsymbol{X}$;
2. Extract patches $\boldsymbol{Y}$ from the TXM image and obtain $\boldsymbol{\alpha}_{txm}$;
3. Compute new patches as $\boldsymbol{Y'} = \boldsymbol{D\alpha}_{txm}$ and reconstruct image from $\boldsymbol{Y}'$.

The steps of computing $\boldsymbol{D}$ and $\boldsymbol{\alpha}$ through Eq. (1) involve the process of solving a multi-variable optimization problem [5]. We used the least angle regression method to solve for the model variables.

We obtained tomographic images of a cathode particle agglomerate ($Li_{1.2}Co_{0.1}Ni_{0.15}Mn_{0.55}O_2$) of a Lithium-ion battery with both the TXM and STEM. The whole particle size used for the TXM is about $6.5\times6.5\times5.5\mu m^3$. The sliced sample for the STEM is about $6.5\times0.4\times5.5\mu m^3$. Figure 1 shows samples of tomographic reconstructed results. The STEM has spatial resolution of 15 nm and is superior compared to 60 nm resolution that TXM offers. Also STEM yields a much higher signal-to-noise ration for the sliced region as compared to TXM. We use STEM image of Fig. 1 to train the dictionary and use other TXM images to learn the dictionary. One of the enhanced images is shown as Fig. 2. The result of dictionary learning shows less noise. The structure becomes more distinguishable than the original image. The dictionary learning brings chance to enhance the structure information of the TXM results from the STEM data.

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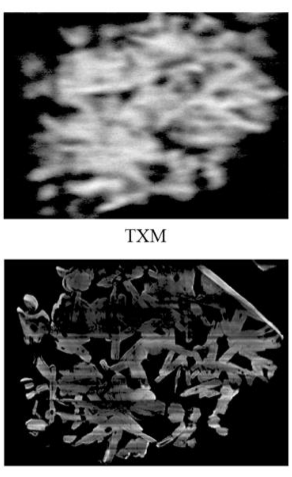

Figure 1. *Tomographic reconstruction results of TXM and STEM for the same slice*

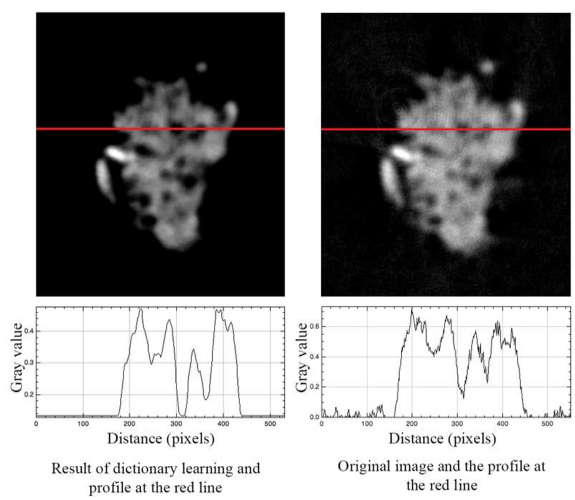

Figure 2. *Comparison of the result from dictionary learning and the original image of TXM.*

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