doi:10.1017/S1431927621001513
Microsc. Microanal. 27 (Suppl 1), 2021
© Microscopy Society of America 2021

Developing Deep Neural Network-based Denoising Techniques for Time-Resolved In Situ TEM of Catalyst Nanoparticles

Joshua Vincent[1], Sreyas Mohan[2], Ramon Manzorro[1], Binh Tang[3], Dev Sheth[4], Mitesh Khapra[4], David Matteson[3], Eero Simoncelli[2], Carlos Fernandez-Granda[2] and Peter Crozier[5]

[1]Arizona State University, United States, [2]New York University, United States, [3]Cornell University, United States, [4]IIT Madras, United States, [5]Arizona State University, Tempe, Arizona, United States

Recent advancements in the realization of highly efficient shot noise-limited direct detectors now enable atomically-resolved *in situ* TEM image time-series to be acquired with temporal resolutions in the millisecond (ms) regime [1]. Many catalysts exhibit turnover frequencies on the order of $10^{-1} – 10^{2}$ sec^{-1}, so the opportunity to visualize atomic behavior with high time resolution holds much promise for understanding the chemical transformation processes occurring on catalyst surfaces. Unfortunately, acquiring *in situ* TEM time-series with ~ms temporal resolution necessarily produces datasets severely degraded by shot noise [2]. For typical atomic-resolution *in situ* TEM imaging conditions, at high frame rates the average dose in each frame can be < 1 e^- per pixel. Following Poisson statistics, counted images with an average dose < 1 e^- per pixel necessarily have a signal-to-noise ratio less than unity, and consequently, ascertaining the structure in the image becomes a major obstacle. There is a pressing need for sophisticated noise reduction techniques that both (1) preserve the temporal resolution of the image time-series and (2) facilitate the retrieval of atomic-level structural features at the aperiodic catalyst surface.

Deep learning-based convolutional neural networks (CNNs) achieve state-of-the-art denoising performance on natural images and are an emerging tool in various fields of scientific imaging, including in electron microscopy [3, 4, 5]. In the context of catalysis, the potentially fluctuating atomic structure at the catalyst surface is of principal scientific interest, and so it is critical to establish methods for evaluating the agreement between the noisy observation and the structure that appears in the network-denoised image. As far as we are aware, such analysis is not found in the literature on CNNs for electron micrograph denoising. Moreover, the mechanisms by which trained networks successfully denoise are often treated as a “black box”. Revealing these mechanisms, however, is a key step towards improving this methodology and understanding its limitations.

We have developed multiple deep CNN denoising techniques for atomic-resolution TEM time-series of catalyst nanoparticles [6, 7, 8]. In one approach (**Figure 1**), we train a supervised CNN on a dataset of simulated images produced through multislice calculations. Then we apply the trained network to an experimentally acquired 25 ms/frame *in situ* ETEM time-series of a Pt/CeO_2 catalyst in N_2 gas, denoising each frame individually. We have developed an approach based on statistical likelihood to quantitatively measure the agreement between the noisy observation and the atomic-level structure present in the network-denoised image, which we call a likelihood map. In another approach, we develop an unsupervised deep video denoising network that is trained just on the noisy time-series itself. This network thus does not rely on the availability of noiseless ground truth images, which can be advantageous in terms of time or required when simulations are not feasible. Additionally, in contrast to the frame-by-frame method, this network explicitly incorporates information present in adjacent frames. Analyzing the network’s equivalent filter, which reveals the mechanisms used by the network to denoise any part of a noisy image, shows that this allows the network to perform spatiotemporal filtering adapted to the local structures and motion of the underlying signal (**Figure 2**). This presentation will discuss our recent work on these networks; the aim will be to facilitate discussion on how to generate methodologies that are generalizable to a variety of materials systems and imaging conditions [9].

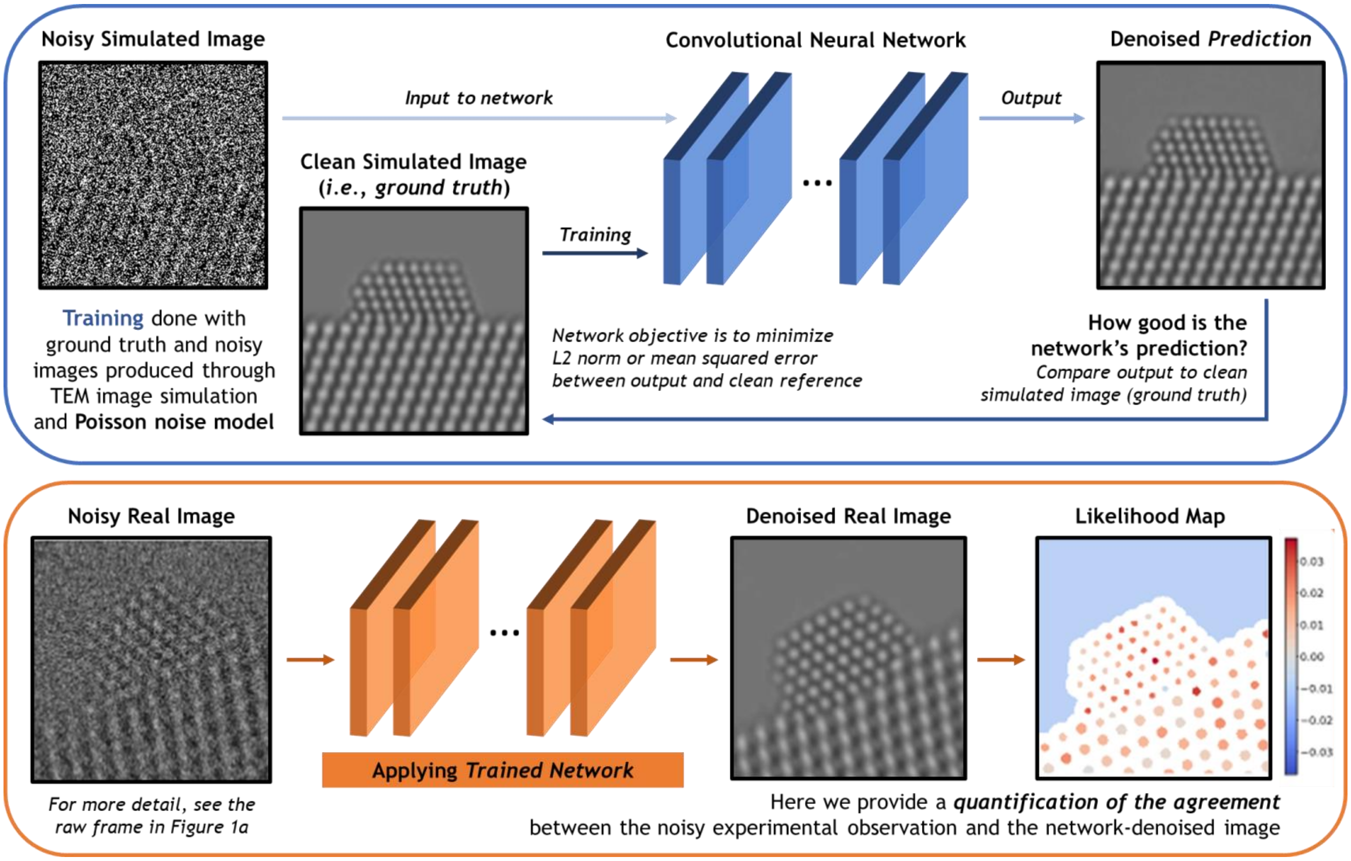

Figure 1. Supervised deep convolutional neural network training, application, and evaluation process. (Top) The network is trained on a large dataset of multislice TEM image simulations. (Bottom) The trained network is applied to experimental data taken under similar imaging conditions. The performance of the network on real images lacking noise-free counterparts can be evaluated through a statistical likelihood analysis, which allows one to quantify the agreement between the noisy experimental observation and the structure in the denoised image.

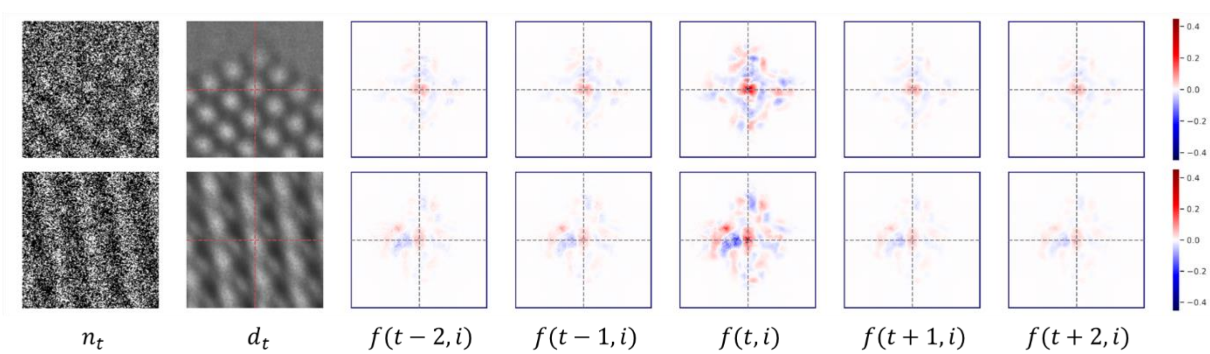

Figure 2. Investigating the mechanisms used by the unsupervised video denoising network to denoise (top) a Pt atomic column near the catalyst surface and (bottom) a Ce atomic column within the CeO2 support. The left column labeled nt shows regions of interest from the experimental 25 ms/frame in situ TEM time-series at a point in time, t. The corresponding denoised frames, dt, are shown next to it. As shown in the next 5 columns, at each point in time the network uses information from two preceding and two consecutive frames. The network's equivalent filter at the central pixel, i, is shown in the color plots, which gives insight into the spatiotemporal regions of the raw input that have the most effect on the denoised output.

References
[1] Faruqi, A. and G. McMullan, *Nucl. Instrum. Methods Phys. Res*, **878** (2018), p. 180-190.
[2] Lawrence, E. L., et al., *Microscopy and Microanalysis*, **26** (2020), p. 86-94.
[3] Liu, B. and Liu, J. *Journal of Physics: Conference Series*, **1176**:022010 (2019).
[4] Tian, C., et al., arXiv:1912.13171v4, (2019).
[5] Ede, J., *Machine Learning: Science and Technology,* (2020), in press.
[6] Vincent, J. L., et al., *in preparation*, pre-print at: arXiv :2101.07770, (2021).
[7] Mohan, S., et al., *in preparation,* pre-print at: arXiv:2010.12970, (2020).
[8] Sheth, Dev Y., et al., *submitted to CVPR*, pre-print at: arXiv:2011.15045, (2020).
[9] We gratefully acknowledge support of NSF grants CBET-1604971, NRT-1922658, CCF-1934985, OAC-1940097, OAC-1940124, and OAC-1940263, and the facilities at ASU's John M. Cowley Center for High Resolution Electron Microscopy.