Automated Crystalline Domain Segmentation in Polycrystalline TEM Images via Machine Learning-Based FFT Analysis

Woojin Bae[1], Shihyun Kim[1], Jinho Rhee[1]

[1]School of Chemical and Biological Engineering, Seoul National University, Seoul, Republic of Korea

Introduction and Methods

Traditional FFT-based crystallographic analysis of HRTEM images relies on manual peak selection, inverse transformation, and domain tracing, making the workflow time-consuming and inherently subjective. Different researchers can extract different crystalline information from identical FFT patterns, leading to inconsistent conclusions—an issue that becomes particularly severe for weakly crystalline and nanoscale-heterogeneous systems such as lithium-metal SEI. While recent 4D-STEM studies have demonstrated that machine learning can enable automated crystallographic segmentation through diffraction-pattern clustering, these approaches require specialized instrumentation, generate massive datasets, and demand substantial computational resources, limiting their accessibility. As a result, conventional HRTEM imaging remains far more practical and widely available in most microscopy facilities.

In our previous work [1], we applied DBSCAN clustering to inverse FFT reconstructions to segment crystalline domains from HRTEM images; however, the critical step of FFT peak selection still relied on manual intervention. In this study [2], we eliminate this remaining bottleneck by fully automating the upstream workflow. We introduce systematic FFT preprocessing to enhance peak visibility, automated detection of individual FFT peaks, and GMM-based classification to distinguish crystalline signal from noise. By integrating this automated peak selection with our established DBSCAN-based domain clustering framework, we remove the most subjective and time-consuming step in FFT-based analysis, enabling consistent, reproducible, and truly high-throughput crystallographic analysis from single HRTEM images.

Workflow: Our pipeline comprises four integrated stages. (1) GMM-Based Peak Classification: After computing the 2D FFT, we employ a two-component Gaussian Mixture Model to automatically classify peaks as signal-like (crystalline) or noise-like. GMM is ideal because FFT peak characteristics—intensity, sharpness, radial profile width—naturally form distinct distributions. Unlike supervised learning requiring labeled training data, GMM performs unsupervised clustering through expectation-maximization. Users adjust classification sensitivity via intuitive sliders, and non-physical peaks can be removed by double-clicking. (2) Automated d-Spacing Calculation: The tool automatically calculates d-spacing values from peak positions using user-provided pixel size calibration, enabling immediate phase identification. (3) DBSCAN-Based Domain Clustering: Selected peaks undergo inverse FFT to reconstruct real-space images showing crystallinity distribution. DBSCAN clustering groups spatially connected high-intensity regions while filtering isolated noise. DBSCAN's ability to discover arbitrarily shaped clusters without specifying cluster count makes it ideal for irregular grain boundaries. Two user-adjustable parameters (neighborhood radius ε and minimum sample threshold) control clustering behavior. (4) Visualization and Quantification: Crystalline domains are color-coded and overlaid onto the original image. The tool automatically quantifies individual domain areas and total crystalline coverage. Non-physical regions can be excluded via double-clicks.

Results

Our tool successfully analyzes polycrystalline HRTEM images from lithium-battery SEI layers and other complex systems where conventional analysis struggles with weak crystallinity and substantial phase heterogeneity. Automated GMM classification achieves robust distinction between crystalline signals and background noise across varying image quality conditions, with adjustable sensitivity accommodating materials ranging from highly crystalline to barely ordered. DBSCAN clustering effectively delineates grain boundaries even in images with irregular domain morphologies and significant amorphous fractions. The complete workflow executes in a fraction of the time required for manual analysis while producing reproducible results independent of analyst identity.

Representative results from SEI analysis demonstrate successful identification of multiple coexisting crystalline phases with varying d-spacings, quantification of their spatial distributions, and measurement of individual crystallite sizes ranging from nanometers to tens of nanometers. The overlay visualization enables immediate assessment of crystallinity distribution relative to morphological features, facilitating interpretation of structure-
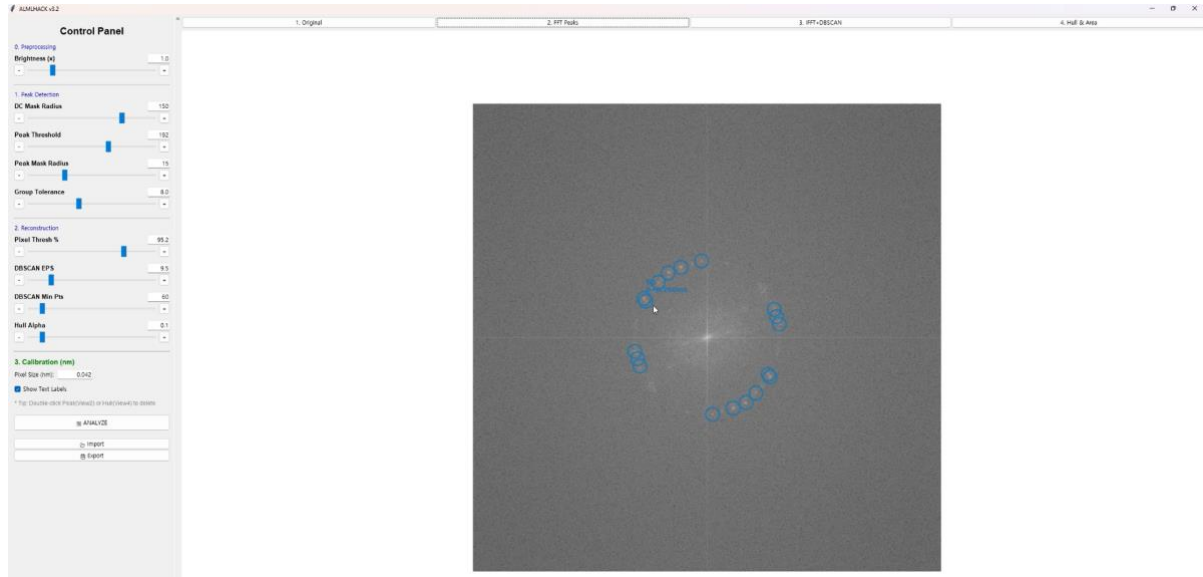
property relationships.

Figure 1. Automated FFT peak classification using Gaussian Mixture Models distinguishes signal-like crystalline peaks (blue) in polycrystalline SEI material, enabling reproducible peak selection.
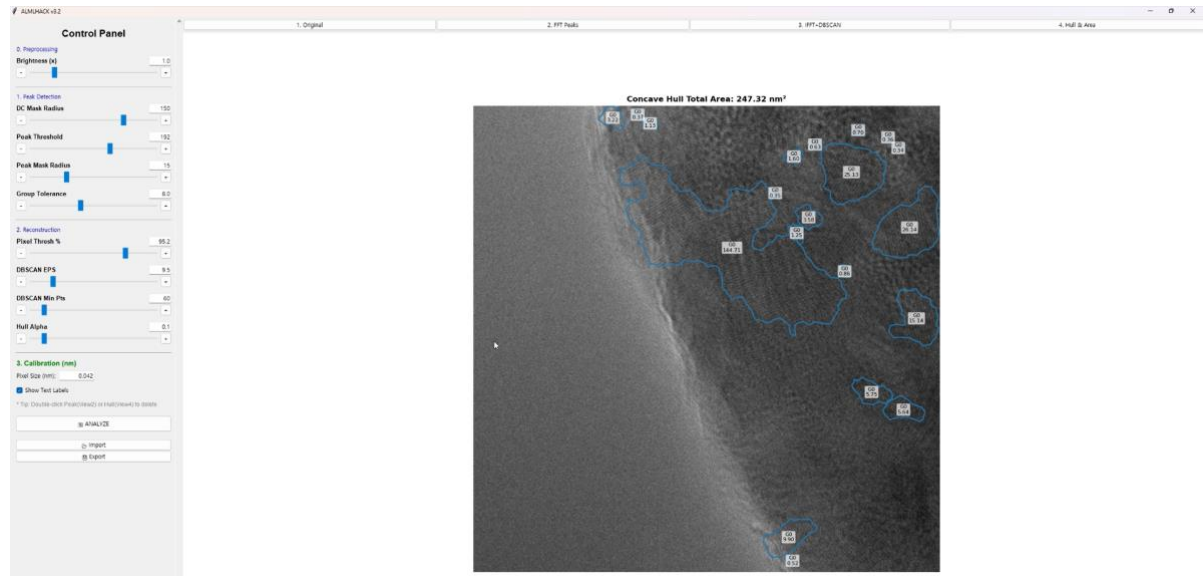

Figure 2. DBSCAN-clustered crystalline domains overlaid onto original HRTEM image following inverse FFT reconstruction.

By combining ML efficiency with flexible parameter control and expert correction, this tool addresses critical bottlenecks in HRTEM characterization. The GUI-based design ensures accessibility without extensive programming experience, democratizing advanced analytical capabilities across the microscopy community. We anticipate particular value for energy storage materials, catalysts, and structural ceramics where nanoscale crystalline heterogeneity understanding is essential.

References

[1] https://github.com/woojinb1/masK-driven-Analysis-for-iNverse-fft-yield-evaluation

[2] https://github.com/woojinb1/AI-ML-for-Microscopy-Hackathon-2025