

Convolutional Neural Networks for Evaluation of Sequential Beam Damage of Beam-Sensitive Solid Electrolytes

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Meeting-report

Convolutional Neural Networks for Evaluation of Sequential Beam Damage of Beam-Sensitive Solid Electrolytes

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Solid-state batteries have gained increasing attention as a promising alternative to conventional Li-ion batteries, owing to their superior safety and stability as well as the potential to achieve higher energy density. The garnet-type $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) solid electrolyte holds great promise due to its excellent stability against lithium and relatively high ionic conductivity [1]. Transmission electron microscopy (TEM) is a powerful tool for materials characterization, especially essential for battery research when in situ and operando techniques are available to provide valuable information for dynamic phenomena and processes [2, 3]. However, TEM characterization of various Li-containing solid electrolyte materials has been very challenging due to the radiation damage induced by high-energy electron beam, which makes the atomic imaging of LLZO difficult to achieve. Despite the advancement of cryo-TEM to mitigate the effect of beam damage [4], it is necessary to control the electron dose below the critical damage threshold to allow for reliable and reproducible characterization results. For accurate quantification of specimen damage, utilizing image dissimilarity as a surrogate measure can be an effective approach. Traditional methods such as Peak Signal-to-Noise Ratio (PSNR) and Structural Similarity Index (SSIM) have limitations: PSNR is overly sensitive to image content variations and lacks considerations for structural integrity, while SSIM may not adequately consider complex perceptual and cognitive elements. Therefore, we propose using pre-trained convolutional neural networks as feature extractors, followed by similarity computation in the feature domain. Pre-trained networks, such as ImageNet that is based on training of large datasets, can extract and compare high-level features and patterns in images, with less interference by human perception and more robustness to image variations than traditional metrics.

In this study, we performed scanning transmission electron microscopy (STEM) characterization of LLZO solid electrolytes at both room temperature and cryogenic temperature. As expected, high-angle annular dark field (HAADF)-STEM images show that cryo-STEM can preserve the LLZO crystal lattice (Fig. 1A), which was substantially damaged when imaged at room temperature (Fig. 1B). However, additional control experiments show that prolonged exposure under identical electron dose conditions would lead to more severe sample damage, even at cryogenic temperature, as depicted in Fig. 1C and D. To evaluate the electron beam damage, we employ ResNet-18 [5], pretrained on ImageNet [6], as the feature extractor. Given the high resolution and local pattern density of HAADF-STEM images, we segmented each frame into non-overlapping 20×20 patches and extracted feature vectors for each patch. Subsequently, we assessed patch-wise similarity between the queried frames (Fig. 1C and D) and the initial frame (Fig. 1A). The results, as illustrated in Fig. 1E and F, respectively, effectively highlight the beam damage in terms of the extent and its localization, offering a meaningful method to quantify electron beam-induced alterations in sequential image series. This dissimilarity measurement approach is essentially useful for improving STEM characterization of beam-sensitive LLZO solid electrolytes, which can be further expanded to quantitatively evaluate beam damage for a variety of sensitive materials, thereby establishing a valuable standard for researchers undertaking analogous investigations [7].

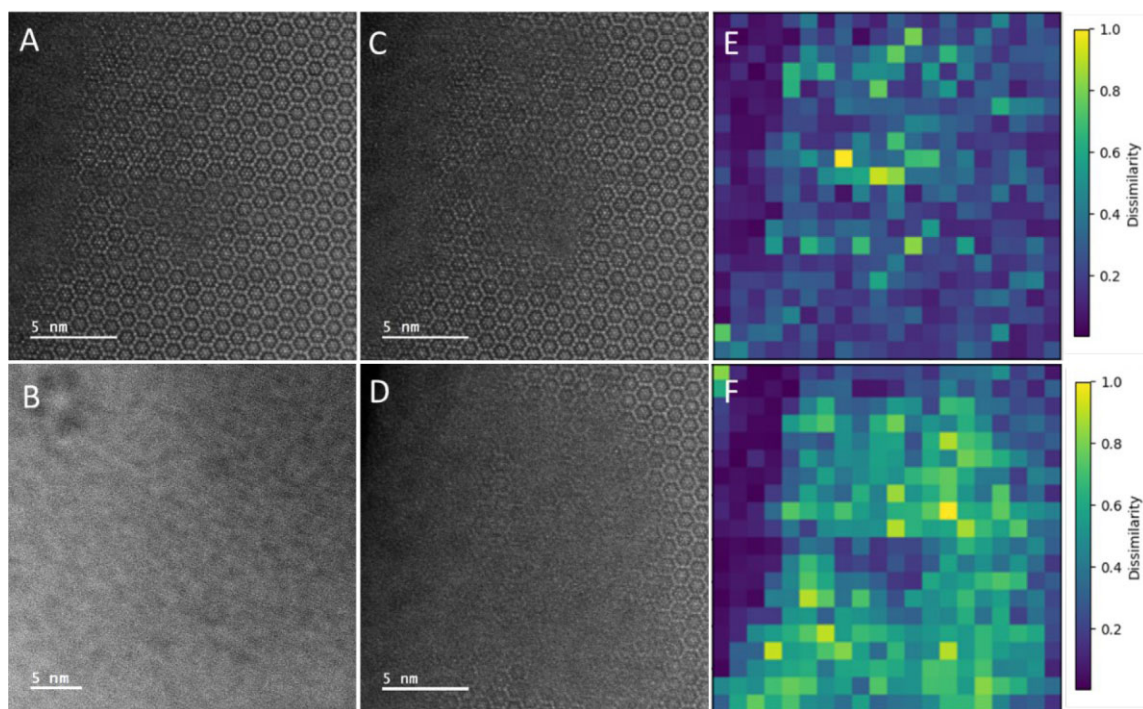


Fig. 1. HAADF-STEM images of LLZO solid electrolytes at (A) cryogenic temperature and (B) room temperature. Sequential HAADF-STEM images obtained after the initial frame (A): (C) the second frame and (D) the third frame, along with the corresponding dissimilarity maps shown in (E) and (F), respectively.

References

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