Predicting Ionic Conductivity of Solid-State Battery Cathodes Using Machine Learning

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Abstract— **Safer and more efficient battery technologies are in soaring demand, with a primary focus on transitioning from flammable lithium-ion batteries to non-flammable solid-state batteries. While solid-state batteries offer enhanced safety features, their power density remains a challenge due to poor ionic conductivity induced by non-optimal cathode microstructures. Laborious experimental processes and time-consuming data analysis algorithms are obstacles to establishing structure–performance correlation and optimizing cathode microstructure. In this paper, we present a machine learning approach to predict the current or resistance of a composite cathode based on scanning electron microscopy (SEM) images, given the inputs as a binary image, a voltage, and a conductivity value. Our results showed that current or resistance can be quickly predicted from input images with high accuracy.**

Keywords—solid-state battery; ionic conductivity; machine learning; image analysis

I. Introduction

Lithium-ion batteries are known to be the most prevalent type of battery in use today, despite their flammable liquid electrolytes and potential combustion hazards. The safety concern underscores the need to develop an alternative, safer type of battery, such as solid-state batteries. Although solid-state batteries offer better safety and higher energy density, their power density is low compared to their liquid-electrolyte counterparts due to highly tortuous ion conduction pathways within cathodes [1]. When the cathode composite possesses an unfavorable microstructure, Li^+ ions need to travel longer to conduct through the cathode, causing higher internal resistance and lower cathode utilization at high cycling rates [2].

Correlation between microstructure, transport properties, and cell performance is often studied with morphological characterization and subsequent data analysis [3]. Although 3D characterizations such as X-ray tomography and focus ion beam (FIB)-assisted SEM offer the most accurate structural insights, they tend to be labor-intensive and time-consuming. In contrast, 2D cross-sectional images are commonly employed for morphological characterization. Hence, having a quick prediction of battery performance from cross-sectional images can be beneficial.

The battery's properties can be predicted using rigorous numerical modeling with cross-sectional images showing the electrode's micro-structure. For example, resistance is an essential indicator of the battery's transport property, and it can be obtained by simulating the current distribution in the electrodes using numerical methods such as the finite different method (FDM). However, conventional numerical methods are oftentimes expensive for this task. Processing 2000 images sized 64×64 pixels using FDM takes approximately 10 minutes on an average laptop, whereas it requires up to 39 hours to process an equivalent number of images sized 400×400 pixels. The time-consuming process of numerical models might not be a desired characteristic in some situations, especially for real-time applications.

In this paper, we present a machine learning approach to quickly predict current or resistance based on cross-sectional SEM images of composite cathodes. Our results showed that current or resistance can be quickly predicted from input images with high accuracy.

II. Methodology

A. Finite Differennce Method

The conventional FDM is applied to solve equation (1) with predefined boundary conditions. Fig. 2 shows an illustration of the input images. The yellow region of the image represents conductive material, and the black region represents non-conductive material. Each region is assigned its corresponding conductivity value. By calculating the potential at each point, the current density is obtained.

$$\nabla(\sigma . \nabla\phi) = 0 \tag{1}$$

Where σ is conductivity [S/m] and ϕ is potential [V].

The conversion between resistance and current can be achieved using equation (2) below.

$$R = V/I \tag{2}$$

Where R is resistance [Ω], V is voltage [V], and I is current [A].

B. Machine Learning Model

The inputs include the generated images, their current or resistance values calculated using FDM with the given conductive material's conductivity (1 [S/m] as default), and the voltage (1 [V] as default). The Output is the predicted current and resistance values.

The general workflow is shown in Fig. 1. The images are inputted into the machine learning model to output the predicted current or resistance values.

Convolutional neural networks (CNNs) are popular choices when working with images. Hence, with some modifications, we selected EfficientNetV2-S as our machine learning model [4]. Yet, the original EfficientNet model is not suitable for our regression task. Consequently, we replaced the original model's last layers with a multilayer perceptron to output a current value. Aside from the regression modification, we also use Warmup Cosine scheduling to efficiently perform transfer learning.

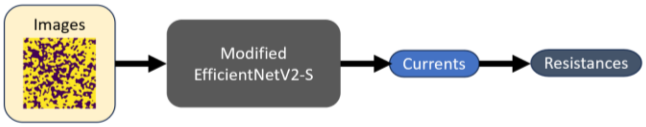

Figure 1. General workflow.

III. RESULTS AND DISCUSSION

Image dataset was generated with electrolyte fraction ranging from 0.6 to 1. Electrolyte fraction is the ratio of conductive region to the overall region. The training : validation : testing proportion is 2500:175:175. An example of the generated images is showed in Fig. 2.

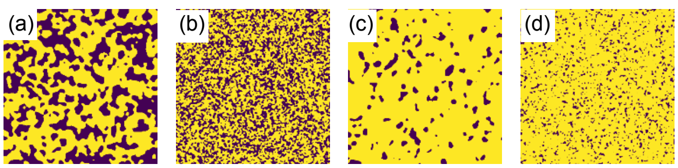

Figure 2. Generated binary images. (a),(b) electrolyte fraction: 0.6. (c),(d) electrolyte fraction: 0.9

Fig. 3 shows the currents of 2500 training images, with the current values exhibiting distinct variations corresponding to different levels of electrolyte fraction.

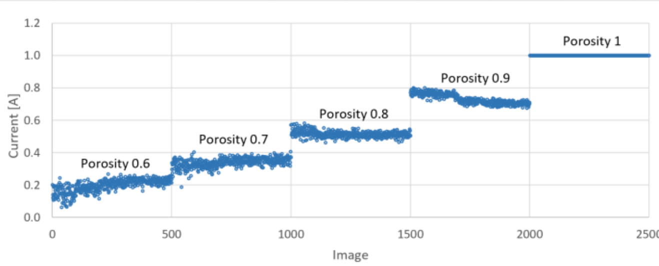

Figure 3. Input current values of 2500 training images.

Our model was able to predict the currents in only 9.7 ± 0.4 seconds for 2500 images of size 64×64. Compared to FDM which requires approximately 12 minutes for small-sized images, the machine learning model demonstrates significantly faster processing time making them particularly advantageous for handling larger images.

Fig. 4 shows the prediction results for the test image set. The results are highly accurate with the mean absolute percentage error of 4.0%. However, the model was only tested on a certain type of image that has similar patterns with electrolyte fraction larger than 0.6. Hence, we will need to work on more cases to generalize the model. To further enhance the accuracy for this specific pattern, our approach involves categorizing images according to their electrolyte fraction and subsequently training the model independently for each distinct electrolyte fraction value.

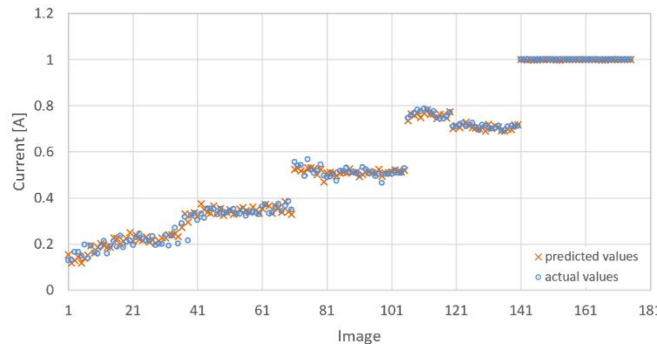

Figure 4. Predicted and actual current values.

The ionic conductivity of composite cathode can be calculated using equation (3). In this work, we assigned $l = A = 1$ so the total conductivity is equal to 1/R, and numerically equals the total current (if we have $V = 1$ [V] in equation (2)). From Fig. 4, where the conductivity of the conductive material equals 1 [S/m], we can see the current also represents the total conductivity. When electrolyte fraction equals 1 (Fig. 4, image 141-175), meaning the cathode contains only conductive material, the output current values are 1 or 100% conductive.

$$\sigma = l/(R.A) \tag{3}$$

Where σ is cathode ionic conductivity [S/m], l is length [m], R is total resistance [Ω], and A is cross-sectional area [m^2].

This paper has proved that ionic conduction properties can be predicted from cross-sectional images, though there is a tradeoff between accuracy and time as FDM gives a more accurate calculation while the machine learning approach is 72 times faster with a mean accuracy of 96%. The next step we want to take in the future is to extend this method to 3D images and test more experimental cases.

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