Deep Learning for the Automation of Particle Analysis in Catalyst Layers for Polymer Electrolyte Fuel Cells

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Abstract

The rapidly growing use of imaging infrastructure in the energy materials domain drives significant data accumulation in terms of their amount and complexity. The applications of routine techniques for image processing in materials research are often *ad hoc*, indiscriminate, and empirical, which renders the crucial task of obtaining reliable metrics for quantifications obscure. Moreover, these techniques are expensive, slow, and often involve several preprocessing steps. This paper presents a novel deep learning-based approach for the high-throughput analysis of the particle size distributions from transmission electron microscopy (TEM) images of carbon-supported catalysts for polymer electrolyte fuel cells. Our approach employs training an instance segmentation model, called StarDist [Schmidt et al. Medical Image Computing and Computer-Assisted Intervention – MICCAI 2018, Lecture Notes in Computer Science, vol 11071. Springer, Cham], which resolves the main challenge in the pixel-wise localization of nanoparticles in TEM images: the overlapping particles. The segmentation maps outperform similar models reported in the lit-

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erature, and the results on particle size analyses agree well with the manual particle size measurements, albeit at a significantly lower cost.

Keywords: High Throughput Screening, Catalyst Layers, Fuel Cells, Microscopy Characterization, Deep Learning, Convolutional Neural Networks

1. Introduction

Relentless exhaustion of non-renewable resources in combination with ever-rising environmental concerns demand escalating efforts to develop and improve clean energy technology [1, 2, 3]. As one of the leading candidates for clean energy applications, especially in the automotive sector, polymer electrolyte membrane fuel cells (PEMFCs) demonstrate remarkable technical characteristics such as high power density, high energy conversion efficiency, instant start-up/shut-down and zero-emission operation [4, 5, 6]. Despite numerous advantages, PEMFC technology suffers from a significant drawback in terms of high costs and insufficient performance of the catalyst layer (CL). The latter is fabricated using expensive Platinum (Pt) nanoparticles (NPs), which are supported on a high-surface area carbon material (Pt/C) [7, 8]. Therefore, further advances in performance and cost reduction of the Pt-based catalyst layer are vital to ensure the widespread deployment of PEMFCs [9, 10, 11].

In the past three decades, a plethora of research activities have focused on developing and evaluating novel alternatives to improve the performance of Pt-based CL through experimental methods, computational studies, and comprehensive characterization [12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22]. High-resolution X-ray and electron microscopy (*in situ/ex situ*) characterization methods are increasingly used for the evaluation of the catalytic activity and stability of the catalyst layer. Examples include ultra-small angle X-ray scattering (u-SAXS), focussed ion beam scanning electron microscopy (FIB-SEM), transmission electron microscopy (TEM), scanning transmission electron microscopy (STEM), or scanning transmission X-ray microscopy (STXM) [23, 24, 25, 26].

To unravel the connection between microstructure properties and performance,

it becomes essential to quantify certain features of the imaging data correctly, e.g., the particle size distribution (PSD), pore size, pore network, or aggregate size - with less reliance on human intervention and interpretation [27, 28]. This quantification may require additional analyses, which can be done either manually by the operator or automatically via processing the images through a set of specialized algorithms to extract the pertinent structural features [29, 30].

The type of post-processing analysis of materials characterization data alluded to be beneficial as numerous studies suggested that the electrochemical performance of CL is strongly dependent on the surface area, size, shape, and distribution of Pt NPs [31, 32, 33, 34]. For example, an increase in mean Pt particle size in an aged cathode and subsequent reduction at anode, associated with Pt^{2+} ion crossover, could indicate degradation of the carbon support [35, 36, 37]. In another study the larger particle size and agglomeration after potential loading was attributed to the dissolution and re-deposition of Pt in the catalyst layer [38]. Smaller Pt particles are not favorable either because of the stronger adsorption of reaction intermediates at the edges and corners of NPs; the latter results in blockage of oxygen reduction reaction path and makes the Pt more susceptible to dissolution [39].

The widely employed technique for particle size analysis entails measuring a set of manually drawn straight lines from the edges of individual particles in the image [40]. This approach employs standard image processing tools like ImageJ [41] and may requires thresholding the image and applying filters for denoising, to segment the particles from the support material or background. Notwithstanding, this essential thresholding step becomes tricky in capturing small particles or when the contrast between particle and support material is not strong. In addition, in a one-at-a-time inspection of particles, the operator usually considers only a subset of the particles for each image, resulting in the loss of information. The long, tedious, and detailed manual procedure makes this approach ineffective for high-throughput or real-time analysis. Moreover, identifying the overlapping particles is strongly dependent on the operator's bias and judgment. These issues generate an unreliable metric for the quantification

and can slow down or even falsify the analysis.

Figure 1: (a) Methodical pipeline for deep learning-based analysis of particle size distribution involving 1. particle annotation, 2. supervised training for the instance segmentation of particles, 3. setting scale for image calibration and particle size extraction with computer vision implemented using OpenCV library [42], 4. Obtaining particle size statistics with visualization libraries, e.g., matplotlib. (b) The left panel shows a TEM image with overlapped Pt particles supported on a carbon agglomerate; the StarDist method used in this work predicts star-convex polygons parameterized by the radial distances $r_{i,j}$, and object probabilities $d_{i,j}$ for the pixel i,j [43]. The right panel shows the U-Net architecture [44] and selected the final instances via the non-maximum suppression (NMS) technique. (c) Example TEM images and their corresponding predicted instance segmentation of Pt particles using StarDist and U-Net.

Semi-automated approaches for the analysis of well-separated and monodisperse NPs have been developed. For example, for magnetite NPs, classical approaches using Otsu’s binarization with Canny edge detection and edge linking algorithms were employed [45]. For the population analysis of Pt NPs on glassy Carbon, an earlier report has introduced the local adaptive threshold (LAT) in the im-

age processing steps [46]. This step improved the segmentation of particles by thresholding the smaller patches of the image, but at the cost of additional computing time [46]. Ponti and co-workers developed PEBBLES [47], the software
65 program for the size measurement and morphological analysis of nanoparticles in TEM micrographs. PEBBLES applies fitting 3D intensity models to relate the contrast of the image and the depth of the dip in the intensity surface to the NP size, making the automatic measurement and analysis of the distribution of morphological parameters possible. However, as pointed out by the
70 authors, the software is limited in accurately fitting the overlapping NPs or the heterostructured NPs (e.g., core-shell NPs) [47].

Over the last decade, deep learning (DL) methods based on the convolutional neural networks (ConvNets) have transformed field of computer vision by out-
75 performing the conventional methods in various image and video recognition tasks such as in verification/identification, classification, object detection, and segmentation, as well in other applications including image reconstruction, denoising, image synthesis, colorization, style transfer and other tasks [48, 49]. To leverage the advancements in DL-based approaches in microscopy image
80 processing, more recently, the ConvNets algorithms have been deployed to several platforms for various analysis tasks like segmentation, object detection, denoising, or super-resolution microscopy. The most exciting example is the recently developed ZeroCostDL4Mic[50] platform that provides the advantage of free computational resources on GoogleColab for training the deep neural network models. Unlike the conventional procedures requiring several preprocess-
85 ing steps for particle analysis to construct suitable features, DL models utilize the information content directly from the raw TEM micrographs [51]; once a suitable model is trained on a representative dataset, they can predict swiftly and accurately without the necessity for the human-based alteration of parameters. Moreover, the robustness of DL models is validated with various metrics
90 like precision, recall, or accuracy, which introduces well-defined quantification metrics for particle analysis.

More recently, Yao *et al.* implemented the popular U-Net model for real-time

nanoparticle segmentation in liquid-phase TEM videos [52]. This novel approach
 95 enabled the statistical analysis of the diffusion, reaction, and assembly kinetics
 of the rod-, prism- and cube-shaped colloidal NPs [52]. Cole and co-workers
 introduced ImageDataExtractor [53, 54], a novel tool to automate the extraction
 and analysis of microscopy imaging data from the scientific literature. The
 main feature of the ImageDataExtractor is its capability in segmentation and
 100 quantification of NPs via Bayesian DL for a diverse set of morphologies [54].
 However, as will be discussed, ImageDataExtractor is limited in the accurate
 localization of overlapping NPs, as is the case for Pt NPs on a carbon support
 material.

This paper presents a novel DL-based approach to automate the PSD analy-
 105 sis from TEM micrographs of catalyst layers in PEMFCs. We employed the
 pre-trained StarDist model [43], and fine-tuned it on our annotated imaging
 dataset for the instance segmentation of Pt NPs on the high surface area Car-
 bon support. The trained model largely resolved the localization of overlapping
 NPs, and the proposed pipeline will enable the automation of PSD analysis.
 110 Moreover, unlike the conventional procedure based on image thresholding, this
 approach adopts definite quantification metrics for evaluating the segmentation
 model.

2. Methods

Samples for TEM imaging were either catalyst powders directly dispersed on
 115 the TEM grid or thin sections of the fuel cell electrodes. The catalyst powders
 were prepared by dispersing catalyst powders in 50 vol% isopropyl alcohol in
 water and dropped on the 200 lacey carbon TEM grid. Thin sections of the elec-
 trodes were prepared by embedding the electrode in a 1:1 mixture of trimethy-
 lolpropane triglycidyl ether resin (Sigma-Aldrich, USA) and 4,4'-methylenebis
 120 (2-methylcyclohexylamine, Sigma-Aldrich, USA) hardener, and sectioning thin
 slices (≈ 100 nm) using Leica UCT ultramicrotome and Ultra 45° DiATOME
 knife, USA. The sections were placed on 200 mesh Cu TEM grids. Images for

particle size distribution were obtained using Talos F200X scanning transmission electron microscope (Thermo Fisher Scientific, USA) with electron accelerating voltage of 200 kV.

Our DL-based approach for the PSD analysis involves the four steps in Figure 1(a): (1) manual annotation of Pt NPs, (2) supervised learning for particle segmentation, (3) automated diameter measurement on the predicted region-of-interest (ROI), and (4) statistics and visualization. The first step for the supervised learning task of segmentation is the pixel-wise annotation of NPs in the TEM images. The quality of the trained model largely depends on the quality of the labeled data; therefore, it is important to choose an efficient and accurate tool for this labor-intensive step. For this purpose, we used QuPath [55], a user-friendly open-source software initially developed for quantitative pathology and bioimage analysis. Forty high-resolution TEM images (1024×1024 and 2048×2048 pixels) of Pt NPs on Carbon support were manually annotated, with 34 used for training purposes and 6 for validation purposes. The image set comprised four different magnification levels, at 10, 20, 50, and 100 nm scales, providing a generalization of the scale range for the training of more robust models. An expert performed the annotations considering visual evidence for the overlapping particles while eluding the strongly crowded Pt aggregates. We make this dataset publicly available.

Figure 2: Demonstration of a TEM image along with the ground truth and predicted masks. Examples of true-positive (TP), true-negative (TN), false-positive (FP), and false-negative (FN) predictions are shown on the predicted mask.

Given the images and corresponding labeled masks as ground truth, for the pixel-level localization of individual Pt particles, we trained the StarDist model [43].

145 StarDist employs the U-Net architecture [44] and overcomes the typical segmentation errors for the dense prediction of merged bordering particles by localizing the singular particles as star-convex polygons (Figure 1 (b)). StarDist was initially developed for the segmentation of biological systems to differentiate individual overlapping cells in crowded images. In this model, each pixel is pa-
150 rameterized by two values, the radial distances and object probabilities. For the former, a star-convex polygon is fitted from the pixel position to the edges of the localized particle. On the other hand, object probability is represented by the normalized shortest distance from the edge of the particle; the latter increases with the decreasing distance of the particle center.

155 Standard architectures for image segmentation usually involve fully convolutional networks that first perform convolution and down-sampling operations to extract essential features from the image (encoding into a latent space), followed by up-sampling and transpose convolution operations that make the activation maps larger until it reaches the input image size (decoding from a latent space).

160 In the U-Net architecture, in addition to this data stream, the up-sampled decoder features are concatenated with the exact corresponding resolution from encoder feature maps (as shown by the gray arrows in Figure 1 (b)). Concatenating encoder and decoder feature maps enables recognizing object boundaries or edges by combining features across all levels, from low to high, leading to more
165 accurate output segmentation maps (Figure 1 (c)). The StarDist implementation of U-Net employs data augmentation to expand the size of the training set by generating modified versions of images in the dataset, where the images are cropped in patches and augmented by rotation, changing intensity, and Gaussian blurring. In this model, the non-maximum suppression (NMS) technique
170 is further used to suppress the multiple detections of individual objects. This way, the predicted box with higher object probability suppresses the overlapped boxes with lower probabilities. The number of epochs and the number of steps per epoch for training were chosen as 400 and 100, respectively.

Figure 3: The left panel shows the TEM image, ground truth mask, and predicted mask. The right panel compares the manually measured particle diameter with the deep learning-based computed particle diameter.

The performance of the trained model was analyzed by plotting standard metrics concerning the Intersection over Union (IoU) threshold. The IoU measures the number of pixels common between the ground truth and prediction masks divided by the total number of existing pixels in both masks. Conventionally, reliable predictions correspond to an IoU value ≥ 0.5 . In our case, a true-positive (TP) represents the case if a prediction-target mask pair for nanoparticles has an IoU score that exceeds 0.5; likewise, a true-negative (TN) is that for support or background. On the other hand, a false-positive (FP) indicates a predicted NP mask with no associated ground truth mask, and a false-negative (FN) indicates a ground truth NP mask with no associated predicted mask. Examples of TP, TN, FP, and FN are demonstrated in Figure 2. This way, the metrics of accuracy, precision, recall, and F1 score are defined as,

$$Accuracy = (TP + TN) / (TP + FP + TN + FN);$$

$$Precision = (TP) / (TP + FP);$$

$$Recall = (TP) / (TP + FN);$$

$$F1 = 2 \times Precision \times Recall / (Precision + Recall)$$

For an image with many NPs, it is not a severe problem to have a few FN predictions because it does not significantly affect the statistical analysis. However, a model with high precision indicates false particles are not introduced to the particle size distribution analysis. Therefore, precision is a reliable measure for
190 the evaluation of the model performance. The accuracy, on the other hand, can be a misleading metric for the images with a small number of NPs as it becomes biased in mainly relating how well the model predicts the background.

Given the model’s prediction for segmentation maps, the ROI for each particle was white-filled, cropped, and saved into a JPEG file. Next, using the contrasted
195 contours, we used the OpenCV library [42] to place a bounding box around the predicted masks and measured the number of pixels along with the X or Y directions (step 3 in Figure 1 (a)). Here, manual image calibration was used for a pixel-to-real-distance conversion. Lastly, The particle size can be estimated as the average values along with the X and Y directions, and the corresponding
200 histogram can be generated for the PSD analysis (step 4 in Figure 1 (a)).

3. Results and Discussion

Figure 3 compares the manually measured particle diameter performed by an expert as a reference and the computed particle diameter values for a test TEM image according to the approach presented. The image is shown in the left panel
205 along with the ground truth and predicted segmentation masks. The strong correlation between the values indicates the effectiveness of our deep learning-based approach for the high-throughput particle size analysis with minimal human interventions. For this image, the fine-tuned StarDist model on our annotated dataset could localize a total of 134 particles out of the 150 particles in the
210 ground truth mask. Interestingly enough, the model captured most of the overlapping particles. The obtained accuracy, precision, recall and F1-score at an IoU threshold of 0.5 were 71%, 0.86%, 0.81%, 0.83%, respectively. Even though the performance of the model can be further improved by expanding the train-

Figure 4: (a) Localization and counting the particles made available by the deep learning approach. (b) Comparison between the predicted segmentation maps of test images at different scales using the StarDist model employed in this work and the ImageDataExtractor 2.0 [54]

ing set, the main origin of discrepancies is for the non-rounded shape particles.

215 This can be explained by general difficulty to predict non-convex shapes that is rooted in StarDist radial model for contour representation. As can also be seen from the predicted map, most of the localized particles are round-shaped or have a convex contour. This would suggest further room for improvements in the correct shape predictions using for example algorithms that use a more

220 flexible contour representation for learning like the Contour Proposal Networks (CPN) in Ref. [56] or PolarMask++ [57].

Figure 4 (a) demonstrates another valuable capability of the deep learning approach to localize particles in the TEM images. The ROI data from the segmentation model makes it possible to assign a number and count the particles

225 in the image. Likewise, with the ROI data, the area of individual particles can be calculated. These abilities can be in particular helpful in estimating the Pt/C ratio or the Pt mass loading from the image. Figure 4 (b) compares the

segmentation maps for the test images obtained in this study at different scales with those from recently developed ImageDataExtractor 2.0 software [54]. The ImageDataExtractor model also uses a deep learning approach for microscopy particle analysis. For our dataset, the StarDist model significantly outperforms the ImageDataExtractor 2.0 in predicting the number of particles and localizing the overlapping particles. As shown in Fig. 4 (b), for the test TEM images at 10, 20, 50, and 100 nm scales, the StarDist predicted 27, 144, 628, and 286 particles, respectively, while the existing model found only 14, 67, 29, and 133 particles. Notably, in ImageDataExtractor 2.0, the lack of capability to separate the overlapping particles leads to a fusion of the near particles in segmentation masks, reducing the total amount of detected particles. Lastly, with the computed particle sizes the histograms for particle size distributions can be generated using typical visualization packages available in various programming languages. An example is shown in Fig. 5. Here, we used a bin size of 0.5 nm and the kernel density estimation (KDE) function for fitting the histogram. Binning can also be accomplished by using conventional statistical methods and expressions including Sturge’s rule, Doane’s Rule, Scott’s Rule, Rice’s Rule or Freedman and Diaconis’s rule [58, 59, 60, 61, 62]. Fitting for particle size distribution histogram can be performed using algorithms based on a variety of distribution functions such as polynomial distribution, Gaussian distribution, Weibull probability distribution, lognormal distribution, or Rayleigh distribution. [63, 64, 65, 66].

Figure 5: Particle size distribution was obtained using the deep learning-based approach of the instance segmentation of nanoparticles in the TEM images of Pt/C catalyst layers.

250 4. Conclusions

In conclusion, this paper presented a deep learning-based approach to automating particle size analysis in the TEM images of catalyst layers for polymer electrolyte fuel cells. The pre-trained StarDist model, fine-tuned on our annotated imaging dataset, could segment round-shaped overlapping nanoparticle instances. Minimizing the human intervention in the imaging-based characterization of catalyst layers in PEFCs enables the high-throughput screening required in the accelerated design and fabrication of novel materials. Unlike the manual approach, the deep learning-based approach introduces a well-defined metric for particle analysis: the model precision for the segmentation of nanoparticles. Future research will focus on developing algorithms for the segmentation of nanoparticles with diverse, non-convex shapes enabling the automation of morphological analyses. While we specifically discussed the particle size analyses, the deep learning algorithms are generic and can learn to automate the pore size or network analyses. Advanced characterization can also be achieved via auxiliary-task learning in an end-to-end training pipeline. The latter allows finding the structure-property relationships directly as the output of the deep learning models for full automation. A pivotal application of this approach is in autonomous materials fabrication using lab-scale stationary or mobile robotic systems [67, 68, 69, 70, 71] which demands a bidirectional and rapid data flow from catalyst fabrication to ink preparation, catalyst coated membrane fabrication, membrane electrode assembly, and a half or complete cell design, including testing and *in situ/ex situ* characterization steps.

Conflicts of interest

The authors declare no competing financial interest.

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