Hierarchically Structured Classification of Carbon Nanostructures from TEM Images by Machine Learning and Computer Vision

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Carbon nanotubes (CNT) and carbon nanofibers (CNF) are two widely used carbon nanomaterials for various industrial applications. The airborne particles released from of these materials during the handling and manufacturing of CNT/CNF products in workplaces has potential health impact on humans when exposed through inhalation [1-2]. In order to evaluate the potential exposure hazards of these materials, the airborne nanoparticulate samples were collected and analyzed by transmission electron microscopy (TEM) to determine their types, sizes, and specific morphological properties. After samples were obtained and imaged, individual particles were identified and classified based on aspect ratios and degree of agglomeration, among other descriptors. However, manual identification and classification of nanoscale structures require significant technical expertise and can be highly time-intensive for complex nanostructures [2-3]. Therefore, we introduced transfer learning-based machine learning algorithms and incorporated computer vision approaches to classify a dataset that consisted of 5,323 greyscale TEM images of airborne carbon/non-carbon nanomaterials (see representative images in Fig. 1) to improve classification accuracies and enable automatic processing of nanostructured micrograph data.

The primary transfer learning training pipeline [4] for our convolutional neural network (CNN) model was constructed with a vgg16 architecture based on ImageNet pre-trained dataset and followed by using hyper-column representation to collect matrix computation results from five convolutional (Conv) blocks. We constructed a K-means library as a grid and quantified it with vector of locally aggregated descriptors (VLAD) as an encoder, and finally trained with the gradient boosting algorithm for an optimized classifier. In addition, data augmentation techniques, by 90-degree rotation transformation to the original images, were used as pre-processing to mitigate the overfitting of the model on the majority classes ($>80\%$) over the minority ones. Other than the classification model, we designed different hierarchical learning structures to reduce the model complexity by dividing an all-in-one task into hierarchical or stepped binary classifications. We designed two different hierarchical structures for 4-class pure carbon sets: (1) easy-to-hard mode that first classifies 4 classes into morphologically similar groups and further identifies each binary group; (2) hard-to-easy mode that applies in an opposite sequence to distinguish the mixed groups. The total probability to correctly predict the labeled images is equal to the probability of making the right decision on the first-level groups, multiplied by the conditional probability of making the correct prediction of the second-level group given the prior first-level determination.

In conclusion, both two-level hierarchical structures achieved comparable classification performance for the 4-class pure carbon dataset, 89.7% and 85.6% respectively, compared to the original vgg16 hypercolumn model with 90.9% cross-validation accuracy (as seen in Fig. 2). And these hierarchically structured learning approaches have shown promising directions in identifying different levels of information, such as specific matrix/matrix-surface structures. Future improvement may be achieved by implementing more combinations of learning structures to either a 4-class or 8-class dataset with approaches based on other Deep Convolutional Neural Network architectures.
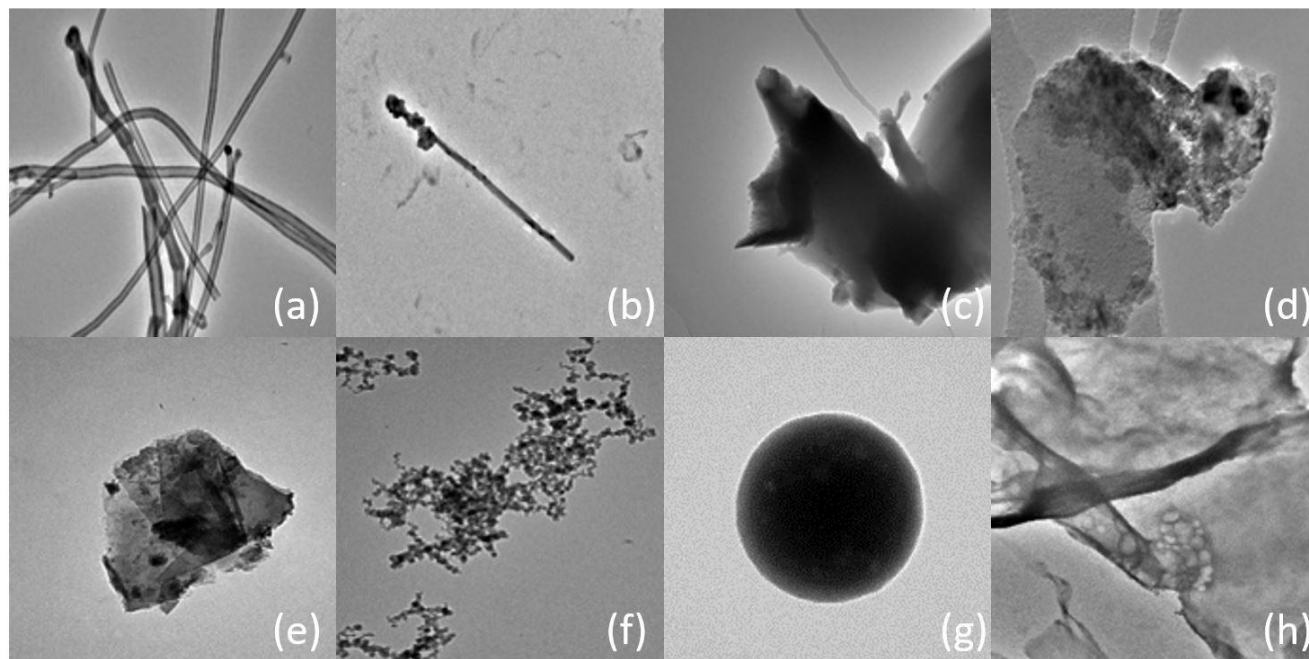
Figure 1. Fig.1 Sample images of 8 types of nanostructures from the TEM dataset. CNT structures are classified as (a) Cluster (Cl), (b) Fiber (Fi), (c) Matrix (Ma) and (d)Matrix-Surface (MS). Non-CNT structures are classified as (e) Graphene Sheet (GS), (f) Soot Particles (SP), (g) High-Density Particles (HDP), and (h) Polymer Residues (PR).
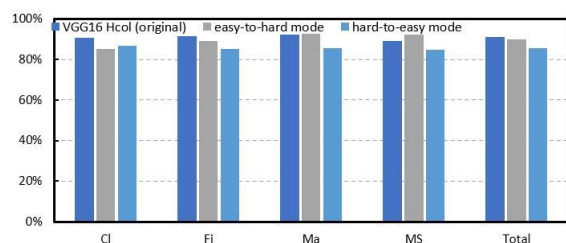
Figure 2. Fig.2. Average classification accuracies of 4 individual types and the pure carbon dataset obtained from three transfer learning approaches: VGG16 Hcol (original), easy-to-hard mode, and hard-to-easy mode.

References

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