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Prediction on the relative permittivity of energy storage composite dielectrics using convolutional neural networks: A fast and accurate alternative to finite-element method

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ABSTRACT

The relative permittivity is one of the essential parameters determines the physical polarization behaviors of the nanocomposite dielectrics in many applications, particularly for capacitive energy storage. Predicting the relative permittivity of particle/polymer nanocomposites from the microstructure is of great significance. However, the classical effective medium theory and physics-based numerical calculation represented by finite element method are time-consuming and cumbersome for complex structures and nonlinear problem. The work explores a novel architecture combining the convolutional neural network (ConvNet) and finite element method (FEM) to predict the relative permittivity of nanocomposite dielectrics with incorporated barium titanite (BT) particles in polyvinylidene fluoride (PVDF) matrix. The ConvNet was trained and evaluated on big datasets with 14266 training data and 3514 testing data generated form a programmatic algorithm. Through numerical experiments, we demonstrate that the trained network can efficiently provide an accurate agreement between the ConvNet model and FEM by virtue of the significant evaluation metrics R^2, which reaches as high as 0.9783 and 0.9375 on training and testing data, respectively. The strong universality of the presented method allows for an extension to fast and accurately predict other properties of the nanocomposite dielectrics.

KEYWORDS

Relative permittivity, nanocomposite dielectrics, convolutional neural networks, finite element method, prediction accuracy.

The permittivity is an essential parameter to characterize the tendency and ability of the local migration of atomic charges in the material under an external electric field. The magnitude of permittivity significantly affects the dielectric performance of the material in many applications, such as capacitive energy storage[1,2], electro-elastomers[3,4], triboelectric nanogenerators[5], piezoelectric sensors[6], flexible electronics technology[7], and insulating packaging[8]. For capacitive energy storage applications, polymer material with higher permittivity is desirable to improve the energy storage density of capacitive film, according to the relationship between permittivity and energy storage density for linear dielectrics[9],

$$U = \frac{1}{2}\varepsilon_0\varepsilon_r E_b^2 \tag{1}$$

where $\varepsilon_0 = 8.854 \times 10^{-12}$ F/m is the vacuum permittivity, ε_r and E_b are the relative permittivity and breakdown strength of the dielectric material, respectively. For the current commercially used biaxially oriented polypropylene material (BOPP), due to its low permittivity ($\varepsilon_r = 2.2$), the energy storage density of the dielectric film is limited [10] in the range of 1–2 J/cm³. Recently, poly(vinylidene fluoride) (PVDF) and its copolymers[11] have gained a lot of attention, owing to their high permittivity originates from its ferroelectric molecular structure. To further improve the permittivity of polymer material, some inorganic ceramic particles, such as barium titanite ($BaTiO_3$)[12–14], boron nitride nanosheets (BNNS)[15], and aluminum oxide (Al_2O_3)[16], have been incorporated, which contribute both the inherent molecular polarizability and interfacial polarization. By adjusting the geometry of the inorganic particles and their distribution in the organic matrix, both the permittivity and the energy density of the composites have been dramatically improved.

Establishing the direct relationship between the microstructure and properties of composite dielectrics is important to understand the microscopic mechanism of energy storage, however the conventional experimental "trial and error" research has the obvious disadvantages of low efficiency and aim blindness. Effective medium theory[17] is helpful for understanding the polarization contribution arises from incorporated particles, but the prediction accuracy is strongly reduced for irregularly shaped particles[18]. The finite element method (FEM)[19], a representative physics-based simulation method, can flexibly establish the arbitrary geometric model of the composite dielectrics and accurately calculate the local distribution of electric field and space charge[20]. Through the bridging effect of these mesoscopic parameters, the connection between the structure and properties of composite materials can be well established and analyzed[21]. Nevertheless, it is still very time-consuming and laborious to model complex composite structures and nonlinear problem such as ferroelectric hysteresis phenomenon[22]. When the composite contains fiber-shape fillers with a large aspect ratio, an extremely fine mesh should be adopted to accommodate the smaller dimension, which will drastically increase the computational workload for the whole model[23]. For nonlinear problems, to ensure the convergence of the calculation, it is necessary to set a very small-time step, result in a significantly increase of the simulation time. In addition, physics-based simulations have shown their limitations in solving inverse design

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problems that is, inversely calculating the microstructure of the material according to the target properties[24], although some optimization algorithms, such as genetic algorithm, annealing algorithm that can help to obtain the optimal material structure.

With the rapid development of computational science, data-driven approaches based on machine learning (ML) techniques, using a variety of statistical and probabilistic methods that allow computers to learn from the training data and explore the hidden patterns, have shown great potential in material design[25,26]. It provides a new means of analyzing the performance of novel materials, establishing quantitative structure-properties relationships, predicting the property parameters, and even disclosing the micro-mechanism of the macro-behaviors of the materials[27]. Lots of machine learning methods, including linear regression, support vector machines, decision trees, and artificial neural networks (ANNs), have demonstrated as effective and efficient methods in the research of dielectric properties of many polymer-based nanocomposites[28,29]. In deep learning, a convolutional neural network (ConvNet) is a class of artificial neural network (ANN), most commonly applied to analyze image-related datasets[30,31]. In a convolutional neural network, the hidden layers include many layers that perform a dot product of the different convolution kernel with the layer's input matrix in each layer. As a result, some hidden information can be extracted separately. Abueidda et al.[32] and Ye et al.[33] have proposed ConvNet models to predict the mechanical properties of composites with checkerboard and real complex microstructures, respectively. Furthermore, due to fundamental microstructure characterization and measurement techniques, including scanning electron microscopy (SEM), tunneling electron microscopy (TEM), atom force microscopy (AFM), optical microscope, and so on, which can provide a large amount of image datasets for ConvNet model training, has great promoted its development prospects in future research.

In this work, a method to predict the permittivity of BT/PVDF composite dielectrics based on convolutional neural network is proposed. Through the connection of geometrically generated codes and finite elements, an adequate number of labeled datasets have been provided for ConvNet model training. The whole work is organized as follows. In Section 1, the development of the ConvNet method is proposed, including dataset construction, finite element method, and convolutional neural network architecture. In Section 2, the effectiveness of the ConvNet method is verified, and the prediction results are illustrated and discussed in details. In the last section the conclusions are derived and the prospect of the present method is introduced.

1 Methods

A convolutional neural network model is development to quantitatively predict the permittivity of composite dielectrics composed of BT spherical nanofillers and PVDF polymer matrix. In order to provide significant amount of data to improve the prediction accuracy, microstructure image generator code coupled with finite element method is adopted, and the overall workflow of the prediction of the relative permittivity of BT/PVDF composites is illustrated in Figure 1. Section 1.1 introduces the construction process of training and testing datasets. Section 1.2 talks about the boundary value problem and calculation of the permittivity of BT/PVDF composites by finite element methods. Section 1.3 scrutinizes the architecture of the ConvNet model including both the home-made model with different layers and model parameters and a transfer learning from a pre-organized ResNet-50 network, while Section 1.4 introduces three evaluation metrics for the model.

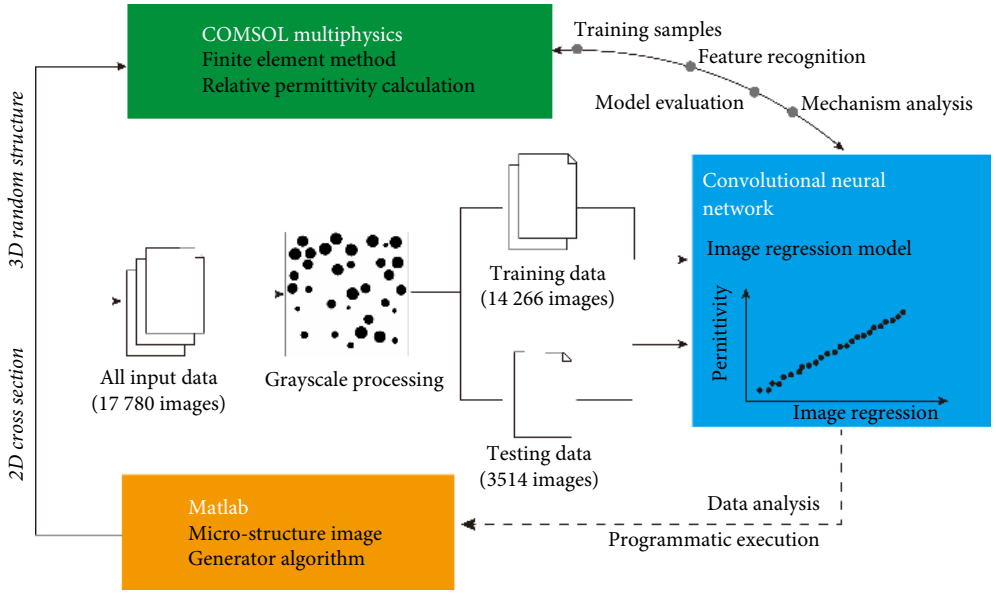

Fig. 1 Schematic of the workflow designed for prediction of the relative permittivity of BT/PVDF composites.

1.1 Datasets preparation

In regression analysis, convolutional neural networks usually require a large number of labeled samples for the training of model parameters to improve the accuracy prediction. For each labeled sample, the feature input is a two-dimensional cross-sectional picture of the BT/PVDF nanocomposites, while the label value is the relative permittivity of the corresponding composites. However, it is obviously time-consuming and labor-intensive to obtain adequate samples for ConvNet model training using the experiment method with respect to high-resolution characterization technique, including SEM, TEM, and AFM. To solve the problem, a structural programming mainly includes composite microstructure generation codes and finite element method is

proposed. The former generates the input required image features, while the latter obtains the label value of the permittivity of the corresponding feature. The random generation algorithm of sample is logically described as in Algorithm 1.

Algorithm 1: The random generation scheme of sample

1: Define the model hyperparameters: length L, width W and height H;
2: Define the particle hyperparameters: radius r, concentration Φ_c;
3: Initialize an empty maxtrix M_k storing the coordinates of each particle;
4: *while* (the concentration Φ_c is not reached) *do*
5: Genetate a random particle spatial coordinate (x,y,z);
6: *if* (New particle coordinates interfere with model boundaries) *do*
7: Go back to step 5 to continue; *end if*
8: *for* i= 1: (the number of elements in the maxtrix M_k) N *do*
9: *if* (New coordinates interfere with other generated paticles) *do*
10: Go back to step 5 to continue; *end if*
11: *end for*
12: Store the new generated coordinate in matrix M_k;
13: *end while*
14: Pass the 3Dmodel into Finit element calculation;
15: Cut to obtain three cross-sectional 2D images at random positions;

A representative cube model with 2 μm side length has been selected. The radius of the incorporated spherical particles is uniformly set as 100 nm, while the filling volume fraction varies from 5 vol.% to 30 vol.%. The particles are randomly generated and gradually added into the model, ensuring that every sample has a random and different internal structural configuration. For each generated 3D spherical particles incorporated composites, the relative permittivity is calculated as the label of the ConvNet model samples by FEM, as depicted in Section 1.2. Meanwhile, three 2D cross-sectional images are obtained as the features of the ConvNet model samples by truncate with three vertical planes at random positions, as illustrated in Figure 2. As a result, 14266 samples are generated as training datasets, while 3514 samples are generated as testing datasets.

1.2 Finite element methods

The description of the behaviors of dielectric phenomenon for space-dependent and time-dependent problems are usually can be expressed in terms of partial differential equations (PDEs). For some regular, simple geometries and problems, these PDEs can be solved with analytical methods, otherwise discretization in space or time is necessary, converting complex structures into many simple structural units, and then approximate solutions for PDEs can be calculated by numerical solutions. The finite element method (FEM) is a popular technique for numerically solving differential equations in engineering and mathematics. In the process of finite element analysis, some mesoscopic parameters giving rise to the dielectric phenomena can directly compute, such as the distribution of electric field, electric potential, current density and so on. It can conveniently contribute to establishing the relationship between the structure and properties of composite materials, and then realizing the numerical prediction of the corresponding dielectric parameters.

For the physical environment of electrostatic energy storage, the electric potential φ satisfies the Laplace's equation, that is

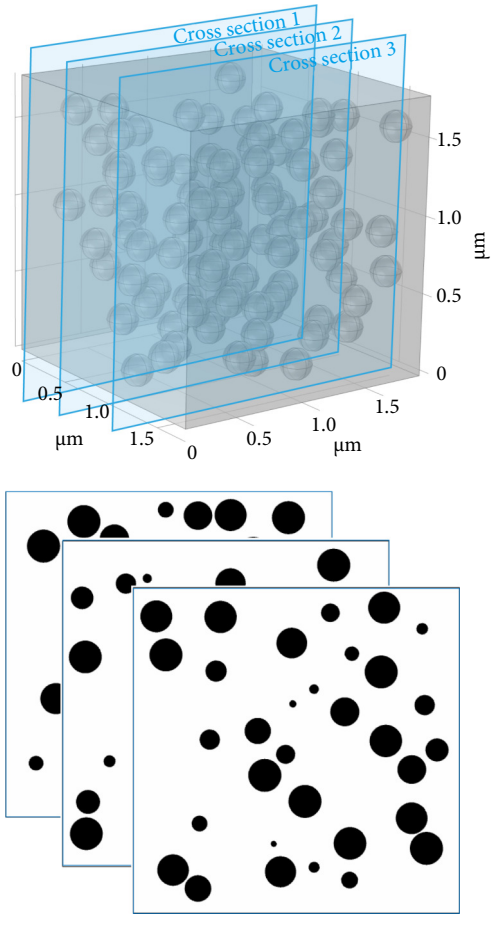

Fig. 2 Images for ConvNet network training.

$$\nabla \cdot (-\varepsilon \nabla \varphi) = 0 \tag{2}$$

where ε is the permittivity of material. We can then obtain the vectors electric field $\boldsymbol{E}$ and electric displacement $\boldsymbol{D}$ through the following equations,

$$\boldsymbol{E} = -\nabla \varphi \tag{3}$$

$$\boldsymbol{D} = \varepsilon_r \varepsilon_0 \boldsymbol{E} \tag{4}$$

where ε_0 is the permittivity of vacuum, andε_r is the relative permittivity of ingredient in the composite material. In terms of Maxwell equations, the total electrostatic energy W storage in the electric field can be obtained by volume integration as

$$W = \iiint \frac{1}{2} \boldsymbol{E} \cdot \boldsymbol{D} \mathrm{d}V \tag{5}$$

The relative permittivity ε_{eff} of the composites can then be calculated by means of definition of capacitive energy storage, that is

$$\varepsilon_{\text{eff}} = \frac{2Wd}{\varepsilon_0 S U^2} \tag{6}$$

where U is the magnitude of applied voltage, S and d are the surface area and the thickness of the material, respectively. In this work for the spherical particle filled BT/PVDF composites, the applied voltage is 2 μV, while the relative permittivity of BT and PVDF is defined to 1700[34] and 10[9], respectively. To improve the solution accuracy, the mesh quality at the surface of particles has been refined. The grid structure and the local distribution of electric potential within the composite material are illustrated in Figure 3.

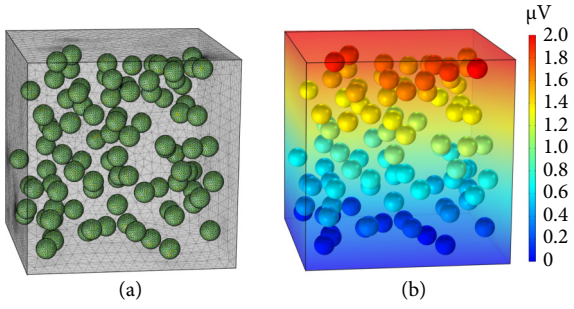

Fig. 3 The FEM to predict the relative permittivity of BT/PVDF composite. **(a)** Mesh configuration and **(b)** electric potential distribution within the material.

1.3 Convolutional neural network

1.3.1 Homemade ConvNet model

A convolutional neural network is a class of artificial neural network (ANN), most commonly applied to analyze visual imagery. It has achieved superior results across a wide range of application domains related to the crystal structure, SEM/TEM images and numerical simulation geometry configuration of nanocomposites. Generally, the ConvNet consists of the following parts: convolutional layer (Conv), pooling layers, fully connected layers (FSLs), and activation functions[35]. The convolution passes the input images through a set of convolutional filters, each of which activates certain features form the images and the following pooling layer is responsible for reducing the spatial size of convolved feature, and thus helps reduce overfitting, extract representative features from the input tensor. For regression or classification analysis, a fully connected layer, that is a simple feed forward neural networks, is often connects after the final pooling or convolutional layer, to obtain the regression or classification labels respectively. The activation function enhances the nonlinear characteristics of the network, allowing for faster and more effective training by mapping negative values to zero and maintaining positive values.

Our convolutional neural network model employs two-dimensional geometric configuration images for predicting the relative permittivity of BT/PVDF nanocomposites. The input of the ConvNet is a 512 × 512 fixed-sized pixel two-dimensional grayscale image. The structure of the ConvNet model consists of three convolutional composite layers and three fully connected layers, as illustrated in Figure 4. Each individual convolutional composite layer also consists of sequentially connected convolutional layer, batch normalization, activation function, and max pooling layer, and the output of the last convolutional composite layer is flattened into a one-dimensional vector for subsequent fully connection layers. Both the convolutional layers and the fully connected layers use ReLU as the activation, and the output of the network is a continuous numeric regression value representing the predicted relative permittivity. A flexible and ease-of-use open-source deep learning framework PyTorch is selected to build and train the model. It combines the efficient GPU-accelerated backend libraries from Torch with the compatibility with the popular Python high-level programming language, and has become a favorite tool for machine learning developers and data scientists. Adam optimizer and MSE (mean squared error) loss function are selected for training the convolutional neural network.

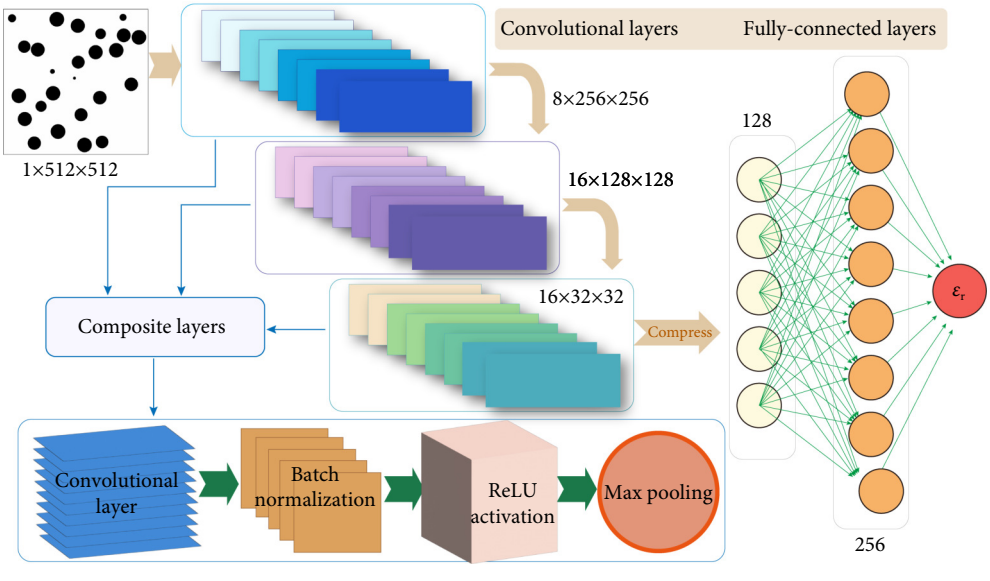

Fig. 4 Illustration of the ConvNet model used in the present study.

Adam optimization is a stochastic gradient descent method that is based on adaptive estimation of first-order and second-order moments.

1.3.2 Transfer learning ConvNet model

Transfer learning is reuse of a pre-trained model on a new problem. It's currently very popular in deep learning because it can train deep neural networks with comparatively little data and enable us to utilize knowledge from previously learned tasks[36]. This is very useful in the scientific world, because many problems existing in different fields are based on similar physical fundamental laws, governed by many partial differential equations[37]. ResNets or Residual Networks are a type of convolutional neural network (ConvNet) architecture introduced by Kaiming He[38] in his paper "Deep residual learning for image recognition" in 2015. This architecture shows a way to train networks with as many as 1000 layers. It can successfully reduce the effect of banishing gradient problem and obtain higher accuracy in network performance especially in image classification. There are many variants of ResNet architecture possessing same concept but with a different

number of layers, among witch, ResNet-50 has been used to predict the relative permittivity of BT/PVDF nanocomposites in the work. The batch size and initial learning rate of the model are provided as 64 and 0.01, respectively.

1.4 Evaluation metrics

To compare the performance of the ConvNet models to predict the relative permittivity of BT/PVDF nanocomposites, there are three metrics in regression have been adopted, that is mean absolute error (MAE), root mean square error (RMSE), and R square (R^2). They are defined[39] as

$$\mathrm{MAE}(y,\hat{y}) = \frac{1}{N}\sum_{i=1}^{N}|y_i - \hat{y}_i| \tag{7}$$

$$\mathrm{RMSE}(y,\hat{y}) = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\|y_i - \hat{y}_i\|_2^2} \tag{8}$$

$$R^2(y,\hat{y}) = 1 - \frac{\mathrm{RSS}}{\mathrm{TSS}} = 1 - \frac{\sum_{i=1}^{N}(y_i - \hat{y}_i)^2}{\sum_{i=1}^{N}(y_i - \bar{y})^2} \tag{9}$$

where $\hat{y}$ represents the predicted values from the ConvNet model, y represents the true values in the FEM dataset, and $\bar{y}$ is the mean value of y for total samples. MAE and RMSE evaluate the average closeness of the prediction result and the real data in the dataset, and the smaller the value, the better the prediction effect. R^2 measures the fraction of the variance in the data that can be explained by the model, which value is between 0 and 1 and a bigger value indicates a better fit between prediction and actual value.

2 Results and discussion

The loss variation of homemade ConvNet model with epoch for training and testing data is indicated as Figure 5, and a total of 100 epochs have been executed for both the training and testing data. The batch size is 64 and the initial leaning rate is set as 0.0001. All the input pictures have been randomly shuffled before training to increase the generality of the model.

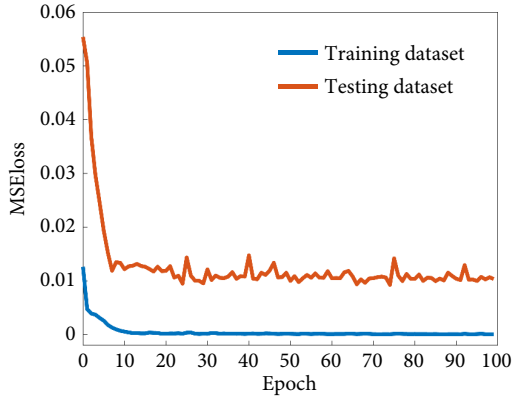

Fig. 5 Loss variation of homemade ConvNet model with epoch for training and testing data.

The MSEloss in training data sharply decreased up to 12 epoch and then gradually diminished to an extremely small value of 0.00001, while the MSEloss in testing data arrives at the stable value of 0.01 until approximately 23 epoch and then fluctuated slightly around a stable level. The performance of the homemade ConvNet model for prediction of the relative permittivity of BT/PVDF nanocomposites is illustrated in Figure 6.

The prediction results of the relative permittivity through ConvNet model are evaluated with respect to the corresponding calculation results by FEM, and the smaller the discrepancy between the two values, the closer the point distribution to the line $y = x$ as in Figure 6. At training dataset, the homemade ConvNet model manifesting an excellent prediction performance, all points are tightly distributed within a very narrow range around the line $y = x$. The standard deviation of relative difference of the prediction relative permittivity of BT/PVDF nanocomposites between homemade ConvNet model and FEM is as small as 0.81% as indicated in Figure 6(b) and the mean value is about zero errors. When the filling concentration of BT particles in the BT/PVDF nanocomposites are 5 vol.%, 10 vol.%, 15 vol.%, 20 vol.%, and 25 vol.%, the relative permittivity predicted by homemade ConvNet model are 11.66, 13.40, 15.67, 18.21, 21.23, which is rather in agreement with values calculated by FEM, that is 11.68, 13.42, 15.60, 18.22, 21.28, respectively. The evaluation metrics of the homemade ConvNet model to predict the relative permittivity of BT/PVDF in training dataset are also illustrated in Figure 6(a). The corresponding MAE, RMSE and R^2 in training data are 0.1064, 0.1396 and 0.9983, respectively. The performance of the homemade ConvNet model on the testing dataset shows somewhat degradation compared to that of training dataset, nonetheless, the prediction results are still acceptable. The relative errors of ConvNet and FEM satisfy the normal distribution as indicated in Figure 6(d), and most of the relative errors are within the range of 12.5%. The prediction accuracy at low volume fraction is significantly better than that at high volume fraction as indicated in Figure 6(c). Take the relative permittivity of BT/PVDF with 5 vol.%, 10 vol.%, 15 vol.%, 20 vol.%, and 25 vol.% concentration as example, the prediction permittivity derived for homemade ConvNet model is 12.488, 13.944, 15.901, 18.001, 20.381, while the related value result from FEM are 11.603, 13.437, 15.589, 18.173, 21.234. The corresponding MAE, RMSE and R^2 in testing data are 1.0435, 1.3842 and 0.8682, respectively.

A transfer learning suing a Pre-trained ResNet-50 model has also been implemented and the prediction performance on the training and testing datasets is indicated in Figure 7. Compared with homemade ConvNet architecture, the prediction accuracy of the ResNet-50 model is slightly weakened, and the MAE and RMSE evaluation metrics have increased to 0.3316 and 0.5063, as shown in Figure 7. Nevertheless, the prediction results of ResNet-50 model are still in good agreement with the calculation results of FEM by virtue of the small relative error in the range of ±10% and high R^2 value as 0.9783. What's more, the distribution the relative error curve is narrower with respect to that of the homemade ConvNet model as illustrated in Figure 7(b), indicating that a larger proportion of the data possess extremely good prediction accuracy with ResNet-50 model. The performance of ResNet-50 model to predict the relative permittivity of BT/PVDF nanocomposites on the testing data has been significantly improved, especially at high volume fraction of particles as illustrated in Figure 7(c). The relative errors between the prediction values arise from ResNet-50 model and the calculation values by FEM are limited in the range of ±10% and the standard deviation is 5.25%. Furthermore, both the MAE and RMSE evaluation metrics have reduced to less than 1, which is 0.7345 and 0.9529, respectively, as shown in Figure 7(c). The R^2 error reaches as high as 0.9375, demonstrating that convolutional neural network is a feasible and efficient surrogate of finite-element method to prediction the dielectric permittivity of BT/PVDF composites.

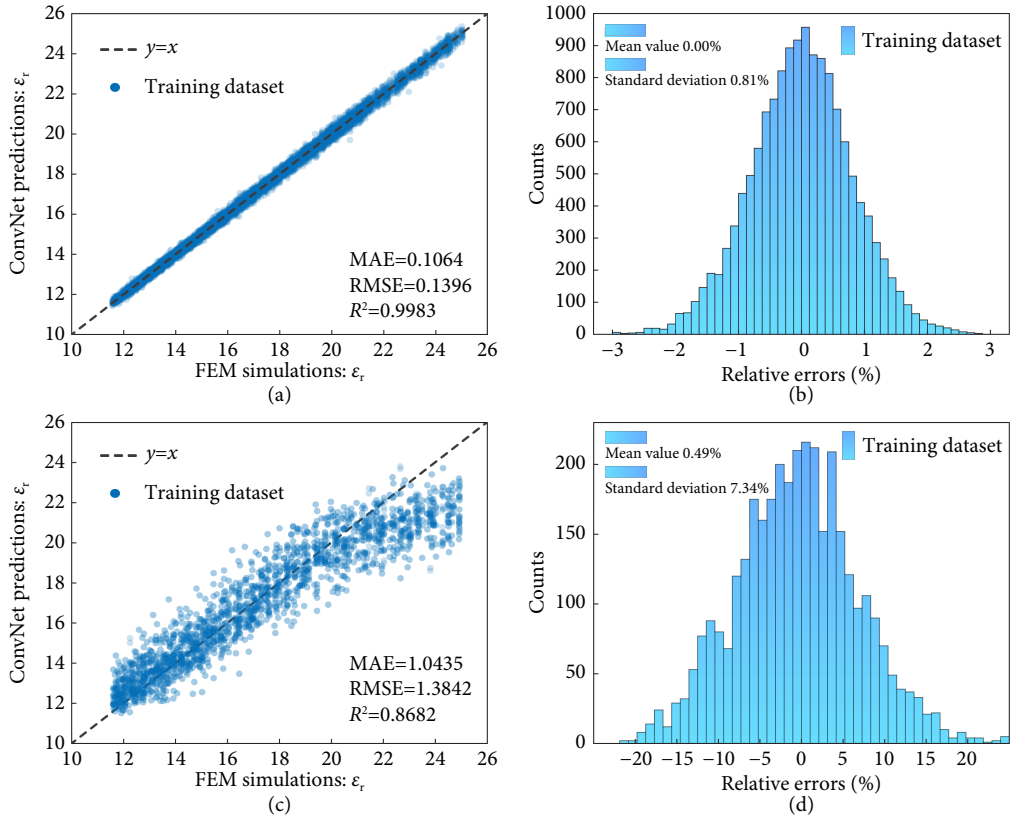

Fig. 6 The performance of homemade ConvNet model for prediction of the relative permittivity of BT/PVDF nanocomposites on the (a) training data and (c) testing data and the corresponding relative errors distribution on the (b) training data and (d) testing data.

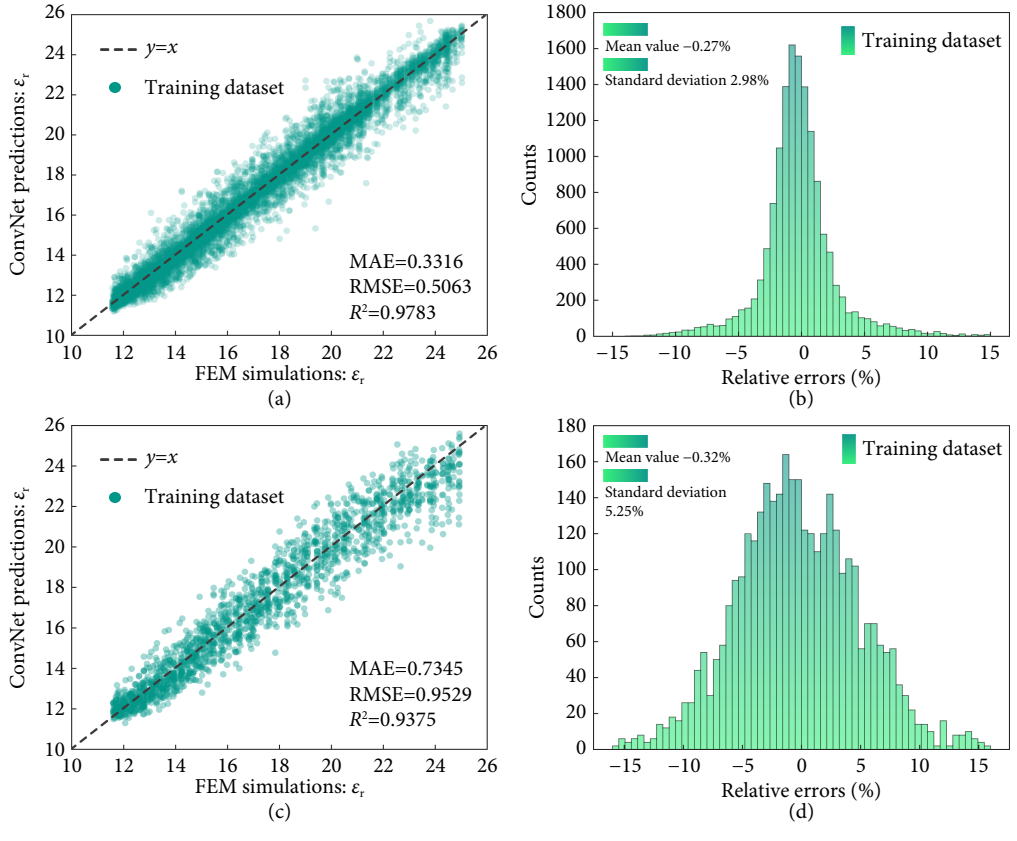

Fig. 7 The performance of Resnet-50 model for prediction of the relative permittivity of BT/PVDF nanocomposites on the (a) training data and (c) testing data and the corresponding relative errors distribution on the (b) training data and (d) testing data.

3 Conclusions

In this work, a generic workflow combining the convolutional neural network and finite element method to predict the relative permittivity of BT/PVDF nanocomposites has been presented. An algorithm for random generation of material microstructure has been proposed, generating a sufficiently large amount of data samples for ConvNet model training. As a result, 14266 samples and 3514 samples were provided for model training and testing, respectively, and each sample constitute of a 2D cross-section picture of the composite as data feature and the corresponding relative permittivity of the composites calculated by FEM as data label. Both a homemade and a transfer learning convolutional neural network architecture have been implemented, and the former includes three convolutional composite layers and followed three fully-connected layers, and each convolutional composite layer compose of convolutional layer, bath normalization, ReLU activation and max pooling, while the latter are a pre-trained Resnet-50 model. As a result, the homemade ConvNet model shows excellent prediction performance on the training data with $R^2 = 0.9983$, and a little weaken on the testing data with $R^2 = 0.8682$. However, the performance on testing data was significantly improved with Resnet-50 architecture and the R^2 evaluation metric reaches 0.9375, while good prediction effect on the training data was maintained with $R^2 = 0.9783$. The proposed workflow and convolutional neural network architectures are not restricted to predict the relative permittivity of particles/polymer composites, but also applicable to prediction of other characteristic and further of structure-property mechanism.

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Additional information

Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

References

[1] Feng, Q. K., Zhong, S. L., Pei, J. Y., Zhao, Y., Zhang, D. L., Liu, D. F., Zhang, Y. X., Dang, Z. M. (2022). Recent progress and future prospects on all-organic polymer dielectrics for energy storage capacitors. *Chemical Reviews*, 122: 3820–3878.

[2] Zhong, S. L., Dang, Z. M., Zhou, W. Y., Cai, H. W. (2018). Past and future on nanodielectrics. *IET Nanodielectrics, 1*: 41–47.

[3] Yin, L. J., Zhao, Y., Zhu, J., Yang, M. H., Zhao, H. C., Pei, J. Y., Zhong, S. L., Dang, Z. M. (2021). Soft, tough, and fast polyacrylate dielectric elastomer for non-magnetic motor. *Nature Communications*, 12: 4517.

[4] Dong, W. Z., Zhao, Y., Yin, L. J., Dang, Z. M. (2022). Fabrication and actuation characterisation of a new UV curing acrylic dielectric elastomer. *IET Nanodielectrics*, 5: 104–111.

[5] Kim, Y. J., Lee, J., Park, S., Park, C., Park, C., Choi, H. J. (2017). Effect of the relative permittivity of oxides on the performance of triboelectric nanogenerators. *RSC Advances*, 7: 49368–49373.

[6] Yildirim, A., Grant, J. C., Es-haghi, S. S., Lee, W., Maruthamuthu, M. K., Verma, M., Sutherland, J. W., Cakmak, M. (2020). Roll-to-roll (R2R) production of large-area high-performance piezoelectric films based on vertically aligned nanocolumn forests. *Advanced Materials Technologies*, 5: 2000553.

[7] Fan, Y. J., Shen, Z. H., Zhou, X. C., Dan, Z. K., Zhou, L., Ren, W. B., Tang, T. X., Bao, S. Y., Nan, C. W., Shen, Y. (2022). Highly sensitive strain sensor from topological-structure modulated dielectric elastic nanocomposites. *Advanced Materials Technologies*, 7: 2101190.

[8] Grill, A., Gates, S. M., Ryan, T. E., Nguyen, S. V., Priyadarshini, D. (2014). Progress in the development and understanding of advanced low *k* and ultralow *k* dielectrics for very large-scale integrated interconnects—State of the art. *Applied Physics Reviews*, 1: 011306.

[9] Cheng, R., Wang, Y. F., Men, R. J., Lei, Z. P., Song, J. C., Li, Y. Y., Guo, M. Q. (2022). High-energy-density polymer dielectrics via compositional and structural tailoring for electrical energy storage. *iScience*, 25: 104837.

[10] Ji, M., Min, D., Wu, Q., Mi, R., Liu, W., Li, S., Qin, S., Zhu, S. (2022). Significantly improved energy storage density of polypropylene nanocomposites via macroscopic and mesoscopic structure designs. *Frontiers in Electronic Materials*, 2: 904405.

[11] Wang, S., Li, Q. (2018). Design, synthesis and processing of PVDF-based dielectric polymers. *IET Nanodielectrics*, 1: 80–91.

[12] Sun, W. D., Lu, X. J., Jiang, J. Y., Zhang, X., Hu, P. H., Li, M., Lin, Y. H., Nan, C. W., Shen, Y. (2017). Dielectric and energy storage performances of polyimide/$BaTiO_3$ nanocomposites at elevated temperatures. *Journal of Applied Physics*, 121: 244101.

[13] Hao, Y. N., Wang, X. H., Bi, K., Zhang, J. M., Huang, Y. H., Wu, L. W., Zhao, P. Y., Xu, K., Lei, M., Li, L. T. (2017). Significantly enhanced energy storage performance promoted by ultimate sized ferroelectric $BaTiO_3$ fillers in nanocomposite films. *Nano Energy*, 31: 49–56.

[14] Yu, K., Wang, H., Zhou, Y. C., Bai, Y. Y., Niu, Y. J. (2013). Enhanced dielectric properties of $BaTiO_3$/poly(vinylidene fluoride) nanocomposites for energy storage applications. *Journal of Applied Physics*, 113: 034105.

[15] Li, Q., Chen, L., Gadinski, M. R., Zhang, S. H., Zhang, G. Z., Li, H. U., Iagodkine, E., Haque, A. M., Chen, L. Q., Jackson, T. N., et al. (2015). Flexible high-temperature dielectric materials from polymer nanocomposites. *Nature*, 523: 576–579.

[16] Zhou, Y., Hu, J., Dang, B., He, J. L. (2017). Effect of different nanoparticles on tuning electrical properties of polypropylene nanocomposites. *IEEE Transactions on Dielectrics and Electrical Insulation*, 24: 1380–1389.

[17] Zakri, T., Laurent, J. P., Vauclin, M. (1998). Theoretical evidence for `Lichtenecker' s mixture formulae' based on the effective medium theory. *Journal of Physics D: Applied Physics*, 31: 1589–1594.

[18] Sihvola, A. (2000). Mixing rules with complex dielectric coefficients. *Subsurface Sensing Technologies and Applications*, 1: 393–415.

[19] Wang, J. C., Shen, Z. H., Jiang, J. Y., Wang, J., Zhang, X., Shen, J., Shen, Y., Chen, W., Chen, L. Q., Nan, C. W. (2022) High-throughput finite-element design of dielectric composites for high-frequency copper clad laminates. *Composites Science and Technology*, 225: 109517.

[20] Zhong, S. L., Dang, Z. M., Zha, J. W. (2018). Prediction on effective permittivity of 0–3 connectivity particle/polymer composites at low concentration with finite element method. *IEEE Transactions on Dielectrics and Electrical Insulation*, 25: 2122–2128.

[21] Cai, Z. M., Wang, X. H., Luo, B. C., Hong, W., Wu, L. W., Li, L. T. (2017). Nanocomposites with enhanced dielectric permittivity and

breakdown strength by microstructure design of nanofillers. *Composites Science and Technology*, 151: 109–114.
[22] Cheng, Y. H., Chen, X. L., Wu, K., Wu, S. N., Chen, Y., Meng, Y. M. (2008). Modeling and simulation for effective permittivity of two-phase disordered composites. *Journal of Applied Physics*, 103: 034111.
[23] Wang, Z. P., Nelson, J. K., Hillborg, H., Zhao, S., Schadler, L. S. (2013). Dielectric constant and breakdown strength of polymer composites with high aspect ratio fillers studied by finite element models. *Composites Science and Technology*, 76: 29–36.
[24] Chen, C. T., Gu, G. X. (2019). Machine learning for composite materials. *MRS Communications*, 9: 556–566.
[25] Mannodi-Kanakkithodi, A., Pilania, G., Huan, T. D., Lookman, T., Ramprasad, R. (2016). Machine learning strategy for accelerated design of polymer dielectrics. *Scientific Reports*, 6: 20952.
[26] Shen, Z. H., Liu, H. X., Shen, Y., Hu, J. M., Chen, L. Q., Nan, C. W. (2022). Machine learning in energy storage materials. *Interdisciplinary Materials*, 1: 175–195.
[27] Shen, Z. H., Bao, Z. W., Cheng, X. X., Li, B. W., Liu, H. X., Shen, Y., Chen, L. Q., Li, X. G., Nan, C. W. (2021). Designing polymer nanocomposites with high energy density using machine learning. *Npj Computational Materials*, 7: 110.
[28] Lin, Y., Guan, Z. D. (2021). The use of machine learning for the prediction of the uniformity of the degree of cure of a composite in an autoclave. *Aerospace*, 8: 130.
[29] Choudhary, K., DeCost, B., Chen, C., Jain, A., Tavazza, F., Cohn, R., Park, C. W., Choudhary, A., Agrawal, A., Billinge, S. J. L., et al. (2022). Recent advances and applications of deep learning methods in materials science. *Npj Computational Materials*, 8: 59.
[30] Ejaz, F., Hwang, L. K., Son, J., Kim, J. S., Lee, D. S., Kwon, B. (2022). Convolutional neural networks for approximating electrical and thermal conductivities of Cu-CNT composites. *Scientific Reports*, 12: 13614.
[31] Bhutada, A., Kumar, S., Gunasegaram, D., Alankar, A. (2021). Machine learning based methods for obtaining correlations between microstructures and thermal stresses. *Metals*, 11: 1167.
[32] Abueidda, D. W., Almasri, M., Ammourah, R., Ravaioli, U., Jasiuk, I. M., Sobh, N. A. (2019). Prediction and optimization of mechanical properties of composites using convolutional neural networks. *Composite Structures*, 227: 111264.
[33] Ye, S., Li, B., Li, Q. Y., Zhao, H. P., Feng, X. Q. (2019). Deep neural network method for predicting the mechanical properties of composites. *Applied Physics Letters*, 115: 161901.
[34] Barber, P., Balasubramanian, S., Anguchamy, Y., Gong, S. S., Wibowo, A., Gao, H. S., Ploehn, H., Zur Loye, H. C. (2009). Polymer composite and nanocomposite dielectric materials for pulse power energy storage. *Materials*, 2: 1697–1733.
[35] Rong, Q. Y., Wei, H., Huang, X. Y., Bao, H. (2019). Predicting the effective thermal conductivity of composites from cross sections images using deep learning methods. *Composites Science and Technology*, 184: 107861.
[36] Li, X. L., Zhang, Y. C., Zhao, H., Burkhart, C., Brinson, L. C., Chen, W. (2018). A transfer learning approach for microstructure reconstruction and structure-property predictions. *Scientific Reports*, 8: 13461.
[37] Xu, Y. J., Weng, H. B., Ju, X. Z., Ruan, H. S., Chen, J. J., Nan, C. Y., Guo, J., Liang, L. H. (2021). A method for predicting mechanical properties of composite microstructure with reduced dataset based on transfer learning. *Composite Structures*, 275: 114444.
[38] He, K. M., Zhang, X. Y., Ren, S. Q., Sun, J. (2015). Deep residual learning for image recognition. *arXiv preprint*: 1512.03385v1.
[39] Wang, W. H., Wang, H., Zhou, J. N., Fan, H. L., Liu, X. (2021). Machine learning prediction of mechanical properties of braided-textile reinforced tubular structures. *Materials & Design*, 212: 110181.