Transfer learning for materials informatics using crystal graph convolutional neural network

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ABSTRACT

For useful applications of machine learnings in materials informatics, one challenge is to overcome inaccuracy of predictions ascribing to insufficient data. We propose a transfer learning using a crystal graph convolutional neural network (TL-CGCNN); here TL-CGCNN is pretrained with big data such as formation energies for crystal structures, and used for predictions of target properties with relatively small data. We confirm that TL-CGCNN can improve predictions for various properties such as bulk moduli, dielectric constants, and quasiparticle band gaps, which are computationally demanding to construct big data for materials. We also quantitatively observe that the more TL-CGCNN pretrains, the more accurate the prediction for the target properties by TL-CGCNN becomes. Finally, we confirm the TL-CGCNN is superior to other regression methods in the performance for the predictions with small data and conclude that TL-CGCNN is promising along with compiling big data for materials which are easy to be accumulated and relevant to the target properties.

Keywords:
transfer learning, crystal graph convolutional neural network, prediction, regression, materials informatics, TL-CGCNN
INTRODUCTION

The usage of machine learning (ML) has been widely applied to materials informatics (MI) field, in particular, for new materials exploration to save time and human efforts. One important challenge for the ML-based material exploration and design is to develop a generic descriptor (representation) that can be flexibly applied to predictions of multiple target properties. Because a material property depends on a crystal structure and constituent chemical elements, the descriptors have been developed based on the structural features such as Coulomb matrix, smooth overlap of atomic positions (SOAP), and reciprocal 3D voxel space (R3DVS) descriptors.

Recently, Xie and Grossman introduced a crystal graph convolutional neural network (CGCNN). As an input for a classification or regression model by CGCNN, only a crystal structure is necessary. The CGCNN constructs crystal graphs from crystal structures and predicts the target property by a deep neural network architecture. Owing to its simplicity and flexibility, many researchers interested in graph-based neural networks have been developed for its improvement or modification. Park and Wolverton incorporated information of the Voronoi tessellated crystal structure into CGCNN to encode three body correlations. Sanyal et al., augmented CGCNN with a multitask learning to predict multiple material properties spontaneously. Chen et al., developed a graph network named MatErials Graph Network (MEGNet) that is extended to treat not only a crystal structure but also a molecule with additional attributes.

Another important challenge for the ML-based materials exploration and design is to overcome unsatisfactory accuracy of predictive models ascribing to insufficient data. Differently from other research fields such as computer vision and natural language processing which are using big data with more than millions of data, many functional or practical properties in MI have not been accumulated much as they are computationally or experimentally demanding for evaluation. A transfer learning is a useful method to help improving predictions suffering from insufficient data.

The transfer learning is to bring knowledge to other predictive models with different tasks (target variables). We focus on applications of transfer learning for a target model with less number of data by loading a pretrained model constructed with enormous number of data. In MI, there have been studies of using the transfer learning. For classifications of Li superionic conductors, Cubuk et al., constructed a large number of labeled data based on crystal structure-based descriptors, and then reconstructed predictive
models based on chemical element-based descriptors by a support vector machine. Yamada et al., developed a pretrained model library called XenonPy.MDL with big data for a multilayer neural network based on fingerprint descriptors, and showed improved prediction of properties of organic or inorganic materials by the transfer learning using the neural network. Jha et al. showed an improved predictions for formation energies of smaller theoretical or experimental database by the transfer learning with a multilayer deep neural network named ElemNet, which uses elemental composition descriptors, and pretrained models for a formation energy with larger theoretical database.

Indeed, in computer vision field, the transfer learning have been more widely used as deep convolutional neural network becomes the mainstream. The convolutional layers work as feature maps that can detect basic features such as vertical and horizontal lines, which are basic elements for recognizing objects. On the basis of the inspirations, CGCNN, because of its neural network architecture, may have potential for effective transfer learning in MI.

In this paper, we investigate the transfer learning using CGCNN (hereinafter called TL-CGCNN). We quantitatively confirm that the predictions for basic materials properties such as a Kohn-Sham band gap ($E_g$) and a formation energy ($\Delta E_f$) can be improved by TL-CGCNN. To confirm a flexible application of the transfer learning, we also investigate whether TL-CGCNN can be useful for improving predictions for properties such as a bulk modulus ($K_{VRIH}$), a dielectric constant ($\varepsilon_r$), and a quasiparticle band gap (GW-$E_g$), which usually suffer from small number of data and heavy computational costs. In addition, we discuss additional issues relevant to TL-CGCNN and its prospects.

RESULTS AND DISCUSSION

CGCNN

First, we reviews background knowledge about CGCNN constructed by Xie and Grossman. Figure 1 shows an overview of CGCNN. The CGCNN consists of a construction of graphs based on crystal structures and a deep neural network architecture including embedding, convolutional, pooling, and fully-connected (FC) layers.
A crystal structure is transformed to a multigraph $G$. In the multigraph $G$, nodes and edges indicate atoms and chemical bonds between the atoms, respectively. The multigraph $G$ consists of set of atoms in the crystal structure, undirected edges, atom features, and bond features. A bond feature vector $(u_{i,j,k})$ denotes the $k$-th bond between atoms $i$ and $j$. An atom feature vector $(v_i)$ contains the properties of $i$-th atom. The information of atoms and bonds in the multigraph $G$ is prepared as discrete representations consist of binary digits for describing constituent elements and bonds such as group and periodic number of atom, and atom distance. The discrete representations are transformed to continuous representations in the embedding layer. Then, the continuous representations are inserted to the convolutional layer.

In the $(t+1)$-th convolutional layer,

$$v_i^{(t+1)} = g \left[ \left( \sum_{j,k} v_j^{(t)} \oplus u_{(i,j),k} \right) W_c^{(t)} + v_i^{(t)} W_s^{(t)} + b^{(t)} \right]$$

(1)

where $\oplus$ denotes the concatenation of atom and bond feature vectors of the neighboring atoms of $i$-th atom. $W_c^{(t)}$, $W_s^{(t)}$, and $b^{(t)}$ indicate optimized parameters in the neural network, namely, convolution weight matrix, self weight matrix, and bias of the $t$-th layer, respectively. $g(\cdot)$ denote a non-linear activation function between layers such as a softplus function. However, because the weight matrix is shared by all neighbors in this convolution formulation, different interaction of neighbors is not distinguished. Therefore, a standard edge-gating technique can be applied for a new type of convolutional formulation as follows. A neighbor feature vector $z_{(i,j),k}$ is used as,

$$z_{(i,j),k}^{(t)} = v_i^{(t)} \oplus v_j^{(t)} \oplus u_{(i,j),k}$$

(2)

and then,

$$v_i^{(t+1)} = v_i^{(t)} + \sum_{j,k} \sigma(z_{(i,j),k}^{(t)} W_c^{(t)} + b^{(t)}) \circ g(z_{(i,j),k}^{(t)} W_s^{(t)} + b_s^{(t)})$$

(3)

where $\circ$ denotes element-wise multiplication (Hadamard product) and $\sigma$ denotes a sigmoid function. Xie and Grossman reported that the latter convolution formalism (equation 3) showed better predictions than the former formalism (equation 1).

The atom feature vectors after the convolutional layers are inserted into the pooling layer. Here, an average pooling is used. After the pooling layer, a crystal feature vector $(v_g)$ can be obtained by,

$$v_g = \frac{1}{N} \sum_i v_i$$

(4)

where $N$ is the number of atoms in the crystal graph.
Finally, $v_g$ is used as input to a network of FC layers with non-linearities which learn to predict a target property value, $y$, for the crystal. Predicted values can be obtained as follows,

$$\hat{y} = f(v_gW_g + b_g)$$  \hspace{1cm} (5)

where $W_g$, $b_g$, and $f(\cdot)$ are weight matrix, bias, and non-linear function of FC layers, respectively.

The parameters such as $W$ and $b$ are optimized to minimize the loss function, which is defined as mean absolute difference of predicted and target values for regressions, by a backpropagation procedure. As an optimizer, stochastic gradient descent is employed.\(^{40}\)

**TL-CGCNN**

Figure 2 shows a comparison of the concept of a conventional machine learning and a transfer learning based on a convolutional neural network. In the case of using a conventional machine learning, when we have different target variables, we prepare each predictive model separately. However, when we construct a "pretrained" (source) predictive model using a big data based on the convolutional neural networks in advance, enormous number of parameters (weight and bias) in the convolutional layer can save the features of the big data. The parameters in the convolutional layer can be transferred to the target predictive model, which have usually smaller number of training data, as "knowledge".

There are two common ways of the transfer learning to treat the loaded parameters for the convolutional neural networks. The first method is a "fine tuning". The parameters of the whole layers of the pretrained model are initially loaded. Then, all of the parameters are optimized by the backpropagation. This method is expected to have better optimization than the case of starting the optimization with randomly generated initial parameters. It is recommended to select adequate learning rate not to lose the information of the pretrained model. The second method is a "layer freezing". This method fixes the parameters of the earlier layers without additional optimizations by the backpropagation. The optimization of parameters is only performed for the latter layers to save the information from the pretrained model. In this study, we mainly used the fine tuning for TL-CGCNN.

In addition, to match the scales of target properties between pretrained and target models in this study, tanh-normalization\(^{41}\) was used for the predictive models as follows,
\[ y' = \frac{1}{2} \left\{ \tanh \left( 0.01(y - \mu) \right) \right\} + 1 \]  

(6)

where \( \tanh \), \( \mu \) and \( s \) are hypertangent function, mean and standard deviation of target variables, \( y \), respectively. After the predictive model is optimized, it was denormalized when the prediction error is summarized.

TL-CGCNN based on CGCNN

Before describing the results of predictions, we note that the names of pretrained and target models are written based on the number of training data and target property in this study. For example, 500-NM-\( E_g \) indicates that the target property of the predictive model is \( E_g \), the number of training data is 500 (both numbers of validation and test data are 125), and only nonmetallic (NM) data is included in the data. Large numbers, namely, 10000, 54000, and 113000 are written as 10k, 54k, and 113k. When the number of training data is greater than 10k, the number of test data is fixed to be 2500, whereas when the number training data is less than 10k, the number of test data is set to its 25%.

Table 1 summarizes material-property dataset that used in this study. A mean absolute difference of the data indicates a baseline for a predictive model.

Figure 3 shows the dependence of prediction error of test data by CGCNN for \( E_g \) and \( \Delta E_f \) on number of training data. The prediction errors decrease with increasing number of training data. When the numbers of training data reach 54k and 113k, the prediction errors for \( E_g \) and \( \Delta E_f \) become 0.358 eV and 0.046 eV/atom, respectively. The prediction error of \( \Delta E_f \) is comparable to the accuracy of the density functional theory (DFT) method with respect to the experiments.29 We expect that the prediction error can further decrease when collecting more training data because the decreasing tendency is not saturated yet. For \( \Delta E_f \), predictive models with only NM materials data were separately constructed. When the same number of training data is used, the prediction error with only NM materials data is smaller than that with metallic and NM material data together.

However, as mentioned in Introduction, we usually suffer from unsatisfactory accuracy of predictive models ascribing to small number of data. Therefore, when we apply a TL-CGCNN to \( E_g \) and \( \Delta E_f \), we intentionally prepared target models with small number of data. The results are shown in Fig. 4, and some of
numerical values are also summarized in Supplementary Table 2. The pretrained models with large number
of data for prediction targets of counterpart properties were used.

When the number of training data is 500, the prediction error for $E_g$ by the CGCNN is 0.978 eV. By
using TL-CGCNN with the 10k-$\Delta E_f$, 54k-$\Delta E_f$, and 113k-$\Delta E_f$ pretrained models, the prediction error
decreases to 0.866, 0.826, and 0.815 eV, respectively. When the same number of training data is used, the
prediction error for $\Delta E_f$ by the CGCNN is 0.239 eV/atom. When we use TL-CGCNNs with 10k-NM-$E_g$ and
54k-NM-$E_g$ pretrained models, the prediction error decreases to 0.236, and 0.222 eV/atom, respectively. In
the both cases, the more training data we use for the pretrained models, the more the prediction powers
are improved.

We also investigated the influence of data type for the transfer learning. By using only NM data, the
predictive models for $\Delta E_f$ were constructed (NM-$\Delta E_f$). When the target model is 500-NM-$\Delta E_f$, the prediction
error by TL-CGCNN with the 54k-NM-$E_g$ pretrained model is 0.149 eV/atom, which is smaller than the case
(0.184 eV/atom) that without TL-CGCNN. In addition, when the target model is 500-NM-$E_g$, the prediction
error by TL-CGCNN with the 54k-NM-$\Delta E_f$ pretrained model is 0.819 eV smaller than that (0.826 eV) with
the 54k-$\Delta E_f$ pretrained model, which used the same number of training data. This implies that when the types
of participating materials in the target and pretrained models are similar to each other, TL-CGCNN for
CGCNN tends to be better.

Then, we tested the dependence of TL-CGCNN on the number of data of target models. With fixing
the 113k-$\Delta E_f$ pretrained model, when the target models are 500-$E_g$, 1000-$E_g$, 5000-$E_g$, and 10k-$E_g$, the
prediction errors decrease from 0.866, 0.752, 0.553 to 0.490 eV, respectively. The percentage improvements
of prediction error with respect to the case without transfer learning become 16.7, 12.0, 9.0 and 3.2%, which
tends to decrease. With fixing the 54k-NM-$E_g$ pretrained model, when the target models are 500-$\Delta E_f$, and
10k-$\Delta E_f$, the prediction errors are 0.222 and 0.089 eV/atom (7.2 and 4.6% improved with respect to
predictions without TL-CGCNN), respectively. This was also tested for the prediction of $\Delta E_f$ for NM data.
With fixing 54k-NM-$E_g$ pretrained model, when the target models are 500-NM-$\Delta E_f$, and 10k-NM-$\Delta E_f$, the
prediction errors are 0.149 and 0.065 eV/atom (19.2 and 2.8% improved with respect to predictions without
TL-CGCNN), respectively. This implies that using TL-CGCNN is more efficient when we have less data.
Figure 5 shows a scatter plot between two properties of $E_g$ and $\Delta E_f$ of 118286 materials. The scatter plot shows that there is not a strong correlation between the $E_g$ and $\Delta E_f$. The linear correlation coefficients ($r_p$) between the two properties for the total data and NM data are only $-0.49$ and $-0.33$, respectively. For 59923 NM materials, the mean absolute error (MAE) of $E_g$ and $\Delta E_f$ by an ordinary linear regression using $\Delta E_f$ and $E_g$ are 1.17 eV and 0.72 eV/atom, respectively. These prediction errors are significantly larger than those by TL-CGCNN performed with only 500 training data. Despite a weak correlation, we confirmed that TL-CGCNN for $E_g$ and $\Delta E_f$ with pretrained models based on the counterpart properties improved the predictions. This implies that atom and bond features from crystal graphs captured in the convolutional layers in the pretrained model with big data are flexibly adapted to predict various material properties.

TL-CGCNN for computationally demanding properties

We also tested TL-CGCNNs for predictive models for other properties, namely, $K_{\text{VRH}}$, $\varepsilon_r$, and GW-$E_g$. They are more computationally expensive, so that there are not so much data as $E_g$ and $\Delta E_f$. Note that the $K_{\text{VRH}}$ and $\varepsilon_r$ values in a log_{10} scale were used considering their broad distributions, and the unit of the $K_{\text{VRH}}$ value inside the log_{10} is GPa.

Figure 6 shows prediction errors of three properties without and with TL-CGCNN. For the prediction for 500-$K_{\text{VRH}}$, the prediction error without TL-CGCNN is 0.123. When the 113k-$\Delta E_f$ pretrained model is used, the prediction error decreases to 0.112 (by 8.7%). For the prediction for 500-NM-$\varepsilon_r$, the prediction error without TL-CGCNN is 0.181. It also decreases to 0.169 (by 7.1%) and 0.163 (by 10.0%) when the 113k-$\Delta E_f$ and 54k-NM-$E_g$ pretrained models are employed, respectively. Despite more training data of the 113k-$\Delta E_f$ pretrained model, the smaller prediction error of prediction based on the 54k-NM-$E_g$ pretrained model is obtained. This may ascribe to the correlation ($-0.48$) between $E_g$ and $\varepsilon_r$ (in log_{10} scale) much stronger than that (0.07) between $\Delta E_f$ and $\varepsilon_r$ (in log_{10} scale), for 4182 NM materials that have $\varepsilon_r$ data. For the prediction for 180-NM-GW-$E_g$, the prediction error without TL-CGCNN is 0.783 eV. The smallest prediction error by TL-CGCNN among various pretrained models is achieved by the 113k-$\Delta E_f$ pretrained model, whose value decreases to 0.591 eV by 24.5%. Therefore, these results suggest that using TL-CGCNN is also useful for such practical properties.
When more data are used for prediction for $K_{VRH}$ and $\varepsilon_r$ (8194 and 2788 training data for $K_{VRH}$ and $\varepsilon_r$, respectively) we obtain decreased prediction error by TL-CGCNN. However, the improvement by transfer learning tends to decrease compared to the case that the target model have less data. The percentage improvements by the transfer learning for $K_{VRH}$ and $\varepsilon_r$ are only 1.7 and 3.7%, respectively. As the case of TL-CGCNN for $E_g$ and $\Delta E_f$ shows, this implies that TL-CGCNN may be more useful for the predictions with small number of data.

Additional issues

We also tested predictions of 500-NM-$E_g$, 500-NM-$\Delta E_f$, 500-NM-$\Delta E_f$, 500-$K_{VRH}$, and 500-NM-$\varepsilon_r$ by partial least squares (PLS), least absolute shrinkage and selection operator (LASSO), support vector regression (SVR), and random forests (RF). These widely-used regression methods were supervised using a generalized set of chemical composition based descriptors. The results are shown in Figures 4 and 6. The numerical values are summarized in Supplementary Table S2.

The RF showed the best performance among the four regression methods. The CGCNN showed better performance on predictions of 500-$\Delta E_f$, 500-NM-$\Delta E_f$, and 500-$K_{VRH}$, but poorer performance on the predictions for 500-NM-$E_g$ and 500-NM-$\varepsilon_r$ than the RF. However, the TL-CGCNN could achieve the smallest prediction errors for the five predictions. The predictions with other widely-used regression methods like the RF can be improved with more specialized descriptors that have strong correlations with the target properties; however, finding additional useful descriptor for each target property is not easy. Therefore, we emphasize that the TL-CGCNN is a powerful and flexible prediction tool with merits; this tool is easy to construct pretrained models with big data, and this tool can systematically capture both features of chemical elements and crystal structures.

Deep neural networks like CGCNN have many hyperparameters. For a good transfer learning, we need to carefully select hyperparameters based on the following considerations; transferred information is to be conserved and flexibly used for the target predictive model, and overfitting is to be reduced. On the basis of the considerations, we optimized a learning rate and a length of vector of FC layer for a case study. The results are described in Supplementary chapter S1.
Until now in this study, the fine tuning has been used as the TL-CGCNN method. We also tested the layer freezing method. We only optimized the parameters of the last FC layer freezing those of the other earlier layers. As a result, for the investigated prediction targets, fine tuning showed much better predictions. The detailed results are described in Supplementary chapter S2. In some previous studies using deep convolutional neural networks with a lot of multiple layers,\textsuperscript{21,31} the concept of fine tuning and layer freezing are suggested to be mixed, namely, only early convolutional layers are frozen and the other layers including convolutional layers are all optimized.

In this study, TL-CGCNNs were applied for the predictive models of the various material properties, which suffer from inaccuracy ascribing to small number of data. We quantitatively confirm that the predictions can be improved by using TL-CGCNN with the pretrained model constructed with big data. Because we usually suffer from insufficient data in many MI issues, we expect that the usage of a combination of general descriptors such as CGCNN and transfer learning can be more widely employed.

**METHODOLOGY**

Data and evaluation of predictive model

Crystal structures, and their corresponding $E_g$ and $\Delta E_f$ (defined with respect to the most stable state of each elemental standard state) have been compiled from a first-principles calculations database, Materials Project database (MPD).\textsuperscript{46} They were calculated based on the exchange-correlation function of Generalized Gradient Approximation (GGA) parameterized by Perdew-Burke-Ernzerhof (PBE) form\textsuperscript{47} in the Vienna \emph{Ab-initio} Simulation Package (VASP).\textsuperscript{48,49} The on-site Coulomb interaction were treated by GGA+U in case the transition metals are included in materials.\textsuperscript{50} We used 118286 computed data (assessed on November 11th, 2019) that is available in MPD to construct crystal graphs without a program error within a cutoff radius of 8 Å. For these data, He, Ar, and Ne were not included. Among them, 59923 data were used as NM materials under the condition, computed $E_g > 0.1$ eV. The CGCNN was applied to predict $\Delta E_f$ value with input of only crystal structures for all materials. For these NM materials, CGCNN was applied to predict $E_g$ as well as $\Delta E_f$ value.

In this study, the ratio of training, validation, and test data was set to 4:1:1 for data selections of the CGCNN. When the number of training data is greater than 10k, the number of validation and test data were
fixed to be 2500. Weights and biases were optimized to minimize the loss function by using training data. Validation data were used to find the best epoch to stop the backpropagation with the minimum loss function, which indicates the threshold point of overfitting. Test data, which do not participate in the modeling, were used to evaluate the prediction error. In this study, a MAE of test data is used to evaluate the prediction error. The prediction errors were evaluated by averaging over several trials with different datasets. When the numbers of training data are 500, greater than 500 and less than or equal to 10k, and greater than 10k, the numbers of trials were set to 10, 5, and 2, respectively. We used random selections of training/validation/test data based on random seeds. Therefore, the same type of data could be used for comparisons of various predictive models when the same number of data are used although training/validation/test data vary among different trials. The maximum iteration of epochs was mainly set to 500 with checking that the minimum MAE of validation data is already obtained at an epoch less than 500. When this is not satisfactory, we increased the maximum iteration of epoch to until finding the minimum MAE of validation data.

The TL-CGCNNs were applied to predict $E_g$ or $\Delta E_f$ with less data using the pretrained CGCNN models that were constructed to predict $E_g$ or $\Delta E_f$ with 10k, 54k, and 113k (only available for $\Delta E_f$) training data. For the target models, the ratio of the number of training, validation, and test sets were also set to 4:1:1.

To investigate the effect of TL-CGCNN on more practical cases that suffer from insufficient data, we used the target properties of $K_{\text{VRH}}$, $\varepsilon_r$, and GW-$E_g$. The $K_{\text{VRH}}$ and $\varepsilon_r$ values were obtained from the MPD. For the $K_{\text{VRH}}$, we prepared 12990 data whose values are ranged from 2 GPa (Cs metal) to 436 GPa (diamonds), and also obtained by Voigt-Reuss-Hill approximation. For the $\varepsilon_r$, we prepared 4182 NM data (with a condition of $E_g > 0.1$ eV). The $\varepsilon_r$ value is prepared with an assumption to be a single scalar polycrystalline value converted from a dielectric tensor. The GW-$E_g$ values were loaded from a previous study. For GW-$E_g$ values, the $G_0W_0$ calculations with Heyd-Scuseria-Ernzerhof hybrid functional ($G_0W_0$@HSE06) were used for 270 binary and ternary materials (inorganic compounds) with optimized crystal structures by PBE-sol functional.

Supplementary Table S1 shows main hyperparameters which were used for the CGCNN models in this study. The defaults of other hyperparameters can be referred to the original code. Among various hyperparameters, the length of feature vector of FC layer and initial learning rate were reduced for the target model when we use TL-CGCNNs.
We compared the prediction of TL-CGCNN with other widely-used regression methods for some prediction models. As the regression methods, PLS,
LASSO,
SVR,
and RF were employed. As descriptors for these regression methods, a generalized set of chemical composition-based descriptors (132 descriptors) obtained by matminer python library were used. The number of training+validation and test data were 625 and 125, respectively. For training and validation data, 5-fold cross validations were used to optimize the hyperparameters relevant to the minimization of overfitting for the regression methods.

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AUTHOR CONTRIBUTIONS

R. A. designed the project. J. L. introduced the transfer learning function for CGCNN and performed the entire simulations in this study. All authors discussed the results and wrote the manuscript.

COMPETING INTERESTS

The authors declare no competing interests.
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Fig. 1 Overview of CGCNN. It predicts a target property with an input of crystal structure, which is transformed into a crystal graph. The CGCNN architecture consists of embedding, convolutional, pooling, and fully-connected layers.
Fig. 2 Overview of transfer learning. Comparison of (a) a conventional learning and (b) its transfer learning. When we have different tasks, we prepare different prediction models separately for a conventional learning case. When we use the transfer learning, the knowledge (parameters, namely, weights and biases) accumulated in the pretrained model with a big data, can be transferred to target prediction model.
Fig. 3 Dependence of prediction error (RMSE of test data). Prediction errors for (a) $E_g$ and (b) $\Delta E_f$ on number of training data. Horizontal axes are in log$_{10}$ scale. Error bars indicate the maximum and minimum values of several trials.
**Fig. 4 Comparison of prediction error of without and with pretrained models.** Prediction errors for (a) $E_g$, (b) $\Delta E_f$, and (c) NM(nonmetallic)-$\Delta E_f$. The predictions by the CGCNN and other regression methods are displayed as scatter plots. The predictions by the TL-CGCNN are displayed as bar graphs. The number of training data of target and pretrained models are displayed as relative size of scatter plot and relative width of bar graph, respectively. Error bars indicate the maximum and minimum values of several trials. The baselines, obtained as the mean absolute differences of the data distribution, for $E_g$, $\Delta E_f$, and NM-$\Delta E_f$ are 1.26, 0.97, and 0.77 (in the same unit as in each subfigure), respectively.
Fig. 5 Scatter plots between the quantities of $\Delta E_f$ and $E_g$ of 118286 materials. The color scale represents the relative density of points generated by Gaussian kernel density estimation. The probability density function is minmax-normalized, and the bandwidth for Gaussian is three times multiplied by Scott’s factor.
Fig. 6 Comparison of prediction error of without and with pretrained models. Prediction errors of (a) $K_{\text{VRH}}$, (b) $\varepsilon_r$, and (c) GW-$E_g$ without and with pretrained models. The predictions by the CGCNN and other regression methods are displayed as scatter plots. The predictions by the TL-CGCNN are displayed as bar graphs. The number of training data of target and pretrained models are displayed as relative size of scatter plot and relative width of bar graph, respectively. The unit of $K_{\text{VRH}}$ in log_{10} is GPa. Error bars indicate the maximum and minimum values of several trials. The baselines, obtained as the mean absolute differences of the data distribution, for $K_{\text{VRH}}$, $\varepsilon_r$, and GW-$E_g$ are 0.30, 0.27, and 2.49 (in the same unit as in each subfigure), respectively.
Table 1. Summary of material-property dataset used in this study.

| Material Property | Number of data | Minimum value | Maximum value | Mean value | Mean absolute difference |
|-------------------|----------------|---------------|---------------|------------|--------------------------|
| $E_g$             | 59923          | 0.100 eV      | 9.721 eV      | 2.179 eV   | 1.256 eV                 |
| $\Delta E_f$     | 118286         | $-4.612$ eV/atom | 5.124 eV/atom | $-1.481$ eV/atom | 0.968 eV/atom          |
| $\Delta E_f$ (NM)| 59923          | $-4.612$ eV/atom | 3.797 eV/atom | $-2.021$ eV/atom | 0.768 eV/atom          |
| $K_{\text{VRH}}$ | 12990          | 0.301         | 2.639         | 1.874      | 0.304                    |
| $\epsilon_r$     | 4182           | 0.063         | 2.992         | 1.096      | 0.265                    |
| GW-$E_g$          | 270            | 0.357 eV      | 14.548 eV     | 4.996 eV   | 2.486 eV                |

\(^a\) The values are in log\(_{10}\).  
\(^b\) The unit in log\(_{10}\) is GPa.
Supporting Information

S1. Optimization of hyperparameters for TL-CGCNN

Deep neural networks like CGCNN have many hyperparameters. For a strict optimization, all the hyperparameters must be optimized. However, it needs heavy computations to optimize such a large set of hyperparameters. In this study, we tried to test the influence of learning rate and length of feature vector of FC layer on the prediction error by TL-CGCNN with fixing other hyperparameters summarized in Supplementary Table S1. We use a case study of predictive model for the 500-NM-$E_g$ by TL-CGCNN with the 113k-$\Delta E_f$ pretrained model.

A learning rate for the optimization of weight and bias is determined by an initial learning rate and a learning rate decay. At early epochs, the initial learning rate becomes the learning rate. Then, the learning rate can be decayed by a multiplicative factor at later epochs. In addition, there can be many combinations relevant to a modifying schedule for the learning rate, such as the frequency of decay and the epochs that decay occurs. In this study, we only check the initial learning rate and multiplicative factor of learning rate decay.

Figure S1a shows the prediction error for the 500-NM-$E_g$ depending on the initial learning rate. The initial learning rate of the pretrained model is 0.01. When the initial learning rate is too large as 1, the prediction errors are larger. This may ascribe to that the optimized parameters loaded from the pretrained models may disappear. The smallest prediction error is obtained when the initial learning rate is 0.005. Therefore, for the "fine" tuning, it is recommended to use an adequately small learning rate, not to lose the transferred knowledge. However, when the learning rate is too small as 0.0001, the prediction error increases again. This means that an effective learning was not performed.

Figure S1b shows the prediction error for the 500-NM-$E_g$ depending on the multiplicative factor of learning rate decay. The multiplicative factor of learning rate decay of the pretrained model is 0.1. The smallest prediction error is obtained when the multiplicative factor of learning rate decay is 0.0001. At the investigated range of 0.0001–10 for the multiplicative factor of learning rate decay, the differences of the prediction errors are not so large as those by change of the initial learning rate. This may ascribe to that
whether transferred knowledge is conserved or not strongly depends on the initial learning rate at early epochs for this predictive model.

Figure S1c shows the prediction error for the 500-NM-$E_g$ depending on the length of feature vector of FC layer of the target model. In the pretrained model, the length of feature vector of FC layer is 128. The smallest prediction error is obtained when the length of feature vector of FC layer is 64. The prediction errors are similar with the values of 0.815–0.836 eV when the lengths of feature vector of FC layer are 4–256. However, we recommend not using too small or large length of feature vector of FC layer, which can result in much larger prediction errors.

Table S1. Main hyperparameters used for CGCNN and TL-CGCNN in this study.

| Hyperparameters                                      | Value                                      |
|------------------------------------------------------|--------------------------------------------|
| Minibatch size                                       | 256                                        |
| Length of atom feature vector $v_i$ after an embedding layer | 64                                         |
| Number of convolutional layers                       | 3                                          |
| Length of feature vector of FC layer                 | 128→64 (in case of transfer learning)      |
| Number of FC layer                                   | 1                                          |
| Optimizer for loss function                          | Stochastic gradient descent                |
| Initial learning rate                                | 0.01→0.005 (in case of transfer learning)  |
| Multiplicative factor of learning rate decay $^a$     | 0.1                                        |
| Dropout                                              | 0                                          |

$^a$ In the CGCNN code, the learning rate decay occurs at the 100th epoch as a default.
Fig. S1. Dependences of prediction error of transfer learning on hyperparameters. Prediction error for the 500-NM-$E_g$ prediction model depending on (a) initial learning rates, (b) learning rate decays, and (c) lengths of vector of FC layer by transfer learning with the 113k-$\Delta E_f$ pretrained model. Closed marks indicate the hyperparameter used in the pretrained model. A vertical indicator shows the optimal hyperparameter with the smallest prediction error. Error bars indicate the maximum and minimum values of several trials.
S2. Test of layer freezing

In this study, the fine tuning has been used as the TL-CGCNN. For a comparison, the layer freezing is also investigated as the TL-CGCNN. We only optimized the weights and biases of the last FC layer with freezing those of the other earlier layers. For the prediction targets, the 500-NM-$E_g$, 500-$\Delta E_f$, 500-NM-$\Delta E_f$, 500-$K_{VRH}$, 500-NM-$\varepsilon_r$, and 180-NM-GW-$E_g$ were used with the 113k-$\Delta E_f$, 54k-NM-$E_g$, 54k-NM-$E_g$, 113k-$\Delta E_f$, 54k-NM-$E_g$, and 113k-$\Delta E_f$ pretrained models, respectively, which showed the best performance when the fine tuning is used. The identical hyperparameters were used for two methods. Supplementary Table S2 summarizes the comparison of the prediction errors by the CGCNN, the fine tuning, the layer freezing, and other regression methods. The prediction errors of training data are also shown.

For all of six predictive models by the TL-CGCNN, the fine tuning shows much smaller prediction errors than layer freezing. In the case of the 500-NM-$E_g$ target model, the layer freezing shows prediction error of 0.956 eV slightly smaller than that of 0.978 eV by the prediction without a TL-CGCNN. However, in the cases of the 500-$\Delta E_f$, 500-NM-$\Delta E_f$, 500-$K_{VRH}$, 500-NM-$\varepsilon_r$, and 180-NM-GW-$E_g$ target models, the layer freezing shows even worse predictions than those without the transfer learning.

Although the prediction improvements are not achieved by the layer freezing, the prediction error differences between the training data and test data are much smaller than those obtained by the fine tuning. This means that the overfitting is reduced. However, relatively larger errors of training data obtained by the layer freezing indicate that the freezing of parameters of the convolutional layers in CGCNN obstructs the optimization of the predictive model to a satisfying level. In some previous studies using deep convolutional neural networks with a lot of multiple layers, similar concept of two methods are suggested to be mixed, namely, only early convolutional layers are frozen and the other layers including convolutional layers are all optimized.
**Table S2.** Comparison of prediction error (as MAE) of training and test data by the CGCNN, fine tuning, layer freezing, and other regression methods. The bold model emphasizes the smallest error of test data. The value in the parentheses is standard deviation of MAE of ten trials. Note that different training/validation/test data are used for each trial.

| Target model | Pretrained model | Method | Prediction error of training data | Prediction error of test data | Unit |
|--------------|------------------|--------|-----------------------------------|-------------------------------|-------|
| 500-NM-$E_x$ |                  | CGCNN  | 0.576 (0.129)                     | 0.978 (0.113)                 | eV    |
| 500-NM-$E_x$ | 113k-$\Delta E_f$ | Fine tuning by TL-CGCNN  | **0.511 (0.144)**              | **0.815 (0.090)**             | eV    |
| 500-NM-$E_x$ |                  | Layer freezing by TL-CGCNN | 0.919 (0.046)                   | 0.956 (0.099)                 | eV    |
| 500-NM-$E_x$ |                  | PLS    | 0.940 (0.021)                     | 1.051 (0.066)                 | eV    |
| 500-NM-$E_x$ |                  | LASSO  | 0.850 (0.021)                     | 1.078 (0.084)                 | eV    |
| 500-NM-$E_x$ |                  | SVR    | 0.720 (0.033)                     | 1.070 (0.087)                 | eV    |
| 500-NM-$E_x$ |                  | RF     | 0.367 (0.038)                     | 0.872 (0.060)                 | eV    |
| 500-$\Delta E_f$ |                 | CGCNN  | 0.092 (0.018)                     | 0.239 (0.026)                 | eV/atom |
| 500-$\Delta E_f$ | 54k-NM-$E_x$    | Fine tuning by TL-CGCNN         | **0.083 (0.030)**             | **0.222 (0.024)**             | eV/atom |
| 500-$\Delta E_f$ |                  | Layer freezing by TL-CGCNN      | 0.443 (0.021)                   | 0.482 (0.055)                 | eV/atom |
| 500-$\Delta E_f$ |                  | PLS    | 0.268 (0.016)                     | 0.338 (0.038)                 | eV/atom |
| 500-$\Delta E_f$ |                  | LASSO  | 0.301 (0.026)                     | 0.323 (0.044)                 | eV/atom |
| 500-$\Delta E_f$ |                  | SVR    | 0.224 (0.071)                     | 0.313 (0.051)                 | eV/atom |
| 500-$\Delta E_f$ |                  | RF     | 0.103 (0.007)                     | 0.249 (0.033)                 | eV/atom |
| 500-NM-$\Delta E_f$ |                 | CGCNN  | 0.081 (0.022)                     | 0.184 (0.023)                 | eV/atom |
| 500-NM-$\Delta E_f$ | 54k-NM-$E_x$    | Fine tuning by TL-CGCNN         | **0.059 (0.006)**             | **0.149 (0.021)**             | eV/atom |
| 500-NM-$\Delta E_f$ |                  | Layer freezing by TL-CGCNN      | 0.371 (0.018)                   | 0.424 (0.033)                 | eV/atom |
| 500-NM-$\Delta E_f$ |                  | PLS    | 0.211 (0.010)                     | 0.257 (0.022)                 | eV/atom |
| 500-NM-$\Delta E_f$ |                  | LASSO  | 0.237 (0.025)                     | 0.262 (0.023)                 | eV/atom |
| 500-NM-$\Delta E_f$ |                  | SVR    | 0.222 (0.045)                     | 0.332 (0.042)                 | eV/atom |
| 500-NM-$\Delta E_f$ |                  | RF     | 0.093 (0.004)                     | 0.215 (0.020)                 | eV/atom |
| 500-KV/atom |                  | CGCNN  | 0.067 (0.020)                     | 0.123 (0.015)                 | a,b   |
| 500-KV/atom | 113k-$\Delta E_f$ | Fine tuning by TL-CGCNN         | **0.075 (0.022)**             | **0.112 (0.012)**             | a,b   |
| 500-KV/atom |                  | Layer freezing by TL-CGCNN      | 0.149 (0.008)                   | 0.161 (0.011)                 | a,b   |
| 500-KV/atom |                  | PLS    | 0.132 (0.008)                     | 0.166 (0.013)                 | a,b   |
| 500-KV/atom |                  | LASSO  | 0.145 (0.007)                     | 0.160 (0.009)                 | a,b   |
| 500-KV/atom |                  | SVR    | 0.075 (0.039)                     | 0.149 (0.024)                 | a,b   |
| 500-KV/atom |                  | RF     | 0.059 (0.008)                     | 0.132 (0.012)                 | a,b   |
| 500-NM-$\varepsilon$ |                 | CGCNN  | 0.120 (0.032)                     | 0.181 (0.024)                 | a     |
| 500-NM-$\varepsilon$ | 54k-NM-$E_x$    | Fine tuning by TL-CGCNN         | **0.095 (0.016)**             | **0.163 (0.024)**             | a     |
| 500-NM-$\varepsilon$ |                  | Layer freezing by TL-CGCNN      | 0.181 (0.008)                   | 0.193 (0.032)                 | a     |
| 500-NM-$\varepsilon$ |                  | PLS    | 0.165 (0.006)                     | 0.200 (0.020)                 | a     |
| 500-NM-$\varepsilon$ |                  | LASSO  | 0.175 (0.005)                     | 0.189 (0.021)                 | a     |
| 500-NM-$\varepsilon$ |                  | SVR    | 0.074 (0.031)                     | 0.200 (0.024)                 | a     |
| 500-NM-$\varepsilon$ |                  | RF     | 0.079 (0.010)                     | 0.171 (0.019)                 | a     |
| 180-NM-GW-$E_x$ |                  | CGCNN  | 0.302 (0.140)                     | 0.783 (0.102)                 | eV    |
| 180-NM-GW-$E_x$ | 113k-$\Delta E_f$ | Fine tuning by TL-CGCNN         | **0.196 (0.082)**             | **0.591 (0.063)**             | eV    |
| 180-NM-GW-$E_x$ |                  | Layer freezing by TL-CGCNN      | 0.525 (0.178)                   | 0.867 (0.152)                 | eV    |

* The values are in log_{10}.

* The unit in log_{10} is GPA.

* Note that number of test data is 25% of that of training data.
Supplementary Reference

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