Machine learning-aided search of enhanced elastocaloric effect in graphene kirigami

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Two-dimensional materials, particularly graphene, have captivated scientific interest due to their extraordinary physical properties. The versatility of graphene has led to a wide range of derivatives and geometrically engineered modifications. One promising structural paradigm is graphene kirigami (GK), where precise cuts are introduced into the graphene lattice to form complex architectures inspired by the Japanese art of paper cutting [1]. Kirigami structures significantly alter the mechanical properties of graphene, enabling high stretchability and tunable stiffness. In addition, kirigami patterns also allow control of thermomechanical properties, including the elastocaloric effect, which relates a temperature change to an applied strain. Recent studies have examined the elastocaloric response of graphene kirigami (GK) and shown how it may be tailored through geometric design [2]. This tunability makes GK a promising platform for applications in nanoscale solid-state thermal devices. In this work, we combineD molecular dynamics (MD) simulations and machine learning (ML) models to explore how GK geometries affect the elastocaloric coefficient (ECC), defined as the adiabatic ratio between temperature change and applied tensile stress. To this end, we generated GK structures from a pristine graphene sheet partitioned into a 3x5 grid, where each grid cell could remain pristine or contain a vertical cut. The binary choice for each of the 15 cells resulted in 32,768 possible configurations. However, some configurations are not valid, such as when three vertically aligned cuts completely severed the structure. Those configurations were excluded, restricting the number of valid configurations to 16,807, with a number of cut cells ranging from 0 (pristine graphene) to 10. Further considering periodic boundary conditions along the in-plane directions reduced the number of unique GK configurations to 1,123. Therefore, MD simulations were performed for each one of those unique configurations at room temperature, and used to train ML models. In order to predict the ECC of GK configurations, we framed the problem as a supervised regression task and employed traditional and deep-learning models using the 3x5 array representation of the GK as inputs, with 1 denoting a defect and 0 a pristine cell. The data set was divided into three subsets: 70% for training, 10% for validation (model evaluation and hyperparameter tuning), and 20% for the final testing. We considered several ML models: linear regression, random forest, gradient boosting (XGBoost), multilayer perceptron (MLP), and convolutional neural networks (CNNs). The performance of each model was evaluated considering their root-mean-squared error (RMSE) and coefficient of determination (R^2) on the test data set. We found that CNN achieved the best performance (RMSE = 0.064 K/GPa; R^2 = 0.96), shown in Figure 1. Finally, as an application of this best-performing CNN architecture, we verified whether this model could be employed to efficiently search the available design space for structures with high-ECC. As a baseline, we considered a purely random search strategy, in which structures were randomly selected from the available pool and added to the data set. We then computed the mean ECC of the top 50 structures found thus far. This random search required 54 generations to converge, corresponding to almost total exploration of the entire data set. In contrast, our accelerated search began with the same structures used in the random search, and a CNN corresponding to the best-performing architecture found previously was trained on this initial set. The model predicted the ECC for all remaining structures, then new structures with the highest predicted values were selected in each subsequent generation, and the process was repeated. The accelerated search significantly outperformed the random baseline, reaching the true mean ECC of the top 50 structures in only 5 generations, which is approximately ten times faster than the random search, demonstrating the power of ML-assisted strategies for the accelerated discovery of advanced elastocaloric materials [3].

References

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Figures

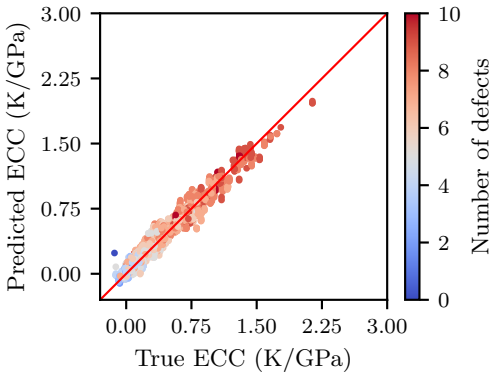

Figure 1. Elastocaloric coefficient (ECC), defined as the adiabatic ratio between temperature change and applied tensile stress, predicted by a convolutional neural network versus the true value obtained from molecular dynamics simulations.