



Deep learning based design of porous graphene for enhanced mechanical resilience

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ABSTRACT

Fracture behaviors of brittle materials are a crucial problem when it comes to reliability, especially for nanoscale devices and systems such as those built based on graphene. This study aims to use a new deep learning model, successfully incorporating data from molecular-level modeling, to predict the fracture path of graphene under the presence of various defects. In the process to build the model we first perform tensile test simulations on various graphene systems using molecular dynamics. The results are then transferred into image-based data for processing in the deep learning model. Based on this dataset we then construct multiple ConvLSTM-based models to learn the spatial-temporal information about crack propagation for each graphene system, respectively. The results show that our ConvLSTM-based models can predict the fracture path of graphene with 99 percent accuracy on a system of different crystallinity and 98 percent accuracy on different sets of defects, demonstrating excellent generalizability and transferability. These models demonstrate the power of exploiting deep learning for nanoengineering, and to specifically confer desired properties of materials based on defect engineering, which has great potential for next-generation materials by design.

1. Introduction

Graphene is an excellent material for its exceptional properties, including high thermal conductivity, high electrical conductivity, and outstanding mechanical properties [1–4]. As a result, it has shown excellent potential for various applications such as graphene-based glucose biosensors [5], gas sensors [6], membrane for water desalination [7], and drug delivery for biomedical application [8]. Furthermore, in terms of the mechanical performance of engineering applications, the ultimate tensile strength of graphene is up to 130 GPa, and Young's modulus is about 1 TPa [9], which indicates that graphene has a great potential to serve as the next-generation material considering its relatively low mass density compared with other thin materials.

However, despite its high strength, the fracture toughness of graphene is only around $4 \text{ MPa}\cdot\sqrt{m}$, measured as a stress intensity factor [10], and such a level of resistance to crack causes its functional strength to be lower than expected. Moreover, during the deformation of

graphene, defects and impurities may result in stress-concentration around the defective regions [11]. This heterogeneous local stress distribution changes the way graphene can endure deformation and thus lead to a higher likelihood to break atomistic bonds, which may limit the application because fracture behaviors are imperative in material design concerning the safety and reliability of the materials, especially when they rupture in a brittle manner such as graphene.

Directed nanoengineering of graphene could bring a new solution to improve the weakness of defective graphene materials. There used to be a one-sided understanding that defects in structural materials are inferior features since they tend to decrease the overall strength of materials; however, in nanoscale, defects can lead to benefits if they are just placed ingeniously. Furthermore, recent studies in both simulations and experiments have demonstrated the capability of defect engineering to adjust the material properties, such as reducing thermal conductivity [12], introducing magnetism [13] and increasing toughness [14,15]. As a result, scientists now have an excellent opportunity to design novel

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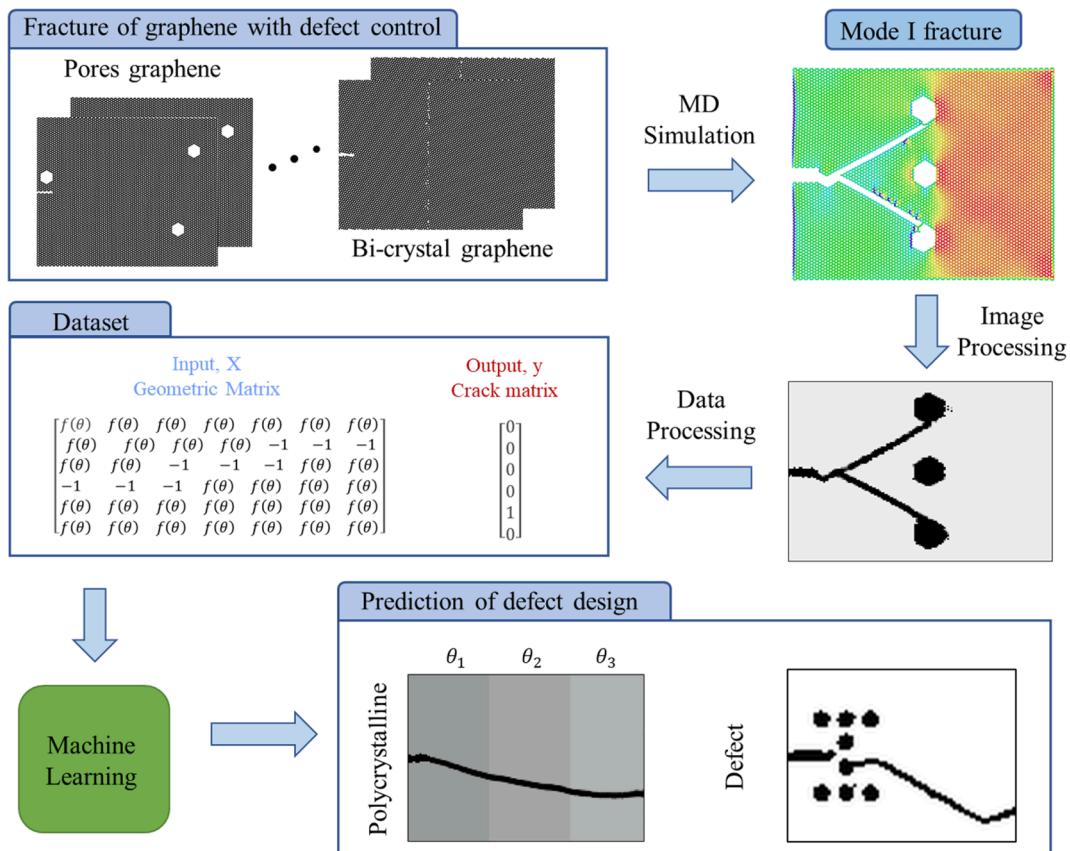


Fig. 1. Overall flowchart of the fracture prediction model in defected graphene. We perform several cases of uniaxial tensile simulations of different defective graphene materials including pores or grain boundaries to study their subtle fracture behaviors. Before applying deep learning skill, we need to represent these simulation results in array-based format so the geometries can be recognized by our predictive model. First, the MD simulation results are converted into gray-scale images to lower the dimensionality of the dataset. Then, the images are mapped into input matrices and output matrices for our deep learning model. Finally, our deep learning model can predict fracture of graphene with different orientations or different patterns of pore defects.

materials with high strength and high ductility by systematically manipulating the nano-defects.

With defect engineering drawing more and more attention in recent years, researchers have investigated several key positive effects of defects, instead of the classical focus on material deterioration [12,16]. However, broad-scope computational studies on fracture prediction of graphene under defect conditions are still lacking. The greatest challenge is the intensive computational cost needed to deal with the sophisticated combination of various defects. Therefore, in this study, we aim to offer an efficient way to explore the fracture behaviors of graphene under the massive design space of defect conditions, which can contribute to novel graphene design.

In recent years researchers have applied artificial intelligence techniques to solve complex physics problems in various fields, such as finding out wave function for quantum many-body problem [17], estimating gravitational lensing parameters [18], and predicting the folding structures of proteins using deep learning [19]. The material problems such as predicting fracture growth [20–22] or fracture stress [23], estimating mechanical properties [24,25] and designing tougher nanocomposite material [15,26] can now be addressed well via machine learning. In this study, we follow the footprint of the deep learning model for two-dimensional materials in our previous study to predict crack propagation of graphene with complicated geometries, including different crystalline orientations and various types and combinations of defects. Combined with a specialized representation method for the MD simulation results, our predictive model can learn from the spatiotemporal relation between fracture process and the geometry of graphene systems with various defective conditions and then predict their possible crack patterns that imply the mechanical performance against large

deformation and fracture. Hence, the proposed method using machine learning techniques to substitute MD simulation has a significant potential in developing novel graphene-based materials.

2. Results

In this section, we will present the results of the proposed method under various defective conditions predicted by our deep learning model and compare them with the counterpart of MD simulation results. A summary of the training process is shown in Fig. 1. Our dataset includes different graphene systems, such as single crystal graphene, bicrystal graphene, and porous graphene, with the same size of 20×16 nm. The dataset is preprocessed by our customized image mapping and data representation of MD simulation results to learn crack propagation more precisely. Moreover, we expanded the ML model compared with our previous work [20,22], including part of geometry matrices where cracks have not reached our model's input and the regions where the cracks have passed through (details, see Methods). As a result, our model provides a more comprehensive scope of fracture propagation mechanism and thus make a more precise prediction on more various geometric conditions that graphene might have.

2.1. Machine learning prediction on graphene with different orientations

To systematically investigate the effect of orientation on crack growth, we select ten different orientations ranging from 0° to 54° in every 6° for single crystal and bicrystal graphene simulation cases. The orientations of the graphene are defined as the angle between the

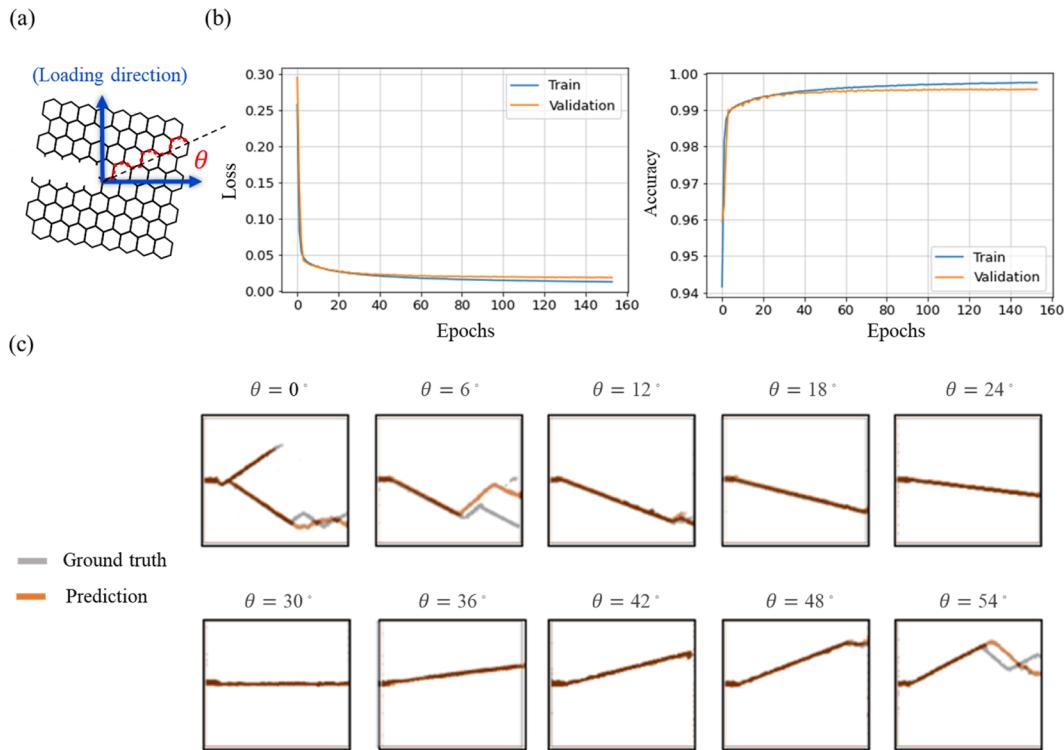


Fig. 2. Model performance tested on single crystal graphene. (a), Definition of the surface orientation of graphene system. (b), Training history of the model. (c), Fracture prediction of single crystal graphene with 10 different orientation.

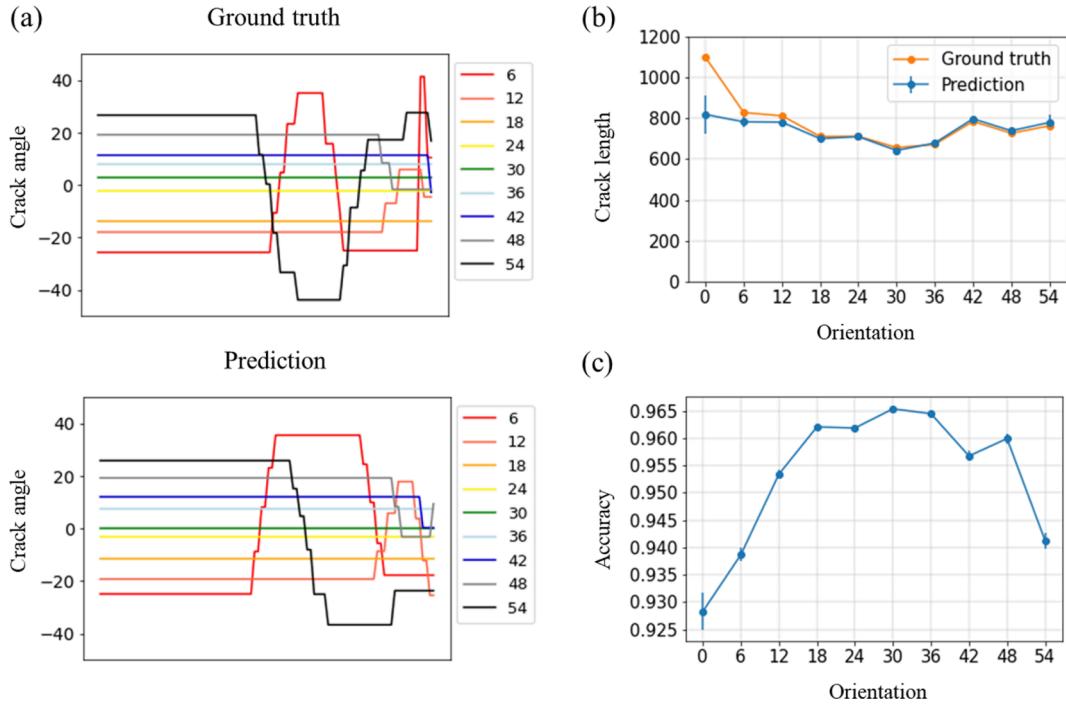


Fig. 3. Prediction on single crystal graphene with different orientations. (a), Comparison of crack angle between ground truths and predictions. (b), Comparison of crack length between ground truths and predictions. (c), Pixelwise accuracy of fracture predictions.

armchair direction and the horizontal direction (Fig. 2(a)). As shown in Fig. 2(b), the binary cross-entropy loss of our ConvLSTM-based model is 0.0143, and the accuracy reaches 99.45%, which demonstrates the capability of our model to learn the fracture path of graphene in different orientations. Moreover, the comparison between ground truths and ML predictions is shown in Fig. 2(c). One can see cracks develop in

different angles according to different surface orientations of graphene sheets, which shows the strong relation between surface orientation and angle of fracture path. The results are consistent with the atomistic perspective from the previous study that graphene is strongly depend as the bonding between carbons tend to break along the zigzag direction.

The changes of crack angles along crack lengths are collected and

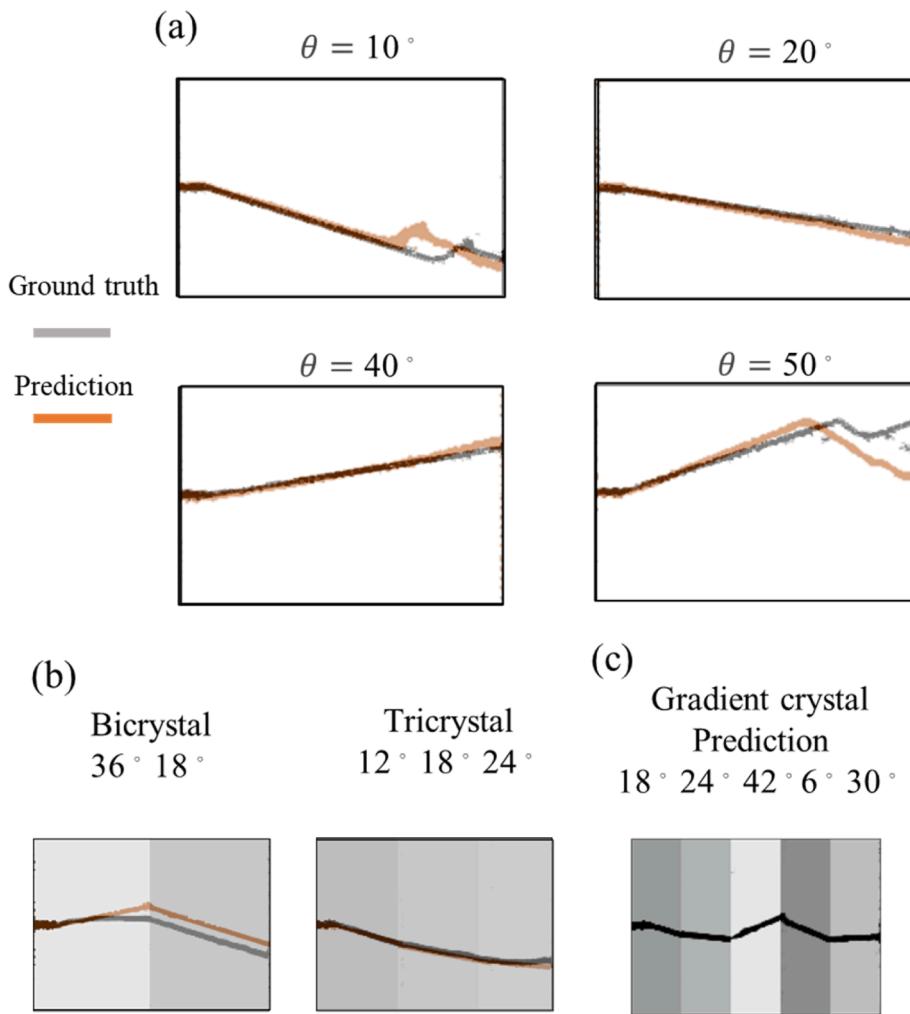


Fig. 4. Fracture predictions on various conditions of different orientations. (a), Fracture prediction of graphene with novel orientations compared to the ground truth. (b), Fracture prediction of polycrystal graphene compared to the ground truth. (c), Prediction of the fracture behavior of a gradient crystal graphene material.

shown in Fig. 3, where we take the average of five results from MD simulations as our testing data and compared them to ML predictions. Please note that in Fig. 3(a), each crack angle is calculated pixelwise. One can see from both prediction and ground truth that cracks are easier to change its direction in $\theta = 6^\circ$ and $\theta = 54^\circ$ (loading along zigzag direction) while the cracks are perfectly straight in cases of $\theta = 30^\circ$ (loading along armchair direction). Notice that the ground truths from MD simulations are essentially random due to the random state of initial velocity, while machine learning model predicts the most likely result according to the training data. We can see that as shown in Fig. 3 (b), the crack lengths (which are determined as the total number of black pixels) from prediction and ground truth are in great agreement except for the $\theta = 0^\circ$ case. As can be seen from Fig. 3(c), higher accuracy of fracture prediction occurs at $\theta = 30^\circ$ (loading along armchair direction), while lower one occurs at $\theta = 0^\circ$ (loading along zigzag direction), which results from the variance of MD simulation results. Specifically, as the strong preference of crack development along zigzag edge, crack propagates in horizontal direction every time in $\theta = 30^\circ$ cases, while in cases of $\theta = 0^\circ$, crack goes either upwards or downwards in the same probabilities because of the symmetry of the zigzag edge.

Further, as can be seen in Fig. 4(a), the model can predict fracture for orientations of 10° , 20° , 40° , and 50° , which were not included in the training dataset. For the bicrystal cases, the crack changes its direction when passing through the grain boundary, which again indicates that crystal orientation around the crack tip dominates crack

propagation of graphene. The key point of predicting fracture path in polycrystal graphene is to learn the direction change at the grain boundary. As can be seen in Fig. 3(b), training with 33 bicrystal cases, our model can predict fracture path of bicrystal and tri-crystal graphene of arbitrary surface orientation. We can further make predictions on gradient crystal graphene shown as Fig. 3(c). Using a sliced image instead of an entire image to make predictions on fracture evolution overcomes the restriction on image length, which means it is possible to predict fracture of graphene with infinite length and infinite grains. This is a significant improvement over earlier work [20,22].

2.2. Machine learning prediction on graphene with defects

The crack path becomes more complex in defected graphene compared to those of pristine ones as the defects greatly affect the stress distribution around the crack tips. To investigate the effect of nano-defects on crack growth, we introduced defects to our simulation cases and then built another model trained with additional 295 cases of tensile tests of graphene. All graphene sheets in the training dataset involve three sets of porous defects. The 128 out of the 295 cases are built with 2×2 hierarchical defects (Fig. 7) to see whether our model is sensitive to different types of defects. All tensile tests are performed in zigzag direction in all cases ($\theta = 0^\circ$) and finally 80% of the results are for training and 20% of them are for validation. To fully represent the features of the porous graphene, we use three labels in the input matrices to describe the geometrical features of cracks, pores and non-cracked

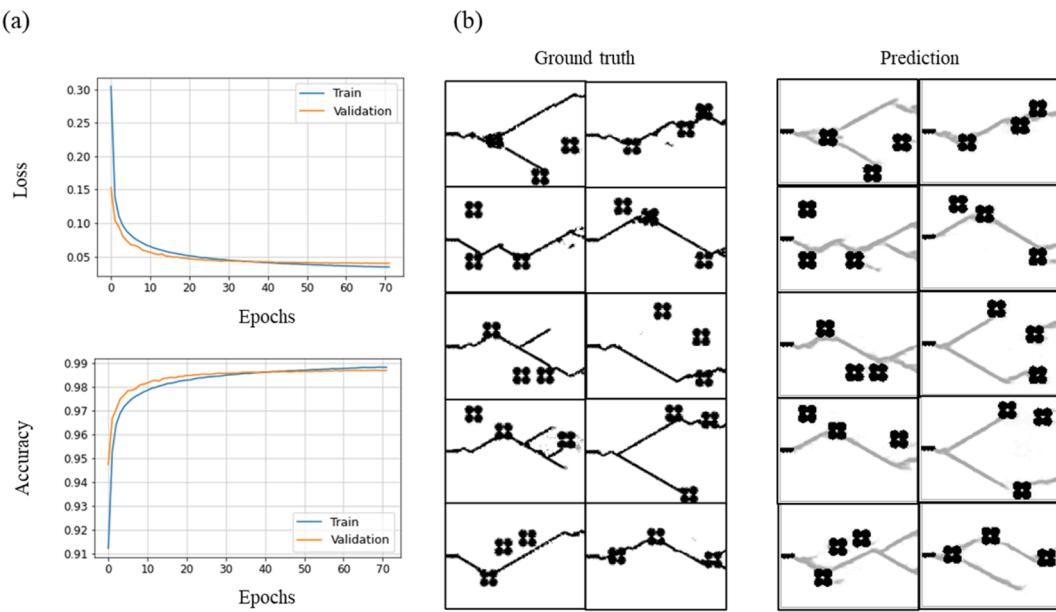


Fig. 5. Model performance on graphene with random defects. (a), Training history of our extended model trained with simulation results of defective cases. (b), Fracture prediction of defective graphene compared to the ground truth.

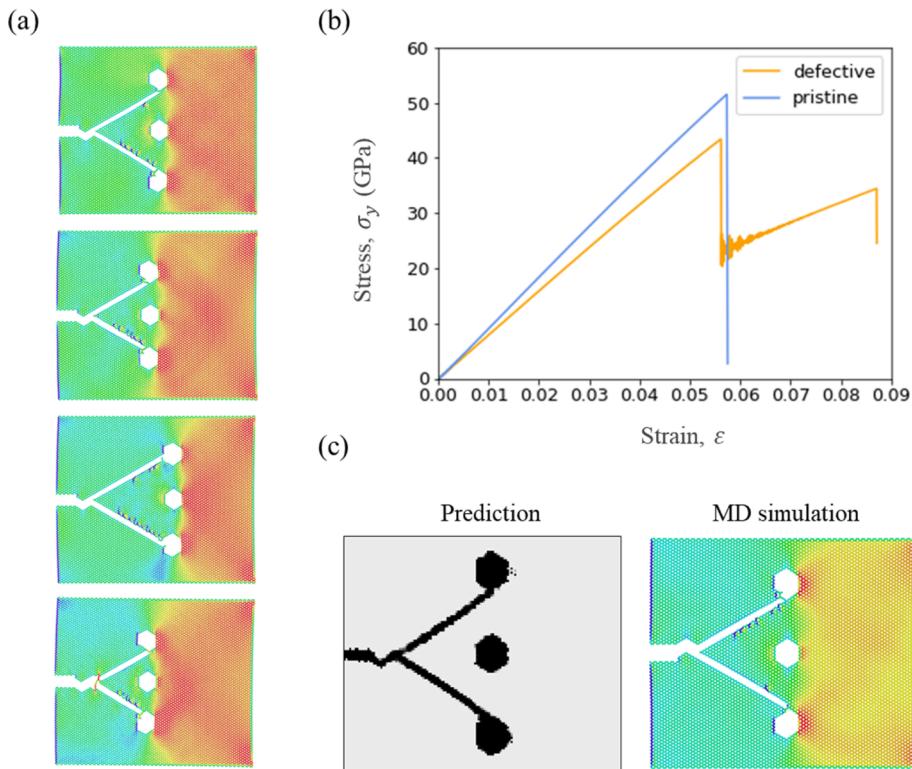


Fig. 6. Analysis of model results showing that defects postpone crack propagation. (a), Defective graphene possessing the capability to postpone the crack propagation in MD simulations. (b), The stress strain curve of the pristine graphene and the defective graphene, whose toughness are 1.50 GPa and 2.13 GPa respectively. (c), Fracture prediction of defective graphene in comparison of the ground truth.

regions. Specifically, we use -1 for cracks, -2 for pore defects and 0 for non-crack regions in the input matrices. On the other hand, we treat the prediction task as a multi-classification problem. Here we use another three labels in output matrices, where 1 is for crack, 2 is for defect and 0 is for non-cracked region (detail sees Material and methods).

In this way, our model shows the capability of capturing the complex spatiotemporal information of defected graphene. As can be seen from

Fig. 5(a), the training and validation accuracy are about 98%, However, it is good enough to make fracture predictions on the cases of graphene with random defects which are shown in Fig. 5(b). It is worth noting that even with data of less than 300 images, which are much smaller than the enormous combination of total defect patterns, our model still predicts cases well that are not involved in the training dataset. This demonstrates generalizability of the resulting predictions.

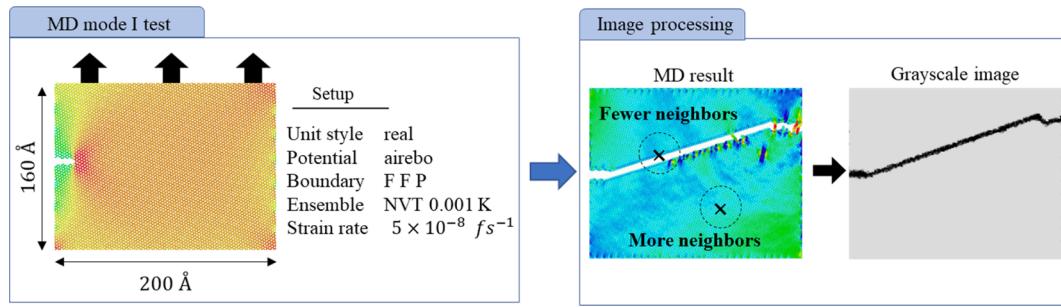


Fig. 7. Data pre-processing used in dataset generation. Molecular dynamics simulations of the uniaxial tensile tests of graphene are carried out to obtain graphene fracture. Subsequently, these data are converted into grayscale images. The color is based on the number of the neighbors of every atom. One with fewer neighbors (crack) is black whereas one with more neighbors (non-crack) is white.

As a few specific examples, we now demonstrate a set of special cases that the presence of the nano-defect could delay crack propagation shown in Fig. 6(a). In those special cases, the crack initiates from notch and then splits into two branches propagating at nearly the same speed. As the cracks reaches the undefective regions, they stop growing for a certain period of simulation time while the graphene is still under tensile deformation. One can see the stress drop to a certain value when the cracks reach the nano-defects shown as in Fig. 6(b). At this stage, the strain energy is partly converted into kinetic energy during the process of bond rupture and the maximum local stress is released down, which enables the stop of crack propagation. At this point graphene could be further stretched, which results in a toughness that is 42% greater than that of pristine graphene (see Fig. 6(b)). These results suggest that by introducing nano-defects, we can indeed enhance the durability of the material. More specifically, defects are capable of reducing the stress concentration during fracture evolution. A comparison of the ground truth and our model prediction is shown in Fig. 6(c). Although the defect patterns are included in the training dataset, the successful predictions on those patterns shows the capabilities of this model to learn such crack stop phenomenon as it can recognize the possibilities of crack propagation in the certain condition to be sufficiently low to induce crack stopping. More data or other physical information such as stress field could raise the predictive power of this ConvLSTM-based model, as could be explored in future work.

3. Discussion

In this study, we leverage the use of a ConvLSTM-based deep learning model, expanding on our previous work [20,22] to predict fracture growth of graphene with complex geometry. Apart from predicting crack propagation of graphene with different surface orientations, we can also make predictions of fracture of porous graphene, featuring certain types of defects. To improve the predictive power of this model over earlier work, we first extended the geometric matrices from the region behind the crack tip (cracked part) to the region ahead of crack tip (non-cracked part). We considered the region *ahead* of crack to be critical as they significantly affect the stress distribution, and directly reflect *where* the bonds will rupture. Second, we try to use multi-labels to represent the cracks and defects. Our results show that it is feasible to label our data in this way and to treat the problem as a multi-classification task. We successfully extended the method to be applied on defective cases by only adding another label for defects. This indicates our method has the flexibility on a more complex system, with even more labels or atom types, or other parameters. For example, we might use a similar model to handle doping of a material. Third, we replaced our earlier 1D ConvLSTM-based model [20,22] with a 2D ConvLSTM-based model. Although the model based on convolutional 1D layers and LSTM is powerful enough to predict fracture of undefective graphene, it performs not well on predicting defective cases because it can only sees a limited scope of spatial information. Hence, 2D

convolutional LSTM layers are adopted here to extract more complex spatiotemporal features which can focus on both input-to-state and state-to-state transitions of information between geometries and cracks. The use of 2D ConvLSTM improve the performance of the predictive model with the categorical cross entropy loss and categorical accuracy from 0.067 and 0.977 to 0.040 and 0.987, respectively. This is a significant improvement.

The success of fracture prediction on defective graphene further suggests the feasibility to apply this method on fatigue of graphene, which is a significant challenge in fracture mechanics. According to previous studies, graphene as well as GO with point defects display a fatigue phenomenon under cyclic loading, which is demonstrated by both experiments and MD simulations using ReaxFF [27,28]. We may be able to accomplish this prediction task by adapting the algorithm reported here. First, we would need to generate a set of data using MD simulation (e.g. using the ReaxFF potential). Second, we should change our model output from one column to two columns. In this study we assume that cracks propagate from left edge to right edge. However, in the fatigue cases (as shown in previous work), the cracks grow from the middle point to the left and the right sides. This means we would need to predict two columns in every fracture evolution. It is worth noting that the way we label our data and the central idea of the making predictions on the fracture evolutions remains the same. We believe this may work well because the dataset consists of cracks and defects only, which shares the same structure as in the dataset used here.

Through these new developments, we were able to show that the model is capable of discovering the effect of nano-defects to postpone crack growth in graphene by investigating the simulation results of defective graphene sheets. The necessary condition to trigger this mechanism in graphene nanosheets is to guide the cracks into *branches* so that the branched cracks can reach the symmetric defects shown as Fig. 6(a). The symmetric defects will then disperse the stress that was being concentrated at the crack tips to the region ahead of the crack. On the other hand, the leading cracks propagating to specific defects becomes critical as it can trigger the release of stress concentration, which contrasts with the general view that crack propagation causing ultimate failure of materials, especially in brittle ones. This finding also provides another perspective of increasing the durability of materials by isolating the cracked region controlled by putting defects deliberately, which in traditional process of manufacturing should be avoided. This type of mechanistic insight is important in terms of generalizability of the modeling results for other cases and expands the scope of defect engineering.

Altogether, we demonstrated the capability of our model to predict the crack path of graphene under various defect conditions and geometries. As defect engineering is increasingly applied in various fields, this model holds the potential to be a powerful tool in the general area of defect design. It is worth noting that our model does not take mechanical properties as input to predict the fracture behavior of graphene, which means the model can learn directly from the subtle relation between

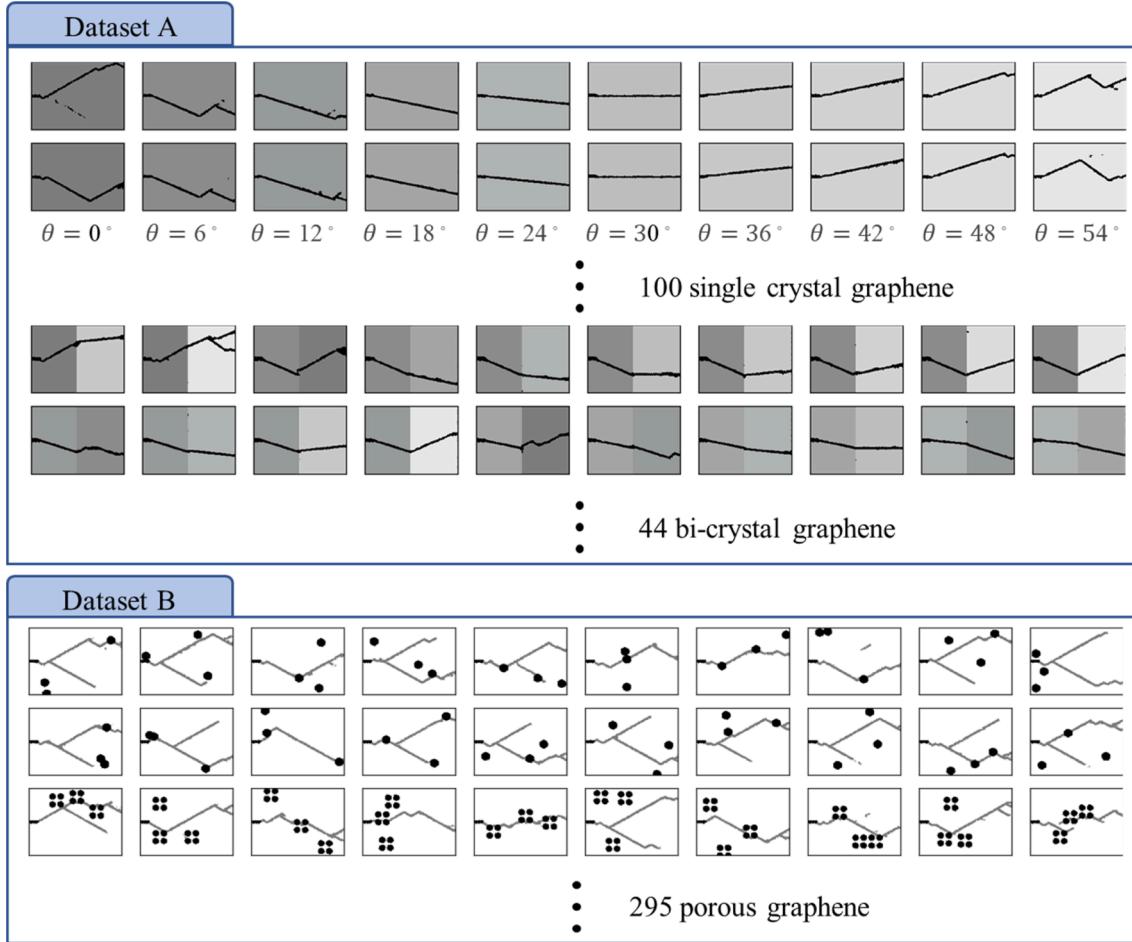


Fig. 8. Training dataset illustration. After image processing, we organize two collections of training data. One is graphene system with different orientation (involving single crystal and bi-crystal), and the others is porous graphene system (involving hierarchical defects).

geometries, defects, and cracks. From this viewpoint, even though we did not use experimental data in this study due to the expense, we can infer that the image-based method utilized here would work as well on experimental dataset because it is feasible to process those raw data into images. This may also be combined with transfer learning where experimental data could inform an already trained model, to adapt it to make predictions suitable for experimental environments. The core value of this model is that it can quickly provide detailed and useful information about fracture, for instance, recognizing whether or even where the crack might stop. This method bypasses the complex description on the physics and make prediction more efficiently, which is potential for next-generation material design.

4. Materials and methods

4.1. MD simulations

We select a $200 \text{ \AA} \times 160 \text{ \AA}$ graphene sheet with a 20 \AA -long initial crack at the left edge to be the simulation model. The cases of single crystal graphene are created by rotating the lattice at intervals of ten degrees of orientation angle from 0 to $\pi/3$. And the cases of bicrystal graphene are created from the combination of two different single crystal graphene with size of $100 \text{ \AA} \times 160 \text{ \AA}$ each. To investigate the effect of defects on crack propagation, we introduce defect of different sizes and hierarchical structures into graphene. Free boundary condition is applied along all planar direction. Molecular dynamics simulations are performed using the Large-Scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) in this study, and we use OVITO to process the

simulation results into images. The adaptive intermolecular reactive bond order potential (AIREBO) is used since it is a credible potential which can characterize the elastic mechanism of graphene-based materials [29]. The cutoff parameters of the switching function are both set to 2.0 \AA for more realistic measurement of bond breaking [30]. Uniaxial tensile tests are performed in y direction with strain rate of $5 \times 10^{-5}/\text{ps}$ in 0.001 K NVT . It is worth noting that the strain rate adopted here is much higher compared to those in experiments due to the computational constraints of molecular dynamics simulation. Still, the strain rate is slow enough (within the computational limitations) to avoid overestimating the energy release rate [31] and is widely used in related studies [32]. Moreover, the low temperature here lessens the impact of strain rate on the fracture properties of graphene [33].

4.2. Data statistics

We prepare two collections of data for two different machine learning models respectively (see Fig. 8). Dataset A is for predicting fracture growth of graphene in different orientation, and Dataset B is for graphene with defects. Dataset A consist of 100 cases of single crystal graphene and 44 cases of bicrystal graphene. Single crystal graphene is selected with 10 different orientations (of every 6° ranging from 0° to 54°) in 10 sets. The cases of bicrystal graphene are randomly selected with the correlation coefficient of the two orientations in graphene sheets to be -0.17 . Dataset B consists of another 295 cases of graphene, among which, 128 out of the 295 are introduced with three sets of 2×2 hierarchical pore defects and the others are introduced with three sets of single pore defects. All of the cases in dataset B are in the same

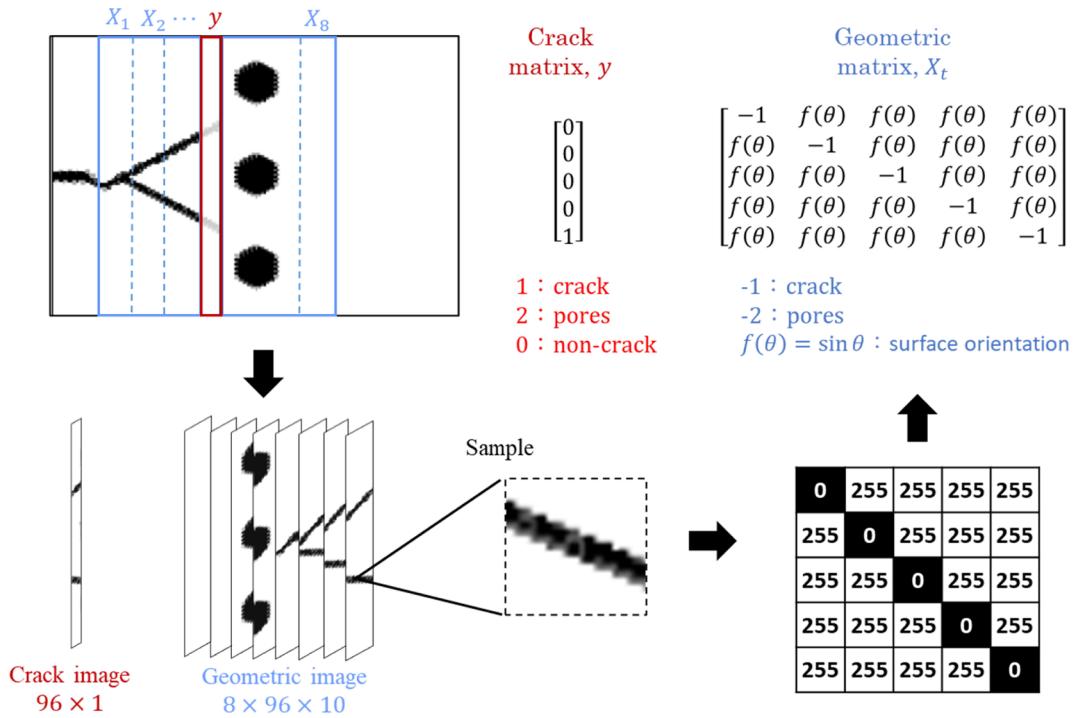


Fig. 9. Data processing for the machine learning dataset. An image is captured forwards and backwards to the corresponding crack image for every timestep, soliciting key geometric parameters. We then slice the fracture process into sequences of time series. After transformed to downscaled images, both geometric image and crack image are then converted into geometric matrix, X and crack matrix, y , respectively.

orientation of $\theta = 0^\circ$.

4.3. Data processing

To obtain a better feature representation for crack propagation of graphene with different orientations and defect patterns, MD simulation results are first transformed into grayscale images with length of 128 pixels and width of 96 pixels. As can be seen from Fig. 9, black regions represent for crack and defect, while white regions represent for the undamaged parts. The images are then further mapped into input matrices and output matrices. Instead of using the whole image to predict the corresponding crack image, we only take the region forwards and backwards around where the actual predicted crack matrix y is undertaken and then slice them into sequences that we call geometric matrix X .

For the inputs, geometric matrices X , the black pixels are mapped with the value of -1 and -2 in representation of cracks and pore defects respectively. In contrast, the white pixels are mapped with the value of $\sin \theta$ according to its orientation angle. For the output, crack matrices y , the pixels where crack happens are labeled as the value of 1, defect pixels are labeled as 2, while non-crack pixels are labeled as 0. By doing so, we are able to capture the complicated geometric information including cracks, defects and surface orientations as much as possible.

4.4. Machine learning model

A ConvLSTM-based model is adopted to predict graphene propagation. In our previous work we used convolutional 1D layer combined with LSTM to predict fracture path of graphene of different orientation [22]. In this work, the LSTM layer is replaced with ConvLSTM layer to extract more subtle spatial features. ConvLSTM is a layer based on fully

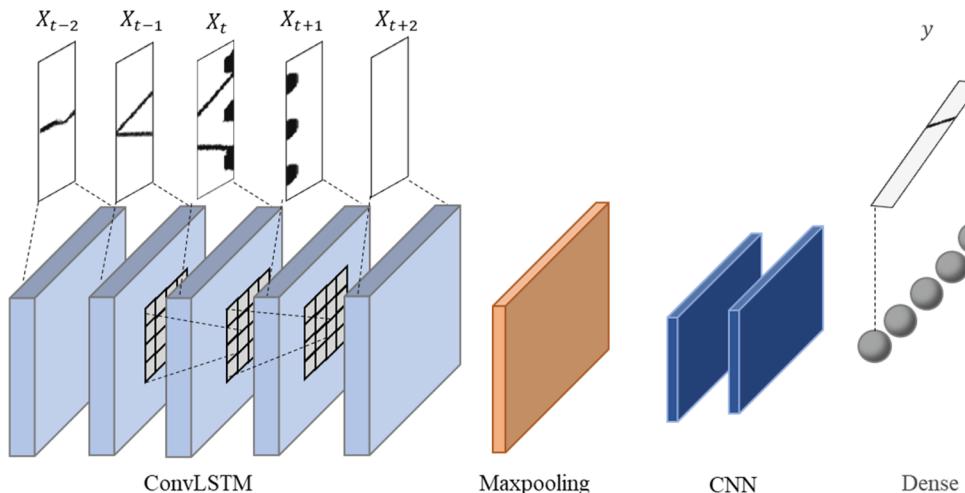


Fig. 10. Architecture of the ConvLSTM-based model used in this work. The model predicts crack matrix using the geometric matrices with eight time sequences. Feature extraction in both input-to-state and state-to-state are implemented using ConvLSTM along with max pooling. The feature maps further pass convolutional layers to learn detailed spatial dependencies. Finally, a dense layer is adopted for classification of every pixel. The output matrix is then used to update the geometric matrix for the next prediction. By repeating the process, we can ultimately obtain a whole picture of the crack image.

connected LSTM which further contains the convolutional structures to extract the spatial information. Compared to general LSTM, ConvLSTM overcomes the limitation of vector-variant representation of the inputs, which can remain more spatial information. The architecture of our ML model is shown in Fig. 10, which consists of one ConvLSTM layer, one pooling layer, two convolutional layers and a dense layer. In this paper we predict the crack image of the next time steps using sequence of geometric matrices.

The input to our model is a four-dimensional tensor whose first dimension is time sequences, second and third dimension are rows and columns of images, and the last dimension is the color channel. The ConvLSTM layer extracts the spatial features with convolutional operators, then temporal dependencies between feature maps are achieved from input-to-state and state-to-state transition in LSTM cells. Max pooling layers are used to intensify the feature of crack and defect as they are relatively small compared to untracked region. Convolutional layers are applied to further capture overall spatiotemporal features followed by a dense layer classify crack or non-crack pixels. We adopt Adam optimizer with learning rate 0.005 with decay of 0.001. Categorical cross entropy is selected as loss function. All training is implemented in Python with TensorFlow and Keras packages [34], and using NVIDIA GeForce RTX2080Ti to perform GPU computation. We note that 80% of data is used for training and 20% is for validation.

CRediT authorship contribution statement

Chi-Hua Yu: Conceptualization, Funding acquisition, Methodology, Project administration, Supervision, Validation, Writing - original draft, Writing - review & editing. **Chang-Yan Wu:** Formal analysis, Validation, Writing - original draft. **Markus J. Buehler:** Conceptualization, Funding acquisition, Supervision, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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