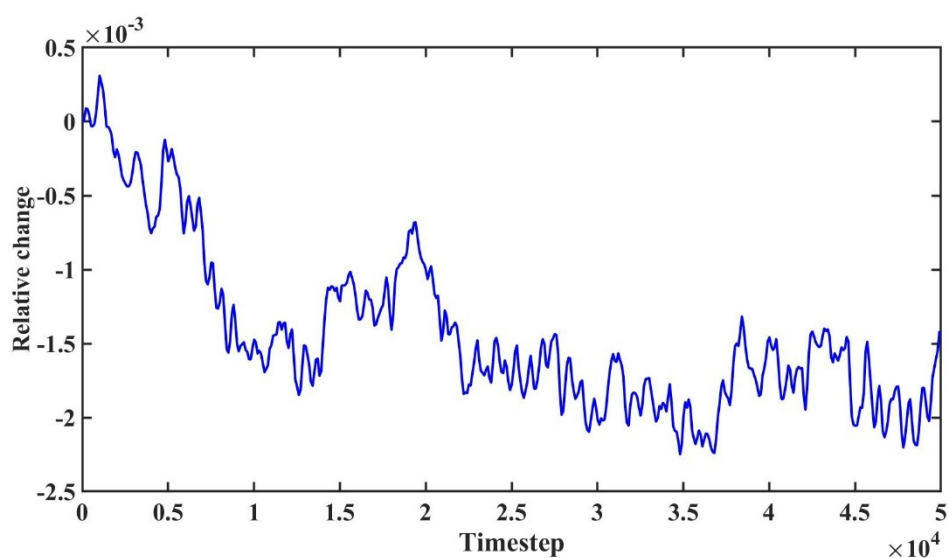


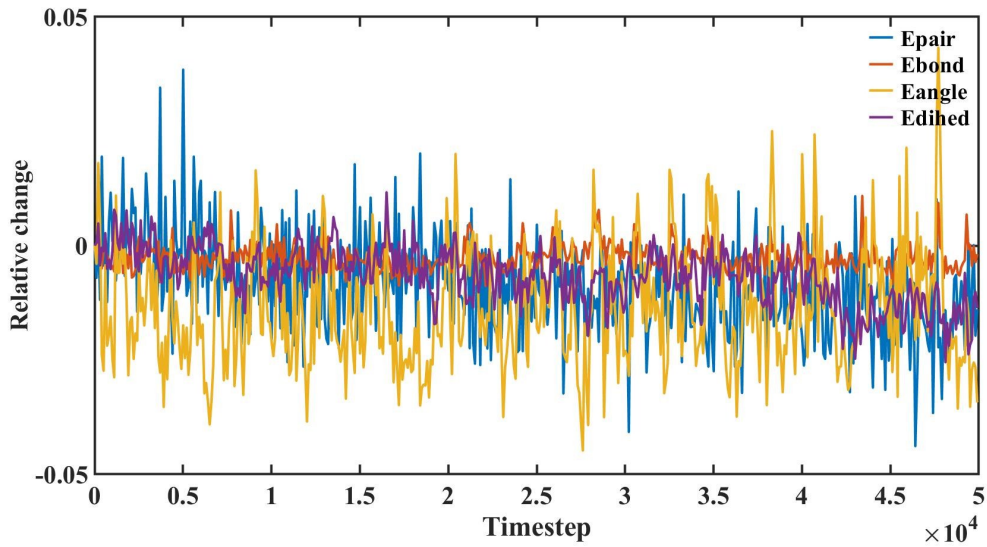
### Supplementary information

Figure S1 shows the relative change of the system size with 60 graphene sheets randomly distributed in rubber matrix during the last stage of relaxation at 300 K. It can be seen that the change amplitude of the system size is smaller than 0.3%, indicating the equilibrated state of the simulation system.



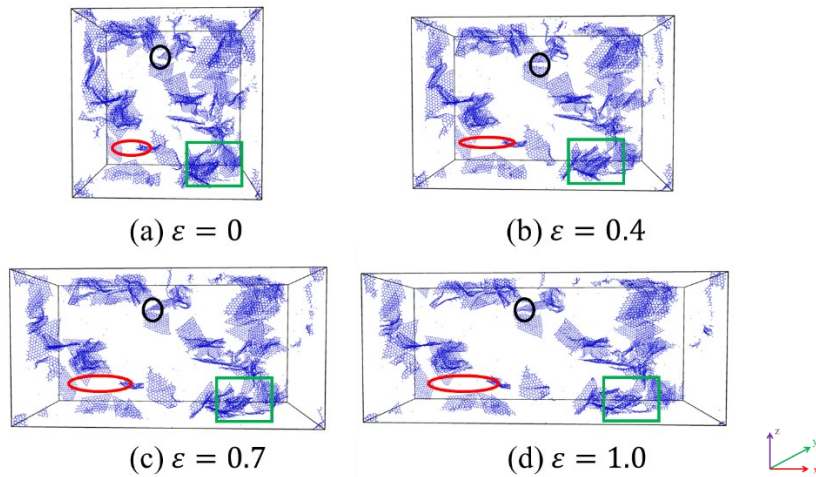
**Figure S1. Relative change of the system size with 60 graphene sheets randomly distributed in rubber matrix**

The systems are adjusted to reach the equilibrium state through a sequence of relaxation that has been described in the first paragraph of Section 3.2. In this study, the simulation box length extends from 100 Å to 113 Å~125 Å with a final rubber's density of 0.78 g/cm<sup>3</sup> ~ 1.06 g/cm<sup>3</sup>, varying among the simulation systems with different graphene filling volume fractions, graphene dispersion patterns and interfacial interaction strengths, which is closed to the actual density of rubber. Besides, all the system energies reach a stable value with a relative change less than 5% after the equilibration process as shown in Figure S2. Thus, it can be affirmed that the system has reached an equilibrium state and the configuration has been fully relaxed.



**Figure S2. Relative change of the system energies with 60 graphene sheets randomly distributed in rubber matrix**

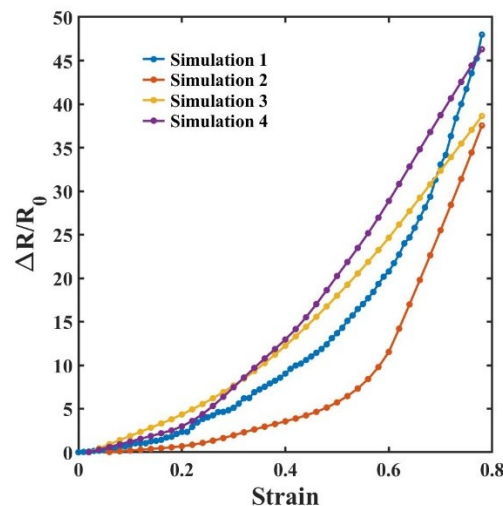
As for the mobility of graphene, it is true that the graphene sheets move relative slow during the relaxation. However, the displacements of the graphene sheets are not small at all over the tensile session. Figure S3 (Figure 3 in the manuscript) displays the movement of the graphene sheets while the system is stretched to the strain of 0.1. It can be seen that the locations of certain graphene molecules change a lot during the deformation which leads to the evolution of the conductive network. Considering that the object of this study is to capture the dynamic and instantaneous strain-resistance behavior of graphene rubber systems, there is no need to decide whether the systems reach the equilibrium state during the deformation. Therefore, the stretching process is continuous without any relaxation or interruption, during which the coordinate information of graphene sheets is recorded to calculate the system resistance.



**Figure S3. Configurations of graphene rubber during tension ( $v_f = 7.59 \text{ vol\%}$ )**

The simulation results depend much on the initial dispersion state of graphene. In this study, the graphene sheets were generated in the simulation box with random locations and orientations. The distances between graphene sheets were calculated to decide whether the configuration was reasonable without any overlap or intersection. Afterwards, the initial locations of graphene are adjusted to form another two distribution patterns, i.e., the diagonal distribution and the intermediate distribution. The strain-resistance behaviors of systems with graphene of the three different initial dispersion states have been studied in this paper. The results show that the initial graphene distribution discrepancy can lead to different strain-resistance sensitivity of the composites due to the different original spacing ratios.

Meanwhile, it should be noted that the specific locations and orientations of graphene molecules can be different each time they are regenerated in the simulation box, even for the same kind of graphene distribution pattern. Figure S4 shows the relative change of the system resistance for four simulation systems, which all contain 60 graphene sheets with random locations and orientations but are different from each other. It can be seen that the curves show basically similar increasing trends despite of the difference in specific values. Therefore, it is believed that the configurations chosen in this paper are able to represent the responses of the systems with the same type of dispersion.



**Figure S4. Relative change of the system resistance with 60 graphene sheets randomly distributed in rubber matrix**

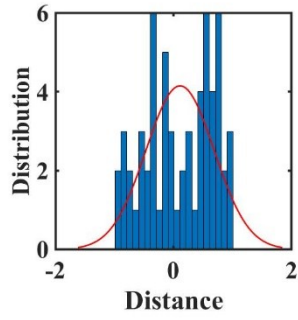
In this study, the simulation box length was initially set as 100 Å, extending to 113

$\text{\AA} \sim 125 \text{\AA}$  after an equilibration sequence. The force field parameters of rubber used in this study were determined according to the research of Uddin et al.<sup>2</sup> on a coarse-grained molecular model of natural rubber, where the molecular dynamics (MD) simulations were conducted in an orthogonal simulation box size of  $(54.01 \text{\AA})^3$  and showed a good agreement with experimental results in the prediction of macroscopic responses. Therefore, it is reasonable to believe that the simulation box of  $(\sim 100 \text{\AA})^3$  is large enough to reveal the true mechanical response of rubber matrix, which plays a fundamental role in the simulations of graphene rubber's strain-resistance behavior.

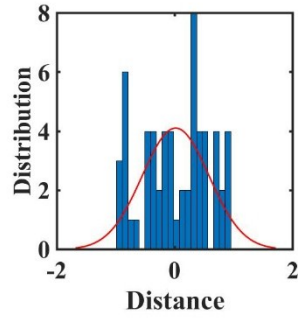
In addition, a literature survey on MD simulations of graphene polymer nanocomposites has been made to decide the proper size of graphene sheet, as listed in Table S1. It can be concluded that the size of graphene sheets is chosen comparable in certain dimensions to the simulation box only when the study is aimed at investigating the properties of multilayer graphene like the work of Li et al.<sup>3</sup> and Alian et al.<sup>4</sup>, or a single graphene molecule in the box like the work of Rahman et al.<sup>5</sup>. In our study, the system of graphene rubber was generated as a mixture of rubber chains and numerous monolayer graphene sheets, similar to the work of Rissanou et al.<sup>6</sup> and Li et al.<sup>7</sup>. Therefore, the size of graphene sheets in the simulations of this study are taken as  $27.06 \text{\AA} \times 23.43 \text{\AA}$ , about 1/4 of the box length.

**Table S1. A literature survey on MD simulations of graphene polymer nanocomposites**

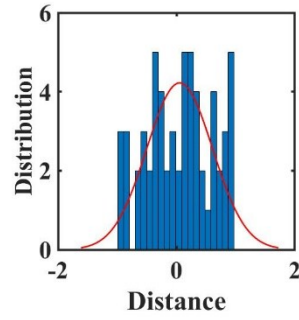
Researcher	System	System/RVE size	Graphene size
Li et al. <sup>3</sup>	Multilayer graphene/epoxy	93.33 Å × 93.33 Å × 20.088 Å	93.588 Å × 93.744 Å × 19.648 Å
Alian et al. <sup>4</sup>	Multilayer graphene/polyethylene	unprovided	a) 100 Å × 100 Å b) 49 Å × 51 Å
Rahman et al. <sup>5</sup>	Graphene/amorphous polyethylene	$18.6 \times 10^6 \text{ Å}^3$ $\sim 19.93 \times 10^6 \text{ Å}^3$	$211.62 \text{ Å} \times 9.84 \text{ Å}$
Rissanou et al. <sup>6</sup>	Graphene/polyethylene	unprovided	a) 19 Å × 20 Å b) 49 Å × 51 Å c) 84 Å × 86 Å
Li et al. <sup>7</sup>	Graphene/NBR	$50 \text{ Å} \times 50 \text{ Å} \times 50$ Å	$22.6 \text{ Å} \times 34.4 \text{ Å}$



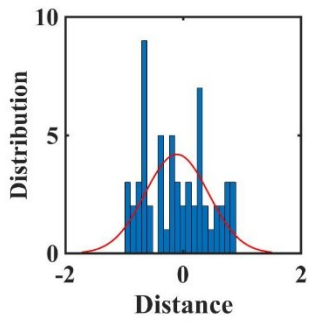
(a) group 1-x



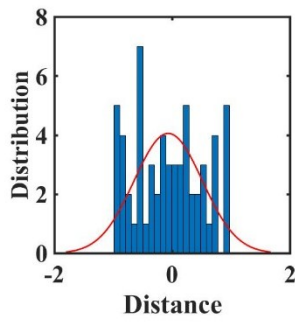
(b) group 1-y



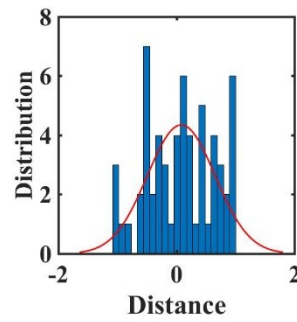
(c) group 1-z



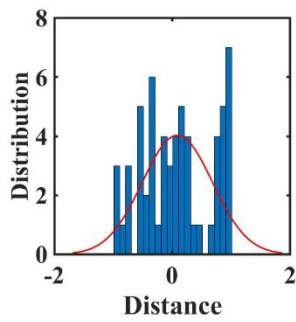
(d) group 2-x



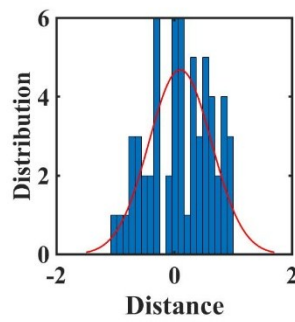
(e) group 2-y



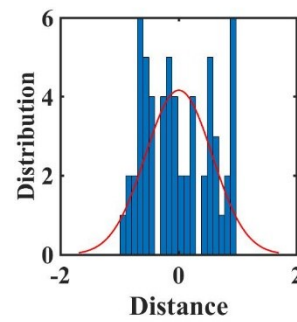
(f) group 2-z



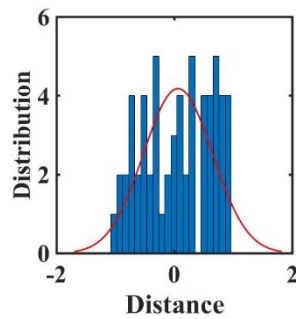
(g) group 3-x



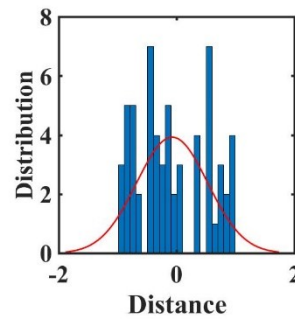
(h) group 3-y



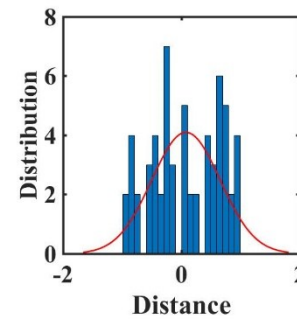
(i) group3 -z



(j) group 4-x



(k) group 4-y



(l) group 4-z

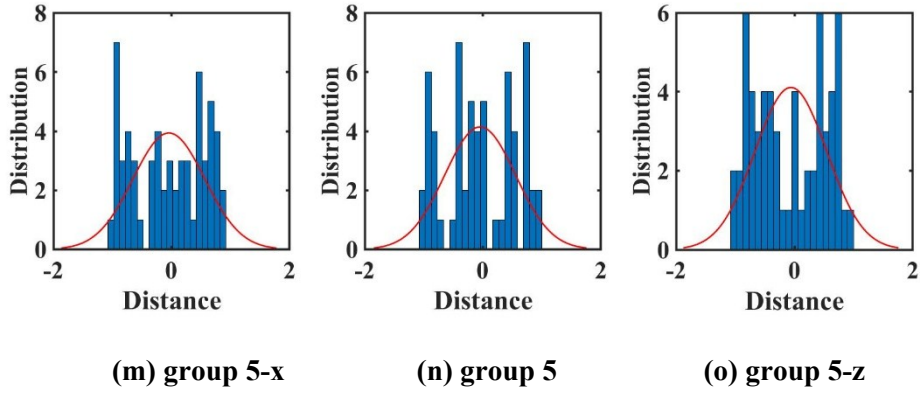
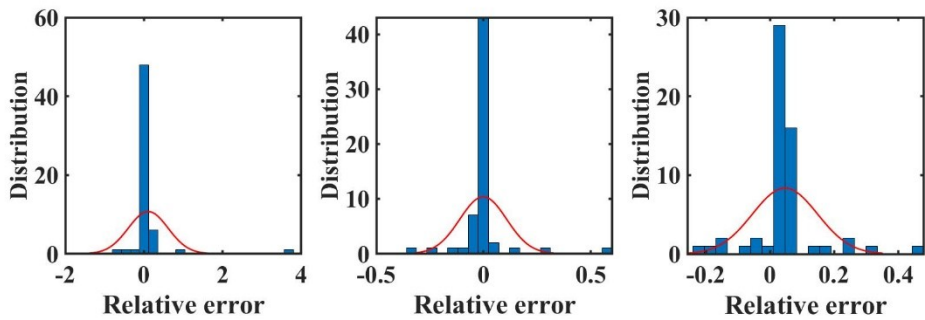
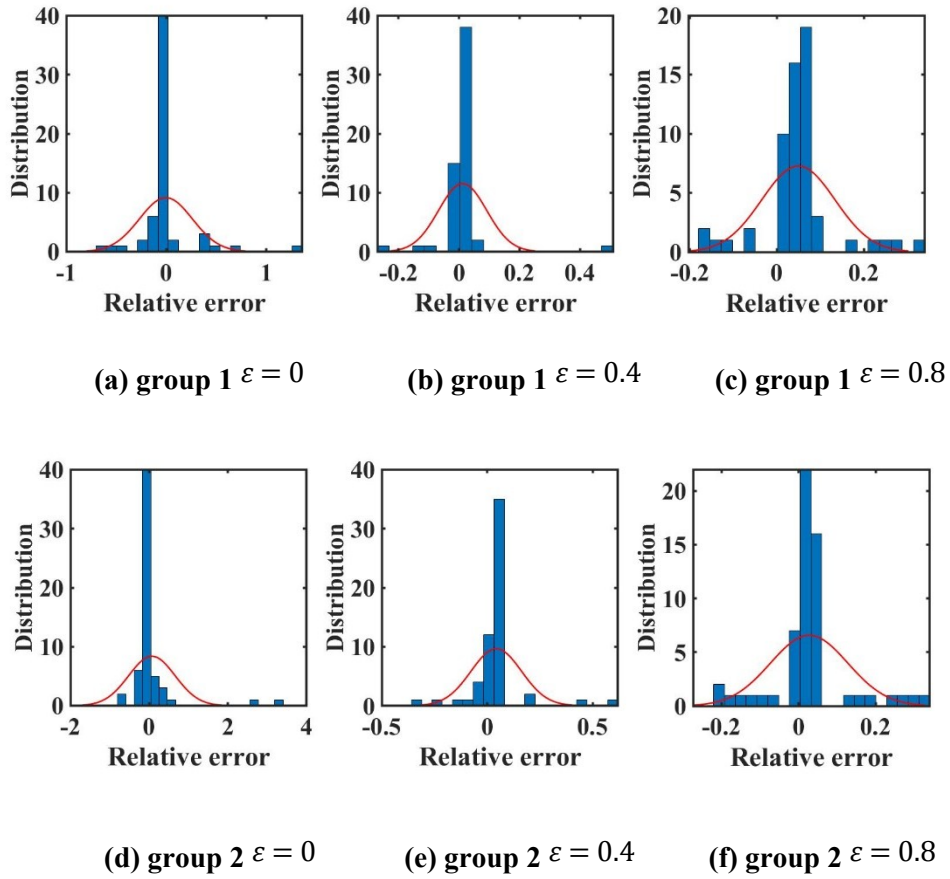


Figure S5. Distribution of deviations in different directions at the initial moment

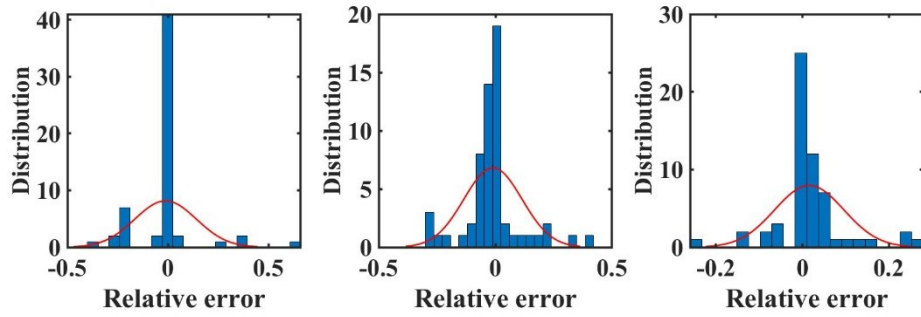




(g) group 3  $\varepsilon = 0$

(h) group 3  $\varepsilon = 0.4$

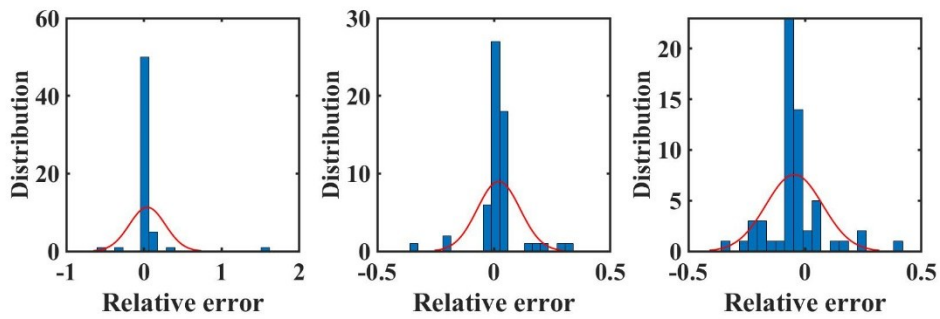
(i) group 3  $\varepsilon = 0.8$



(j) group 4  $\varepsilon = 0$

(k) group 4  $\varepsilon = 0.4$

(l) group 4  $\varepsilon = 0.8$



(m) group 5  $\varepsilon = 0$

(n) group 5  $\varepsilon = 0.4$

(o) group 5  $\varepsilon = 0.8$

Figure S6. Distribution of the relative errors of node voltages at different strains

The atomic configurations were dumped to compute the average distance  $d$  between graphene sheets during the deformation by the following way:

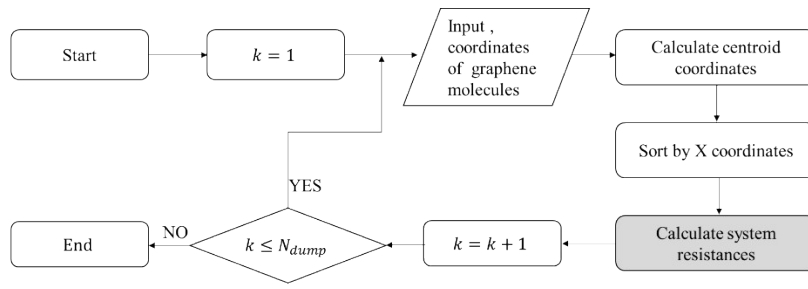
$$d = \frac{1}{N(N-1)} \sum_{i,j=1, i \neq j}^N \|\vec{x}_i - \vec{x}_j\|$$

, where  $N$  is the number of graphene molecules,  $\vec{x}_i$  and  $\vec{x}_j$  are the centroid coordinates of the  $i$ th and the  $j$ th graphene sheets, respectively.

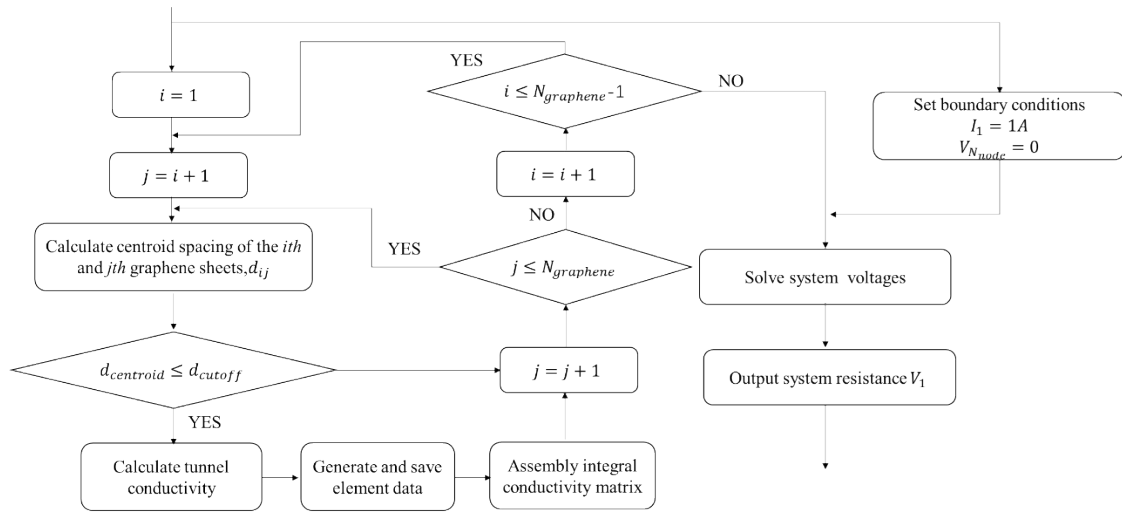
The strain-resistance responses of the system were studied in accordance with the algorithm presented in Scheme S1 (a), where  $N_{dump}$  is the number of atom configurations during the deformation. Based on the Kirchhoff's current law, the total current  $I$  under an applied voltage can be calculated with the incomplete Cholesky conjugate gradient method (ICCG). The details of calculating system resistances are shown in Scheme S1 (b), where  $d_{centroid}$  is the distance between the centroids of adjacent graphene sheets,  $d_{cutoff}$  is the upper bound distance of tunneling effects,  $N_{graphene}$  is the number of graphene sheets,  $N_{node}$  is the number of conductive nodes calculated while assembling the integral conductivity matrix,  $I_1$  and  $V_1$  are the total current and the voltage applied in x direction of the system, respectively. According to the Ohm's law, the macroscopic electrical resistance of the composites can be evaluated

$$R = \frac{V_1}{I_1} = V_1$$

as



(a)



(b)

**Scheme S1. (a) Flowchart to calculate the strain-resistance responses of the system. (b)**

**Detail on the box to calculate system resistances with grey background.**

## References

1. Yuan Q, Zhao Y P . Hydroelectric Voltage Generation Based on Water-Filled Single-Walled Carbon Nanotubes[J]. *Journal of the American Chemical Society*, 2009, 131(18):6374-6376.
2. Uddin M S, Ju J. Multiscale modeling of a natural rubber: Bridging a coarse-grained molecular model to the rubber network theory[J]. *Polymer*, 2016, 101: 34-47.
3. Li C , Browning A R , Christensen S , et al. Atomistic simulations on multilayer graphene reinforced epoxy composites[J]. *Composites Part A: Applied Science and Manufacturing*, 2012, 43(8).
4. Rahman R , Foster J T . Deformation mechanism of graphene in amorphous polyethylene: A molecular dynamics based study[J]. *Computational Materials Science*, 2014, 87:232-240.
5. Alian A R , Dewapriya M A N , Meguid S A . Molecular dynamics study of the reinforcement effect of graphene in multilayered polymer nanocomposites[J]. *Materials & Design*, 2017, 124:47-57.
6. Rissanou A , Power A , Harmandaris V . Structural and Dynamical Properties of Polyethylene/Graphene Nanocomposites through Molecular Dynamics Simulations[J]. *Polymers*, 2015, 7(3):390-417.
7. Li Y , Wang S , Wang Q . A molecular dynamics simulation study on enhancement of mechanical and tribological properties of polymer composites by introduction of graphene[J]. *Carbon*, 2017, 111:538-545.
8. Vu-Bac N , Lahmer T , Zhuang X , et al. A software framework for probabilistic sensitivity analysis for computationally expensive models[J]. *Advances in Engineering Software*, 2016, 100:19-31.
9. Hamdia K M , Silani M , Zhuang X , et al. Stochastic analysis of the fracture toughness of polymeric nanoparticle composites using polynomial chaos expansions[J]. *International Journal of Fracture*, 2017, 206(2):215-227.