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Multiple component correlation model for elastic modulus of single layer graphene sheets

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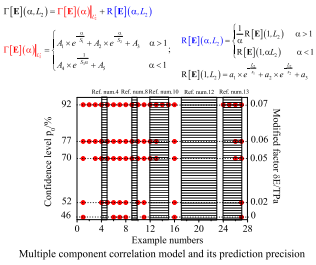
HIGHLIGHTS

- We present a multiple component correlation model to predict the elastic properties of SLGS.
- The correlation of geometric size, chirality and aspect ratio is considered in the model.
- The model has a high precision with a lower relative error and a higher confidence level.
- The model may be directly used to predict the mechanical properties of SLGS accurately.

GRAPHICAL ABSTRACT

Multiple component correlation model and its prediction precision.



Multiple component correlation model and its prediction precision

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ABSTRACT

A multiple component correlation model is first proposed to give an accurate prediction for the elastic modulus of single layer graphene sheets (SLGS). This model reflects the correlation of geometric size, chirality and aspect ratio by introducing two parameters, the graphene size and a coupling factor. The coupling factor may be used to reflect the chirality and aspect ratio simultaneously. Determination of the model coefficients is by using MD atomistic simulation. Precision of the prediction model is evaluated by relative error and confidence. The reliability of this model is verified by comparing with published data. It provides a convenient and effective prediction for elastic modulus of SLGS. It is beneficial to evaluate the complex macroscopic mechanical behavior of SLGS accurately.

1. Introduction

The single layer graphene sheets (SLGS) have unique physical properties especially the highest elastic modulus, thus various practical applications pay a lot of attention to them [1]. The elastic modulus of SLGS depends on chirality, geometric size and aspect ratio and so on [2–8]. Although a number of studies have been carried out on the elastic modulus of SLGS, they only take a single factor into account and are mostly qualitative research. Thus an accurate multi-factor correlation quantitative representation for the elastic modulus of SLGS is absent.

In recent research, the calculated values for the elastic modulus of SLGS range from 0.659 TPa [9] to 5.189 TPa [10]. The large discrepancy is due to different thicknesses [11]. In addition, different analysis methods, such as ab initio method [12–16], Molecular Dynamics (MD) simulation [17–22], Molecular Mechanics [23–26] and the tight-binding and bond-potential method [27–30], also may lead to different results. The possible reasons are analysis model inconformity, various force field and different force field equivalent models.

Lee et al. [31] acquired the elastic modulus of 1.024 TPa for a monolayer graphene sheet by using atomic force microscope nano-indentation experiment. Then some researchers recognized

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the elastic modulus of SLGS may be close to 1 TPa. However, Grantab et al. [32] presented the anomalous mechanical characteristics for graphene sheet with large-angle tilt boundaries. Thus, determination of the SLGS elastic modulus is a difficult problem. Dealing with this outstanding problem, it is important to research on macroscopic mechanical behavior of SLGS, such as thermal, vibration and structural instability characteristics of SLGS [33–36].

A unified model for the elastic modulus of SLGS is needed, which can avoid repeating experiments and simulation calculations. In this paper, a multiple component correlation model for the elastic modulus of SLGS is presented first. The coefficients of the model are determined by MD atomistic simulation. Then, this scheme is extended to predict Poisson's ratio and shear modulus. This model can reflect the parameters sensitivity effectively using relative error. In the end, compared with published data, the validation of this model is evaluated by confidence level.

2. Multiple component correlation model of elastic modulus

Based on previous references, the elastic modulus of SLGS is relevant to the chirality, structural size and aspect ratio. Thus we establish a function of elastic modulus associated with the above three structure characteristic parameters. The structure and chirality of SLGS are shown in Fig. 1.

We define a coupling factor $\alpha = L_1/L_2$. This coupling factor can reflect the chirality and aspect ratio simultaneously. $\alpha < 1$ is the Armchair-chirality and $\alpha > 1$ is the Zigzag-chirality. In addition, we use E_1 and E_2 to denote the elastic modulus along the Zigzag-chirality and Armchair-chirality directions respectively.

The elastic modulus of SLGS is expressed as a mathematical function $\Gamma[\boldsymbol{E}](\alpha, L_2)$, where $\boldsymbol{E} = (E_1, E_2)$. It was reported that the influence of chirality on elastic modulus may be much stronger [5–8]. So the function of elastic modulus is divided into two terms. One term is the univariate function $\Gamma[\boldsymbol{E}](\alpha)|_{L'_2}$, which denotes the elastic modulus of the standard SLGS with a fixed width L'_2. The other is a multivariate function $R[\boldsymbol{E}](\alpha, L_2)$, also named residual elastic modulus, for arbitrary structures size L_2. The residual elastic modulus is the elastic modulus of SLGS with different sizes and fixed aspect ratio minus the elastic modulus with special size L'_2 and the same fixed aspect ratio. Then we have

$$\Gamma[\boldsymbol{E}](\alpha, L_2) = \Gamma[\boldsymbol{E}](\alpha)|_{L'_2} + R[\boldsymbol{E}](\alpha, L_2) \tag{1}$$

We compute the elastic modulus of SLGS with different aspect ratios, chirality and structural size to determine the expression of Eq. (1). The MD simulation incorporating a software MS (Materials Studio by Accelrys Inc.) is used, which has been approved by ab initio computations [15] and nano-indentation measurements [37]. The COMPASS force field in Forcite module is used to simulate the interatomic interactions. The atomistic structure is optimized first. The elastic modulus of SLGS is computed by applying a strain amplitude of 0.003 with four steps for each strain to the cell.

The elastic modulus of SLGS with different structural sizes and aspect ratios is simulated using the above method. The elastic modulus versus structural size is plotted as Fig. 2. It shows that with the increase of structural size, elastic modulus increases first. Then it tends to be stable after reaching a critical value. This variation reveals a minimum size corresponding to a stable elastic modulus of SLGS. These variations agree well with the results in published literatures [17,19,23].

Thus, the elastic modulus of SLGS with different aspect ratios and chirality are calculated by MD simulation for fixed effective width size (1.968 nm according to Fig. 2). We determine $\Gamma[\boldsymbol{E}](\alpha)|_{L'_2}$ by fitting MD results in Appendix Table 1(a), expressed as

$$\Gamma[\boldsymbol{E}](\alpha)|_{L'_2} = \begin{cases} A_1 \times e^{-(\alpha/S_1)} + A_2 \times e^{-(\alpha/S_2)} + A_3 & \alpha > 1 \quad (a) \\ A_4 \times e^{-(1/S_3\alpha)} + A_5 & \alpha < 1 \quad (b) \end{cases} \tag{2}$$

Eq. (2) denotes a nonlinear exponential function. A_i and S_i are fitting coefficients with a precision 0.97.

Fig. 3 shows the residual elastic modulus for different coupling factors versus the standard SLGS with $\alpha = 1$. It indicates that the variation of residual elastic modulus versus different dimension sizes for arbitrary α has a special relation to $\alpha = 1$.

Thus the determination of $R[\boldsymbol{E}](\alpha, L_2)$ depends on $R[\boldsymbol{E}](1, L_2)$, that is, $\alpha = 1$. We have

$$R[\boldsymbol{E}](\alpha, L_2) = \begin{cases} (1/\alpha) R[\boldsymbol{E}](1, L_2) & \alpha > 1 \quad (a) \\ R[\boldsymbol{E}](1, \alpha L_2) & \alpha < 1 \quad (b) \end{cases} \tag{3}$$

Here $R[\boldsymbol{E}](1, L_2) = a_1 \times e^{-(L_2/s_1)} + a_2 \times e^{-(L_2/s_2)} + a_3$. The parameters a_i and s_i may be determined by fitting the data in Appendix Table 1(b) with a precision 0.97.

Further we have

$$\Gamma[\boldsymbol{E}](\alpha, L_2) = \begin{cases} A_1 \times e^{-(\alpha/S_1)} + A_2 \times e^{-(\alpha/S_2)} + A_3 + (1/\alpha)\left(a_1 \times e^{-(L_2/s_1)} + a_2 \times e^{-(L_2/s_2)} + a_3\right) & \alpha > 1 \quad (a) \\ A_4 \times e^{-(1/S_3\alpha)} + A_5 + \left(a_1 \times e^{-(\alpha L_2/s_1)} + a_2 \times e^{-(\alpha L_2/s_2)} + a_3\right) & \alpha < 1 \quad (b) \end{cases} \tag{4}$$

In addition, we also determine the functions of Poisson's ratio and shear modulus of SLGS with different chirality, structural sizes and aspect ratios.

$$\Gamma[\nu](\alpha, L_2) = \begin{cases} B_1 \times e^{-(\alpha/M_1)} + B_2 \times e^{-(\alpha/M_2)} + B_3 + (1/\alpha)\left(b_1 \times e^{-(L_2/m_1)} + b_2 \times e^{-(L_2/m_2)} + b_3\right) & \alpha > 1 \quad (a) \\ f\left[\frac{B_4}{(1+(1/M_3\alpha)^p)} + B_5\right] + g\left[B_6 \times e^{-((1/\alpha)-\omega)^2/M_4)} + B_7\right] + \left(b_1 \times e^{-(\alpha L_2/m_1)} + b_2 \times e^{-(\alpha L_2/m_2)} + b_3\right) & \alpha < 1 \quad (b) \end{cases} \tag{5}$$

$$\Gamma[\boldsymbol{G}](\alpha, L_2) = \begin{cases} C_1 \times e^{-(\alpha/N_1)} + C_2 \times e^{-(\alpha/N_2)} + C_3 + (1/\alpha)\left(c_1 \times e^{-(L_2/n_1)} + c_2 \times e^{-(L_2/n_2)} + c_3\right) & \alpha > 1 \quad (a) \\ C_4 + \left(c_1 \times e^{-(\alpha L_2/n_1)} + c_2 \times e^{-(\alpha L_2/n_2)} + c_3\right) & \alpha < 1 \quad (b) \end{cases} \tag{6}$$

The parameters B_i, M_i, C_i, N_i, b_i, ω, b_i, m_i, c_i and n_i are determined by fitting the data in Appendix Table 1(a) and (b) with a precision 0.95.

3. Results and discussions

3.1. Example 1

We have $L_1 = 8.096$ nm and $L_2 = 3.936$ nm, that is, $\alpha = 2 > 1$. So we may determine the elastic modulus of SLGS based on Eq. 4 (a) and the coefficients in Appendix Table 2(a), $E_1 = 1.134$ TPa and $E_2 = 1.158$ TPa. In a similar way, we have the Poisson's ratio and shearing modulus, $\nu_{12} = 0.439$, $\nu_{21} = 0.449$ and $G_{12} = 0.397$ TPa based on Eqs. 5(a) and 6(a) and the data in Appendix Table 2 (b) and (c).