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Wrinkling and wrinkling-suppression in graphene membranes with frozen zone

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ABSTRACT

The stretch-induced wrinkling behaviors in graphene membranes with frozen zone are investigated in molecular dynamics simulations and suppressed by adopting optimal designs obtained in topology optimization. The graphene membranes with optimal designs under stretching are wrinkle-free, this is meaningful to ensure the exceptional performances of graphene-nanoelectronics. Both the wrinkling and wrinkling-suppression mechanisms are qualitatively and quantitatively elucidated with finite element analyses, which are helpful to understand the stretch-induced wrinkling in graphene membranes with frozen zone and to provide useful guidance to the design of the large-area graphene-nanoelectronics.

1. Introduction

Graphene [1,2] has promising applications in nanoelectronics due to its excellent mechanical, optical and electronic properties. However, as nature's thinnest material with one-atom thickness, graphene has an ignorable out-of-plane stiffness and is prone to wrinkling. The interplay between the wrinkled morphology and the electronic property [3,4], the thermal conductivity [5], the electrical property [6] and the charge transport [7] shows that wrinkles considerably affect performances of graphene and hinder its applications in nanoelectronics. Therefore, it is highly desired to systematically investigate the wrinkling behaviors of graphene, and more importantly, to deliberately tune wrinkling for achieving the wrinkle-free graphene-nanoelectronics.

To study the wrinkling behaviors of graphene, extensive researches have been carried out. Different mechanical wrinkling behaviors related to compression [8,9], shear [10–12], torsion [13], bending [14], local tension [15,16], local uplift [17], folding [18] are considered. However, with these extensive mechanical wrinkling studies, the stretch-induced wrinkling behavior with both ends clamped, commonly observed in macroscale membranes [19–24], has rarely been addressed for graphene membranes. Moreover, in these existing wrinkling studies, graphene membranes are mostly freestanding. On the contrary, in nanoelectronics, some regions of a graphene membrane might act as functional components or are directly attached to the substrate [25]. These regions, referred to as frozen zone, restrict deformations in their vicinities to ensure the performance of nanoelectronics, while the rest regions exhibit deformations to release strain. Whether and how wrinkles occur in graphene membranes with frozen zone are still unclear.

Tuning wrinkling is an important issue in the wrinkling studies of graphene membranes. For example, predefined defects [26], guiding paths [27] and external harmonic excitations [28] have been adopted to generate wrinkles of desired patterns. Optimized transfer process [29,30], thermal excitation [31–33], surface smoothing [29,30,32,34] and interfacial adhesion modification [29,35] have been utilized to suppress wrinkling for implementing the wrinkle-free graphene-nanoelectronics. However, these approaches either introduce new wrinkle patterns or require additional physical/chemical treatments. Recently, Bonin and Seffen [36] mitigate wrinkles and obtain maximal reflectance area of a membrane by pressuring the membrane into the out-of-plane parabolic shape. Yan et al. introduce holes [37] and square rigid elements [38] into membranes and report that the wrinkling patterns are tunable by considering the distributions and parameters of holes and rigid elements. These findings indicate that that wrinkling is tunable through structural design without requiring additional treatments. Therefore, it is promising to tune wrinkling in graphene membranes by finding optimal designs through design approaches, such as the well-known topology optimization [39].

In this study, as illustrated in Fig. 1, the stretch-induced wrinkling behaviors of graphene membranes with frozen zone is explored in molecular dynamics (MD) simulations. Then, using the topology optimization technique, we propose the optimal designs of considered graphene membranes with frozen zone, the related wrinkle-free capabilities are verified with MD simulations. Finally, both wrinkling and wrinkling-

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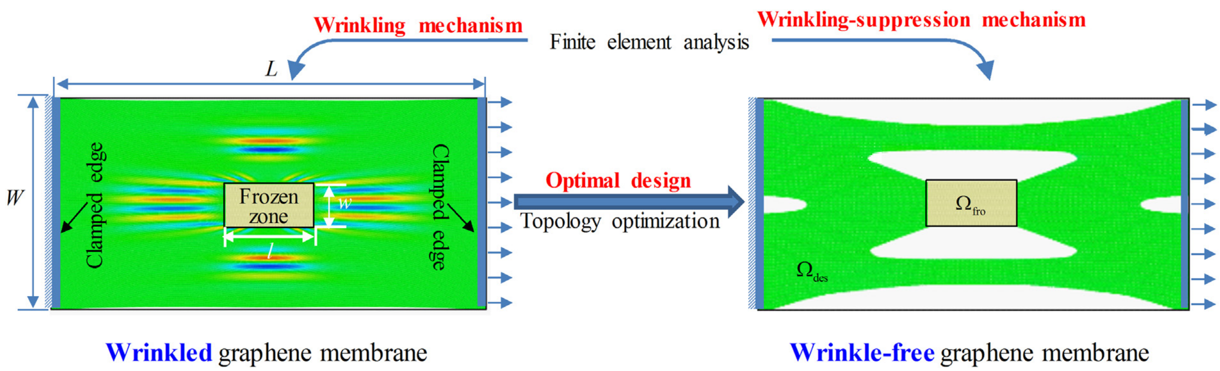

Fig. 1. Wrinkling and wrinkling-suppression in graphene membranes with frozen zone under in-plane stretch.

suppression mechanisms for graphene membranes of the original and the optimal designs are qualitatively and quantitatively elucidated with the finite element analysis (FEA), respectively.

2. Methods

2.1. Molecular dynamics (MD) simulations

The single-layer graphene membranes with a constant length $L = 200$ nm are stretched with LAMMPS [40] and analyzed in OVITO [41]. The C-C interactions are characterized by the AIREBO potential [42–44], in which $\varepsilon = 2.4$ meV and $\sigma = 0.34$ nm. As illustrated in Fig. 1, both the clamped edges (2 nm in the stretching direction) and the frozen zone Ω_{fro} are fixed and modeled with NVE ensembles, while the rest atoms in the design domain Ω_{des} are described by NVT ensembles to keep the temperature at 0 K to avoid severe thermal oscillations. The out-of-plane deformations at the clamped edges and the frozen zone are prohibited during whole process to avoid undesired perturbations. The in-plane longitudinal stretch with a loading rate of 0.01 Å/ps and a time step of 0.05 fs is applied, this rate is sufficiently low to reflect the quasi-static wrinkling behaviors.

2.2. Finite element analysis (FEA)

We perform FEA with ABAQUS [45] to elucidate both the underlying wrinkling and wrinkling-suppression mechanisms. Graphene membranes, with the Young's modulus $E_g = 0.95$ TPa and the Poisson's ratio $v_g = 0.17$ [46], are modeled with plane-stress (CPS4R) elements under the wrinkle-free assumption. With frozen zone Ω_{fro} fixed, nonlinear deformation analyses are carried out under stretching. The obtained distributions of $\sigma_{\min}$, the minimum in-plane principal stress, are utilized to assess the onset of wrinkling, since compressive $\sigma_{\min}$ is the prerequisite of wrinkling according to the wrinkling criterion [47] as

$$\begin{aligned} &\text{If } \sigma_{\min}>0, \text{ taut} \\ &\text{If } \sigma_{\min}\le 0 \text{ and } \sigma_{\max}>0, \text{ wrinkled} \\ &\text{If } \sigma_{\min}<0 \text{ and } \sigma_{\max}\le 0, \text{ slack} \end{aligned} \tag{1}$$

where $\sigma_{\max}$ is the maximum in-plane principal stress. This wrinkling criterion implies that no wrinkles occur if the magnitudes of $\sigma_{\min}$ are positive for all the elements in the considered graphene membranes.

2.3. Topology optimization

The topology optimization problem is to find the optimal material distributions in the design domain Ω_{des} of a graphene membrane, while Ω_{fro} is frozen. Using the solid isotropic material with penalty (SIMP) approach [48,49], we relate the relative density of each element in Ω_{des} with a continuous variable $\rho_e \in [0, 1]$. Here $\rho_e = 1$ indicates that the eth element is occupied by the graphene membrane material, while $\rho_e = 0$ means void. Hence, the Young's modulus of the eth element E_e can be expressed as

$$E_e = {\rho_e}^p E_g \quad (e = 1, 2, \ldots, N) \tag{2}$$

where p is the penalization factor, N is the total number of elements in Ω_{des}. It is noted that the intermediate densities $0<\rho_e<1$ have no real physical significance in structural optimization, they are employed to relax the discrete topological problems with continuous design variables, which can be solved using continuous mathematical programming. To discourage the formation of intermediate densities, penalty scheme is employed to drive the solution to the 0/1 layout and the macroscopic elasticity tensor of element is expressed as a power-law interpolation function of the relative density ρ_e as stated in Eq. (2). The usual value $p = 3$ is chosen for the penalization factor.

The topology optimization formulation for finding the optimal design of a graphene membrane with frozen zone is mathematically stated as

$$\begin{aligned} &\text{Find} \quad \boldsymbol{\rho} = [\rho_1, \rho_2, \ldots, \rho_N] \\ &\text{max} \quad W = \boldsymbol{F}_{\Gamma_T}^{\mathrm{T}} \boldsymbol{u}_{\Gamma_T} \\ &\text{s.t.} \quad {}_{\min,e}(\boldsymbol{u})>0 \qquad (e = 1, 2, \ldots, N) \\ &\qquad A = \sum_{e=1}^{e=N} \rho_e A_e + A_{\text{fro}} \le A^* \\ &\qquad 0<\rho_{\min} \le \rho_e \le 1 \qquad (e = 1, 2, \ldots, N) \end{aligned} \tag{3}$$

where $\boldsymbol{\rho}, W, \boldsymbol{u}, \sigma_{\min,e}$ denote the vector of design variables, the external work done by the reaction forces $\boldsymbol{F}_{\Gamma_T}$ applied on the stretched boundaries Γ_T, the displacement vector retrieved from the plane-stress analysis, and the minimum in-plane principal stress of the eth element, respectively. The stress constraint ${}_{\min,e}(\boldsymbol{u})>0$ is imposed on each element to ensure the taut state of the whole domain to suppress wrinkling. The constraint $A \le A^*$ imposes a restriction on the areal coverage of graphene membrane materials, where A_e, A_{fro} and A^* are the area of the eth element, the area of the frozen zone Ω_{fro}, and the maximum admissible total area, respectively. In this study, A^* is fixed as 75% of the whole structural area. The lower bound of the design variables is defined as $\rho_{\min} = 0.001$ to avoid numerical singularity in FEA.

The optimization model defined by Eq. (3) is solved by using the method of moving asymptotes (MMA) [50]. In each optimization loop, design sensitivity analysis based on the adjoint variable method provides necessary information for updating the design variables. The cosine-type relaxation scheme and the well-known sensitivity filter technique [51] are employed to avoid the stress singularity phenomenon and the checkerboard problem in topology optimization. The optimization iteration procedures will be terminated when the change of adjacent design variables meets a prescribed convergence criterion.