Shape Optimum Design of Graphene Sheets

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1. Abstract
Graphene sheet (GS) is a monolayer of carbon atoms arranged in a honeycomb lattice and is the strongest material ever measured and the thinnest material ever synthesized in the universe. Due to its unique mechanical, structural and electronic properties, GS is supposed to be a base material for nanoelectromechanical systems (NEMS), given that lightness and stiffness are the essential characteristics sought in NEMS for sensing applications. In this study, shape optimum design of GS is carried out to improve its stiffness for these applications. At first, we model C-C bond as an equivalent continuum beam by means of molecular mechanics (MM) method. So that GS can be adopted as a continuum frame structure. Then, we optimize the shape of the atomistic finite element model based on a free-form optimization method for frame structures. In the optimization process, we use the compliance as objective function and minimize it under the volume constraint. Each equivalent continuum beam is assumed to vary in the off-axis direction to the centroidal axis and we derive the shape gradient function for determination of the optimal design velocity field based on the free-form optimization method. Using the derived optimal design velocity field, the shape optimum design of GS can be carried out without shape parametrization. The numerical results show that, using the proposed shape optimization method, the compliance of GS can be significantly reduced that would be helpful for designing GS used in NEMS.
2. Keywords: Compliance; Free-form; Graphene sheets; Molecular mechanics; Shape optimum design.

3. Introduction
Due to its large specific surface area, high intrinsic mobility, high Young's modulus and thermal conductivity, Graphene sheet (GS), a one-atomic-thick monolayer of graphite, has been proposed to be used in nanoelectromechanical systems [1]. For a broad range of industrial applications of GSs, the prediction of mechanical properties for the perfect 2D nanostructure, such as the stiffness, vibration characteristics and buckling analysis, have been carried out by means of experiment [2], *ab initio* energy calculation [3] or molecular dynamics (MD) simulation [4], molecular mechanics (MM) method [5] and continuum mechanics [6]. Conduction experiments with nano-size systems are difficult and expensive, while MD methods are time consuming and have convergence problems. Thus, developing appropriate continuum mathematical models based on MM method and continuum mechanics for nanostructures is important for the development of GSs [7].
Here, we need to introduce MM method for modeling frame-like structure of GSs. For a GS can be treated as a large array of molecule consisting of C atoms, MM method has played important roles for modeling GSs. From the viewpoint of MM method using the equivalent atomistic based continuum mechanics, MM method depicts the forces between individual atoms as typical beam elements (shown as Fig. 1). According to the Tersoff-Brenner force field theory [8] and a link between molecular and solid mechanics of C-C bond, we assume the equivalent C-C beam with circular cross-section of diameter d and initial length 1.42 Å, and get the proposed material constants are Young's modulus E_b = 5.53 TPa, shear modulus G_b = 0.871 TPa and d = 1.46 Å [9].

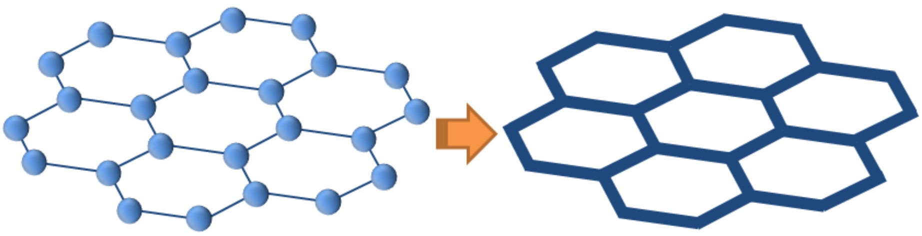

Figure 1 Molecular mechanics method for graphene sheets

Recently, the shape of GSs can be controlled by an external electric field [10] or chemically modifying the adherence of GSs on metal [11]. Hence, shape optimum design of GSs can make an effective role to improve their mechanical behaviors.

Shape optimum design of traditional continuum structures such as shell, solid or frame structures have been carried out based on the free-form optimization method. The free-form optimization method is a gradient method with a P. D. E. (Partial Differential Equation) smoother in the Hilbert space for shape optimization of continua, which is also called H^1 gradient method or traction method and does not require any shape parameterization. This method was firstly proposed by Azegami [12] and Shimoda et al. developed this optimization method for designing frame [13], shell [14] and solid structures [15]. The free-form optimization method is a parameter-free or a node-based shape optimization method that treats all nodes in the body as design variables. The advantages of this method include efficiency for treating large-scale problems and the ability to obtain a smooth shape. Using the free-form optimization method, it is possible to obtain the optimized shapes of frame-like structures of GSs.

The present work is arranged as following. In section 4, we introduce the MM method for GSs and build the frame-like continuum model of GSs at first. Then, we use a developed free-form shape optimization method for the frame structure and build the shape optimization system for designing the shape of GSs. Using the shape optimization method, we carry out two examples to do shape optimum design of GSs in section 5. At last, conclusions are presented in section 6.

4. Shape optimum design of graphene sheets

Based on the MM method, we assemble the frame-like GS finite element models for shape optimization in the present work. Thus, shape optimum design of GSs can be simplified to a shape optimization problem of frame structures as shown in Fig. 2.

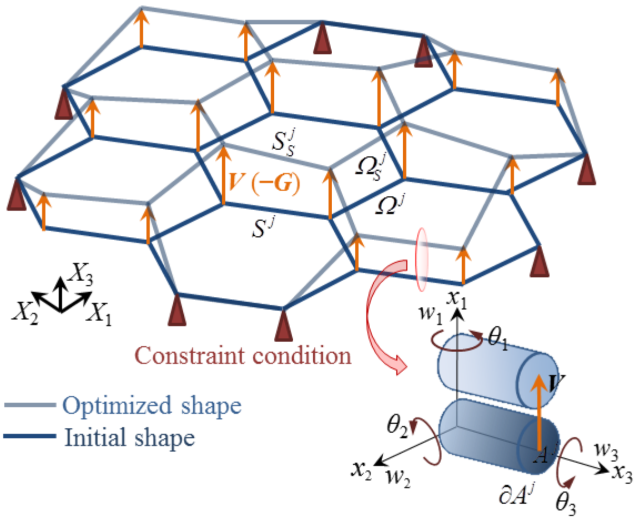

Figure 2 Shape variation of frame structure of graphene sheets

4.1. Domain variation of frame structure of graphene sheets

As shown in Fig. 2, members $\Omega^j, j=1,2,...,N$ consisting of Timoshenko beams compose a frame structure of GSs that can be represented by a bounded domain $\Omega \subset \mathbb{R}^3$, where N is the number of beams and $\mathbb{R}$ is a set of positive real numbers. The notations (x_1,x_2,x_3) and (X_1,X_2,X_3) indicate the local coordinate system with respect to the beam and the global coordinate system, respectively. Hence, we have

$$\Omega^j=\left\{\left(x_1^j,x_2^j,x_3^j\right)\in\mathbb{R}^3\left|\left(x_1^j,x_2^j\right)\in A^j\subset\mathbb{R}^2, x_3^j\in S^j\subset\mathbb{R}\right.\right\},\quad \Gamma^j=\partial A^j\times S^j \qquad \Omega^j=A^j\times S^j \tag{1}$$

where S^j, Γ^j and Ω^j express the centroidal axis, circumference surface and whole domain of member *j*, respectively. A^j and ∂A^j express the cross section and its circumference of member *j*, respectively. The subscript *j* shall be frequently omitted to avoid the complexity of expression in the sequel. $\boldsymbol{w}=\{w_i\}_{i=1,2,3}$ and $\boldsymbol{\theta}=\{\theta_i\}_{i=1,2,3}$ express displacement vector and rotation vector in the x_1,x_2,x_3 directions of the local coordinate system, respectively. Then, The weak form governing equation in terms of $(\boldsymbol{w},\boldsymbol{\theta})$ can be expressed as

$$a\left((\boldsymbol{w},\boldsymbol{\theta}),(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})\right)=l\left((\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})\right),\ \forall(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})\in U,\ (\boldsymbol{w},\boldsymbol{\theta})\in U \tag{2}$$

where the notation $\bar{(\cdot)}$ expresses a variation, and U expresses admissible function space in which the given constraint conditions of $(\boldsymbol{w},\boldsymbol{\theta})$ are satisfied. In the frame structure of GSs as shown in Fig. 2, due to the domain variation $\boldsymbol{V}$ (design velocity field) in the out-of-plane direction to the centroidal axis, the initial domain Ω^j and a centroidal axis S^j of member *j* become Ω_s^j and S_s^j, respectively. The subscript *s* expresses the iteration history of the domain variation.

4.2. Compliance minimization problem

We utilize a free-form optimization method for frame structures to minimize the compliance of the frame structure of GSs, the shape optimization problem for finding the optimal design velocity field $\boldsymbol{V}$ can be formulated as

$$\text{Given} \quad \Omega \tag{3}$$

$$\text{Find} \quad \boldsymbol{V} \text{ or } \Omega_s \tag{4}$$

$$\text{that minimizes} \quad l(\boldsymbol{w},\boldsymbol{\theta}) \tag{5}$$

$$\text{subject to} \quad \text{Eq. (2) and } M\left(=\sum_{j=1}^{N}\int_{S^j} A dS\right) \le \hat{M} \tag{6}$$

where M and $\hat{M}$ denote the volume and its constraint value, respectively.

Letting $(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})$ and Λ denote the Lagrange multipliers for the state equation and volume constraints, respectively, the Lagrange functional L associated with compliance minimization problem can be expressed as

$$L\left(\Omega,(\boldsymbol{w},\boldsymbol{\theta}),(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}}),\Lambda\right)=l\left((\boldsymbol{w},\boldsymbol{\theta})\right)+l\left((\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})\right)-a\left((\boldsymbol{w},\boldsymbol{\theta}),(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})\right)+\Lambda\left(M-\hat{M}\right) \tag{7}$$

The material derivative $\dot{L}$ of the Lagrange functional can be derived as

$$\dot{L}=l((\bar{\boldsymbol{w}}',\bar{\boldsymbol{\theta}}'))+l((\boldsymbol{w}',\boldsymbol{\theta}'))-a((\boldsymbol{w}',\boldsymbol{\theta}'),(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}}))-a((\boldsymbol{w},\boldsymbol{\theta}),(\bar{\boldsymbol{w}}',\bar{\boldsymbol{\theta}}'))+\dot{\Lambda}\left(M-\hat{M}\right)+\langle G\boldsymbol{n},\boldsymbol{V}\rangle, \quad \boldsymbol{V}\in C_\Theta \tag{8}$$

where $G\boldsymbol{n}\ (\equiv \boldsymbol{G})$ expresses the shape gradient function (i.e., sensitivity function), which is a coefficient function in terms of $\boldsymbol{V}$. $\boldsymbol{n}$ is defined as an outward unit normal vector on the circumference surface Γ or as a unit normal vector on the centroidal axis *S*. The notations $(\cdot)'$ and $\dot{(\cdot)}$ are the shape derivative and the material derivative with respect to the domain variation, respectively.

The optimum conditions of the Lagrange functional L with respect to $(\boldsymbol{w},\boldsymbol{\theta})$, $(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})$ and Λ are expressed as

$$a\left((\boldsymbol{w},\boldsymbol{\theta}),(\bar{\boldsymbol{w}}',\bar{\boldsymbol{\theta}}')\right)=l\left((\bar{\boldsymbol{w}}',\bar{\boldsymbol{\theta}}')\right), \quad \forall(\bar{\boldsymbol{w}}',\bar{\boldsymbol{\theta}}')\in U \tag{9}$$

$$a\left((\boldsymbol{w}',\boldsymbol{\theta}'),(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})\right)=l\left((\boldsymbol{w}',\boldsymbol{\theta}')\right), \quad \forall(\boldsymbol{w}',\boldsymbol{\theta}')\in U \tag{10}$$

$$\dot{\Lambda}\left(M-\hat{M}\right)=0 \quad M-\hat{M}\le 0 \quad \Lambda\ge 0 \tag{11}$$

When the optimality conditions are satisfied, Assuming that the external forces do not vary with regard to the space and the iteration history s and considering the self-adjoint relationship $(\boldsymbol{w},\boldsymbol{\theta})=(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})$, which is obtained from Eqs. (9) and (10), we get

$$\dot{L}=\langle G\boldsymbol{n},\boldsymbol{V}\rangle=\sum_{j=1}^{N}\left\{\int_{S^j} G_1\boldsymbol{V}\cdot\boldsymbol{n}dS+\int_{S^j} G_0\boldsymbol{V}\cdot\boldsymbol{n}dS\right\} \tag{12}$$

$$G_1=2h_1h_2\left\{Ew_{3,3}\theta_{2,3}-\mu\theta_{3,3}\left(w_{2,3}+\theta_1\right)\right\} \tag{13}$$

$$G_0=\Lambda AH+\left\{2\left(F_iw_i+C_1\theta_1-C_2\theta_2+C_3\theta_3\right)H\right\} \tag{14}$$

where $\boldsymbol{F}=\{F_i\}_{i=1,2,3}$ and $\boldsymbol{C}=\{C_i\}_{i=1,2,3}$ are the force and couple vectors per unit length applied to member *j*, respectively. The notation μ is the Lame constant and E is the Young's modulus. H denotes the curvature of the centroidal axis. Moreover, the tensor subscript notation uses Einstein's summation convention and a partial differential notation for the spatial coordinates $(\cdot)_{,i}=\partial(\cdot)/\partial x_i$.

4.3. Free-form optimization method for frame structures of graphene sheets

The free-form optimization method described here was proposed by Shimoda for solving the shape optimization problem of frame structures [13]. In this method, the negative shape gradient function $-\boldsymbol{G}(=-G\boldsymbol{n})$ is applied as a distributed force to a pseudo-elastic frame structure in the normal direction to the centroidal axis (shown as Fig. 2). This makes it possible both to reduce the objective functional and to maintain smoothness, i.e., mesh regularity simultaneously. The optimal shape variation, or the optimal design velocity field $\boldsymbol{V}$ is determined as the displacement field in this pseudo-elastic frame analysis, and the obtained $\boldsymbol{V}$ is used to update the shape. We call this analysis for $\boldsymbol{V}$ velocity analysis. In other words, this method is a gradient method in a Hilbert space with Laplacian smoother. The stiffness tensor of the pseudo-elastic frame structure has a role of the positive definiteness tensor, which is needed in a gradient method in a function space. The governing equation of the velocity analysis is expressed as

$$a\left((\boldsymbol{V},\boldsymbol{\theta}),(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})\right)=-\left\langle G\boldsymbol{n},(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})\right\rangle,\ \forall(\bar{\boldsymbol{w}},\bar{\boldsymbol{\theta}})\in C_\Theta,\ (\boldsymbol{V},\boldsymbol{\theta})\in C_\Theta \tag{15}$$

$$C_\Theta=\left\{(V_1,V_2,V_3,\theta_1,\theta_2,\theta_3)\in\left(H^1(S)\right)^6 \middle| \text{satisfy Dirichlet condition for shape variation}\right\} \tag{16}$$

In problems where convexity is assured, this relationship definitely reduces the Lagrange functional in the process of updating the shape of GSs using the design velocity field $\boldsymbol{V}$ determined from Eq. (16).

The shape optimization process for frame structure of GSs is built by repeating stiffness analysis, sensitivity analysis for calculating the shape gradient functions, velocity analysis and shape updating, in which stiffness analysis and velocity analysis are conducted using a standard commercial FEM code.

5. Results and discussion

In order to evaluate the shape optimization process for frame structure of GSs, we execute two examples to optimize the shape a rectangular GS and a circular GS. The volume constraint is set to be $M \le \hat{M} = 1.05M_{\text{initial}}$, where M_{initial} is the initial volume of GSs. It should be noted that the constraint conditions utilized in the present work are expressed as 1 (x_1 direction), 2 (x_2 direction), 3 (x_3 direction), 4 (θ_1 direction), 5 (θ_2 direction) and 6 (θ_3 direction).

5.1. Example 1

In this example, we built a frame model of rectangular GS containing 2006 carbon atoms and 2946 equivalent C-C beams. In the structural analysis shown in Fig. 4 (a), three corner points are constrained in 123 and the remaining one point undergoes a concentrated force. In the velocity analysis shown in Fig. 4 (b), all of the four sides of the rectangular GS are constrained in 123 and all of the remained nodes are constrained in 12. The shape optimum design of the rectangular GS is carried out using the proposed shape optimization process. The optimized shape and the iteration history are expressed in Figs. 4 (c) and (d), respectively. The optimized shape of GS is smooth and the iteration history shows that the compliance ratio is reduced by 97.4% normalized to the initial shape. The optimization process converges according to the volume constraint.

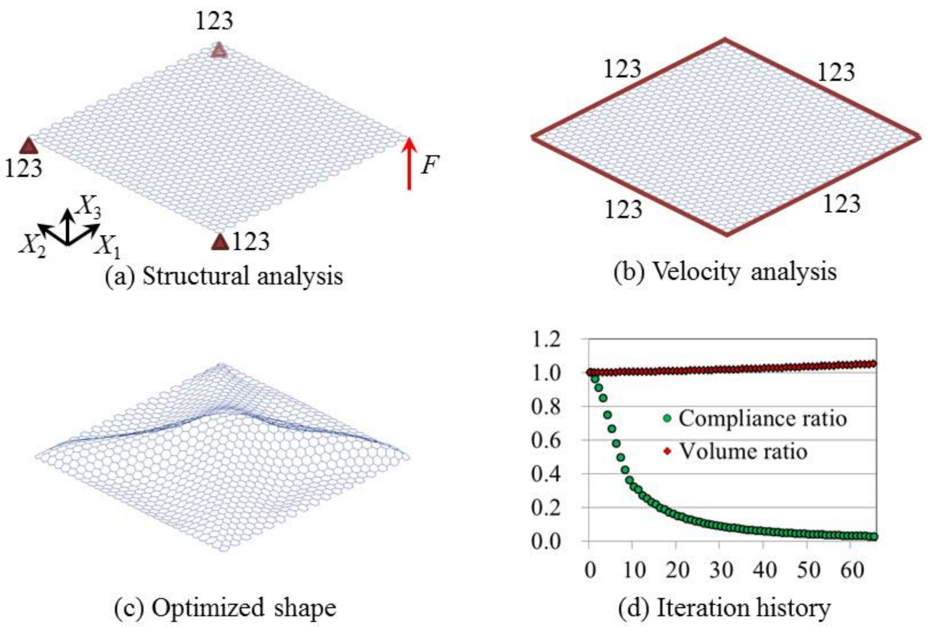

Figure 4 Shape optimization for a rectangular graphene sheet

5.1. Example 2

A frame model of circular GS containing 3120 carbon atoms and 4607 equivalent C-C beams is built in this example. In both of the structural analysis and the velocity analysis shown in Figs. 5 (a) and (b), the edge of the circular GS is constrained in 123. Moreover, all of the remained nodes are constrained in 12 in the velocity analysis. There is a linear nodal forces acting on the surface of the circular GS in the structural analysis. Using the proposed shape optimization process, the shape optimum design of the circular GS is carried out. The optimized shape and the iteration history are shown in Figs. 5 (c) and (d), respectively. The optimization process converges according to the volume constraint, and we obtain a smooth optimized shape of the circular GS .In the iteration history expressed in Fig. 5 (d), the compliance ratio is reduced by 55.5% normalized to the initial shape and the volume ratio goes up to 1.05 M_{initial} .

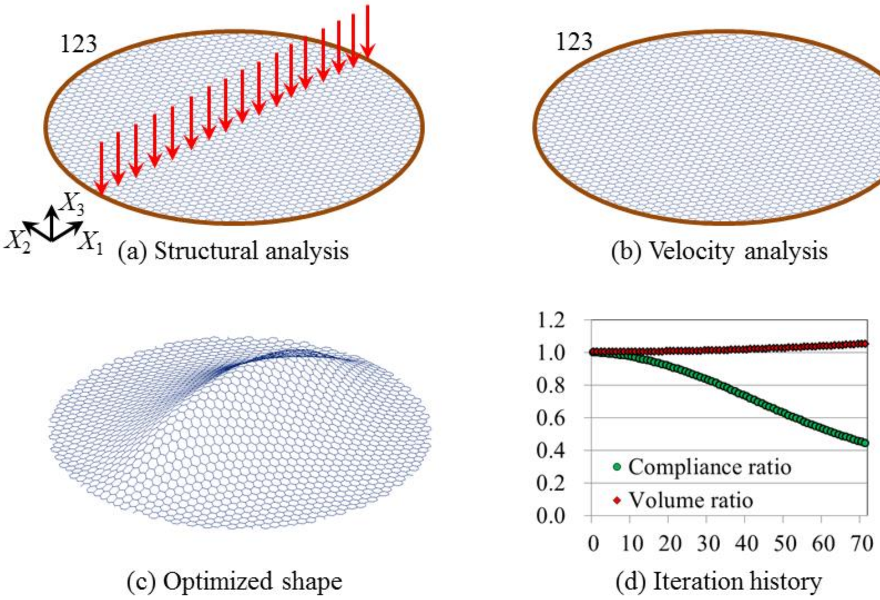

Figure 5 Shape optimization for a circular graphene sheet

6. Conclusions

In the present work, we built the frame-like continuum mechanical model of GSs based on the MM method at first. Then, we formulated the compliance minimization problem of the frame structure of GSs and adopted a shape optimization method that can be used in the optimum design of GSs. This method was developed based on the free-form optimization method, so that the optimized shape of GSs could be determined without re-mesh and requiring shape design parameterization. The objective of the developed optimization method was to minimize the compliance of GSs under the volume constrain. To confirm the effectiveness of the proposed shape optimization method, two examples were carried out using the developed free-form shape optimization method. The results showed that the obtained optimal shapes in both of the two design problems were smooth and the compliance of each case was reduced significantly.

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