Meshfree Simulation of Compressive Buckling of Single-Walled Carbon Nanotubes Using Higher Order Cauchy-Born Rule

Yuzhou Sun and K. M. Liew

***Abstract* — A meshfree computational scheme is developed to study the compressive deformation of single-walled carbon nanotubes (SWCNTs). The continuum constitutive relationship is based on an extended Cauchy-Born rule, the higher order Cauchy-Born rule, in which the atomic-scale deformed lattice vectors are calculated with both the first and second order deformation gradients. The compression of CNTs is simulated, and the buckling behavior is displayed accurately. A numerical simulation is also carried out with the application of the standard Cauchy-Born rule, and the comparison indicates that consideration of the second-order deformation gradient is necessary for the continuum simulation of CNTs.**

***Index Terms*— Carbon nanotubes; Higher order Cauchy-Born rule; Continuum simulation; Meshfree method; Compression buckling.**

I. Introduction

Due to the massive computational cost of the full atomistic simulation, the equivalent continuum models play an important role in understanding the properties of the nano-scale structures. The Cauchy-Born rule [1] is a fundamental kinematic assumption which establishes a connection between the deformation of the lattice vector of an atomistic system and that of a continuum displacement field. The first use of this method at the nano-scale emerged from the quasi-continuum method [2] for two-dimensional problems. Later, Zhang *et al.* [3] and Zhang *et al.* [4] extended the approach to CNTs and proposed a nanoscale continuum theory by incorporating interatomic potentials into a continuum model. However, as Arroyo and Belytschko [5,6] indicated, the application of the classical Cauchy-Born rule to CNTs may be not suitable because a CNT is essentially a curved crystalline sheet with of an atom's thickness, and the curvature effect has to be accounted for. In [5,6], they systematically formulated an exponential Cauchy-Born rule for finite deformation membrane and CNTs. With the direct application of both the first and second deformation gradients, Guo *et al.* [7] and Wang *et al.* [8] recently proposed a higher order Cauchy-Born rule, in which no concept of curvatures is involved and the second deformation gradient is directly considered. By considering a CNT being formed by rolling a graphite sheet into a cylindrical shape, they [7,8] studied the elastic properties of single-walled CNTs.

This paper concerns the computational validity of the constitutive model based on the higher order Cauchy-Born rule in the excellent works [7,8]. In the analysis, the Tersoff-Brenner [9,10] potential is employed as the interatomic potential, and the second set of parameters [9,10] for this potential function is used. A meshfree computational scheme is developed to implement the numerical computation of the constitutive model based on the higher order Cauchy-Born rule. As the second deformation gradient is involved in the present theory, the finite element method generally requires C^1-continuity of the interpolation of displacements [11]. This leads to a challenging difficulty in the establishment of elements and the construction of the interpolation functions. Recently, researchers [11] applied the meshfree method [12-15] to simulate the materials with strain-gradients effects. A distinct advantage of this approach is that the meshfree approximations possess non-local properties and satisfy the higher order continuity requirement [11,13]. This intrinsic non-local property leads to real rotation-free approximation, and displacements can thus be used as the only nodal freedoms [11].

The compression of carbon nanotubes is numerically simulated, and the buckling is studied. The numerical results show a good agreement with those obtained by the atomistic simulation. The obtained buckling pattern also agrees well with that obtained by atomistic simulation. In addition, the constitutive model based the Cauchy-Born rule (whereby no second-order gradient is used) is implemented with the meshfree method, and an unphysical buckling shape is obtained. The comparison reveals that the Cauchy-Born rule cannot accurately display the deformation behavior of CNTs because it

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only considers the first-order deformation gradient and not the effect of bending stiffness.

II. Hyper-Elastic Constitutive Model

A. Higher order Cauchy-Born rule

In general, the Cauchy-Born rule [1] rule describes the deformation of lattice vectors as

$$\mathbf{b} = \mathbf{F} \bullet \mathbf{a}, \tag{1}$$

where $\mathbf{F}$ is the two-point deformation gradient tensor, $\mathbf{a}$ is the undeformed lattice vector, and $\mathbf{b}$ is the corresponding deformed lattice vector.

The Cauchy-Born rule has also been used to study the properties CNTs [3,4]. However, as Arroyo and Belytschko [5,6] indicated, the direct application is not suitable to curved crystalline films of one or more atom in thickness. This is mainly because the deformation gradient tensor $\mathbf{F}$ describes only the change of infinitesimal material vectors that emanate from the same point in tangential spaces of the undeformed and deformed curved manifolds. By incorporating the effect of the curvature, Arroyo and Belytschko [5,6] proposed an exponential Cauchy-Born rule. The numerical simulation of Arroyo and Belytschko [5,6] also indicated that the model based on the Cauchy-Born rule has zero bending stiffness and does not accurately display the buckling behavior of CNTs. From the higher deformation gradient theory, Guo *et al.* [7] and Wang *et al.* [8] proposed a higher order Cauchy-Born rule in which the second deformation gradient is directly considered, and the deformed lattice vector is approximated as

$$\mathbf{b} \approx \mathbf{F(0)} \bullet \mathbf{a} + \frac{1}{2}\nabla\mathbf{F(0)} : (\mathbf{a} \otimes \mathbf{a}), \tag{2}$$

where $\nabla\mathbf{F(0)}$ is the second deformation gradient.

With the involvement of the second deformation gradient, the accuracy of approximation for the deformed lattice vectors is largely enhanced. In particular, the second term of (2) describes the effect of the bending stiffness, and the approximation is thus more reasonable

B. The Constitutive relationship

Considering a representative cell that corresponds to atom i as shown in Fig. 1, the deformed bond vectors $\mathbf{r}_{ij}$ $(j = 1,2,3)$, by using (2), can be calculated as

$$\mathbf{r}_{ij} = \mathbf{F} \bullet (\mathbf{R}_{IJ} + \boldsymbol{\eta}) + \mathbf{G} : ((\mathbf{R}_{IJ} + \boldsymbol{\eta}) \otimes (\mathbf{R}_{IJ} + \boldsymbol{\eta}))/2, \tag{3}$$

where $\mathbf{F} = F_{ij} e_i \otimes e_j$ and $\mathbf{G} = \nabla\mathbf{F} = G_{ijk} e_i \otimes e_j \otimes e_k$ are the first and second order deformation gradient tensors, respectively. $\mathbf{R}_{IJ}$ is the undeformed bond vector. Moreover, an inner shift vector $\boldsymbol{\eta}$ has been added to the undeformed bond vector due to the non-Centro symmetry of the CNT atomic structure [2-6]. This inner shift vector is a function of $\mathbf{F}$ and $\mathbf{G}$, and can be determined by minimizing the strain energy of atom *i* with respect to $\boldsymbol{\eta}$ [2-6].

The strain energy density in this representative cell can be expressed as

$$W_0 = \frac{1}{2\Omega_i}\sum_{j=1}^{3} V_{ij}(\mathbf{r}_{i1}, \mathbf{r}_{i2}, \mathbf{r}_{i3}) = W_0(\mathbf{F}, \mathbf{G}, \boldsymbol{\eta}(\mathbf{F}, \mathbf{G})), \tag{4}$$

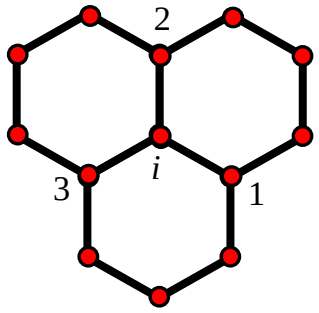

Fig. 1. A representative cell to an atom *i*.

where V_{ij} is Tersoff-Brenner potential[9,10], and Ω_i the average area per atom.

The first Piola-Kirchhoff stress tensor $\mathbf{P}$ and the higher order stress tensor $\mathbf{Q}$ is given by

$$\mathbf{P} = \frac{\partial W_0}{\partial \mathbf{F}}, \quad \mathbf{Q} = \frac{\partial W_0}{\partial \mathbf{G}}. \tag{5}$$

The tangential modulus tensors can be obtained

$$\mathbf{M}_{\mathbf{FF}} = \frac{\partial^2 W_0}{\partial \mathbf{F} \otimes \partial \mathbf{F}}; \quad \mathbf{M}_{\mathbf{FG}} = \frac{\partial^2 W_0}{\partial \mathbf{F} \otimes \partial \mathbf{G}};$$
$$\mathbf{M}_{\mathbf{GF}} = \frac{\partial^2 W_0}{\partial \mathbf{G} \otimes \partial \mathbf{F}}; \quad \mathbf{M}_{\mathbf{GG}} = \frac{\partial^2 W_0}{\partial \mathbf{G} \otimes \partial \mathbf{G}}. \tag{6}$$

The calculations for these tangential modulus tensors are very lengthy [7,8], and their exact expressions are not given here.

III. Mehsfree Method

Based on the constitutive relations in section 2, a computational scheme can be established to implement the numerical simulation of CNTs. A distinct characteristic of the present theory is that the energy is a function of both the first and second derivations of the deformation, and the interpolation of displacement generally requires C^1-continuity. In the conventional finite element, this will lead to difficulties in the establishment of elements and the construction of the interpolation functions [11]. The meshfree method is a newly developed computational technique that has some excellent advantages over the classical finite element method. In particular, meshfree approximations have non-local properties, and satisfy the higher order continuity requirement [12]. This intrinsic non-local property of meshfree interpolation leads to real rotation-free approximation, and displacements can thus be used as the only nodal freedoms [11]. Here, the meshfree method is employed to implement the numerical simulation and test the validity of the higher order deformation gradient theory. Moving least squares approximation [12-14] is used to construct the meshfree interpolation

In the computation, the displacements relative to the undeformed CNT are approximated as

$$\mathbf{u} = \sum_{i=1}^{N} \phi_i \hat{\mathbf{u}}, \tag{6}$$

where ϕ_i is the meshfree shape function and $\hat{\mathbf{u}}$ is the nodal parameter. N is the number of nodes which impact domains involve the evaluated point.

Stable configurations of a CNT are identified with the minimization of the potential energy

$$\Pi = \int_{B_0} W(\mathbf{F},\mathbf{G}) dV - \int_{\partial B_0} \mathbf{u} \cdot \mathbf{t}_0^P dS - \int_{\partial B_0} \nabla_N \mathbf{u} \cdot \mathbf{t}_0^Q dS, \tag{7}$$

where B_0 is the domain of the reference configuration, and $\mathbf{t}_0^P$ and $\mathbf{t}_0^Q$ are the traction and second-order stress traction on the surface of the domain ∂B_0, respectively. Moreover, the trial deformation $\mathbf{u}$ should satisfy the essential boundary condition $\mathbf{u} = \bar{\mathbf{u}}$ on the boundary ∂B_0.

Discretizing (7) in the meshfree framework and differentiating the discretized equation, we have

$$\frac{\partial \prod}{\partial \hat{\mathbf{u}}} = \int_{B_0} (\mathbf{P}\frac{\partial \mathbf{F}}{\partial \hat{\mathbf{u}}} + \mathbf{Q}\frac{\partial \mathbf{G}}{\partial \hat{\mathbf{u}}}) dV - \int_{\partial B_0} \mathbf{t}_0^P(\mathbf{X}) \bullet \frac{\partial \mathbf{u}(\mathbf{X})}{\partial \hat{\mathbf{u}}} dS - \int_{\partial B_0} \mathbf{t}_0^Q(\mathbf{X}) \bullet \frac{\partial \nabla_N \mathbf{u}(\mathbf{X})}{\partial \hat{\mathbf{u}}} dS \tag{8}$$

The Hessian can be derived as

$$\frac{\partial^2 \prod}{\partial \hat{\mathbf{u}}^2} = \int_{\Omega_0} (\mathbf{M}_{\mathbf{FF}} \frac{\partial^2 \mathbf{F}}{\partial \hat{\mathbf{u}}^2} + \mathbf{M}_{\mathbf{FG}} \frac{\partial \mathbf{F}}{\partial \hat{\mathbf{u}}} \frac{\partial \mathbf{G}}{\partial \hat{\mathbf{u}}} + \mathbf{M}_{\mathbf{GG}} \frac{\partial^2 \mathbf{G}}{\partial \hat{\mathbf{u}}^2}) dV. \tag{9}$$

Equilibrium configurations can be solved through the nonlinear Newton's method [16], and the resulting incremental system of equations can be written as

$$\mathbf{K}_{n+1} \Delta \hat{\mathbf{u}}_{n+1} = \Delta \mathbf{f}_{n+1}(\hat{\mathbf{u}}_{n+1}), \tag{10}$$

where the stiffness matrix $\mathbf{K}_{n+1}$ needs to be determined from (9), $\Delta \mathbf{f}_{n+1}$ is the non-equilibrium force to be determined from (8), $\Delta \hat{\mathbf{u}}_{n+1}$ is the incremental solution, and $\hat{\mathbf{u}}_{n+1}$ is the solution at the iterative step $n+1$. $\frac{\partial \mathbf{F}}{\partial \hat{\mathbf{u}}}$ and $\frac{\partial \mathbf{G}}{\partial \hat{\mathbf{u}}}$ can be calculated after the first and second derivatives of the meshfree interpolation shape are determined.

When computing the energy, force, and tangent modulus tensors, the first and second gradients for an evaluated point are computed using the following formulas:

$$\mathbf{F} = \mathbf{F}_{\text{ini}} + \mathbf{F}_{\text{apr}}, \tag{11}$$

$$\mathbf{G} = \mathbf{G}_{\text{ini}} + \mathbf{G}_{\text{apr}}, \tag{12}$$

where $\mathbf{F}_{\text{ini}}$ and $\mathbf{G}_{\text{ini}}$ are the gradients of the evaluated point when no loading is applied, and $\mathbf{F}_{\text{apr}}$ and $\mathbf{G}_{\text{apr}}$ are calculated through the interpolation formulas

$$\mathbf{F}_{\text{apr}} = \sum_{i=1}^{N} \phi_{i,\mathbf{X}} \hat{\mathbf{u}}, \tag{13}$$

$$\mathbf{G}_{\text{apr}} = \sum_{i=1}^{N} \phi_{i,\mathbf{XX}} \hat{\mathbf{u}}, \tag{14}$$

with $\phi_{i,\mathbf{X}}$ and $\phi_{i,\mathbf{XX}}$ being the first and second order derivations of meshfree shape function ϕ_i. Moreover, an imaginative representative cell needs to be used for the evaluated points.

The aforementioned algorithm is a standard Newton-Raphson method [16]. The method is highly efficient before the buckling, but often fails to simulate the buckling behavior of structures because the stiffness matrix becomes non-positive definite around the buckling. As described in Ref. 16, the simplest way to remedy the defection of the non-positive definition of the stiffness matrix is to replace $\mathbf{K}_{n+1}$ with $\mathbf{K}_{n+1} + \alpha \mathbf{I}$, where $\mathbf{I}$ is the identity matrix and α is a positive number that is slightly larger than the magnitude of the most negative eigenvalue of $\mathbf{K}_{n+1}$. This approach is used here.

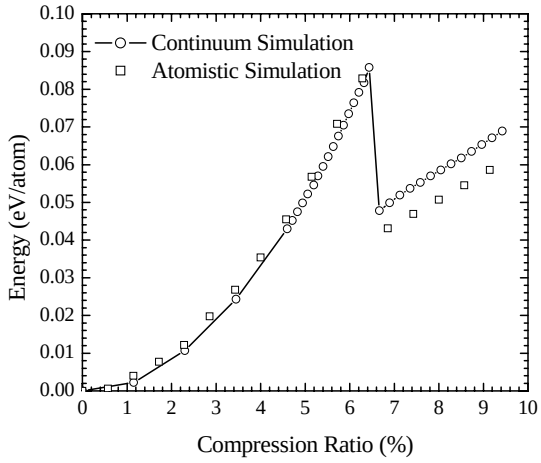

Fig. 2. Comparison of average energy per atom between the continuum and atomistic simulation.

IV. Compressive Buckling of SWCNTs

Using the present meshfree method, we have simulated the compression of a (18, 0) SWCNT. The initial length of the CNT is chosen as 8.7 nm. The problem is treated as quasi-static, and the surface of the undeformed CNT is discretized with 640 particles. The load is imposed through uniformly reducing the length of the CNT by 0.1 nm per loading step at the initial stage and 0.01 nm per loading step near to the buckling. Fig. 2 plots the average energy per atom versus the compression ratio along with the results of molecular dynamic simulation. A distinct energy jump appears at about the compression ratio of 0.065, which corresponds to the buckling. The comparison indicates that the calculated energy before the buckling occurs is in good agreement with that obtained by molecular dynamics

simulation. After buckling the energy calculation becomes less accurate, but this is expectable because the deformation becomes drastically unhomogeneous and it is difficult to obtain a fine simulation with the coarse method.

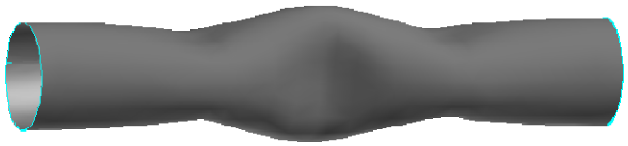

Fig. 3. The buckling patter of a (18,0) CNT of 8.7 nm length using higher order gradient theory.

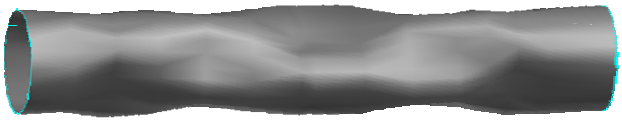

Fig. 4. The buckling patter of a (18,0) CNT of 8.7 nm length using classic Cauchy-Born rule.

Fig. 3 shows the buckling pattern. The numerical simulation displays a true buckling pattern for the CNTs with such a ratio of length to radius [5]. Employing the meshfree method, we also simulated the compression of the same CNT with the constitutive relations based on the Cauchy-Born rule [3,4]. Fig. 4 shows the buckling shape obtained: a series of ridges are scattered on the surface of the CNT. This result has no physical meaning because the Cauchy-Born rule only considers the first deformation gradient (the stretch factor) and does not involve the second deformation gradient (the bending factor).

V. Conclusions

SWCNTs are studied in this paper by considering the effects of the second-order deformation gradient. The meshfree method is employed to implement the numerical computation of the hyper-elastic constitutive model derived from higher order Cauchy-Born rule. Meshfree approximations have non-local properties, and satisfy the requirement of higher order continuity. The meshfree method is thus particularly suitable for problems that involve the higher order deformation gradient (or strain gradient). Good numerical results are obtained for the compression of CNTs, and the buckling behavior is displayed truly. The numerical computation shows that the constitutive model based on higher order Cauchy-Born rule is also computationally efficient.

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