Free Vibration of Defective Nanographene using Molecular Dynamics Simulation and Differential Quadrature Method

Hossein Golestanian*, Ali Khodadadi, Mahmoud Haghighi
Department of Mechanical Engineering,
University of Shahrekord, Iran
E-mail: Golestanian@eng.sku.ac.ir, Alikhodadadi1387@gmail.com, ma.haqiqi@gmail.com
*Corresponding author

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Abstract: In this paper, the free vibration of defective nanographene is investigated using Molecular Dynamics Simulation (MD) and Differential Quadrature Method (DQM). The equations of motions and the related boundary conditions are derived based on the differential constitutive relations in conjunction with the classical plate theory via Hamilton's principle. Then, DQM is used to investigate free vibration of the nanographene with various boundary conditions. At first, in order to determine natural frequencies more realistically, nanographene mechanical properties are determined using MD simulations. The effects of defects are investigated by analyzing pristine and defective nanographenes containing Stone Wales, vacancy, and Adatom defects. According to the results, the non-dimensional fundamental natural frequency parameter converges to the analytical value for N=10×10. Results indicate that graphene with CCCC boundary conditions has the maximum natural frequency. The minimum value corresponds to the graphene with SSSS boundary conditions. In addition, Non-dimensional fundamental frequency parameter of the nanoplate increases with increasing nanoplate aspect ratio. Finally, defects reduce density, position ratio and elastic moduli of nanographene, which causes a decrease in natural frequency. Stone Wales and vacancy defects decrease nanographene natural frequencies by about 8 and 25 percent, respectively.

Keywords: Defective Nanographene, Free Vibration, Molecular Dynamics, Quadrature Method

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Biographical notes: Hossein Golestanian received his PhD in Mechanical Engineering from University of Missouri-Columbia, USA in 1997. He is a Professor of Engineering at Shahrekord University, Iran. His research interest includes modelling nanocomposite mechanical properties. **Ali Khodadadi** is a PhD student of Mechanical Engineering at Shahrekord University in 2021. He received his MSc in Mechanical Engineering from Tarbiat Modares University. His research focuses on MD modelling of nanocomposite mechanical properties. **Mahmoud Haghighi** is a PhD student of Mechanical Engineering at Shahrekord University. He received his MSc in Mechanical Engineering from Isfahan University of Technology in 2021. His research focuses on physical and numerical determination of nanocomposite mechanical properties.

1 INTRODUCTION

Recent studies in the field of nanotechnology have yielded a considerable progress in this field. Specially, introducing Carbon Nanotubes (CNTs) and graphenes with their exceptional mechanical, electronic and chemical properties has been a milestone in nanotechnology development.

The mechanical behavior of CNTs and graphenes has been analyzed through experiments and elastic continuum mechanics. Performing experiments on CNTs and graphenes is a challenging and expensive task due to their nanometer dimensions. It is reported in the literature that continuum mechanics may serve as an alternative method to study CNTs and graphenes by treating them as continuum elastic structures. In the following, nonlocal elasticity theory was presented by Eringen [1]. In contrast to the local theory, where the stress at a point is assumed to be a function of strain at that point, the nonlocal elasticity theory is based on the assumption that the stress at any point is a function of strain at all points in the continuum. Then, this theory has been used for vibration approaches about nanobeam (and nanotube) [2-14] and nanoplate (and nanographene) [15-21].

Nazemizadeh et al. [14] performed vibration sensitivity analysis of Nano-mechanical piezo-laminated beams with consideration of size effects using nonlocal theory. Their results show that the length and the thickness of the piezoelectric layer have prominent effects on the vibration characteristics of the beam. In numerical solutions, DQM is used to investigate free vibration of the nanobeam and the nanoplate. Jalali et al. [21] studied free vibration of rotating functionally graded annular disc of variable thickness using DQM. Recently, Shatt and Abdelkefi [22] found that Eringen's nonlocal theory fails to simultaneously fit both the longitudinal and transverse acoustic dispersion curves of some materials. In addition, defects in the atomic structure of the nanostructures are one of the main reasons for the differences between experimental data and numerical predictions. In modeling nanostructures, it is usually assumed that the nanostructures are perfect and have no defects. It has been reported, however, that different defects such as Stone Wales, vacancy, and Adatom exist in the atomic structure of CNTs and graphenes. Molecular Dynamics simulations can account for these defects in determining mechanical properties of CNTs and graphenes [23-26]. For examples, Aghadavoudi et al. [26] investigate Stone Wales and vacancy defects on mechanical properties of CNT in an article entitled Investigation of CNT Defects on Mechanical Behavior of Cross linked Epoxy based Nanocomposites by Molecular Dynamics.

Thus, combining these two methods (DQM and MD) provides an effective tool for a more realistic prediction of vibration behavior and natural frequencies of these nano structures. In this paper, free vibration of pristine and defective nanographene is investigated using MD simulation and DQM which has not been performed so far. Determination of the mechanical properties such as density, positions ratio and elastic moduli of pristine and defective nanographene accurately using molecular dynamics method and investigating their effects on graphene vibrations is the main novelty of this research. The equations of motion and the related boundary conditions are derived based on the differential constitutive relations in conjunction with the classical plate theory via Hamilton's principle. DQM is used to investigate non-dimensional frequencies. Convergence of the present results was carried out and analysis approach was validated by comparing the predicted results with published data. Mechanical properties of pristine and defective nanographenes are determined using MD simulations. MD-determined properties are used in DQM in order to obtain more realistic natural frequencies of nanographene. In addition, pristine and defective nanographene are considered to investigate the effects of defects on nanographene vibration behavior.

2 THEORICAL FORMULATIONS

Figure 1 shows the nanographene in Cartesian coordinate system. In this figure, a, b and h are length, width and thickness of the nanographene, respectively.

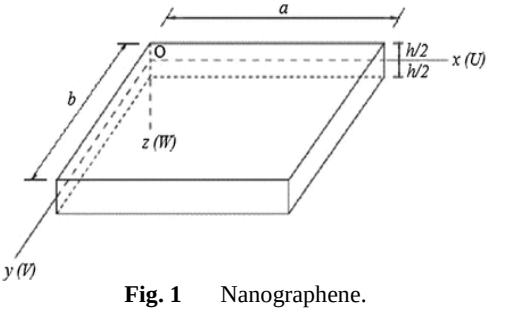

Fig. 1 Nanographene.

2.1. Kinematics

Based on the hypothesis in the classical plate theory, at an arbitrary material point (x, y, z) of the nanoplate, the in-plane displacement components along the x- and y-directions are given by:

$$u(x,y,z,t) = u(x,y,t) - z\frac{\partial w}{\partial x}$$
$$v(x,y,z,t) = v(x,y,t) - z\frac{\partial w}{\partial y} \quad (1)$$

Where, u and v are the in-plane displacement components along the x and y-axis of an arbitrary material point on the mid-surface of the nanoplate and w is the displacement in the z-direction. Therefore, the nonzero components of strain tensor are:

$$\begin{aligned}\varepsilon_{xx} &= \frac{\partial u}{\partial x} - z\,\frac{\partial^2 w}{\partial x^2}\\ \varepsilon_{yy} &= \frac{\partial v}{\partial y} - z\,\frac{\partial^2 w}{\partial y^2}\\ \gamma_{xy} &= \frac{\partial v}{\partial x} + \frac{\partial u}{\partial y} - 2z\,\frac{\partial^2 w}{\partial x\,\partial y}\end{aligned} \tag{2}$$

2.2. Constitutive Relations

Stress-strain relations can be expressed as follows:

$$\begin{Bmatrix}\sigma_{xx}\\ \sigma_{yy}\\ \sigma_{xy}\end{Bmatrix} = \begin{bmatrix}\frac{E}{(1-\upsilon^2)} & \frac{\upsilon E}{(1-\upsilon^2)} & 0\\ \frac{\upsilon E}{(1-\upsilon^2)} & \frac{E}{(1-\upsilon^2)} & 0\\ 0 & 0 & G\end{bmatrix}\begin{Bmatrix}\varepsilon_{xx}\\ \varepsilon_{yy}\\ \gamma_{xy}\end{Bmatrix} \tag{3}$$

Where, E, υ, and G are the material Young's modulus Poisson's ratio and shear modulus, respectively. For a specific strain vector, the stress components are calculated using molecular dynamics simulation to determine nanographene mechanical properties E, G, and ν.

2.3. Equations of Motion

The equations of motion are derived using Hamilton's principle, which takes the following form for the free vibration analysis:

$$\int_0^t (\delta K - \delta U + \delta V)dt = 0 \tag{4}$$

Where, δU is the variation of strain energy, δK is the variation of kinetic energy and δV is the variation of potential energy. Inserting energy variations in (4) and integrating by parts, the equations of motion and the related boundary conditions for transverse displacement can be derived as follows [27]:

$$\begin{aligned}\delta w_b : &\frac{\partial^2 M^{xx}}{\partial^2 x} + 2\frac{\partial^2 M^{xy}}{\partial x\,\partial y} + \frac{\partial^2 M^{yy}}{\partial^2 y}\\ &= \rho\,[A(\frac{\partial^2 w}{\partial^2 t}) - I(\frac{\partial^4 w}{\partial^2 t^2 \partial^2 x^2} + \frac{\partial^4 w}{\partial^2 t^2 \partial^2 y^2})]\end{aligned} \tag{5}$$

Where, Mαβ with (α, β = x, y) are the stress resultants which are defined as:

$$M^{\alpha\beta} = \int_{-h/2}^{h/2} z\,\sigma^{\alpha\beta} dz \tag{6}$$

And ρ is the mass density of the nanographene, and (A, I) are:

$$(A, I) = \int_{-h/2}^{h/2} (1, z^2) dz \tag{7}$$

Boundary conditions:

$$\begin{aligned}&F_b \text{ or } w_b = 0\\ &M_b^{xx} \text{ or } \frac{\partial w_b}{\partial x} = 0\\ &M_b^{yy} \text{ or } \frac{\partial w_b}{\partial y} = 0\end{aligned} \tag{8}$$

Substituting (2) into (3) and the subsequent results into (6), the stress resultants are obtained as follows:

$$\begin{aligned}M^{xx} &= -D(\frac{\partial^2 w}{\partial x^2} + \upsilon\frac{\partial^2 w}{\partial y^2})\\ M^{yy} &= -D(\frac{\partial^2 w}{\partial y^2} + \upsilon\frac{\partial^2 w}{\partial x^2})\\ M^{xy} &= -D(1-\upsilon)(\frac{\partial^2 w}{\partial x\,\partial y})\end{aligned} \tag{9}$$

Using (5) and (9), the equations of motion for the transverse free vibration of the nanographene, in term of w, can be expressed as:

$$\begin{aligned}&-D(\frac{\partial^4 w}{\partial x^4} + 2\frac{\partial^4 w}{\partial x^2 \partial y^2} + \frac{\partial^4 w}{\partial y^4})\\ &= [\rho A(\frac{\partial^2 w}{\partial t^2}) - \rho I(\frac{\partial^4 w}{\partial x^2 \partial t^2} + \frac{\partial^4 w}{\partial x^2 \partial t^2})]\end{aligned} \tag{10}$$

Where:

$$D = \frac{Eh^3}{12(1-\upsilon^2)} \tag{11}$$

Simply supported (S) and Clamped (C) nanographene, along the edge n=0 or ln (n=x or y, lx=a and ly=b) are considered in this investigation. The corresponding boundary conditions are as follows.

Simply supported (S) along the edges *n=0 or* l_n:

$$w = 0, \frac{\partial^2 w}{\partial n^2} = 0 \tag{12}$$

Clamped (C) along the edges *n=0 or* l_n:

$$w = 0, \frac{\partial w}{\partial n} = 0 \tag{13}$$

2.4. Solution Pocedure

It is impossible to find an analytical solution for the nanographene free vibration equations of motions exposed to arbitrary boundary conditions. Therefore, an approximate method should be used to solve the problem. DQM, as an efficient and accurate numerical solution method, is used in this investigation.

First, a set of non-dimensional terms are introduced as follows:

$$\begin{aligned} &\zeta = \frac{x}{L} \quad , \quad \eta = \frac{y}{L} \ , \ W = \frac{w}{h} \quad , \ m2 = \frac{I}{Ah^2} \\ &\iota = \frac{t}{L^2}\sqrt{\frac{D}{\rho h}} \quad , \quad \Omega_{mn} = \omega_{mn} L^2 \sqrt{\frac{\rho h}{D}} \end{aligned} \tag{14}$$

Using (14) along with (10), the nonlocal equations of motion for transverse free vibration of nanographene can be written as:

$$\begin{aligned} &-(\frac{\partial^4 W}{\partial \zeta^4} + 2\frac{\partial^4 W}{\partial \zeta^2 \partial \eta^2} + \frac{\partial^4 W}{\partial \eta^4}) \\ &= \Omega_{mn}{}^2[(\frac{\partial^2 W}{\partial \iota^2}) - m2\ (\frac{\partial^4 W}{\partial \zeta^2 \partial \iota^2} + \frac{\partial^4 W}{\partial \zeta^2 \partial \iota^2})] \end{aligned} \tag{15}$$

In DQM, the nth-order partial derivative of a continuous function f(x, y) with respect to x, y at a given point x_i, y_i can be approximated as a linear sum of weighted linear sum of the sampling points at all discrete points in the domain of x.

$$\begin{aligned} \left(\frac{\partial \mathrm{f}(\mathrm{x},\mathrm{y},\mathrm{z})}{\partial \beta}\right)_{(x_i, y_j)} &= \sum_{m=1}^{\mathrm{N}_\beta} \mathrm{A}_{\mathrm{im}}^{\beta}\ \mathrm{f}_{mj} \\ \left(\frac{\partial^2 \mathrm{f}(\mathrm{x},\mathrm{y},\mathrm{z})}{\partial \beta^2}\right)_{(x_i, y_j)} &= \sum_{m=1}^{\mathrm{N}_\beta} \mathrm{B}_{\mathrm{im}}^{\beta}\ \mathrm{f}_{mj} \end{aligned} \tag{16}$$

Where, N_β is the number of sampling points, and A_{im} and B_{im} are the weighting coefficients.

In this study, sampling points are chosen according to the following relation:

$$\begin{aligned} &\beta_i = \frac{l_\beta}{2}\left(1 - \cos\frac{(m-1)\pi}{N_\beta - 1}\right) \\ &\beta = x \ or \ y \end{aligned} \tag{17}$$

The weighting coefficients of the second order derivative can be obtained as:

$$\left[B_{ij}^{\beta}\right] = \left[A_{ij}^{\beta}\right]\left[A_{ij}^{\beta}\right] \tag{18}$$

Using DQM, "Eq. (15)" can be written as:

$$\begin{aligned} &-(\overline{D}\ )[\sum_{n=1}^{N_x}\sum_{m=2}^{N_x-1} B_{im}^x B_{mn}^x W_{nj} + 2\sum_{n=1}^{N_y}\sum_{m=1}^{N_x} B_{im}^x B_{jn}^y W_{mn} \\ &+\sum_{n=1}^{N_y}\sum_{m=2}^{N_y-1} B_{im}^y B_{mn}^y W_{jn} + B_{i1}^x \kappa_{1j}^x + B_{iN_x}^x \kappa_{N_x j}^x \\ &+B_{1j}^y \kappa_{1j}^y + B_{iN_y}^y \kappa_{iN_y}^y)] - \Omega^2[(W_{ij}) \\ &-m2\ (\sum_{m=1}^{N_x} B_{im}^x W_{mj} + \sum_{m=1}^{N_y} B_{jm}^y W_{im})] = 0 \end{aligned} \tag{19}$$

3 MOLECULAR DYNAMICS SIMULATION

In order to determine nanographene natural frequencies more realistically, one must use exact mechanical properties for the structure under investigation. In this research, mechanical properties of pristine and defective nanographenes were determined using MD analysis. Materials Studio software with Condensed-phase Optimized Molecular Potentials for Atomistic Simulation Studies (COMPASS) force field was used for this purpose. COMPASS has been assessed using different calculation methods as well as molecular dynamics simulations of liquids, crystals, and polymers [28]. The process of simulating molecular dynamics in modeling graphene could be found in a previous investigation performed by the co-authors [29]. MD Models of pristine and nanographene containing various defect types were created and analyzed to determine their mechanical properties. These models are shown in "Fig. 2". The MD obtained mechanical properties are then used in the governing equations to investigate nanographene free vibration behavior more realistically.

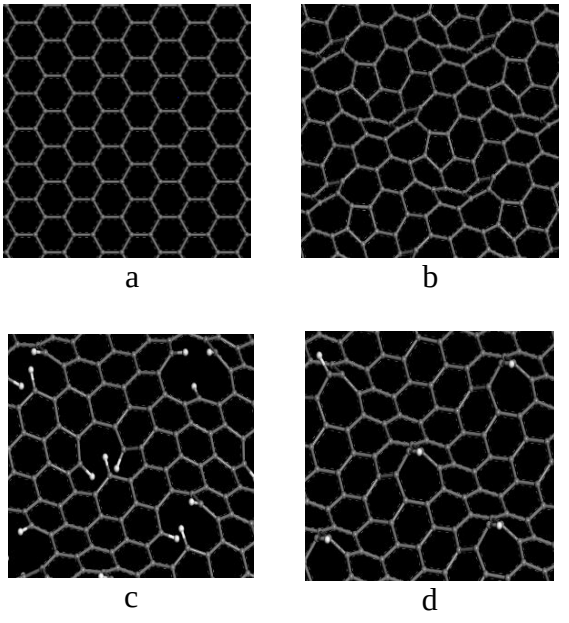

Fig. 2 Graphene with various defects: (a): Pristine graphene, (b): Graphene with 42 Stone Wales defects, (c): Graphene with 42 vacancy defects, and (d): Graphene with 42 Adatom defects.

4 RESULTS AND DISCUSSION

First, non-dimensional natural frequencies of SSSS nanographene were determined using different number of sampling points in DQM. The results were compared with analytical solution results presented in Ref [27] in order to validate the solution procedure. This comparison is presented in "Table 1". According to these results, for N=10×10 (number of sampling points), the non-dimensional fundamental natural frequency parameter converges to the analytical value presented by Pradhan and Phadikar [27].Therefore, N=15×15 was used in the analysis of nanographene in this investigation.

Table 1 The convergence behavior of non-dimensional fundamental natural frequency parameter of the simply supported nanographene with different aspect ratios (L = 10 nm)

L/h	Method			
	DQM			Analytical [27]
	(5×5)	(10×10)	(15×15)	
10	18.9801	19.0653	19.0653	19.0653
20	19.4728	19.5625	19.5625	19.5625
50	19.6196	19.7105	19.7105	19.7105
100	19.6409	19.7320	19.7320	19.7320

To establish validity of the present work, results are compared with the presented results in Ref [30] for nanoplate in "Tables 2 and 3".

Table 2 Comparison of the higher order non-dimensional natural frequency parameters of the simply supported nanographene (L = a = b = 10 nm, (L/h = 10))

Mode	Investigator	
	[30]	Present
$\Omega(1,1)$	0.0963	0.0963
$\Omega(2,2)$	0.3853	0.3853
$\Omega(3,3)$	0.8669	0.8670

Note in "Table 2" that, non-dimensional natural frequency results obtained in this study are in good agreement with the results presented by Karami and Malekzadeh [30].

Table 3 The non-dimensional fundamental frequency parameter of the nanoplate with different types of boundary conditions (L = a = b = 10 nm, (L/h = 10))

L/h	Boundary conditions				
	SSSS	SSCS	SCSC	CCCS	CCCC
10	19.0653	22.536	27.1729	29.6286	32.8859
20	19.5625	23.3039	28.4682	31.2665	35.1164
100	19.7376	23.6464	28.0541	31.8008	35.9852
100 Ref [30]	19.7376	23.6463	29.0541	31.8008	35.9852

The results presented in "Table 3" show that non-dimensional fundamental frequency parameter of the nanoplate increases with increasing nanoplate aspect ratio.

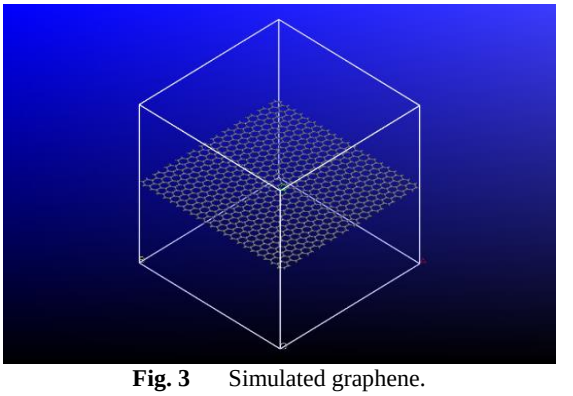

Fig. 3 Simulated graphene.

Once the modeling technique and the solution procedures were verified, models were created to investigate free vibration of the nanographenes under investigation. First, pristine and defective nanographenes were modeled in Materials Studio software to determine their mechanical properties. Nanographene with dimensions L = a = b = 50.5 Å, and h = 3.35 Å were considered in all models. Figure 3 shows graphene simulation box in Materials Studio software.

MD-determined mechanical properties for pristine and defective graphene are presented in "Table 4". The accuracy of the MD models was also verified by comparing the results with those found in the literatures [31-32]. Their results show that pristine nanographene elastic moduli is about 500 GPa. Note that the difference between our result with those presented is below 1 percent.

Table 4 Graphene properties

Material	Properties		
	ρ (kg/m3)	E (GPa)	υ
Pristine graphene	2368	505	0.32
Graphene with 42 Stone Wales defects	2319	460	0.15
Graphene with 42 Vacancy defects	2109	280	0.14
Graphene with 42 Adatom defects	2134	475	0.24

Figure 4 shows the first and higher modes of natural frequencies for pristine graphene with SSSS boundary conditions. Note in this figure that, natural frequency of pristine nanographene increases by mode number increasing.

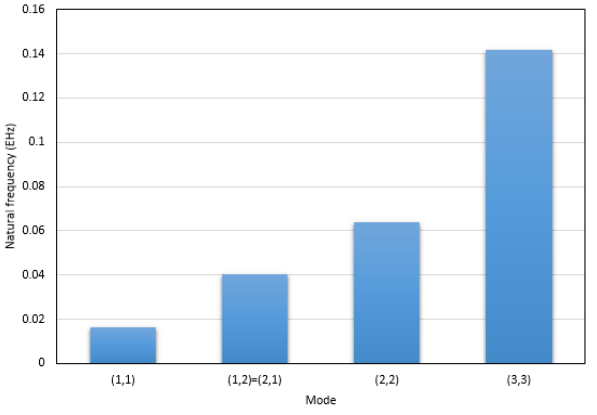

Fig. 4 Natural frequencies of pristine nanographene.

Figure 5 shows the first vibration mode of pristine graphene natural frequency with five different boundary conditions.

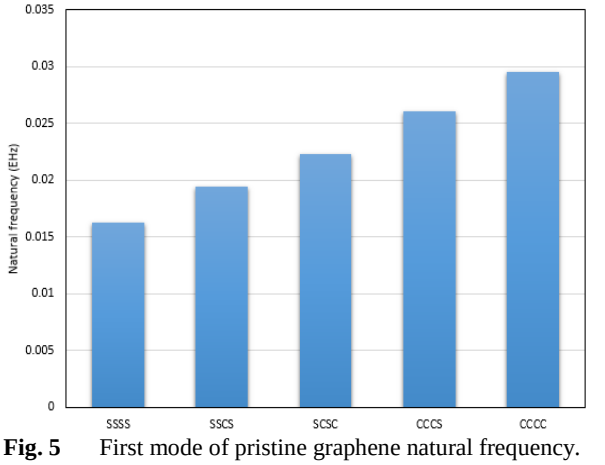

Fig. 5 First mode of pristine graphene natural frequency.

It can be observed that graphene with CCCC boundary conditions has maximum natural frequency whereas minimum value corresponds to the graphene with SSSS boundary conditions. The trend suggests that the graphene stiffness increases as the number of boundary constraints is increased. In addition, to investigate the effects of defects on the natural frequency of graphene, the first frequency mode was determined for pristine and defective graphenes. In this model, SSSS boundary conditions were imposed. The results are presented in "Fig. 6".

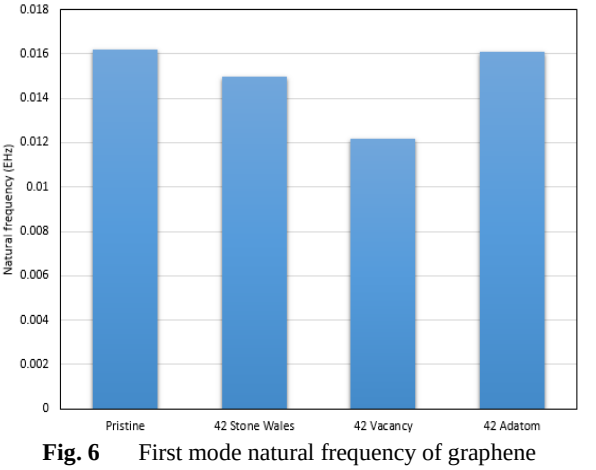

Fig. 6 First mode natural frequency of graphene

As can be observed in this figure, Stone Wales and vacancy defects reduce all natural frequency modes of graphene, but Adatom defect has a negligible effect on natural frequency of graphene. The largest decrease is related to Vacancy defect. The reductions in natural frequencies are due to the weakening of the nanographene structure and stiffness as a result of defects. Vacancy defects eliminate a number of bonds from the pristine nanographene, which results in a pronounced decrease in its elastic modulus that decreases natural frequencies. The Stone Wale defect is

a crystallographic defect which involves the change of connectivity of two bonded carbon atoms. This defect type leads to a bond rotation by 90 degrees with respect to the midpoint of the bond and results in a reduction of nanographene natural frequencies. This defect type has an effect on nanotube stiffness compared to the flat graphene sheets causing natural frequencies decrease. The Adatom defect interrupts the sequencing in the nanographene structure. This type of defect has a negligible effect on graphene modulus, and in turn, natural frequencies.

5 CONCLUSIONS

In this paper, free vibration of pristine and defective nanographenes is investigated using molecular dynamics simulation and differential quadrature method. The equations of motion and the related boundary conditions were derived based on the differential constitutive relations in conjunction with the classical plate theory via Hamilton's principle. MD-determined properties are used in DQM in order to obtain more realistic natural frequencies of nanographene. In addition, pristine and defective nanographene are considered to investigate the effects of defects on nanographene vibration behavior. Stone Wales, vacancy, and Adatom defects were modeled. Finally, vibration behavior of nanographene with various boundary conditions was studied. Results indicate that graphene with CCCC boundary conditions has the maximum natural frequency. The minimum natural frequency corresponds to the graphene with SSSS boundary conditions. Finally, Stone Wales and vacancy defects have large effects on nanographene natural frequencies. Adatom defect type has a negligible effect on graphene natural frequency.

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