www.ietdl.org

Published in IET Nanobiotechnology
Received on 9th April 2013
Revised on 21st June 2013
Accepted on 24th June 2013
doi: 10.1049/iet-nbt.2013.0020

ISSN 1751-8741

Boron nitride nanotube-based biosensing of various bacterium/viruses: continuum modelling-based simulation approach

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Abstract: In this study, the feasibility of single walled boron nitride nanotube (SWBNNT)-based biosensors has been ensured considering the continuum modelling-based simulation approach, for mass-based detection of various bacterium/viruses. Various types of bacterium or viruses have been taken into consideration at the free-end of the cantilevered configuration of the SWBNNT, as a biosensor. Resonant frequency shift-based analysis has been performed with the adsorption of various bacterium/viruses considered as additional mass to the SWBNNT-based sensor system. The continuum mechanics-based analytical approach, considering effective wall thickness has been considered to validate the finite element method (FEM)-based simulation results, based on continuum volume-based modelling of the SWBNNT. As a systematic analysis approach, the FEM-based simulation results are found in excellent agreement with the analytical results, to analyse the SWBNNTs for their wide range of applications such as nanoresonators, biosensors, gas-sensors, transducers and so on. The obtained results suggest that by using the SWBNNT of smaller size the sensitivity of the sensor system can be enhanced and detection of the bacterium/virus having mass of 4.28×10^{-24} kg can be effectively performed.

1 Introduction

In recent years, biosensors have been increasingly demanded in fundamental biological studies, health science research, drug discovery and clinical diagnosis [1–3]. Depending on the nature of the transduction signal, biosensors can be classified into optical, electrical and mechanical biosensors [4]. In mechanical biosensors, the resonance frequency is measured and related to the mass change induced by the attached additional biological object to the sensor system. A nanomechanical biosensor, is the one which capitalise on the nanoscale size of at least one of their dimensions [5]. The mass detection of biomolecules has become a growing field in biological and biomedical applications. It is recognised as one of the key technologies for predictive and preventive medicine [6]. The common strategy for prediction of disease is based on sensing the corresponding biomolecules [6]. It is reasonable to believe that biosensors with real-time sensing capability and ease of use can change the future of disease detection and health monitoring.

Boron nitride nanotubes (BNNTs) are more thermally and chemically stable than carbon nanotubes (CNTs) [7]. Such properties together with excellent mechanical property and thermal conductivity make BNNT a potential candidate for applications like biosensors [7]. Chopra and Zettl [8] had reported mechanical measurement of BNNTs, where the amplitude of thermally induced vibration of a cantilevered BNNT was examined at room temperature inside transmission electron microscope (TEM) and the elastic modulus of a single BNNT was estimated to be 1.22 ± 0.24 TPa. Ciofani *et al.* [9] had exploited the use of BNNTs in the nanomedicine field and reported that BNNTs are more suitable for development of sensors and transducers for detection of biological entities because of their chemical stability. In the later study, Ciofani *et al.* [10] highlighted that BNNT-based nanomaterials require further experiments for accurate assessment of their biocompatibility.

Several literatures can be found addressing biosensing using nanomaterials. Elishakoff *et al.* [11, 12] have used exact solution as well as finite difference method to explore cantilever single walled CNTs and cantilever double walled CNTs as mass sensors considering various bacterium or viruses. Gupta *et al.* [13] have used continuum mechanics-based analytical and finite element method (FEM)-based simulation approach to analyse the dynamic behaviour of fixed-free single walled CNT as a biosensor for mass-based detection of various bacterium or viruses. The BNNTs are theoretically analysed, considering Euler-Bernoulli beam theory-based analytical approach and molecular mechanics-based simulation approach, as biosensors considering biological objects; alanine amino terminal residue (a type of α-amino acid with simple structure) and deoxyadenosine free residue (a nucleoside component of DNA composed of adenosine and deoxyribose) [14]. The BNNTs of small diameter comprising boron and nitrogen atoms can be approximated as continuum columns [15]. Using Euler-Bernoulli beam theory and continuum modelling-based FEM simulation, the authors have investigated the mass sensitivity limits 10^{-23} g for single walled BNNT (SWBNNT)-based nanomechanical resonators [16, 17].

In the present work, the feasibility of SWBNNT as a biosensor has been explored considering the effect on the resonant behaviour because of various adsorptions of various bacterium/viruses at the free end of the cantilevered configuration of SWBNNT. The SWBNNTs of diameter 0.52 nm are considered same as that considered by Gupta *et al.* [13] for single walled CNTs. The resonance-based detection grants continuous scaling down of a cantilever to nanometer length scales. This can be assigned to the fact that scaling down leads to an increase of the resonant frequency of a nanomechanical mass sensor, which increases detection sensitivity. In other words, smaller the size of nanomechanical sensor, more sensitivity it owns [16, 17]. Based on this conception, the lengths of SWBNNTs are considered as 400, 200, 150 and 100 nm. As a systematic analysis approach, the simulation results based on FEM, are compared with the continuum mechanics-based analytical results and found in excellent agreement for mass-based detection of various bacterium/viruses and, it can be considered as one of the toolkits to design SWBNNT-based biosensor systems.

2 Mass-resonant frequency shift relation for SWBNNT-based biosensors

The rationale of mass detection using nanomechanical biosensors is based on the fact that the resonant frequency is very much sensitive to the mass of attached biomolecules, as over all effective mass of the sensor system varies. The adsorption of the biomolecule on the sensor system imparts some stiffness to it and this is only true when the adsorption takes place at the free end. In the present work, the effect of stiffness because of adsorption of various bacterium/virus is neglected, as the detection of various bacterium/viruses is considered based on the change in resonant behaviour of the sensor system because of change in attached mass. The key issue of mass-based sensing using present SWBNNT-based sensor system is in quantifying the change in the resonant frequency because of attached mass. Based on this rationale, the development of computational tools adequate to simulate the vibrational behaviour of BNNT-based biosensor is important.

For an SWBNNT of length L, with the attached mass at the free end of the cantilevered configuration, the fundamental frequency based on Euler-Bernoulli theory can be given as [18]

$$f = \frac{1}{2\pi}\sqrt{\frac{k_{eq}}{M_{eq}}} \tag{1}$$

where k_{eq} and m_{eq} are the equivalent stiffness and mass of the cantilevered configuration of SWBNNT, respectively.

The equivalent stiffness (k_{eq}), and equivalent mass (m_{eq}) for the cantilevered configuration of nanotube can be obtained as [14, 16]

$$k_{eq} = \frac{3EI}{L^3} \tag{2}$$

and

$$m_{eq} = \frac{33}{140}\rho AL + M \tag{3}$$

Substituting (2) and (3) in (1), the resonant frequency can be obtained as

$$f = \frac{1}{2\pi L^2}\frac{\alpha^2\beta}{\sqrt{1+\Delta M\mu}} \tag{4}$$

where

$$\alpha 2 = \sqrt{\frac{144}{11}}, \quad \beta = \sqrt{\frac{EI}{\rho A}}, \quad \Delta M = \frac{M}{\rho AL} \text{ and } \mu = \frac{140}{33} \tag{5}$$

Resonant frequency with no attached mass for cantilevered configuration of sensor is expressed by considering $\Delta M = 0$ in (4)

$$f_0 = \frac{1}{2\pi L^2}\alpha^2\beta \tag{6}$$

and resonant frequency shift Δf because of attached mass M can be defined as

$$\Delta f = f_0 - f = f_0 - \frac{f_0}{\sqrt{1+\Delta M\mu}} \tag{7}$$

rearrangement gives expression

$$\Delta M = \frac{1}{\mu\left(1-\left(\Delta f/f_0\right)\right)^2} - \frac{1}{\mu} \tag{8}$$

Using (5), (8) can be used to obtain the relationship, between the attached mass and resonant frequency shift as

$$M = \frac{\rho AL}{\mu}\frac{\left(\alpha^2(\beta/L^2)\right)^2}{\left(\alpha^2(\beta/L^2) - 2\pi\Delta f\right)^2} - \frac{\rho AL}{\mu} \tag{9}$$

This is a general equation, which relates the attached mass and resonant frequency shift.

3 Finite element modelling

The forcefield consisting of a combination of strong interatomic forces is acting within the nanotube. The inequality between the magnitude of interatomic forces leads to a highly anisotropic constitutive relation [19]. Assuming the linear bonding forces along the length of nanotube and considering it as a transversely isotropic material (which means isotropic about one axis) [20], the compliance matrix of nine terms; having only five of them are independent can be considered to define the overall stiffness properties of SWBNNT. The simulation based on FEM is performed by considering constitution of SWBNNT in a cylindrical coordinate system and the stress–strain

doi: 10.1049/iet-nbt.2013.0020

relationship can be given as

$$\begin{Bmatrix} \sigma_r \\ \sigma_\theta \\ \sigma_z \\ \tau_{\theta z} \\ \tau_{rz} \\ \tau_{r\theta} \end{Bmatrix} = \begin{bmatrix} D_{11} & D_{12} & D_{13} & 0 & 0 & 0 \\ D_{12} & D_{22} & D_{23} & 0 & 0 & 0 \\ D_{13} & D_{23} & D_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & D_{22}-D_{23} & 0 & 0 \\ 0 & 0 & 0 & 0 & D_{55} & 0 \\ 0 & 0 & 0 & 0 & 0 & D_{66} \end{bmatrix} \times \begin{Bmatrix} \varepsilon_r \\ \varepsilon_\theta \\ \varepsilon_z \\ \gamma_{\theta z} \\ \gamma_{rz} \\ \gamma_{r\theta} \end{Bmatrix} \tag{10}$$

where D_{ij} represents the stiffness coefficients. Considering the elastic modulus of 1.22 ± 0.24 TPa, transverse modulus of 30 GPa and Poisson's ratio of 0.35; the values of stiffness coefficients for SWBNNT as an anisotropic material have been used as per the given Appendix [16, 17].

The commercial finite element numerical package ANSYS is used to analyse the resonant behaviour of the cantilevered SWBNNT. ANSYS provides an efficient and readily implementable tool for anisotropic coupled field element analysis. A three-dimensional (3D) continuum model has been used for resonant frequencies-based analysis of SWBNNT-based biosensor system for mass-based detection of various considered bacterium/viruses. SWBNNTs are modelled using a 3D FEM and attached mass is modelled using Boolean operation. In this analysis, the attached mass needs to be changed in every iteration, therefore the GLUE, Boolean operator is used for modelling to the SWBNNT with attached mass, as it allows to modify the solid model construction more easily. The element type SOLID 64 is used for 3D modelling of anisotropic solid structures. This element type is defined by eight nodes having three degrees of freedom at each node. Furthermore, this analysis has been used to analyse frequency variation caused by addition of different bacterium/viruses at the free end of the cantilevered configuration of SWBNNT.

As indicated in Fig. 1, it is expected that the effective wall thickness of a continuum model of SWBNNT must be smaller than the theoretical diameter of boron atom (~0.145 nm) or that of nitrogen atom (~0.150 nm). A cross section of a real nanotube contains only a limited number of atoms. In a continuum mechanics model, on application of external load, the stresses are transmitted through the continuous wall area, whereas in the nanotube the same stresses are transmitted through its atoms. As a result, to maintain tube equilibrium, the wall thickness must be smaller than the atom diameter. In this sense, calculations using continuum models that apply wall thickness greater than or equal to the diameter of a boron atom or a nitrogen atom, can give incorrect answer. The 3D continuum model of the nanotube with thickness of 0.065 nm, 44% of the average of theoretical diameters of boron atom (~0.145 nm) and nitrogen atom (~0.150 nm) has been considered for the FEM simulation-based analysis to analyse the resonant behaviour of SWBNNT-based sensor systems.

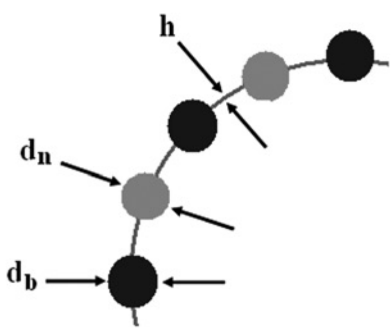

Fig. 1 *Partial cross section of an SWBNNT, where d_b is the diameter of boron atom, d_n diameter of nitrogen atom and h is the equivalent thickness of the nanotube [16, 17]*

4 Results and discussion

The aim of the present study is to check the feasibility of SWBNNT-based biosensor so as to be able to identify the bacterium or virus that may be attached at the tip of the nanotube. In order to perform vibration analysis of the sensor system 'SWBNNT and bacterium/virus', information about the masses of various bacteria/viruses is required. These are available in monographs by Fraenkel-Conrat [21], Gunsalus and Stanier [22] and Elishakoff *et al.* [11]. Information about masses of various bacterium and viruses in increasing values of the mass is shown in Table 1. The bacterium/virus can be treated as light mass when its mass is much less than the mass of the nanotube for specified length of the sensor, whereas the bacterium/virus should be treated as heavy mass when its mass is either comparable with mass of nanotube or exceeds it [11].

As mentioned in the earlier section nanotube of different lengths 400, 200, 150 and 100 nm are considered such that various bacterium/viruses considered in terms of light mass can be analysed. The properties of SWBNNTs of diameter 0.52 nm having different considered lengths are listed in Table 2. Fig. 2 shows the variation in fundamental frequencies of SWBNNTs having lengths 400, 200, 150

Table 1 List of bacteria/viruses and associated masses

Name of the bacterium/virus	Symbolic representation	Mass, kg
ESAT-6	V1	9.96×10^{-24}
Parvoviridae largest	V2	4.28×10^{-24}
Parvoviridae smallest	V3	1.29×10^{-23}
Picornaviridae	V4	1.37×10^{-23}
Nodaviridae	V5	1.71×10^{-23}
Tymovirus group	V6	1.81×10^{-23}
Diantovirus group	V7	1.94×10^{-23}
Nudaurelia group	V8	2.90×10^{-23}
Caliciviridae	V9	3.94×10^{-23}
Togaviridae largest	V10	4.19×10^{-23}

Table 2 Properties of SWBNNT of considered lengths having wall thickness of 0.065 nm

Diameter, nm	Length, nm	Mass, kg	Fundamental frequency, Hz
0.52	400	4.34×10^{-23}	$1.45 \times 10^{+07}$
0.52	200	2.17×10^{-23}	$5.80 \times 10^{+07}$
0.52	150	1.63×10^{-23}	$1.03 \times 10^{+08}$
0.52	100	1.08×10^{-23}	$2.32 \times 10^{+08}$

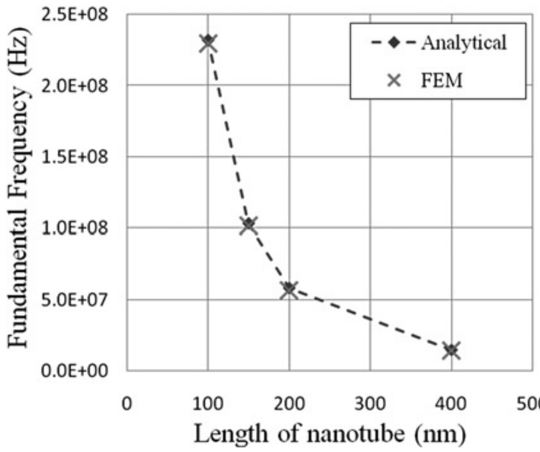

Fig. 2 *Variation in fundamental frequencies of SWBNNTs of different considered lengths calculated analytically using (6) and FEM approach*

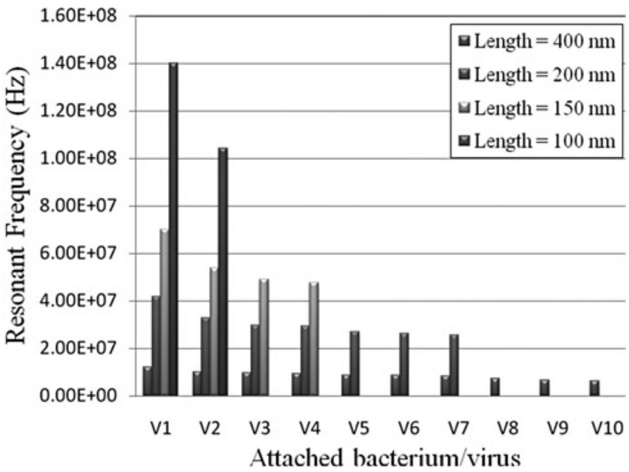

Fig. 3 *Variation in resonant frequency because of attached different considered bacterium/virus at the free end of the cantilevered SWBNNT-based biosensors of different lengths: 400, 200, 150 and 100 nm, calculated using continuum modelling-based FEM simulation*

and 100 nm, calculated using (6) and present continuum modelling-based FEM approach. The analytical results are obtained considering SWBNNT as an isometric material having material properties: Young's modulus 1.22 ± 0.24 TPa and density 2180 kg/m^3 [8, 16, 17]. The FEM simulated results are in excellent agreement with the analytical results. The obtained results indicate that as the length of the nanotube increases the fundamental frequency of the nanotube decreases as the mass of the sensor system increases.

Various bacterium/viruses are considered as light mass, which means, mass of bacterium or virus is less than the mass of SWBNNT, and the resonant frequency variation-based analysis is performed for SWBNNT of different considered lengths. Fig. 3 shows the obtained FEM simulation results of resonant frequency variation because of various considered bacterium/viruses attached at the free end of the cantilevered configuration of SWBNNT of considered different lengths. It is depicted from the obtained results that as the mass of bacterium or virus increases the resonant frequency for given length of nanotube decreases. For the particular attached bacterium/virus, as the length of nanotube decreases the resonant frequency because of the attached particular bacterium or virus increases. Table 3 shows the summarised data regarding percentage error in the FEM-based simulation results compared with the analytical exact solution based on (4). It is evident from Table 3 that continuum modelling-based FEM simulated results are in excellent agreement with the exact solution. The variation in percentage errors in FEM simulated results is in the very narrow range of 0.78–1.38, with close proximity with the exact solution for nanotube lengths of 400, 150 and 100 nm. For nanotube length of 200 nm the percentage errors in FEM simulated results vary in the range of 2.57–3.62. Thus, the FEM simulated results are acceptable for continuum modelling of SWBNNT with effective wall thickness of 0.065 nm. Hence, with the consideration of the obtained results it can be concluded that the FEM model used for the present study is correct and can be used for further investigation for wide range of applications of BNNTs such as sensors, transducers and actuators.

Fig. 4 shows variation in frequency shift Δf for the considered bacterium/virus attached at the free end of the cantilevered configuration of SWBNNT-based biosensors of

Table 3 Percentage error in FEM simulated results compared with exact solution

Bacterium/virus	Mass, kg	% Error in FEM simulated results			
		L = 400 nm Nanotube mass 4.34×10^{-23} kg	L = 200 nm Nanotube mass 2.17×10^{-23} kg	L = 150 nm Nanotube mass 1.63×10^{-23} kg	L = 100 nm Nanotube mass 1.08×10^{-23} kg
ESAT-6	4.28×10^{-24}	1.38	2.57	1.19	1.35
Parvoviridae largest	9.96×10^{-24}	1.16	3.06	1.13	0.78
Parvoviridae smallest	1.29×10^{-23}	1.04	3.62	1.13	–
Picornaviridae	1.37×10^{-23}	1.28	3.31	1.18	–
Nodaviridae	1.71×10^{-23}	1.24	3.59	–	–
Tymovirus group	1.81×10^{-23}	1.15	3.30	–	–
Diantovirus group	1.94×10^{-23}	1.16	3.21	–	–
Nudaurelia group	2.90×10^{-23}	1.15	–	–	–
Caliciviridae	3.94×10^{-23}	1.21	–	–	–
Togaviridae largest	4.19×10^{-23}	1.20	–	–	–

doi: 10.1049/iet-nbt.2013.0020

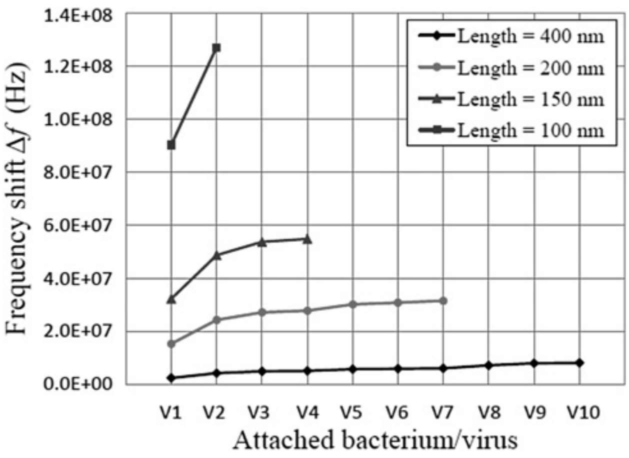

Fig. 4 *Variation in frequency shift Δf, for various attached considered bacterium/virus at the free end of the SWBNNT of different lengths: 400, 200, 150 and 100 nm*

different considered lengths. As the mass of the bacterium/virus increases the frequency shift Δf increases. Equation (9) is the governing equation for estimation of the attached mass, as it relates the attached mass M and the frequency shift Δf. From (9) it is clear that for sensing the particular bacterium/virus in terms of its mass, frequency shift Δf because of attachment of that particular bacterium/virus must be measurable. From Fig. 4 it is clear that for the considered lightest bacterium/virus (ESAT-6, V1) the frequency shift Δf is measurable (or detectable). As the length of nanotube decreases frequency shift Δf for the lightest bacterium/virus (ESAT-6, V1) increases. Such variation in resonant frequency shift Δf against variation in the size of nanotube suggests that the sensitivity of the cantilevered SWBNNT-based biosensors can be improved by scaling down their size.

5 Conclusions

Using continuum modelling-based FEM simulation approach the feasibility of the SWBNNT-based biosensors for detection of various bacterium/viruses (as mentioned in Table 1) is checked. The present FEM-based simulation approach considering effective wall thickness of SWBNNT is found in excellent agreement with the exact solution. Resonant frequency-based analysis is reported for possible detection of various bacterium/viruses in terms of their masses. Also, the variation in frequency shift Δf for different considered lengths of nanotube is performed to find out the possible enhancement in sensitivity of the overall sensor system. The obtained results indicate that effective detection of various considered bacterium/viruses is possible using SWBNNT-based biosensor system. Also, the present continuum modelling-based FEM simulation approach can be used as a systematic analysis approach as one of the toolkits for design of the SWBNNT-based biosensors. The obtained results may be useful to practically realise future SWBNNT-based biosensors having primary advantages of biocompatibility and bio safety compared with CNT as a counterpart.

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7 Appendix

Mass density of SWBNNT is 2180 kg/m^3 and stiffness coefficients for anisotropic material behaviour of SWBNNT are

(Stiffness $\times 10^9$ N/m^2)	
D_{11}	1376.3
D_{12}	16.308
D_{13}	487.4
D_{14}	0
D_{15}	0
D_{16}	0
D_{22}	30.285
D_{23}	16.308
D_{24}	0
D_{25}	0
D_{26}	0
D_{33}	1376.3
D_{34}	0
D_{35}	0
D_{36}	0
D_{44}	14.9
D_{45}	0
D_{46}	0
D_{55}	444.4
D_{56}	0
D_{66}	14.9

doi: 10.1049/iet-nbt.2013.0020