Computational Materials Science 90 (2014) 50–55

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

Molecular separation with carbon nanotubes

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ARTICLE INFO

Article history:
Received 14 February 2014
Received in revised form 4 April 2014
Accepted 5 April 2014
Available online 4 May 2014

Keywords:
Molecular separation
Carbon nanotube
Graphene
Impulse wave propagation
Molecular dynamics simulations

ABSTRACT

The feasibility of molecular separation with carbon nanotubes is studied by investigating a separation of water molecules and lead nanoparticles with a carbon nanotube-based nanopore system from molecular dynamics simulations. The nanopore system is comprised of a single-walled carbon nanotube and two graphene sheets placed at two ends of the nanotube. The nanotube is subjected to impact loads on a local portion on the wall to generate the propagation of impulse waves in the tube for pumping the encapsulated water molecules out of the tube, while blocking lead nanoparticles by the two graphenes. Studies indicate that the separation is a result of van der Waals interactions between the nanotube and the encapsulated molecules through the propagation of impulse waves in the nanotube. The efficiency of the molecular separation is found to be dependent on the amount of impact loads and the size of the impacted portion of the tube.

1. Introduction

The remarkable electrical, mechanical, and thermal properties of carbon nanotubes (CNTs) and graphene sheets (GSs) [1,2] have made them among the most promising materials in a wide range of applications such as nanocomposites and nano-sensors [3,4]. In addition, the morphology of the nano-materials provides an excellent opportunity to build a new generation of membranes for separating atoms and molecules without any phase changes [5,6], which significantly reduces the energy consumption compared to traditional methods. The atomic and molecular separation has great potential in areas of clean energy, medicine, and chemical industries including ammonia purge gas, helium and hydrogen recovery, nitrogen generation, and water purification [7,8].

The principle of molecular separations with nano-membranes is based on the fact that large molecules are not allowed to permeate the membranes due to the size restriction. A defect-free membrane is, however, impermeable to molecules, but a selective permeability can be achieved by designing precise nanometer pores in the membranes, and controlling their size and shape for a successful separation [9,10].

One of such selectively permeable membrane structures is porous graphenes [11–13]. A molecular dynamics (MD) simulation study on functionalized graphene nanopores serving as ionic sieves with high selectivity was conducted by Sint et al. [11]. The research showed that graphene nanopores terminated by negatively charged nitrogen and fluorine atoms are favoring the passage of cations, while the pores terminated by positively charged hydrogen atoms are favoring the passage of anions. Jiang et al. [12] investigated the permeability of porous graphene membranes for separating hydrogen and methane mixtures by using first principles density functional theory calculations. Du et al. [13] theoretically proposed a series of porous graphenes as the separation membrane for H2/N2 by adjusting the pore size and shape. Their simulation results revealed that the selectivity and permeability of porous graphenes could be controlled by drilling various nanopores with different shapes and sizes. According to the mechanism of separation behaviors using graphene membranes, the control of pore sizes is indispensable for an efficient permeability. However, controlling a precise size and shape of pores as well as decorating pores still remain a challenge.

In view of the problems, applications of CNTs in filtering processes have drawn attentions on achieving more practical designs of separation membrane systems. The cylindrical hollow structure and smooth walls of CNTs provide an excellent opportunity of building nanopumping and nanofiltering devices for water desalination, petroleum filtration, and even kidney dialysis [14,15]. Different methods for transportation of atoms and molecules by CNTs have been introduced by imposing chemical potential and thermal gradients, electrical fields [16–18]. Since the relatively low efficiency of the proposed transportation methods, nanopumping effects on the activation of atomic/molecular flows in CNTs by applying mechanical loads were investigated [19,20]. Recently, a novel transportation of atoms and molecules with CNTs subjected to torsion was proposed and developed [21]. Based on the method,

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http://dx.doi.org/10.1016/j.commatsci.2014.04.012
0927-0256/

a CNT subjected to a torsional load beyond the critical value undergoes a collapsed state and formation of torsional kinks, and the propagation of the kinks along the CNT becomes a driving force for moving atoms inside the nanotube [21]. The transportation method with CNTs in torsion was then extended for separating encapsulated helium and carbon atoms [5]. It was shown that the torsion rate and torsion angle can be adjusted to enable effective separations of different species of atoms using CNTs [22]. Although the separation of atoms with CNTs in torsion was shown to be successful in design of filtration systems, the realization of a successful separation is found to be very sensitive to the amount of torsional load applied to the CNTs. Therefore, a more feasible and technically sound method for an efficient separation process is indispensable.

Recently, interests have been orientated toward applications of terahertz physics of nano-materials in design of nanodevices [23,24], which opens a new topic on nano-materials wave characteristics. Arash and Wang [25] investigated properties of impulse wave propagations in CNT-based sensors using molecular simulations, and provided a new method for detection of noble gases. The impulse waves propagating along the axial direction of a CNT are generated by applying impact loading on a local portion of the wall of the tube in the transverse direction of its axial axis [25]. The mechanical waves would induce the change of van der Walls (vdW) potential between the nanotube wall and molecules encapsulated in the CNT. It is expected that the impulse wave propagation method could be extended in molecular transportations to fulfill a more feasible and practical molecular separation.

In this work, we report and examine a separation of water molecules and lead nonaparticles encapsulated in a CNT subjected to a periodically impact load for the potential in filtration applications. The impacts, subjected on a local portion of the tube, generate impulse waves along the axial direction of the nanotube, which induce the change of van der Walls (vdW) potential between the tube and the encapsulated molecules for pumping water molecules out of the CNT. Two GSs are placed at two sides of the nanotube in the longitudinal direction to build a nanopore based instrument, which enables the feasibility of the molecular separation by blocking the ejection of the lead nanoparticles. In the present method, the rate of the impact applied to the nanotube is not sensitive for the separation process. Hence, the main limitation of the previously proposed separation method using CNTs in torsion [5,22] is resolved, and a more practical separation could be realized for filtration applications. The dependence of the amount of impact loads and the size of impacted portion on the effectiveness and efficiency of the separation process is investigated.

2. Methods

In order to examine the feasibility of the method, the separation of 1600 water molecules and a lead nanoparticle encapsulated in an armchair (22, 22) CNT with a length of about 20 nm illustrated in Fig. 1(a) and (b) is investigated with molecular dynamics. The mass density of water molecules is 1 g/cc, and the average diameter of the lead nanoparticle is taken to be around 0.8 nm. Two fixed GSs are also placed at a distance of 0.8 nm from two ends of the nanotube as a nanopore filtering system as illustrated in Fig. 1(a). The interaction potential in simulations is modeled by the universal force field (UFF) [26], which is a purely diagonal and harmonic force field. In the UFF, The potential energy of an arbitrary geometry of a molecule is provided as a superposition of various two-body, three-body, and four-body interactions. The total potential energy, E, is expressed by $E = E_R + E_\theta + E_\varphi + E_\omega +$

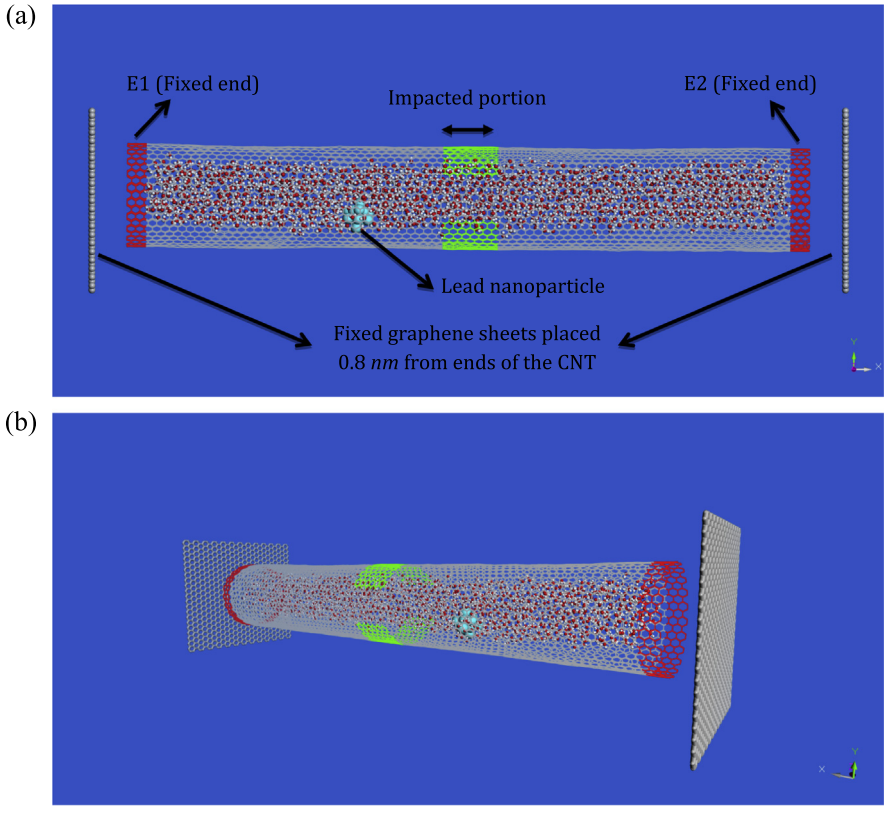

Fig. 1. A (22, 22) CNT with a diameter of 3 nm and a length of 20 nm filled with 1600 water molecules with a mass density of 1 g/cc and a lead nanoparticle with an average diameter of around 0.8 nm: (a) initial configuration from side view, and (b) initial configuration from perspective view. The length of impacted portion is 1.5 nm.