Computational Materials Science 154 (2018) 122–131

Contents lists available at ScienceDirect

Computational Materials Science

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

Design and mechanical characterization of a novel carbon-based hybrid foam: A molecular dynamics study

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ARTICLE INFO

Keywords:
Nano-structures
Hybrid
Mechanical properties
Computational modelling

ABSTRACT

Graphene and fullerenes are impressive nanostructures having great potential be used in many areas due to their unique physical and chemical properties. By hybridizing these nanostructural units, it is possible to obtain superior composite nanostructures with novel properties. In this study, a new hybrid nanostructure that will be named as graphene nanoribbon-fullerene (GNR-F) network is introduced by providing its tensile and compressive mechanical characteristics through molecular dynamics simulations. The proposed nanostructure is mainly an interconnected network of GNR units on which fullerenes are covalently grafted. Three different atomistic models are generated using different types of fullerenes (i.e. C60, C180 and C320) to investigate the effects of the fullerene types on the mechanical properties. After examining the thermodynamic stability of the specimens -by monitoring the free energy profiles and morphologic evolution within a sufficiently long period of time, deformation mechanisms of the specimens subjected to tensile and compression tests are investigated. Simulation results indicate that the proposed GNR-F network nanostructure has a ductile nature in tensile loading while its compressive response is foam-like with remarkable energy absorbing capacity. In addition to the expected effect of the fullerenes on the compressive behavior, a significant effect is also observed for the tensile response.

1. Introduction

Research efforts on nanostructured materials science are of great importance for the development of novel materials with superior properties. By using different manufacturing techniques, it has been demonstrated that the development of new advanced nanomaterials with remarkable properties for a wide range of applications can be achieved [1–7]. Among different kinds of nanostructured materials, carbon-based nanomaterials such as graphene, fullerenes and carbon nanotubes (CNTs), which are very popular and extensively studied over the last two decades, are widely used in various applications including photovoltaic[8,9], field emission transistors [10,11], fuel cells [12,13], supercapacitors [14], and environmental remediation [15,16] owing to their unique morphology and superior thermal [17,18], electrical [19–22], optical [23–25], mechanical [26,27] and chemical properties.

Majority of the studies is on the application of carbon-based nanostructures in the areas such as composite materials, scanning probe microscopy, field emission, nanoelectronics and solar cell technology [18–32]. All these superior features of graphene and other carbon-based nanomaterials offer great opportunities for the synthesis of new, effective and multi-functional nanostructured composites by combining different types of carbon based nanostructures. At this point, numerical experiments performed by means of simulations play an important role for the computational design of new materials because the synthesis trials and experimental studies on nano materials are both more expensive and difficult to study desired properties in certain ambient conditions. One of the numerical methods used in the study of the nanostructures is the molecular dynamics (MD) simulations that are widely used to study the mechanical behavior of carbon-based nanostructures [32,33].

In literature, vast amount of experimental and theoretical studies investigating the behavior of carbon-based materials such as graphene, fullerenes and carbon nanotubes can be found. While the majority of these studies focuses on the properties of the individual carbon-based nano units [27,32–34], in recent years a rising interest on the design and analysis of hybrid foam nanomaterials constructed with different carbon-based units is noticed [3,29,30,35,36].

Cellular structures, namely foams, are special materials and attract great attention owing to their superior properties such as tunable thermal characteristics, high energy absorbing capacity, high shear and fracture strengths compounded with low density [37–40]. Among many different foam structures, carbon based foams have an important place

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https://doi.org/10.1016/j.commatsci.2018.06.039
Received 4 February 2018; Received in revised form 6 June 2018; Accepted 25 June 2018

due to exceptional characteristics of carbon material. The behavior of carbon-based foams depends on the micro-level morphology, which can be controlled via process parameters (i.e. temperature, pressure, pressure drop time, etc.) used in manufacturing, as well as the properties of the base material. Therefore, in order to determine the characteristics of carbon-based foams through numerical simulations, the nanostructural geometry and the distribution of the material over this geometry must be well-established [38]. The fact that the properties of the foams vary in a wide range constitutes an important potential for engineering applications such as designing lightweight sandwich structures, biocompatible and large portable structures with high strength [39]. For instance, carbon based nanoporous foams are studied for spongiform bone tissues to understand the bone injuries and to develop equivalent replacing materials for the damaged bone tissues [40].

Carbon based nanoporous foams can be synthesized from two-dimensional carbon based building blocks such as graphene by template assisted growth or self-assembly in solution through van der Waals forces or chemical cross-linking [41–43]. In order to develop the 3D architecture of foam structures, both chemical vapor deposition (CVD) and solution processing methods are used [44–48]. Chakravarty et al. [40] demonstrated the use of spark plasma sintering (SPS) technology as a possible method for welding graphite sheets onto 3D skeletons for use in biological applications. They also experimentally investigated the mechanical properties of 3D network structures with porosity exceeding 40% and performed molecular dynamics (MD) simulations to investigate the bonding between adjacent graphene layers. These studies revealed that for the purpose of developing three dimensional carbon based nanoporous foams with good structural stability, strong intermolecular bonds between building blocks such as graphene and fullerene units are required to form junctional regions. These junctions can be formed by covalent or weaker van der Waals (vdW) forces. While covalent bonds provide highly stable and strong junctions, non-covalent van der Waals interaction, which are caused by electrostatic interactions between the fluctuations in the electronic charge density [49], supplies much less coherence to the network. For instance, in the formation of a 3D network of the graphene sheets, a significant reduction in the mechanical properties is observed due to weak π-π stacking and vdW interactions between the graphene sheets [50]. Therefore, it is necessary to establish high coherence between the carbon-based building blocks (i.e. grapheme, CNTs, fullerenes) to ensure their potential use in practical applications and to avoid reductions in high strength values [51]. A possible technique to achieve this by protecting nanostructures is to weld nanosheets, nanolayers or nanotubes by applying high energy through electric field, radiation, heating or chemical modifications [52,53]. These 3D carbon-based foam structures formed with covalent bonds are used in many areas. For instance, these carbon-based foam hybrid nanostructures are proposed for several applications including hydrogen storaging [54] and supercapacitors [55], special vertical thermal transport materials [56] and nano/micro integrated devices [57,58].

In order to increase the mechanical performance, structural stability, thermal and electrical conduction of the hybrid structures, maintaining a strong coherence between the different carbon-based units are particularly important. Several experimental techniques have already been developed to fabricate hybrid nanostructures consisting of covalently bonded carbon-based units. For example, a new carbon-based hybrid nanomaterial, called Nanobud, was synthesized by combining single-walled carbon nanotubes (SWCNTs) and fullerenes into a single structure via forming covalent bonding [59]. In that study reporting the synthesizing of nanobuds, the hybrid structure of fullerene-functionalized SWCNTs was demonstrated by utilizing transmission electron microscopy (TEM) images, spectroscopic and spectrometric investigations [59]. In another study of carbon nanotube-fullerene (i.e. C60) hybrid nanostructures [60], formation of nanobuds through fullerene bombardment on CNTs was investigated theoretically by molecular dynamics (MD) simulations. In the study, different types of nanobuds generated by C60 bombardment on CNTs structure are presented and the junctions formed between C60 and CNTs are reported to be remarkably stable. Moreover, it was also found that the nanobuds usually decrease the ultimate tensile strength and Young's modulus of CNTs.

In addition to the hybrid nanostructures made of CNTs and fullerenes, nano-materials formed by covalent bonding of graphene and CNTs are also studied in literature. One of the first studies to present the idea of combining graphene with CNT units investigated the hydrogen storage capacity of a new three-dimensional network material consisting of CNTs vertically positioned between the graphene layers [61]. This theoretical study for the proposal of such a CNT-graphene hybrid material is followed by a number of studies investigating the synthesis methods for the formation of CNT-graphene hybrid nanostructures. Among them, a new technique enabling to produce CNT-graphene films, which consists of vertical CNT units grown on graphene sheet, with significant flexibility and stretchability is described by Lee et al. [62]. In another study, Yang et al. [36] presented a 3-D hierarchical design of a CNT-graphene nanocomposite to enhance the electrochemical capacitive performance of supercapacitors. They systematically investigated the effects of microstructure and capacitive characteristics of GS-KNT composites by changing the weight ratio of reduced graphene oxide (rGO) to multi-wall carbon nanotubes (MWCNTs). As a new approach to design and manufacture of carbon-based three-dimensional nanostructures with seamless C-C connections, Niu et al. [63] simulated the growth of junctions between CNTs on graphene sheets by employing Fe nanoparticles as a catalyst through quantum mechanical molecular dynamics (QM/MD) simulations. They showed that pure covalent CsbndC bonds could be obtained by moving the catalyst during CNT growth and annealing.

Along the same line with CNT-graphene and fullerene-CNT hybrid nanocomposites, graphene-fullerene hybridization is also under investigation. For instance, Yu et al. presented a successful attempt to bond C60 fullerenes onto the graphene layers covalently [64]. Resultant fullerene grafted graphene nanosheets were shown to be significantly enhanced for the electron transport in photovoltaic devices resulting in higher device efficiency [64]. In a similar vein, there are other experimental studies presenting novel synthesis methods for manufacturing fullerene/graphene (FG) nanocomposites to utilize their superior properties in electrochemical applications such as supercapacitors [65,66]. Regarding numerical studies on fullerene-graphene nano-hybrid structures, Kirca [67] presented the computational design of a new carbon-based hybrid nanostructured material consisting of fullerene units covalently sandwiched between graphene sheets and examined its mechanical characteristics by performing compression tests with MD simulations [67]. Furthermore, Ozturk et al. investigated the hydrogen storage capacity of the same hybrid nanostructure by underlying its remarkable hydrogen storage potential due to its micro- and mesoporous morphology with high surface-to-weight ratios and superior structural stability [68].

Considering the recent developments and studies outlined above, hybridization of carbon-based nanostructures (i.e. CNTs, fullerenes, and graphene) through chemical bonding is an effective way to expand extraordinary properties of these nanostructures into larger scales. In this regard, production of FG-like nanoporous hybrid materials appears to be achievable in the near future, since fullerene grafted monolithic graphene sheets can be synthesized successfully with several new techniques presented in literature [64,69]. Although there are many studies on the design and manufacturing of new nanostructured materials formed by combining carbon nanotubes and graphene structures in the literature, fewer works on the graphene-fullerene nanocomposites are noticed in literature. In this work, the design of a new nanoporous material consisting of randomly dispersed graphene nanoribbons on which a number of fullerenes are covalently bonded is presented. For this purpose, following the atomistic modeling process, thermodynamic stability of the proposed hybrid nanostructure is examined to show its stability under real physical conditions. After checking the