FEM analysis of metal matrix nanocomposites reinforced with off-line atomistically-informed equivalent nanofillers

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Abstract

With ever increasing the applications of metal matrix nanocomposites (MMNCs) reinforced with carbon nanofillers, demand for accurate and effective modeling techniques in this area has been increased in recent years. To provide such a route, this study is dedicated to apply information from interfacial atomic interactions in modified continuum models implemented to analyze the mechanical behavior of these nanocomposites. In the proposed methodology, the metal matrix is considered as a continuous medium. Meanwhile, to capture the atomic-scale phenomena governing the problem, the carbon nanofiller, local metal atoms neighboring it, and the nanofiller/metal interface are analyzed using molecular dynamics (MD) simulations. Subsequently, employing the equivalent continuum modeling (ECM) concept, geometrical and mechanical characteristics of the equivalent nanofiller is obtained in each case and introduced in a finite element analysis (FEA). It is demonstrated that the atomistically informed FEA can successfully reproduce the results of full atomistic MD simulations with ignorable run-time compared to that of this method. Finally, constructing several computational cells with different metal matrices and reinforcing agents, the developed MD/FE multiscale approach is utilized to capture the influences of temperature, nanofiller geometry and orientation, its volume fraction and interfacial interactions on the axial elastic constants of these nanocomposites.

Keywords:

Metal matrix nanocomposite (MMNC); Equivalent nanofiller; Molecular dynamics simulation; Finite element analysis; Axial elastic constant; Equivalent continuum modeling

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1. Introduction

Metal matrix composites reinforced with nanofillers are novel materials suitable for a large number of applications including automobile, aerospace, and electronic industries. Among various nanofillers employed to reinforce metals, carbon nanofillers such as single-layered graphene sheets (SLGSs) and carbon nanotubes (CNTs) have been extensively utilized in recent years [1-3]. This was due to their exceptional mechanical and physical properties in combination with their good dispersion in various metal matrices particularly in the case of SLGSs [4]. Previous experimental and numerical investigations have shown that adding these nanofillers can improve the base metal in terms of mechanical strength, wear and corrosion, and also coefficient of friction. The subject has been extensively reviewed in our recent publication [5]. It is noted that while several experimental works have been conducted informatively to study the mechanical properties of metal-based nanocomposites (MMNCs), they are costly at the nano-scale and also need complicated equipment. Accordingly, to facilitate the development of MMNCs, modeling is an alternative and suitable approach.

Numerical studies for modeling nanocomposites can be divided into two main categories including atomistic modeling and continuum mechanics. Although, continuum-based methods have been known as an effective way to study the overall mechanical behavior of nanocomposites, they are unable to capture the atomic-scale phenomena occurred at the interface. Meanwhile, these interfacial interactions play an important role in the amount of load transfer between nanofiller and matrix as thoroughly studied by Wernik et al. in the case of CNT/polymer nanocomposites [6]. On the other hand, atomistic-based methods such as molecular dynamics (MD) simulations can be successfully implemented to take into account the matter. However, their applications are limited due to time and length scales associated with them. Consequently, it is necessary to establish a modified continuum analysis in a way that essential characteristics in atomistic/nanoscales can be involved in the constitutive

relationships of MMNCs. To construct these modified continuum-based models it should be noticed that the volume fraction (VF) of nanofillers implemented to reinforce the metal matrices is ignorable in many cases compared to the whole nanocomposite. Additionally, although the dimensions of the nanofillers are within nanoscale and thus, they should be modeled at this length scale, the metal itself is dense enough to be considered as a bulk matrix material. In other words, it is only necessary to employ intense atomic calculations near the core of atomic-scale inhomogeneity and use continuum-based solutions in the homogeneously deforming regions.

To provide such a route for suitably characterizing properties of nanocomposites using the conventional continuum theory, Odegard et al. [7] were the first who proposed a method called as equivalent-continuum modeling (ECM). The method was introduced to develop constitutive models for polymeric systems reinforced with single-walled carbon nanotubes (SWNTs). To this end, they implemented the effective fiber concept, which consists of the nanotube, the local polymer near the CNT, and the CNT/polymer interface. The methodology was based on applying an external loading to an SWCNT and its local surrounding polymer chains and then computing the interatomic potential energy changes as a function of the imposed load. A direct transformation to its equivalent continuum model was then made by equating the potential energy density of discrete atomic interactions and the strain energy density of its continuous substance counterpart. The effective fiber was then successfully included in a micromechanical analysis based on the Mori-Tanaka method to predict the mechanical properties of these nanocomposites in an accurate and efficient way [8].

Regarding the afore-mentioned works, it was concluded that the method is applicable to a wide range of problems that require the accuracy of atomistic level descriptions coupled with the general applicability of continuum-level models. Accordingly, ECM would principally be useful for the field of predictive modeling of nanocomposites. Regarding this matter, Tsai et

1 al. [9] employed the idea of equivalent fiber in a three-phase continuum micromechanical
2 model to examine the role of considering interphase for better prediction of the mechanical
3 behavior of CNT/polymer composites. Employing several micromechanical models,
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5 Lacarbonara et al. introduced a new equivalent elastoplastic constitutive model to explore the
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7 specific damping capacity of polymer composites reinforced with unidirectional CNTs [10].
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9 Furthermore, the same group proposed an updated version of Eshelby-Mori-Tanaka method
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11 to take into account the actual morphological features of CNTs recognized via experimental
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13 analyses. These include the actual variability of the CNTs aspect ratio and their degree of
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15 dispersion within the host polymer in real samples [11]. In a similar way, elastic properties
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17 for silica nanoparticle/polyimide composites with various nanoparticle/polymer interfacial
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19 treatments were investigated through an effective interface model [12]. Additionally, the
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21 authors utilized the ECM method to study the effect of SWCNT orientation on the shear
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23 modulus of polymeric matrices reinforced with this type of carbon nanofillers [13]. In the
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25 proposed method, first, a combined MD/continuum elasticity theory was introduced to obtain
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27 the geometrical and mechanical characteristics of the equivalent fiber representing the CNT
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29 and its neighboring polymer. Consequently, the defined fiber was then embedded in a finite
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31 element simulation to construct the required model.
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41 As mentioned before, ECM approach has been effectively used in recent years to study the
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43 behavior of polymer nanocomposites, especially for composites reinforced with CNTs.
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45 Meanwhile, in spite of the accuracy of the results proposed by this method and also its
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47 efficiency, this concept has been rarely used in the case of other composites having different
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49 matrices and also nanofillers. With ever increasing the applications of metal matrix
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51 nanocomposites reinforced with carbon nanofillers and the subsequent demands for better
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53 modeling techniques in this area, this article aims to develop an MD/FE multiscale approach
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55 based on the ECM concept for this type of nanocomposites. In the introduced method, the
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metal matrix was considered as a continuous medium. Also, to examine various factors affecting the mechanical behavior of MMNCs, distinct computational cells including a metal matrix infused with SLGS and SWCNT, were examined under uniaxial tensile loadings via MD simulations. Accordingly, based on these results and employing the ECM approach, the equivalent fiber was obtained in each case and introduced in a finite element simulation to build the whole FE model. This paper is organized as follows. In Section 2 we introduce our analysis methodology and details of the computational models. Verification of the MD simulation and numerical results for defining the equivalent nanofillers are presented in Section 3. Additionally, employing the equivalent nanofiller in each case, finite element results of the examination of the effects of temperature, nanofiller geometry and orientation, and interfacial interactions on the axial Young's modulus of these nanocomposites are discussed in this section. Section 4 contains the concluding remarks.

2. Modeling

2.1. Analysis Methodology

As discussed before, due to the different nature of metal matrix and nanofillers leading to different length and time scales for them, multiscale modeling was raised in the literature as the main methodology implemented to model the mechanical behavior of this type of nanocomposites. Accordingly, in the present work, a newly hierarchical multiscale modeling approach was developed to evaluate the effects of temperature, geometrical characteristics, and also interfacial interactions on the elastic constants of metal matrix nanocomposites infused with carbon nanofillers. In the proposed model, the matrix deformation was analyzed at the macroscopic scale by the continuum-based finite element method. Meanwhile, the nanofiller (i.e., Graphene and Carbon nanotube), the local metal near the nanofiller, and the metal/filler interface were modeled employing MD simulation. To this end, considering copper as the matrix material, first, two distinct computational cells, i.e., SLGS/Cu and

CNT/Cu samples, were constructed and examined under uniaxial tensile loadings via MD simulations. Having the results obtained from MD, in the next stage, an effective continuum fiber was obtained in each case based on the Cauchy-Born rule and equivalent continuum concept. The effective fiber was then implemented in an FE analysis to predict the macroscopic mechanical properties of these nanocomposites. Particularly, the developed model was employed to investigate the effects of nanofiller geometry on the elastic constants of the aforementioned nanocomposites. Moreover, the influence of temperature variations and type of interfacial interactions on the results was studied for the sample reinforced with SLGS. Additionally, the orientation dependence of the results was also examined in this case. As will be shown later, the proposed method can take advantages of BOTH the efficiency of continuum mechanics and the accuracy of atomistic simulation methods.

2.2. Equivalent fibers representing nanofiller/surrounding metal

2.2.1. *Cauchy-Born Rule*

The Cauchy-Born rule has been used to concurrently relate the deformation of a continuum media to its atomistic structure through the local deformation gradient $\mathbf{F}$, as shown in Fig. 1. It stems from Cauchy [14] who revealed an approach to coupling atomistic and continuum models through an energy-based method. As seen in this figure, each point in the continuum is enclosed within a computational cell in which the deformation is uniform. Accordingly, during any uniform loading case, the energies of all of the interatomic bonds of the cell can be utilized to compute the strain energy density of the continuum level. With developing this idea by Born [15], the common standard method in elastic molecular theories of crystals was created, which is known as the Cauchy-Born rule. Based on this rule, in the proposed multiscale model developed here, an effective continuum fiber was obtained in the case of each carbon nanofiller. This was achieved through equating the strain energy density of the

continuum model under uniaxial tensile loading and the potential energy changes associated with its atomic structure obtained via MD simulations performed in the similar conditions.

Fig. 1. Schematic representation of the Cauchy-Born rule.

2.2.2. Molecular dynamics simulation

The procedure for preparing computational cells of copper-based composites reinforced with SLGSs and CNTs is similar to those used in our recent publication [5]. MD simulations were accomplished by open-source LAMMPS code, developed by the Sandia National Laboratory [16]. Here, in accordance with previous studies of the mechanical properties of carbon nanostructures, The AIREBO potential was used to model the energetics and dynamics of the covalent bonds between carbon atoms. To simulate Cu-Cu interatomic forces, EAM potential was employed [17-18]. Additionally, to describe the non-bonded van der Waals interactions between carbon nanofillers and copper matrix, the Lennard-Jones (LJ) 12-6 potential was utilized. It is noted that as mentioned before, to consider the effects of metal/nanofiller interactions on the results, Ni/SLGS nanocomposite sample was also examined. Detailed explanation about the values of LJ parameters for the C-Cu and C-Ni interface can be found in Ref. [5].

Simulation were conducted using an NVT ensemble at the room temperature of $T=300$ K. Accordingly, the Nose-Hoover thermostat [19] was used to maintain the simulation

temperature at the mentioned value. Also, the velocity Verlet algorithm [20] was employed to integrate the equations of motion with a time step of 0.5 fs for nanocomposite samples. Finally, imposing appropriate boundary conditions, axial tension was utilized to obtain the desired elastic constants of the samples. To this end, keeping one side of the computational cell fixed, the other side was displaced incrementally when time evolves until the desired strain was achieved. It is noted that in each sample, the system was initially relaxed for 30000 steps, while after imposing each incremental displacement, the relaxation time was taken as 1 ps. Figure 2 illustrates a typical computational cell under the uniaxial tension test.

Fig. 2. Schematic illustration of the uniaxial tensile test.

2.3. Finite element simulation

As mentioned before, continuum-based finite element formulation was implemented to analyze the equivalent continuum fiber and the metal matrix. Here, an isoparametric cubic element was employed for modeling the continuum fiber and matrix. The element was defined by 20 nodes having three degrees of freedom per node, i.e. translations in x and y directions. Due to the small size of the nanofiller dimensions compared to those of the RVE, it is very important that elements become smaller around the nanofillers gradually without any sudden distortion. The metal matrix selected in this work, was copper. The Young modulus of this isotropic metal was obtained to be 56.7 GPa according to our MD

simulations, which is in good agreement with the value reported by previous works [21] at the same temperature. Also its Poisson's ratio was determined to be 0.3.

The equivalent continuum fiber was then symmetrically placed in the RVE as an inclusion surrounded by metal to determine the macroscopic properties of nanocomposite samples via the finite element method. It is noted that the geometrical characteristics and elastic constants of the effective fiber was established based on the numerical results of MD simulation and employing the Cauchy-Born rule. The continuum fiber and metal matrix were assumed to be isotropic linear elastic materials. As discussed before, to have a better comparison, two RVEs were constructed in the shape of a rectangular box having square cross-section including two distinct equivalent continuum fibers i.e., SLGS and SWCNT. Based on the dimensions of the mentioned fibers (as will be presented in the next section), the geometrical characteristics of the RVEs was set so that the similar nanofiller VF was achieved in both cases. Imposing the uniaxial tensile test in the FE software, the Young's modulus was directly determined through the stress-strain curve with the strain $<5\%$ using linear regression.

3. Results and Discussion

3.1. Verification of the MD simulation

As discussed before, to define the dimensions of the equivalent fiber and its elastic constants in the methodology introduced here, first, MD simulations should be performed. It is noted that to accompany with the validation of the developed tensile code in LAMMPS, several case studies were examined for pure carbon nanofillers, pristine metal, and metal-based nanocomposites embedded with SLGSs and CNTs in our recent publication [5]. Accordingly, the determined elastic constants of the samples were found to be in agreement with the ones presented in the literature stating the correctness of the procedure implemented in the MD simulations and also the necessary parameters and dominant assumptions. As the main

1 results, it was shown that due to the rippled structure of SLGSs, these carbon nanofillers
2 could better reinforce the copper matrix compared to SWCNTs. Also, it was concluded that
3 the presence of thermal fluctuations causes a sharp decrease in the Young's modulus of this
4 type of nanocomposites. Additionally, comparing the elastic constants of SLGS/Ni
5 nanocomposite with the corresponding values of SLGS/Cu sample indicated that due to the
6 strong Ni-C bonding in the former case, graphene could enhance the Young's modulus of
7 nickel in a better way. For the sake of simplicity, the reader may refer to [5] that documents
8 in detail the explanation of the MD simulation, the utilized parameters, and the relevant
9 numerical results.
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25 3.2. Construction of equivalent nanofillers

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27 To define the geometric configuration associated with equivalent fiber representing the
28 nanofiller and metal matrix surrounding it, the methodology developed by Odegard et al. [7]
29 was implemented. Based on the equivalent continuum concept, three main steps of this
30 method are defined as follows. First, RVE of an equivalent continuum model identical to its
31 molecular structure in geometry is established. Second, potential energy associated with the
32 deformation of each model under identical boundary conditions is determined. Finally, by
33 equating energies of deformation of the two models, the unknown dimensions of the
34 equivalent continuum fiber are obtained. It should be noted that, here, the concept of cut-off
35 radius was considered in defining the geometrical dimensions of computational cells
36 representing the MD models of nanocomposite samples. This was done to speed up the
37 calculations and keep the accuracy of the results simultaneously. It comes from the fact that
38 for each type of interfacial interactions in nanocomposites, there is a distance (i.e., cut-off
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radius), above which, the interactions between the nanofiller and the host material approaches to zero.

With respect to the abovementioned point, to construct the atomistic model of the CNT/Cu sample, first, a (5,5) armchair nanotube with the diameter and length of 0.678 and 9.95 nm respectively was selected. As seen in Fig. 3a, the nanotube was then embedded in a cylindrical RVE having the same length as the CNT and a diameter of 4.3 nm to achieve a sample having 5% VF of CNT. As the second RVE, since the spatial dimensions of both of the nanofillers should be the same, an SLGS with the same length as the CNT and width of 2.12 nm ($\pi \times d_{\text{SWCNT}}$) was selected. Considering the similar VF for nanofiller in this case and keeping the geometrical shape of the SLGS, the size of the RVE was obtained as 9.95×4.38×3.07 nm in the x, y, and z directions, respectively (See Fig. 3b).

Fig. 3. (a) CNT/Cu and (b) SLGS/Cu nanocomposite sample with 5% VF of nanofiller.

Following construction of the molecular models in LAMMPS (Figs. 3a and 3b), to obtain the geometrical dimensions of the equivalent fiber, first, the deformation energy variations of these RVEs should be obtained in the desired loading condition. This energy was determined by subtracting the energy of the system in its equilibrium position from the corresponding value after imposing the uniaxial tensile loading. In this study, both of the bonding and non-

bonding atomic interactions were considered. Accordingly, total potential energy of metal-based nanocomposite samples was defined as follows:

$$U^m = \sum U^r + \sum U^\theta + \sum U^{vdW} \quad (1)$$

where U^r , U^θ and U^{vdW} are bond stretching, bond-angle bending, and van der Waals interactions energy, respectively (Fig. 4).

Fig. 4. Schematic illustration of interatomic interactions in atomic structures.

Fig. 5. Bar with uniform cross section under uniaxial loading.

On the other hand, assuming the equivalent continuum fiber as an elastic homogenous bar under uniaxial tensile loading, the corresponding energy density could be defined as [22]:

$$U = \frac{P^2 L}{2EA} \quad (2)$$

where P , L , and A are the uniaxial load, length of the bar and its cross-sectional area respectively, as illustrated in Fig. 5.

Considering this equation and having obtained the potential energy variations of the atomistic structure of the equivalent continuum fiber obtained from MD analysis, the dimensions of the nanofiller in each case could be determined through the Cauchy-Born rule as follows:

$$\frac{\sum U^m}{V_{cell}} = \frac{P^2 L}{2AE AL} \quad (3)$$

where V_{cell} is volume of the atomistic RVE, and E is its Young's modulus determined from MD simulation. As seen, in this equation, all of the parameters were specified except A determined by substituting the known values from MD results. Accordingly, to model the CNT-reinforced copper case, as seen in Fig. 6b, an equivalent continuum fiber in the cylindrical shape was obtained with the length of the RVE (i.e., 9.95 nm) and the diameter of 3.7 nm. Also, for the other case, i.e. the SLGS/copper sample, the equivalent fiber achieved was in the shape of rectangular box with the dimensions of 3.485×2.448×9.95 nm, in the x, y, and z directions, respectively. The aspect ratio (i.e., width to thickness) of the equivalent continuum SLGS, was considered to be the same as the corresponding ratio for its atomistic model. It should be emphasized that for achieving high-accuracy data, these dimensions were reported from the average results of the MD simulations performed in three strains of 0.02, 0.03 and 0.04. As expected, due to the compactness of the equivalent continuum fiber, its geometrical dimensions are smaller than the similar atomistic model (e.g. 3.7 nm for equivalent SWCNT diameter vs. 4.3 nm for its atomistic counterpart as illustrated in Fig. 6a). In the next stage, employing the equivalent nanofillers that accurately account for the structure/property relationships at the nanoscale, the macroscopic mechanical properties of metal composites infused with carbon nanofillers could be determined via the effective finite element method.

Fig. 6. CNT/Cu RVE: (Up) atomic structure and (Down) equivalent continuum fiber.

3.3. Finite element results

Once the process of defining the equivalent-continuum nanofiller was established, then the model was implemented in a subsequent finite element analysis. The FE analysis was carried out on the five main factors influencing on the elastic constants of the nanocomposites that are geometry of the nanosized inclusion, its volume fraction, temperature effects, interfacial interactions, and nanofiller orientation. Following are the brief summary and conclusion of individual analyses conducted for these parameters. To create and analyze the FE models, the commercial code ANSYS was utilized.

3.3.1. Geometrical effect

As the first issue, the multiscale MD/FE multiscale approach developed here was employed to compare the role of SLGSs and CNTs in enhancing the elastic constant of metal matrix composites. It should be noted that since the production technique of SLGSs got common in 2004 [23], the question that raised was: how important is the contribution of nanofiller *geometry* on the nanocomposite mechanical properties? To overcome this issue, here, two distinct RVEs were built and numerically examined under identical tensile loading conditions. Accordingly, employing the equivalent nanofillers obtained in the previous

section, the SLGS/copper and CNT/copper nanocomposite samples were constructed as illustrated in Figs. 7 and 8. As is seen, here, the discontinuous reinforcement model in which the nanofiller is entirely embedded in the matrix was used that is more practical assumption compared to the continuous one [24]. The computational cell used in the FEM analysis was in shape of a rectangular box having square cross-section. Based on the obtained equivalent nanofiller, the size of the RVE containing it and the copper matrix was set so that, in each case, a sample with 1.5% VF of carbon nanofiller was achieved. In this regard, in the case of SLGS-reinforced composite, the size of the RVE and its embedded continuum fiber was $5.227 \times 3.808 \times 20$ nm and $3.485 \times 2.448 \times 14$ nm, in the x, y, and z directions, respectively (Fig. 7a). Additionally, to model the CNT-reinforced copper case, a cylindrical equivalent continuum fiber with the length of 14 nm and the diameter of 3.7 nm was placed in the rectangular RVE with the dimensions of $5.016 \times 5.016 \times 20$ nm in the x, y, and z directions, respectively (Fig. 8a). Afterwards, for each sample, to impose the uniaxial tensile loading, one side of RVE was held fixed and the displacement of 0.6 nm imposed on the other side of the RVE until the desired strain of 0.03 was attained (Figs. 7b and 8b).

Fig. 7. (a) SLGS/Cu RVE containing equivalent continuum fiber with 1.5 % SLGS
(b) Deformed SLGS/Cu sample

Fig. 8. (a) CNT/Cu RVE containing equivalent continuum fiber with 1.5 % CNT
(b) Deformed CNT/Cu sample

Upon completion of loading, the total value of uniaxial forces exerted on the end plate was obtained and effective stress was then calculated in each case. Having the tensile stress at the strain of 0.03, the Young's modulus of SLGS/Cu and CNT/Cu nanocomposite sample was determined to be 67.20 and 63.72 GPa, respectively. Comparing these results with the corresponding value for the pristine copper sample (i.e., 56.7 GPa) demonstrated that at 1.5% VF of SLGSs, the Young's modulus of the nanocomposite was 18.5% greater than the copper matrix. Meanwhile, CNTs showed only 12.4% increase in the base-line copper modulus for the same VF. Therefore, it is concluded that similar to the results of our MD simulation [5] and also previous experimental investigations on polymer-based nanocomposites [25-27], under identical conditions, graphene sheets performed significantly better than CNTs in terms of enhancing the elastic constants of these nanocomposites. This superiority was ascribed to the wrinkled surface of SLGS resulting in enhancing the interlocking between graphene and matrix. It should be emphasized that, the rippled topography of these nanostructures were mainly due to the thermal fluctuations that could be captured successfully by the MD part of the presented method. In other words, the ripples manifested themselves in the MD simulations by altering the load transfer occurred at the interface leading to the enhanced mechanical properties of the equivalent nanofiller.

3.3.2. *Effect of volume fraction*

In the next section, attention was devoted to the analysis of nanofiller VF on the results. To this end, considering copper as the matrix material, six case studies involving 0.625%, 2.25% and 3% VF of SLGSs and CNTs were also investigated under uniaxial tensile loading similar to the aforementioned cases. The corresponding results of these samples can be found in Table 1. As seen, higher magnitude of enhancement was achieved in nanocomposites with more VF of nanofillers. Surprisingly, this trend was much more noticeable in the case of SLGS/Cu sample. For example, percentage increase in Young's modulus of the copper matrix infused with 0.625% and 3% VF of SLGSs was from 9.3% to 35.1%. Meanwhile, this enhancement factor showed a smaller growth from 8.2% to 22.8% at the mentioned volume fraction of CNT.

To verify the accuracy of the results estimated by the proposed method, two sets of numerical experiments were performed. As the first one, employing the results listed in Table 1, we followed the variations of nanocomposite axial Young's modulus in terms of the nanofiller VF. For visual representation of the dependence, the reader is referred to Fig. 9. As seen, these parameters were linearly dependent to each other from which, the axial Young's modulus for the samples with 5% VF of SLGS and CNT were predicted to be 89.09 and 74.46 GPa, respectively. These values implied a negligible deviation from the MD results of completely similar samples reported as 90.1 and 76.8 GPa in our recent publication [5]. Meanwhile, the run-time of the introduced method was negligible compared to that of the MD simulation (e.g. 10 days for MD against only 3 minutes for the developed method on the same hardware). Additionally, as the second test, a computational cell representing copper sample infused with 3% VF of SLGS was examined through full atomistic MD simulation. In this case, the Young's modulus of the nanocomposite was achieved to be 73.1 GPa which is

in good agreement with the corresponding value obtained using the equivalent nanofiller concept (76.6 GPa) introduced in the present research.

Table 1. Young's modulus of nanocomposite samples at different VF of nanofillers.

Cases	0.625%	1.5%	2.25%	%3
SLGS/copper	61.97 (GPa)	67.20 (GPa)	72.09 (GPa)	76.60 (GPa)
CNT/copper	61.36 (GPa)	63.72 (GPa)	66.77 (GPa)	69.61 (GPa)

Fig. 9. Axial Young's modulus of samples vs. nanofiller VF

3.3.3. Temperature effect

After examining the accuracy of the MD/FE multiscale method developed based on the equivalent continuum concept, the next step involved the analysis of temperature fluctuations influence on the mechanical properties of above-mentioned composites. To further investigate the temperature effect, in addition to the room temperature, the Young's modulus of SLGS/Cu composites infused with 1.5% VF of SLGSs, was also determined at two other temperatures (i.e., 1K and 500K). It should be noticed that to define equivalent fiber in each case, MD simulation was performed individually at the desired temperature. Consequently,

employing the obtained results, the equivalent nanofiller was established at each temperature, and hence, new RVE was created for FE analysis. These RVEs were examined under the uniaxial tension test similar to the previous cases. Table 2 illustrates the corresponding elastic constants of these nanocomposite samples along with the results of the pure copper determined at the mentioned temperatures via MD simulation. As revealed, presence of thermal fluctuations caused a decrease in the axial Young's modulus of samples (e.g., from 78.19 GPa at 1 K to 52.57 GPa at 500 K) which is in accordance with the previous researches. It comes from the fact that the main reason for superlative mechanical properties of SLGS is the sp^2 hybridization of carbon atoms in its structure that was changed due to the temperature enhancement.

Table 2. Young's modulus of SLGS/Cu nanocomposite samples at different temperatures along with the results of the pristine copper case.

Cases	1 K	300 K	500 K
Pristine copper	68.72 (GPa)	56.71 (GPa)	42.21 (GPa)
SLGS/copper	78.19 (GPa) (13.8%)	67.20 (GPa) (18.5%)	52.57 (GPa) (24.5%)

Additionally, the percentage of increase in the Young's modulus of each case compared with the pristine copper has been given by numbers in parentheses in this Table. As seen, at higher temperatures, better reinforcement role for SLGSs was achieved. This was ascribed to the rippled structure of SLGS at these temperatures leading to the better interlocking between carbon and metallic atoms at the interface.

3.3.4. Effect of graphene/metal interaction

Previous numerical and experimental investigations have shown that the tremendous mechanical properties of nanofillers used to reinforce different type of matrices can be

captured only if an efficient load transfer exists in the interfacial region. In other words, the reinforcement role of nanofillers is chiefly controlled not only by their superior properties but also by the nature of interactions happening at the interface. In the case of graphene/metal composites there are different types of interactions ranging from strong couplings (e.g., for Ni, Pd and Co) to weaker conjunctions (e.g., for Cu and Al) in the interfacial zone [28,29]. In this section, the effect of interaction between metallic matrix and nanofiller was also studied as one of the most important factors governing the mechanical behavior of this type of nanocomposites.

Table 3. Young's modulus of Ni and Cu based nanocomposite reinforced with 1.5% VF of SLGS at T=300 K.

Pristine Ni	Ni/SLGS	Pristine Cu	Cu/SLGS
90.1 (GPa)	103.2 (GPa)	56.7 (GPa)	67.2 (GPa)

In order to check out the matter, the sequential procedures described previously were carried out in the case of Ni/SLGS nanocomposite sample. To this end, first, MD simulation was conducted to determine the energy required to deform the sample in the uniaxial tensile test. To have a better comparison, the geometrical characteristics, loading conditions, volume fraction and simulation temperature were defined completely similar to the Cu/SLGS case. Afterwards, the dimension of the equivalent continuum fiber was calculated based on the MD results. Finally, a nickel-based RVE embedded with this equivalent nanofiller at the total VF of 1.5% SLGS was constructed and examined via FE analysis. The amount of change in the axial Young's modulus of nickel due to this reinforcement can be found in Table 3. To capture the role of interfacial interactions, the previous results of neat copper and Cu/SLGS sample has also been illustrated. An interesting observation is that, due to the strong cohesion in the interfacial zone, addition of 1.5% VF of SLGS caused a large influence on the Young's

modulus of Ni matrix (~13 GPa increase). Meanwhile, this enhancement was approximately only 10.5 GPa in the case of copper-based sample. Consequently, the specific interactions at the interface should be taken into account when analyzing the mechanical properties of these nanocomposites.

Fig. 10. Schematic illustration of randomly oriented nanofillers in a metal matrix.

3.3.5. Nanofibers orientation effect

In all of the cases examined up to now, it was assumed that the axis of the equivalent nanofiller is the same as the tensile loading direction. However, as schematically illustrated in Fig. 10, it has been experimentally shown that in the production process of nanocomposites, not all of the nanofillers have been oriented in the same direction. Therefore, in practical samples, most of the fillers are loaded in the direction diverse from their longitudinal axis. Considering the fact and knowing the different axial and lateral response of carbon nanofillers [30], the modeling procedure was directed to capture the effects of equivalent nanofiller orientation on the results. To this end, the effective fiber obtained in the case of SLGS/Cu sample was served as a means to construct RVEs containing nanofillers distributed at different schemes. This is given in Fig. 11. These RVEs included eight equivalent fibers with 1% VF of SLGS at the temperature of 300 K. As the first case (Fig. 11a), an ideal

sample was analyzed, in which, all of the fibers were located in the loading axial direction. This was chosen as a suitable criterion for evaluating the orientation effects. Table 4 features the axial elastic modulus of this sample along with the results of other three RVEs. Similar to the previous sections, to impose uniaxial tensile loading in each case, one side of the RVE was held fixed, while the appropriate displacement was imposed on the other side until the desired strain of 0.03.

Table 4. Young's modulus of the samples shown in Fig. 11 infused with 1% VF of SLGS at T=300 K along with the corresponding value determined through the linear equation in Fig. 9.

Case I	Case II	Case III	Case IV	From Fig. 9
64.40 (GPa)	58.62 (GPa)	61.21 (GPa)	63.71 (GPa)	64.24 (GPa)

Fig. 11. RVEs representing Cu matrix containing 8 SLGS equivalent fibers oriented at different schemes: (a) Case I: Coaxial (b) Case II: at the angle of 10° relative to the loading axis, (c) Case III: Randomly oriented, and (d) Case IV: Symmetrically oriented at the angle of 10° and -10° .

As expected before, the maximum modulus occurred for the fibers aligned parallel with the loading direction (i.e., Case I shown in Fig. 11a). According to this table, the Young's modulus achieved in this case was approximately 64.40 GPa which has a good agreement with the value predicted by the linear equation found in Fig. 9 for SLGS reinforced samples. Substituting graphene VF as 1% in the afore-mentioned equation gives the stiffness of the sample as 64.24 GPa stating the validity of the modeling procedure and results of new RVEs utilized in this section. As mentioned previously, to investigate the orientation dependency of the results, three RVEs representing several arrangements of fibers were also considered keeping all of the other parameters fixed (See Fig. 11). According to the results listed in Table 4, arrangement of fibers at the angle of 10° (referred to as Case II) caused a decrease of 9% in the Young's modulus from 64.40 to 58.62 GPa. It comes from the fact that, in this case, a smaller part of the each fiber was loaded in the axis loading direction. Therefore, these fibers had a less role in load-bearing contrary to the coaxial fibers in Case I.

As the next case, the RVE including randomly oriented fibers was examined (Fig. 11c). Compared to the previous case, the greater Young's modulus (61.20 GPa) was attained for this sample that was attributed to the counteracting negative effects of fibers by each other. It seems that the resultant forces tolerated by some of the fibers positioned at different directions (e.g. 10° and -10°) would align with the loading direction resulting in a greater stiffness in this case. The validity of this assumption was proved based on the results of the last sample (Case IV). As seen in Fig. 11d, in this RVE, all of the equivalent fibers were located symmetrically to each other at the angle of 10° and -10° . Therefore, as expected before, the Young's modulus of nanocomposite was approximately 63.71 GPa, which has a negligible disparity ($\sim 1\%$) with the results of the first sample. It should be noted that this analysis will be important for the development of metal-based nanocomposites where the nanofiller orientation distribution cannot be ignored in practical applications.

4. Conclusion

From the research that has been carried out, a method was introduced for linking atomic-scale interfacial interactions in metal-based nanocomposites to continuum models governing the macroscopic behavior of the corresponding bulk material. The linkage was done through equivalent nanofiller replaced for the nanofiller, its surrounding metal atoms and the interface. To extract the geometrical and mechanical characteristics of these equivalent fillers, the concept of equivalent continuum modeling was employed. Accordingly, conducting several case studies, the elastic constants of this type of nanocomposites were determined via finite element analysis of RVEs including these atomistically informed nanofillers and also, by MD simulation of their counterpart full atomistic models. It was concluded that the equivalent nanofiller is an effective way to transfer atomic-scale information providing a route to investigation of the nanocomposites in less time compared to MD simulations with the same accuracy. Subsequently, the hierarchical MD/FE multiscale methodology developed in the present work was utilized to examine the crucial factors affecting the mechanical behavior of MMNCs with the advantage of the accuracy of MD and speed of FEM, simultaneously.

It is worthy to emphasize that as long as the metal matrix, type of nanofiller, simulation temperature, and interfacial interactions are kept fixed, the equivalent continuum fiber is still valid in terms of its geometrical and mechanical characteristics. Accordingly, the complex and time-consuming part of the approach (i.e., MD simulation) can be carried out once for each case of study. The obtained equivalent fiber can then be implemented simply in an FEM analysis to study the effects of different parameters on the elastic constants of the samples. As the main results, it was shown that under identical conditions, due to the better interlocking between SLGSs and metal matrix, these carbon nanostructures performed significantly better than CNTs in terms of enhancing the elastic constants of these nanocomposites. This was due

1 to the ripples produced on the surface of SLGSs that can be captured by the MD part of the
2 presented method. Furthermore, we also examined the influences of nanofiller VF on the
3 results and found a linear trend governing the dependency in each case. Additionally,
4 existence of MD simulations in the proposed approach enabled us to investigate the
5 temperature and metal/nanofiller interaction effects on the elastic constants of these MMNCs.
6
7 Finally, it was observed that the mentioned properties are highly dependent to the nanofiller
8 orientation distributions. The proposed method can be readily used to design of this new class
9 of nanocomposites in a detailed way without compromises of the computational cost.
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CNT/Cu RVE: (Up) atomic structure and (Down) equivalent continuum fiber