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Research Article

SYNERGETIC EFFECTS OF GRAPHENE AND CARBON NANOTUBES AS FILLERS IN REINFORCED EPOXY NANOCOMPOSITES: A MULTISCALE APPROACH

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ABSTRACT

In this study, a two-step modeling method has been proposed to obtain the mechanical properties of epoxy nanocomposites containing single-wall carbon nanotubes (SWNTs) and graphene nanoplatelets (GNPs). Initially, by using molecular dynamics simulation with Lammps software, the mechanical properties of epoxy resin and two nanoparticles of single-walled carbon tubes and graphene nanoplatelets were calculated. Then, the mechanical properties of the composites derived from these materials were extracted by applying the finite element method with Abacus software. The synergistic effect of GNPs and SWNTs on improvement of the mechanical properties of epoxy resin has been investigated. It was found that adding 1 wt.% of these nanoparticles to the epoxy matrix had a higher increase in the Young's modulus and shear modulus compared to the epoxy matrix when the CNTs / GNPs volume fraction ratio was equal to 25:75 (41% increase).

Keywords: nanocomposites, graphene, Synergetic

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INTRODUCTION

Nanomaterials are extensively used, due to their high surface to volume ratios and remarkable mechanical properties. One of the most significant aims of the usage of nanomaterials is the reinforcement of the mechanical properties of various materials. Addition of a small number of nanomaterials to polymers may lead to a considerable increase in mechanical properties.

In the last few years, there have been growing investigations on the mechanical properties of nanocomposites which include polymers such as epoxy, polyethylene or polypropylene as the base matrix and nanomaterials like carbon nanotube and graphene nanoplatelet as the reinforcement. Robert et al. [1] have comprehensively studied the mechanical properties of nanocomposites which have been used nanographene in their structure as the reinforcing material. Also, in 2015, Garima et al. [2] summarized the main results of the researches on reinforcing properties of polymer nanocomposites by using carbon nanotube or graphene.

In the last years, using a hybrid of nanoparticles in polymers reinforcement has attracted much attention from researchers. In this literature, researches demonstrated that due to the unique and disparate properties of each nanoparticle, their combination and synergism to reinforce the mechanical properties of polymers have a better impression in comparison to their separate usage.

Jing et al. [3] experimentally studied the effects of hybrid carbon nanotubes (CNT) and graphite nanoplatelets (GNPs) on the mechanical properties of epoxy nanocomposites.

K.Eswar et al. [4] studied the effects of binary combinations of nanodiamond, graphene and carbon nanotubes on the reinforcement of mechanical properties of polyvinyl alcohol.

Using the combination of carbon nanotubes and graphene nanoplates on the reinforcement of mechanical properties of polymers have been widely researched rather than other nanoparticles combinations [5-15]. Moreover, numerous experimental studies have been done on the effect of reinforcement of the combination of particles.

The previous researches have shown that the combination of carbon nanotubes and carbon nanoplates for the reinforcement of polymers aims to the formation of a connected network of nanoparticles in the whole of the material structure and the existence of the network has a tremendous effect on the improvement of mechanical properties of nanocomposites[10].

Using the graphene nanoplates to reinforce the polymers leads to an enhanced mechanical interlocking of filler with the polymer chains and enlarged interphase zone at filler–polymer interface due to the high specific surface area and nanoscale 2-D flat surface.

While this effect is less pronounced in CNTs based nanocomposites because the CNTs have a reduced surface area and can interact with the polymer only at 1-D linear contact [16, 17].

On the other hand, the addition of one-dimensional CNTs to nanocomposites which include polymer and two-dimensional GNPs leads to better dispersion of GNPs in the base polymer and also, will reduce the π-π stacking and agglomeration of the graphene nano-plates in the polymer.

Longfei et al. [7] experimentally studied the reinforcement effect of the mechanical characteristics of polyamide 6 by using the combination of CNTs and GNPs. In the investigation of nanocomposite samples, no obvious aggregation of CNTs and GNPs was observed. Also, this research demonstrated that the tensile strength of the nanocomposite which consists of 0.2 % of GNPs and 0.3 % of CNTs (weight percent) is 2.4 times higher than the tensile strength of pure polyamide 6.

Liang et al. discussed the synergy effects and the dispersion of CNTs and GNPs to reinforce the epoxy polymer. The results illustrated that for the constant value of 0.1% for the total weight percent of CNTs and GNPs, the most value of flexural modulus of nanocomposite will be at the combination of CNT and GNP in a ratio 8:2. Enkeleda et al [5] studied the usage of different concentration of CNT and GNP for the reinforcement of mechanical properties of a biocompatible polymer. The results showed the effect of the combination of these nano-fillers in the reinforcement of elastoplastic properties of nano-composites such as modulus of elasticity and amount of stress and load required to break is much more than their separate use. This is more significant at lower concentration values of nano-composites in comparison to higher concentration counterparts. The researchers proposed that due to superior elastoplastic properties, the hybrid nano-composites are usable in a number of areas including military complex composites, energy, automotive industry (lighter and stronger car parts), and nano-medicine (as scaffolds for bone formation/regeneration).

Weikang et al [9] performed an experimental study on the reinforcement of epoxy with the combination of CNT and GNP. The results indicated that the embedding of CNT–GNP hybrids into pristine epoxy endows optimum dispersion of CNTs and GNPs as well as better interfacial adhesion between the carbon fillers and matrix, which results in a significant improvement in load transfer effectiveness. Remarkably enhanced mechanical properties in the CNT–GNP/epoxy composite were achieved at ultralow hybrid loading (0.5 wt.%).

Chatterjee et al. [10] investigated the effect of GNPs size and their combination with CNTs for improvement of mechanical properties of epoxy resin. They concluded

that the combination of the high aspect ratio of CNTs and larger surface area of GNPs contribute to the synergistic effect of the hybrid samples.

Yang et al. [12] examined the Synergistic effects of graphene platelets and carbon nanotubes on the mechanical and thermal properties of epoxy composites.

Most of the previous studies on the reinforcement of mechanical properties of polymers with various types of nanofillers have been carried out experimentally. In the present research, by conducting a theoretical study and also, by using multi-scale research, the effect of the combination of CNT and GNP in the reinforcement of mechanical properties was assessed. Firstly, by conducting molecular dynamics and atomic modeling of two disparate representative volume element (RVE) which include one of the nanofillers with their around polymer, the homogenized elastic properties of RVEs, including the influence of the interphase region was determined and then, by applying finite element method, the random distribution of nanofillers with different concentrations in the polymer and their effect of mechanical properties will be discussed.

MOLECULAR DYNAMIC SIMULATION

General

Molecular dynamics (MD) simulation is a powerful tool for numerical experiments of nanocomposites [18,19]. The fundamental concept behind molecular dynamics is to apply Newton's equations of motion in solving multi-body interaction system (i.e. polymers, crystals, nanocomposites, etc.). The molecular interactions play an important role in MD simulation and here, they are defined by an appropriate force-field. The key components of a force-field are bonded interactions and non-bonded interactions such as van der Waals force. This makes molecular dynamics simulation to be a primary tool for assessing the mechanical properties of polymer nanocomposites.

In the present study, to perform a molecular design simulation, a large-scale atomic/molecular massively parallel simulator (LAMMPS) software was utilized. On the other hand, an epoxy resin (EPON 862) with its hardener (TETA) was considered as the matrix. The combination of the epoxy resin with nanoparticles have been used in applications such as aerospace and aviation structures. In order to provide a sufficient number of chemical reactive sites for the crosslinking reaction, the resin/hardener ratio in the modeled unit cell is 3:1

During the cross-linking process, reactive sites in EPON 862 (epoxide groups) form new bonds with their counterparts in TETA hardener (sites containing NH and NH2), which result in a highly cross-linked epoxy structure, as depicted in Fig. 3. If they get close enough to each other within the range of 4–10 Å [20], the cross-linking occurs.

CNT and GNP were selected as the matrix reinforcement nanofillers. Periodic boundary conditions were imposed on all directions of the MD unit cells. The interatomic (Carbon) interactions in CNT and graphene are defined with the adaptive intermolecular reactive empirical bond-order (AIREBO) [21] potential function, while the Lennard–Jones Potential is adopted for characterizing the interatomic interaction between the polymer monomers and the carbon nano-fillers and also, among carbon nano-fillers. All the potential parameters for polymer beads and hybrid reinforcements and their interaction are defined by Zhang et al. [22]. Since the potentials for the bond break are not available, covalent bonds between polymer and carbon nano-fillers are not regarded in the present study.

Also, the interactions between the atoms of polymer chains are modeled by using the modified AMBER potential functions [23]. The van der Waals interactions were modeled with an interaction cutoff radius of 10Å [24,25]. Conjugate gradient algorithm was applied to minimize the total potential energy of the initial configurations, while the velocity Verlet algorithm was used to integrate the equations of motion in all the MD simulations.

Modeling of pure epoxy resin

The initial cross-linked polymer molecular structure was established consisting of 3 EPON 862 molecules cross-linked with 1 molecule of TETA curing agent (see Fig.1). The cross-linked structure was utilized to form a 3D structure, as shown in Fig. 1. Then, this cross-linked epoxy structure was used to build the epoxy system in the subsequent MD simulations for neat epoxy, CNT-epoxy, and GNP-epoxy composites.

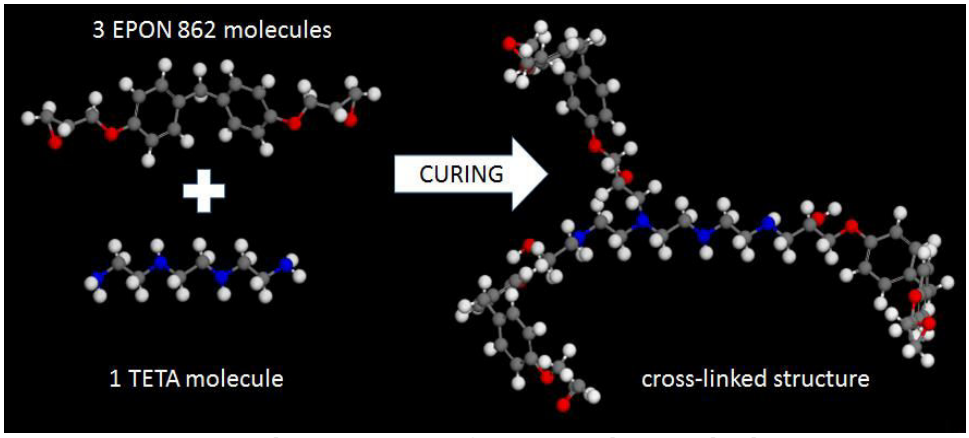

Fig. 1. Schematic structure of EPON 862 and TETA molecules

For generating the neat epoxy model, fifty cross-linked structures were replicated and placed in a cubic simulation box with the size of 61 Å × 61 Å × 61 Å and the replicated structures were randomly rotated and then translated along the three Cartesian axes and form a system containing 150 chains of EPON 862 and 50 chains of TETA, as shown in Fig. 2a. The total number of atoms in the simulation box is 7850.

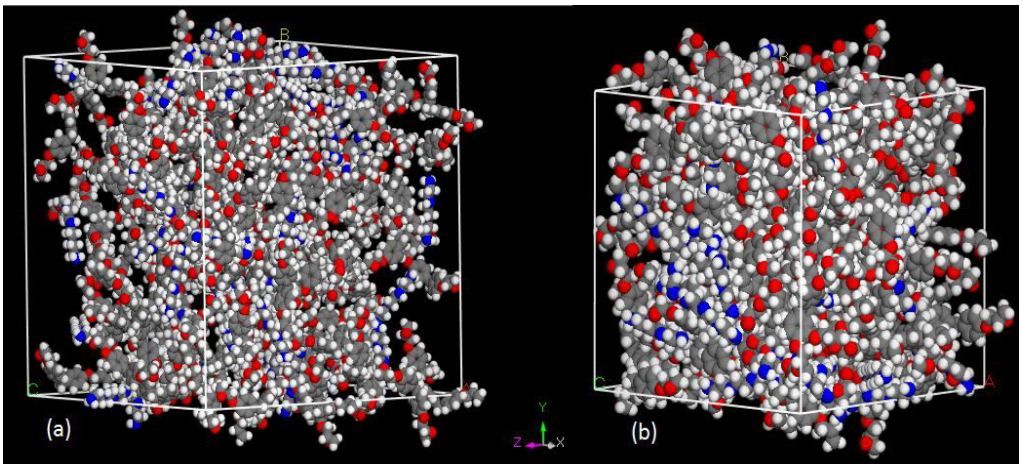

Fig. 2. Simulation boxes: (a) the initial structure with 0.4 g/cm³ density (b) the final structure of the system with 1.11 g/cm³ density

The simulation box was compressed gradually through 20 steps from its initial size of 61 Å × 61 Å × 61 Å to the dimensions of 42 Å× 42 Å × 42 Å (see Fig. 2b). At each stage, the atoms coordinates were remapped to fit inside the compressed box, then a minimization simulation was conducted to relax the coordinates of the atoms. The system was considered to be optimized once the change in the total potential energy of the system between subsequent steps is less than 1.0×10^{-4} kcal/mol. The optimized system was then equilibrated at room temperature in the constant temperature and volume canonical (NVT) ensemble over 200 ps by using a time step of 1 fs.

The compressed system was equilibrated for another 400 ps in the isothermal-isobaric (NPT) ensemble at 298 K and 1 atm to generate an epoxy system with the correct density and, to reduce the induced residual stresses which originate from the volume reduction. This equilibration step resulted in an equilibrated amorphous structure with an average density of 1.11 g/cm³ (The diagram is shown in Fig. 3). That this density corresponds to the actual value of the resin density [26] indicates the accuracy of molecular dynamics simulations. At the end, in order to achieve a fully relaxed structure, the system is again equilibrated for 1 ns in the canonical (NVT) ensemble at 500 K, followed by NVT annealing to 298 K at the cooling rate of 10 K/10 ps.

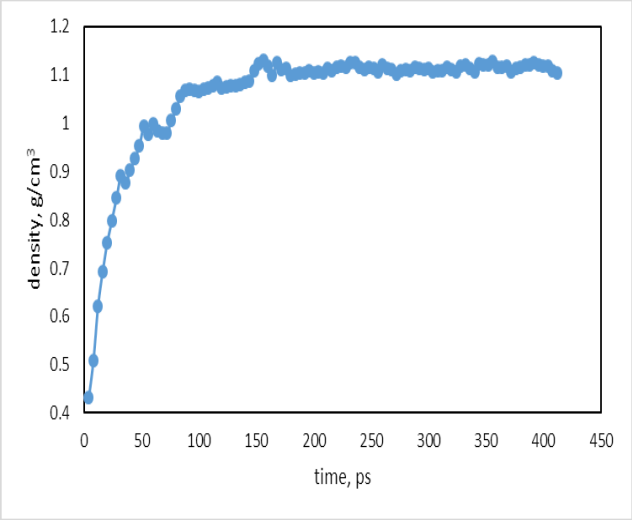

Fig. 3. density of EPON 862 /TETA system

The simulation box was subjected to uniaxial mechanical deformations to predict its elastic mechanical responses. Previous studies of molecular dynamics simulations report that the calculated Young's modulus in polymeric materials are not sensitive to strain rate changes [27-29]. So at a certain strain rate the model was deformed with uniaxial 2.5% strains in tension and compression over a period of 0.5 ns. Additionally, shear deformations of 2.5% were performed along the x-y plane. The values of Young's modulus, the shear modulus in the x-y plane (Gxy), and the Poisson's ratios for the simulation box are given in Table 1. These moduli are consistent with the experimentally measured moduli of a similar epoxy resin [30]. This is also a proof of the correctness of the simulation.

Table 1. Elastic moduli of the epoxy simulation box

Young's modulus (GPa)	Shear modulus (GPa)	Poisson's ratio
3.456	1.452	0.387

MD simulation of graphene with its surrounding epoxy polymer

Graphene nanoplatelet (GNP) is a two-dimensional nanostructure which consists of carbon atoms bonded by sp^2 hybridized electrons. The carbon atoms are arranged in a hexagonal pattern with the shortest distance of 1.42 Å between atoms and with a bond angle 120^0 (shown in Fig. 5). According to up-to-date research, GNPs offer more advantages towards highly improving mechanical properties in the polymer composites [16, 31]. Because their planar structure and ultrahigh aspect ratio are expected to endow better stress transfer from GNPs to the matrix during loading.

In this section, by modeling a representative volume element (RVE) consisting of a GNP and surrounding epoxy polymer, the effect of graphene in improving the mechanical properties of the polymer is investigated.

For generating the RVE model, a two layers zigzag GNP was placed in the center of a cubic simulation box of size 24.6 Å × 24.6 Å × 178.4 Å and 20 number of cross-linked polymer molecular structures were replicated and placed on each side of GNP as shown in Fig. 4. In the simulation box, x and y represent graphene length and width direction and z corresponds to the thickness direction. The energy minimization, equilibration and density adjustment of the RVE were performed as the same steps as described above for pure epoxy. The final dimensions of the equilibrated RVE will be 24.6 Å × 24.6 Å × 92.85 Å.

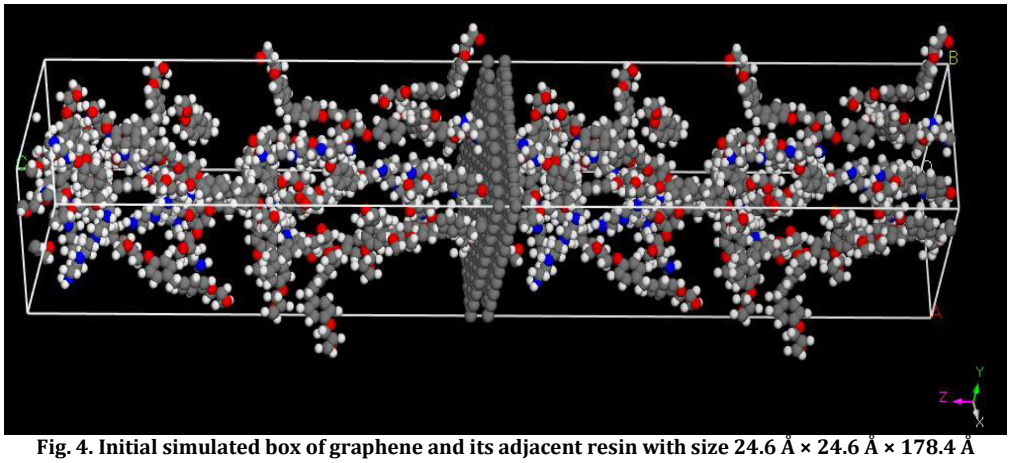

Fig. 4. Initial simulated box of graphene and its adjacent resin with size 24.6 Å × 24.6 Å × 178.4 Å

The RVE was subjected to uniaxial mechanical deformations to predict its elastic mechanical responses. The model was deformed with uniaxial 5% strains in tension and compression along the x-, y-, and z-axes over a period of 500 ps. The Poisson contraction was allowed in the transverse directions for the direct calculation of Young's modulus and Poisson's ratio. Additionally, shear deformations of 5% were performed separately along the x-y, y-z, and x-z planes over a period of 500 ps for the model. The values of Young's modulus in the three orthogonal directions (Ex, Ey, Ez), the shear modulus in the x-y plane (Gxy), and the Poisson's ratios for the model are given in Table 2.

Table 2. Elastic moduli of the graphene with its surrounding epoxy

Young's modulus in X direction, E_X (GPa)	Young's modulus in Y direction, E_Y (GPa)	Young's modulus in Z direction, E_Z (GPa)	Shear modulus in X-Y plane, GXY (GPa)	Poisson's ratio υ_{XY} and υ_{YX}
1251.8	1251.8	56.7	145.01	0.12

The shear modulus values in the y-z and x-z planes are not included in Table 2 because they were nearly zero-valued based on the dominance of the van der Waals bonds between the graphene sheets and polymer and the periodic boundary conditions. As expected, the value of E_z are much lower than those of E_x and E_y, due to the significance of the van der Waals forces in that direction and also, because the graphene is aligned in the x-y plane. From Table 2 it can be seen that Ex and Ey are nearly identical, as is expected given the material symmetry (Fig. 5). Ez was much lower in magnitude than Ex and Ey, because the graphene sheet was oriented in the x-y plane.

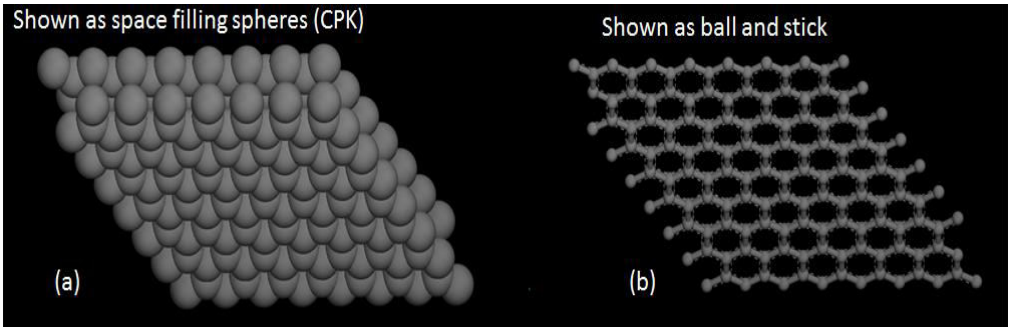

Fig. 5. Schematic of GNPs in two forms: (a) space filling spheres and (b) ball and stick

The mechanical properties of the resin are much weaker than the properties of GNP nanoparticles. Therefore, the stress tolerance is the main task of the GNP nanoparticles and the adjacent resin of nanoparticles plays no role in stress tolerance and mechanical properties. By comparing the mechanical properties obtained in molecular dynamic simulation of GNP nanoparticles (Table 2) with experimental data and theory [32-35] it was determined that the results are in very good agreement with the references mentioned, and the stress tolerance is entirely on the nanoparticles and the properties mentioned in Table 2 are completely related to the particles. The resin in this case only does the job of nano-particle sizing (surface modification).

MD simulation of SWCNT with its surrounding epoxy polymer

CNTs could be defined as relatively long and thin fullerene structures. Their tubular walls are made of hexagonal carbon cells. They have been used in an extensive range of applications including ultra-strong composite materials and nanofibers. Their good chemical stability, large surface area, small size, low density, high stiffness, high strength, and excellent thermal properties have demonstrated high potentials in advanced material and Nanomechanical devices. The outstanding mechanical properties of carbon nanotubes (CNTs) make them highly desirable as potential reinforcing constituents in structural composites.

In this section, we obtain the mechanical properties of a representative volume element (RVE) consists of an embedded single-walled CNT in epoxy polymer by using the MD simulations. In MD simulations, an armchair type CNT (as shown in Fig. 6) with a chiral vector of (10,0) is

chosen as reinforcement and was placed in the center of a Cubically simulation box of size 40 Å × 40 Å × 80 Å and 40 number of cross-linked polymer molecular structures were placed in sides of CNT.

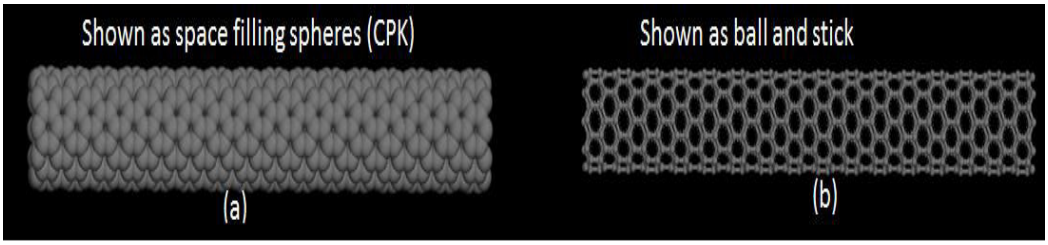

Fig. 6. Schematic of SWCNT (10,0) in two forms: (a) space filling spheres and (b) ball and stick

The steps involved in the MD simulations, energy minimization, equilibration and obtaining the elastic coefficients of the RVE are the same as adopted in the above sections. The final dimensions of the equilibrated RVE containing the CNT and the surrounding polymer are equal to 27 Å × 27 Å × 80 Å, and its mechanical properties are presented in Table.3.

Table 3. Elastic moduli of the SWCNT with its surrounding epoxy

Young's modulus in X direction, E_X (GPa)	Young's modulus in Y direction, E_Y (GPa)	Young's modulus in Z direction, E_Z (GPa)	Shear modulus in X-Y plane, GXY (GPa)	Poisson's ratio		
				υ_{YX} And υ_{XY}	υ_{XZ} and υ_{YZ}	υ_{ZY} and υ_{ZX}
28.12	28.95	1017.08	35.51	0.76	0.025	0.10

Similar to the preceding section, the mechanical properties of the resin over CNT nanoparticle are much weaker and negligible. Then again, the nanoparticle adjacent resin plays no role in stress tolerance and mechanical properties. By comparing the mechanical properties obtained in molecular dynamic simulation of CNT nanoparticles (Table 3) with experimental data and theory [36-38] it was determined that The results are in very good agreement with the references mentioned, and the stress tolerance is entirely on the nanoparticles and the properties mentioned in Table 3 are completely related to the particles. The resin in this case only does the job of nano-particle sizing (surface modification).

It was shown that nanocomposites with low carbon nanotubes content possess a homogeneous microstructure, the nanotubes being efficiently dispersed into the polymer matrix [18, 39-42]. At relatively high carbon nanotubes contents (above 1 wt.%) a tendency for agglomeration was observed due to the tendency of nanotubes to interact with each other. In the current study, for reinforcing the polymer, a low volume fraction of CNTs will be used and thus the effects of the local aggregation of the CNT are excluded.

MICRO MECHANIC MODELING

After determining the mechanical response of the molecular models, their mechanical properties were used as input to the next higher length-scale (continuum) analysis.

In this section, a number of 3D Finite Element (FE) models were developed to predict mechanical properties of epoxy composite reinforced with the different volume fraction of SWCNTs and GNPs.

Each generated model for nanocomposite was comprised of a cubic volume of epoxy polymer with randomly dispersed nanofillers inside the polymer. Here, five separate models that each of them has certain volume fractions of one of the SWCNTs or GNPs or a combination of them were generated and their mechanical properties were determined. The sum of the concentrations of the GNPs and SWCNTs in all models was selected equal to 1 wt.% as above this concentration there can be increased in nanofillers agglomeration in the composites. The values of volume fractions ratio of the GNPs to CNTs in the 5 above models were presented in the Table 4.

Table 4. Volume fractions ratio of GNPs: CNTs

Model nom.	1	2	3	4	5
GNPs:CNTs	100:0	75:25	50:50	25:75	0:100

Models Generation

The previous models have shown that the nanocomposite has been created from random dispersion of nanofillers throughout a cubic volume of the epoxy polymer. These models were generated by using the visual basic software. Dimensions of all the models in the x, y and z directions are similar and equal to 4 μm. (The models were generated by using the Python-based software code.)

In the 3D models generation, the RVEs of the nanofillers which have been created and described in the previous sections represent nanofillers. As described above, each RVE consist of a single GNP or SWCNT, surrounding polymer epoxy and the interphase region between the nanofiller and polymer. Although each of RVE's part has certain mechanical properties, during the process of placing the RVEs into the model, only a homogeneous volume with the averaged elastic properties which has been calculated and presented in the last sections are inserted into the model.

In the embedding process of random nanofillers within the base polymer and creation of models, firstly, planar GNPs are embedded and then, rod-like wavy SWCNTs are embedded.

Embedding of the GNPs inside the 3-D model of the nanocomposite was carried out randomly. In this process, firstly, the position and the embedding orientation of the initial GNP inside the boundary of nanocomposite was selected randomly.

The next GNPs enter the model one by one, respectively, with coordination and random orientation. After inserting each GNP, the intersection of GNP with previous GNPs became controlled. If there is an intersection between the GNP and the previous GNPs, it will be eliminated and another GNP with unique coordination and orientation will be embedded in the model. This process will continue until it reaches its regarded volume fraction for the GNP inside the model.

In order to maintain the periodicity condition of the model, parts of GNP that exceeds the boundary of the model are cut and shifted to the opposite boundary. [43].

In the next stage, CNTs were embedded in the model wavy and randomly. Considering the waviness of CNTs in the model results that the model becomes more realistic and the CNTs can be embedded in the model more easily.

The modeled CNTs have a length equal to 600 nm. In order to achieve wavy modeling, each CNT was divided into 10 cohesive segments with

an equal length which can be embedded in various orientations. Every two neighboring segments were joined together with a 3D spline.
Each of these segments was modeled as a straight cylinder with a 60nm length and 7.83 Å diameter and their mechanical properties were inserted to the model according to calculated values in section 2.4.
The first step for building the model of CNT is the random choice of the start point's coordination and the spatial orientation of the first segment of the CNT. After embedding the segment inside the Nano composite's model, the possibility of intersection between the CNT and the other CNTs and GNPs which have been entered in the model was controlled. If there is an intersection, the coordinates of the start point and the other spatial orientation will be randomly chosen for that segment. After determining the definite position of the first segment inside the Nanocomposite, the coordinates of the endpoint of the first segment will be calculated and then, this point will be regarded as the start point of the next segment. By choosing the spatial orientation for the second segment, its embedding condition will be determined. Again, the possibility of collision of the segment with the previously entered nanoparticles will be controlled and this method will be repeated for embedding the next segments in the model. The embedding of the CNTs inside the Nanocomposite will be kept on until reaching the defined volume fraction.

Models Analysis

The analysis of the models which have been created in the above section was done by the Finite Element analysis method and using the Abaqus software.
If we intend to carry out the finite element analysis for the model which includes polymer and its inside nanoparticles by applying the common and classic method, for analyzing an integrated classic model which consists of the random distribution of a large number of CNTs and GNPs, we have to perform heavy and time-consuming computational calculations or we have to model and analyze a smaller counterpart which may lead to reduction of response accuracy.
So, in this problem, instead of applying the classic analysis model, technique Embedded Regions (ER) was used that results into the reduction of computational calculations and its error will be an acceptable value. In this method, firstly, the meshes of CNT and GNP and the host polymer will be created independently and then, the built meshes will be linked together and their interaction will be considered. For each node of a given CNT or GNP, the closest matrix element face is defined. Then, each of its nodes and the given nanofiller node are constrained by the equations which are expressing the equality of their interpolated displacements. By the procedure, the ER technique introduces stiffness of each nanofiller into the model by constraining degrees of freedom at the nanofiller nodes to the matrix nodes [44].
It should be noted that in modeling with the ER technique, the elements describing nanofillers and polymer are physically overlaid on each other, causing an approximation in terms of the phase volumes into the models. The resulting errors are minor at the low nanofiller volume fractions considered here [45].
The three-dimensional 4-node linear tetrahedron solid element C3D4 is used for meshing the matrix and nanofillers. In the modeling, an assumption was made that there is a perfect bonding at the interface of nanofillers and base polymer.
Because of the homogenous and randomly dispersion of nanofillers in the polymer matrix, the created models of nanocomposite have isotropic elastic properties. By subjecting the 3D models to uniaxial mechanical deformations, their elastic mechanical responses were determined.
Uniaxial deformations of 2% strain in tension were performed along the x-axes direction for each model. Separately the models were deformed with shear deformations of 2% strains along the x-y plane. These values of strain correspond to the level of the deformation prior to which no damage yet appears in the nanocomposites. Representative stress vs. strain curves for the tensile deformation along the x-axis and shear in the x-y plane is shown in Fig. 7 and Fig. 8. The values of Young's modulus (E), the Poisson's ratios and the shear modulus (G) for all 5 models are presented in Table 5.

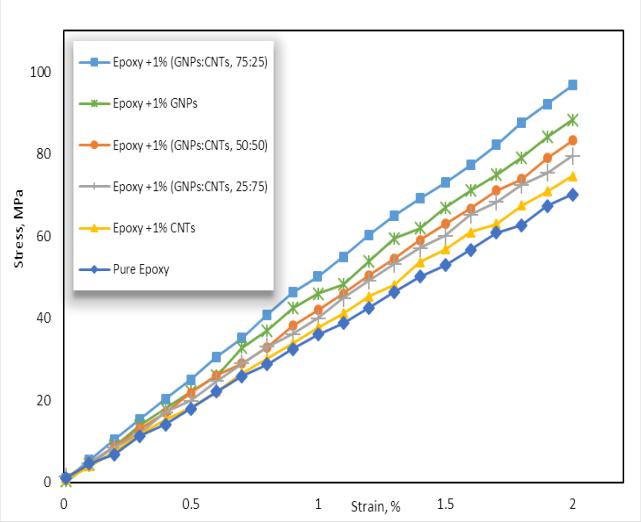

Fig. 7. stress vs. strain curve for tensile mode in x-axis

Figures 7 and 8 show the stress-strain diagram in the tensile and shear modes. As you can see in these graphs, by adding 1% by weight of nanoparticles to the epoxy matrix, the mechanical properties of the resin are improved. This increase in properties is generally more evident in composites with higher GNP content. And in a composite containing a CNTs / GNPs volume fraction ratio was equal to 25:75, it has the highest slope of the stress-strain graph, or in other words, it has the highest Young's modulus and shear modulus. The reason for this increase in mechanical properties, in this case, can be attributed to the resulting composite microstructure and the optimal arrangement of the nanoparticles together, which ultimately leads to a better stress transfer mechanism than the matrix to the nanoparticles. In this case, the nanoparticles will undergo more stress.
On the other hand, in sections 2.3 and 2.4, Young's modulus and shear modulus values for the nanoparticles were obtained and it was found that these properties were higher for GNP nanoparticles than for CNT nanoparticles. This has led to higher mechanical properties in composites where graphene nanoplatelets content is higher than carbon nanotubes (Excluding the GNPs / CNTs volume fraction ratio of 100: 0 where the Young's modulus and shear modulus is less than the 75: 25 volume fraction ratio).

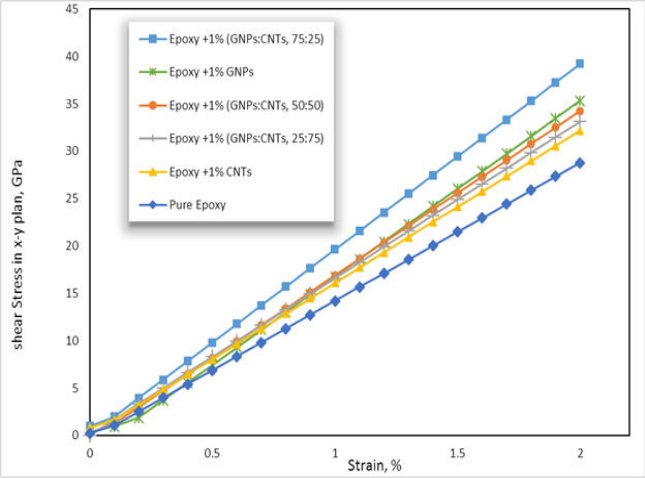

Fig. 8. stress vs. strain curve for shear mode in x-y plane

Table 5 and Figure 9 show the shear modulus and Young's modulus values for the 5 types of composites with different volume fraction of graphene nanoplatelets to carbon nanotube. The maximum values of Young's modulus and shear modulus are 4.89 GPa and 1.949 GPa, respectively, for nanocomposites modeled by adding GNPs to CNTs with a 75:25 volume ratio. That's a 41% increase over pure epoxy.

On the other hand, the lowest Young's modulus value among the nanocomposites is that of a composite containing 1 wt.% of carbon nanotube. But compared to pure epoxy, the nanocomposite Young's modulus increased by 7%. In other experimental and theoretical work, researchers have confirmed this increase in mechanical properties. [48-50]

As you can see from the results in Table 5, the addition of nanoparticles to the epoxy matrix did not change the Poisson ratio significantly. Also, since composites are considered to be isotropic. The relationship between the shear modulus and Young's modulus will be straightforward

according to the E = 2G(1 + U) relation. Here, too, there was a direct relationship between the increase in values obtained for both shear modulus and Young's modulus.

Table 5. Mechanical properties of 5 composite types with 1 wt.% of total nanoparticles

	GNPs /CNTs ratio:					Epoxy pure
	100:0	75:25	50:50	25:75	0:100	
E (Gpa)	4.444	4.890	4.130	3.972	3.697	3.456
v	0.356	0.367	0.361	0.371	0.341	0.387
G (Gpa)	1.846	1.949	1.728	1.658	1.598	1.452

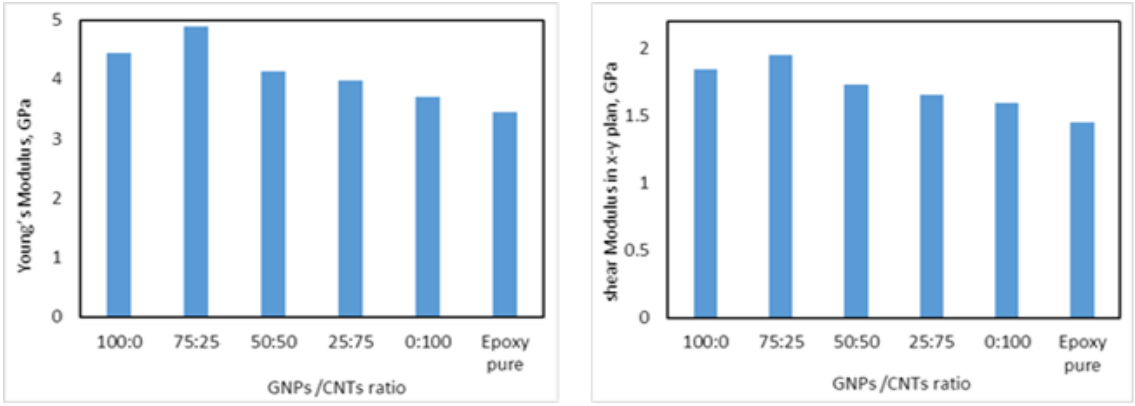

Fig. 9. Elastic modulus vs. GNPs: CNTs ratio

CONCLUSION

In this research, the mechanical properties of epoxy nanocomposites have been studied in a multiscale Approach by molecular dynamics simulation and finite element method. The following results are obtained from this study.

Through the molecular dynamics simulation method, the properties of the particles are obtained. For GNPs, the Young's modulus was 1251.8GPa and the shear modulus was 145.01GPa. Also for the CNT nanoparticles, the Young's modulus of 1017.08GPa in the longitudinal direction and the shear modulus of 35.51GPa are calculated.

The synergistic effect of two GNPS and SWNTs on improving the mechanical properties of epoxy resin has been investigated. It was found that adding 1 wt.% of these nanoparticles to the epoxy matrix had a higher increase in the Young's modulus and shear modulus compared to the epoxy matrix when the CNTs / GNPs volume fraction ratio was equal to 25:75. The reason for this increase in the mechanical properties of this composite can be attributed to the stress transfer mechanism better than the matrix to the nanoparticles.

As for the Poisson's proportion, the addition of nanoparticles to the epoxy matrix did not cause a significant change in this parameter.

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