

## Viscoelastic behavior of Silica nanoparticle/polyimide nanocomposites using finite element approach

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### Abstract

A three-dimensional micromechanical finite element model is developed to study the viscoelastic behavior of the silica nanoparticle/polyimide nanocomposites. The representative volume element (RVE) of the model consists of three phases including silica nanoparticle, polyimide matrix and interphase which surrounds the nanoparticle. The interphase region is created due to the interaction between the silica nanoparticle and the polymer matrix. The effects of different important parameters such as interphase material properties and thickness, silica nanoparticle volume fraction and geometry as well as type of nanoparticles distribution are investigated. It is found that the interphase significantly affects the viscoelastic behavior of the nanocomposites. Also, the results reveal that with decreasing the nanoparticle diameter or increasing volume fraction, the creep strain of the nanocomposite reduces. Moreover, the creep strain of the nanocomposites decreases with the uniform distribution of the nanoparticles inside the polymer matrix. It is shown that for the elastic properties of the nanocomposites, while the predictions without interphase are far from the reality, the predicted mechanical properties with interphase demonstrate very good agreement with experimental data.

**Keywords:** Finite element; Interphase; Nanocomposite; Silica nanoparticle; Viscoelastic behavior.

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## INTRODUCTION

Demands on structural materials in the various fields of engineering for better performance under more severe loads and environmental conditions are increasing. Accordingly, extensive efforts have been made to improve the performance of the polymer materials by adding particles as reinforcement with different sizes at micro- and nano-scale. It has been shown that the effective properties of the nanoparticle-reinforced polymer composites which is called nanocomposites, are better than those of the composites reinforced by microparticle [1-6]. It may be due to the larger interfacial area between the nanoparticles and polymer matrix in the nanocomposites as compared with that the interfacial area between the microparticles and polymer matrix in the conventional composites. At a specified particle volume fraction, the nanoparticles have a greater surface area-to-volume ratio

when compared to microparticles because of the higher number density of nanoparticles in the composites. Consequently, the interaction between the nanoparticle and polymer matrix plays a more important role in nanocomposites [7-12]. An equivalent solid continuum interphase is usually considered between the nanoparticle and the polymer matrix which characterizes this interaction [13-15]. Fig. 1a and 1b illustrate a polymer composite coupon reinforced by microparticles and nanoparticles, respectively, at the same particle volume fraction. The interphase that covers the outer surface of the particle has distinct properties from that of the bulk polymer matrix. The interphase in the nanocomposites is considered as the third phase with a constant thickness [14-19].

Silica at nano-scale is among the most applied nanoparticles in the polymer materials. Numerous investigations have revealed that

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the effective properties of silica nanoparticle/polymer nanocomposites are considerably improved even at very low nanoparticle volume fraction [9, 13, 15-16]. There are some studies on the elastic properties of silica nanoparticle/polymer nanocomposites. For example, the elastic modulus of epoxy nanocomposites reinforced by silica nanoparticle with different volume fractions was investigated by Sun *et al.* [9] using two- and three-dimensional FE analysis. It was found that the results of the three-dimensional FE model are closer to experiment than those predicted by two-dimensional FE model. Boutaleb *et al.* [13] employed both analytical and two-dimensional FE micromechanical models to study the effect of interphase on the elastic modulus of silica nanoparticle/polymer nanocomposites. Odegard *et al.* [15] used a continuum-based constitutive method to extract the elastic modulus of silica nanoparticle/polyimide nanocomposites. Also, they studied various nanoparticle/polyimide interfacial treatments in the nanocomposite. Tang *et al.* [20] experimentally evaluated the elastic modulus of silica nanoparticle/epoxy nanocomposites with different weight fractions. Jang *et al.* [21] investigated the stress-strain curves of polyurethane nanocomposites containing silica nanoparticles. The experimental results revealed that both the elastic modulus and strength of polymer nanocomposite increase with adding silica nanoparticles. Lee *et al.* [22] also reported that silica nanoparticles incorporated into the polyurethane can significantly improve the mechanical properties including ultimate strength and yield strength of polymer nanocomposites. Moreover, Dong *et al.* [23] studied the mechanical response of shape memory epoxy matrix reinforced with silica nanoparticles. According to the experimental observations, the mechanical properties of the polymer composite were

improved with the addition of silica nanoparticles into the polymer nanocomposite. Hassanzadeh-Aghdam *et al.* [24] investigated the coefficients of thermal expansion (CTEs) of polyimide nanocomposites containing silica nanoparticles. To this end, a three-dimensional unit cell-based micromechanical model was proposed. The interphase region between silica nanoparticle and polyimide matrix was considered as an equivalent solid continuum [24]. Hassanzadeh-Aghdam and Ansari [25] presented a micromechanics-based analytical model to evaluate the effective thermo-elastic properties of polymer matrix nanocomposites containing silica nanoparticles. Wang and Zhao [26] reported the creep response of polyimide nanocomposites reinforced with silica nanoparticles under tensile tests. They analyzed the influences of nanoparticle weight fraction, stress level and force pattern in the experiments. Their results indicated that the existence of silica nanoparticles can improve the creep resistance of polyimide nanocomposites [26]. Dorigato *et al.* [27] investigated the creep compliance of poly(lactic acid) nanocomposites containing various kinds of silica nanoparticles, different in terms of specific surface area and surface functionalization. Other investigations on the elastic properties of silica nanoparticle/polymer nanocomposites are available in the literature [28-34]. However, due to the viscoelastic response of the polymer under mechanical and environmental conditions [34-35], the polymer nanocomposite behaves as a viscoelastic material. It should be noted that the viscoelastic behavior of silica nanoparticle/polymer nanocomposites is an important part of the long-term mechanical behavior of these materials. To the best of authors' knowledge, the viscoelastic behavior of silica nanoparticle/polymer nanocomposites was not studied in the literature.

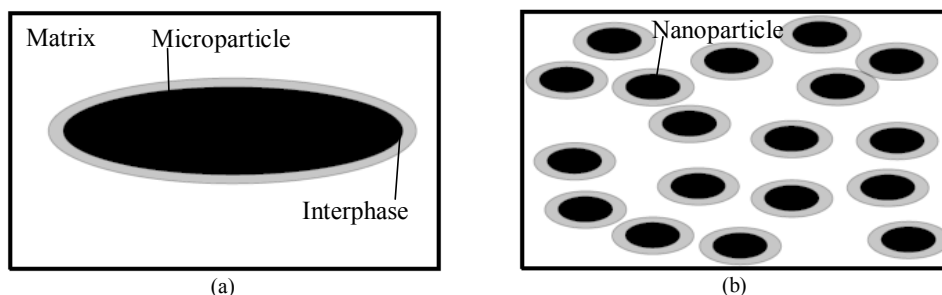


Fig. 1: A polymer composite coupon reinforced by (a) microparticles and (b) nanoparticles.

Many attempts have been made to develop the computational modeling techniques based on molecular dynamics (MD) approaches to predict the nanocomposite properties [15, 36]. However, such techniques are very time-consuming. On the other hand, except for the conventional micro-scale composite materials [37-39], the finite element (FE) method has been employed to investigate the behavior of the composite materials at nanoscale [9, 13, 15, 40-45]. The objective of this paper is to examine the viscoelastic behavior of silica nanoparticle/polymer nanocomposites using a three-dimensional FE micromechanical approach. It should be noted that the two-dimensional FE method could not accurately consider the nanoparticles geometry and distribution into the matrix. The interphase generated due to the interaction between the nanoparticle and polymer is modeled as an equivalent solid continuum. The interphase and silica nanoparticle are assumed to be elastic. The polymer matrix is assumed to behave as a viscoelastic material. The effects of nanoparticle diameter and volume fraction as well as interphase material properties

and thickness on the viscoelastic behavior of the nanocomposites are studied. Moreover, the type of nanoparticles distribution on the viscoelastic behavior of nanocomposites is examined. In the case of elastic analysis, the results of the model are compared with those obtained directly from the experimental tests.

## EXPERIMENTAL

### Computational Method

In this section, the details of the FE simulation are introduced. Silica nanoparticles are modeled as spherical nanoparticles. Fig. 2 illustrates a sample of three-dimensional cubic RVE with randomly dispersed nanoparticles into the polymer matrix. It is assumed that the effective properties of the nanoparticle-reinforced polymer nanocomposite are similar to the properties of the RVE.

The interphase is considered as the covering layer of the outer surface of nanoparticle with a specific thickness, as shown in Fig. 3. The perfect bonding conditions are applied between the constituents of the nanocomposite RVE including nanoparticle, interphase and matrix.

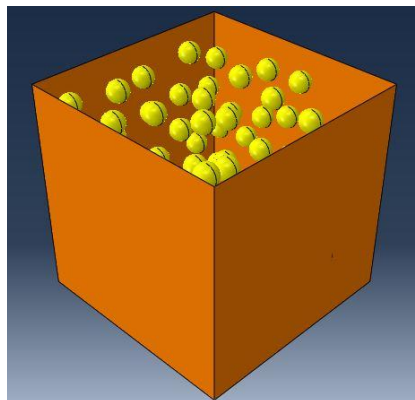


Fig. 2: A sample of three-dimensional cubic RVE containing randomly dispersed nanoparticles.

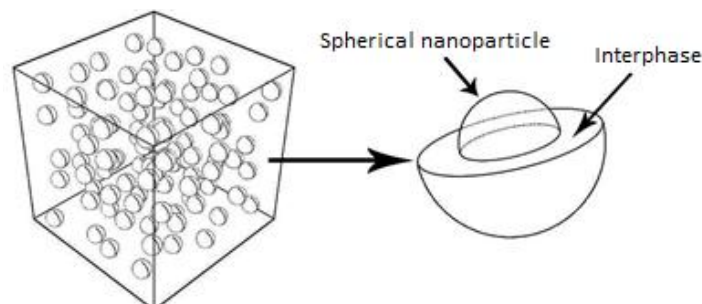


Fig. 3: Spherical nanoparticle and surrounding interphase.

The RVE is meshed using 10-node linear tetrahedron shape elements. For evaluating the mechanical properties of the nanocomposite, three-dimensional stress (C3D10) elements are employed. Fig. 4 illustrates the FE meshing for the RVE containing randomly dispersed nanoparticles. It should be noted that the element size is chosen so that the results become mesh-independent.

To predict the elastic modulus, a uniform certain positive displacement about 1% of the RVE length is applied to a surface of the RVE, while the opposite surface is fixed along the displacement direction and other surfaces condition is free to move but remain parallel to their initial position without rotation. Fig. 5 displays the boundary and load conditions on the RVE for predicting the mechanical properties of nanocomposite. By measuring the fixed boundary reaction force and

calculating the stress value ( $\sigma$ ), the equivalent elastic modulus ( $E_{NC}$ ) of the nanocomposite is derived through the Hook's law, as follow:

$$E_{NC} = \frac{\sigma}{\varepsilon} \quad (1)$$

where  $\varepsilon$  is the RVE strain in the load direction. For the modeling of nanocomposite elastic properties, it is considered that all phases of the RVE to be time-independent. In the other words, for the elastic analysis, three phases of the nanocomposite are considered as the elastic materials. The material properties of silica nanoparticle and polyimide matrix are listed in Table 1 [13, 32]. The values of interphase elastic modulus ( $E_i$ ) and Poisson's ratio ( $\nu_i$ ) are taken as 16.79 GPa and 0.4, respectively [13]. The interphase thickness ( $t_i$ ) and silica nanoparticle diameter ( $d$ ) are considered to be 25 nm and 50 nm, respectively [13].

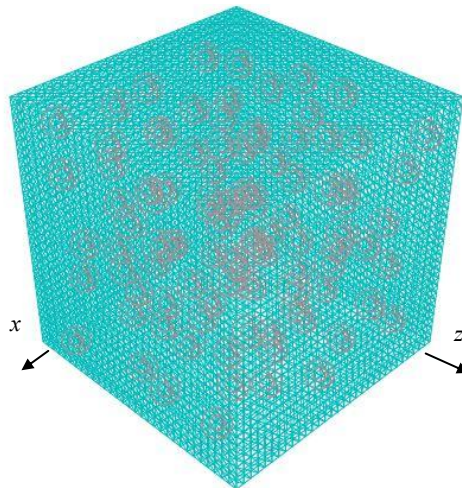


Fig. 4: FE meshing for RVE containing randomly dispersed nanoparticles.

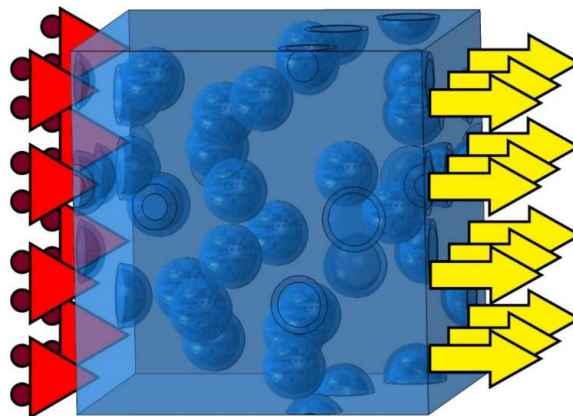


Fig. 5: Boundary and load conditions in obtaining the mechanical properties of nanocomposite.

The viscoelastic behavior of the nanocomposite is dependent on that of the polymer matrix. Assuming that the polymer matrix is a linearly viscoelastic material [46], its creep compliance  $M_0(t)$ , can be characterized by a power-law model, as  $M_0(t) = D_0 + D_1 t^n$  (2)

where  $D_0$  is the initial elastic compliance.  $D_1$  and  $n$  are constant values and experimentally determined. The viscoelastic parameters and Poisson's ratio of polyimide matrix are given in Table 2 [46]. Silica nanoparticle and interphase are treated as the elastic materials. The material properties of silica nanoparticle are listed in Table 1. The values of interphase elastic modulus and Poisson's ratio are taken as 26.66 GPa and 0.367, respectively. To obtain the viscoelastic response of the RVE, a constant stress is applied on one surface along the z-direction for a definite time.

**RESULTS AND DISCUSSION**

Firstly, the elastic moduli of the silica nanoparticle/polyimide nanocomposite estimated

by the present FE method are compared against those from the experiment [32]. The comparison of the elastic moduli of the nanocomposite estimated by the present FE approach with those of the experimental results [32] is depicted in Fig. 6. It should be noted that the elastic modulus of the nanocomposite ( $E$ ) is normalized with respect to the matrix ( $E_m$ ). The elastic modulus of the nanocomposite is plotted versus silica nanoparticle volume fraction ( $V_f$ ). The results of the FE method with interphase are found to be in a good agreement against the experiment [32]. Fig. 6 reveals that the FE model without interphase underestimates the elastic moduli of the nanocomposite as compared with those predicted by FE method with interphase. Moreover, it is found that the elastic modulus of the nanocomposite increases as the silica nanoparticle volume fraction increases. Another comparison is accomplished between the results of the present model and experiment [13] in the case of effective elastic properties of silica nanoparticle-reinforced nylon-6 nanocomposites.

Table 1: Material properties of silica nanoparticle and polyimide [13,32].

Material	$E$ (GPa)	$\nu$
Silica nanoparticle	88.7	0.23
Polyimide	1.679	0.4

Table 2: Viscoelastic properties and Poisson's ratio of polyimide [46].

Material	$\nu$	$D_0$ (GPa <sup>-1</sup> )	$D_1$ (GPa <sup>-1</sup> hour <sup>-1</sup> )	$n$
Polyimide	0.367	0.375	0.051606	0.4103

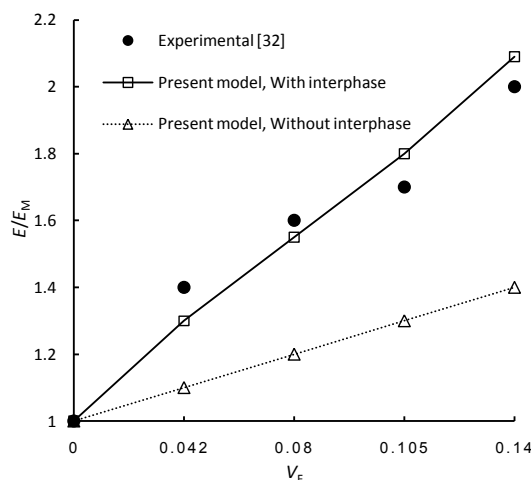


Fig. 6: Elastic modulus of silica nanoparticle/polyimide nanocomposite versus nanoparticle volume fraction.

Fig. 7 shows the variation of elastic modulus with silica nanoparticle volume fraction. The effective elastic moduli of the silica nanoparticle-reinforced nylon-6 nanocomposite estimated by the present model and the experimental data [13] are in excellent agreement to each other.

In the case of viscoelastic behavior, the predictions by the present micromechanics modelling approach are compared with available experimental data of polyimide nanocomposites containing silica nanoparticles [26], as illustrated in Fig. 8. The volume fraction of nanoparticle is equal to 0.5044% [26]. Also, the average of nanoparticle diameter is assumed to be about 50 nm [26]. The values of thickness, and for the interphase are assumed to be equal to = 25 nm, =  $0.1 \times 0.831 \text{ GPa}^{-1}$  and = 0, respectively. Also, the values of and for the interphase are considered as =  $0.5 \times 0.057 \text{ GPa}^{-1} \text{ s}^{-1}$  and = 0.152, respectively. Comparison between the results of the present model in the presence of interphase shows a good agreement with existing experiment [26].

Fig. 9 illustrates the creep strain of the silica nanoparticle/polyimide nanocomposite as a function of time ( $t$ ). The interphase thickness and silica nanoparticle diameter are considered to be 25 nm and 50 nm, respectively. The nanoparticle volume fraction is equal to 3%. Also, the effect of interphase on the creep strain of the nanocomposite is shown in the figure. The results reveal that the interphase between silica nanoparticle and the polymer matrix

significantly affects the viscoelastic behavior of the nanocomposite. It can be observed from Fig. 9 that at a given time  $t$ , the FE method with interphase underestimates the value of creep strain as compared to that predicted by FE method without interphase. For example, when  $t=10$  hour, the creep strains for the nanocomposite without and with interphase are 0.454 and 0.496%, respectively, which corresponds to about 8.5% reduction. The effect of interphase elastic modulus on the creep strain of the nanocomposite is depicted in Fig. 10. It is found that at a given time  $t$ , as the interphase elastic modulus increases, the creep strain significantly decreases. However, when the interphase elastic modulus becomes larger than 36.66 GPa, its effect on the creep strain of the nanocomposite is negligible.

Fig. 11 demonstrates the effect of interphase thickness on the creep strain of the nanocomposite. Three different interphase thicknesses including 15, 20 and 25 nm are used for this analysis. The figure indicates that the variation of  $t_i$  has a significant influence on the creep strain of the nanocomposite. It can be clearly observed that the creep strain of the nanocomposite decreases with increasing the interphase thickness. It may be attributed to the fact that the elastic modulus of interphase is higher than that of polymer matrix. Consequently, increasing the interphase thickness can lead to an increment in the creep resistance of the silica nanoparticle-reinforced polymer nanocomposites.

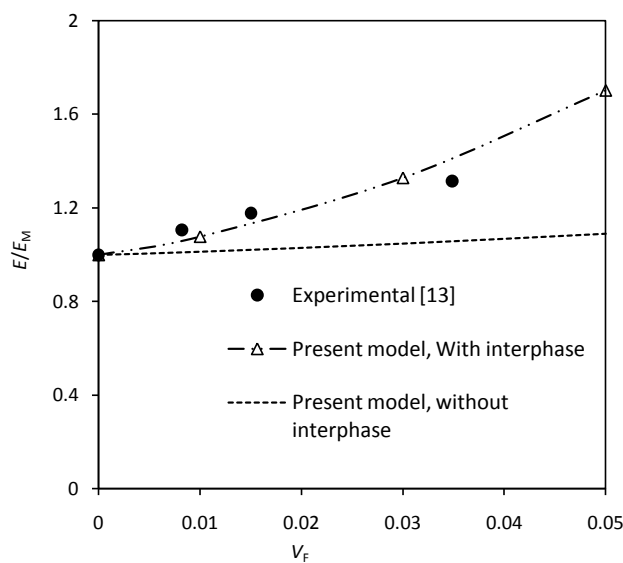


Fig. 7: Elastic modulus of silica nanoparticle/ nylon-6 nanocomposite versus nanoparticle volume fraction.

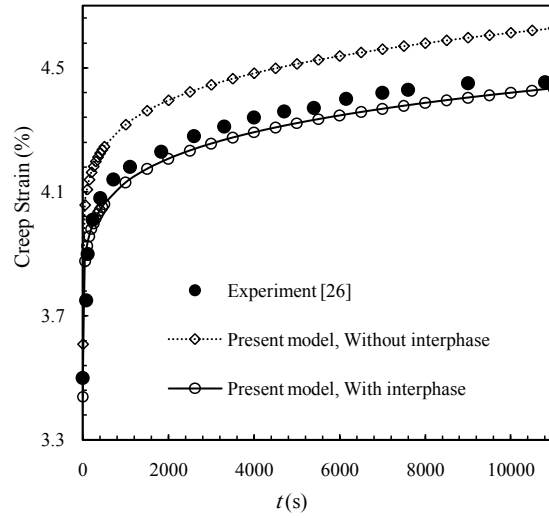


Fig. 8: Viscoelastic response of silica nanoparticle/polyimide nanocomposite.

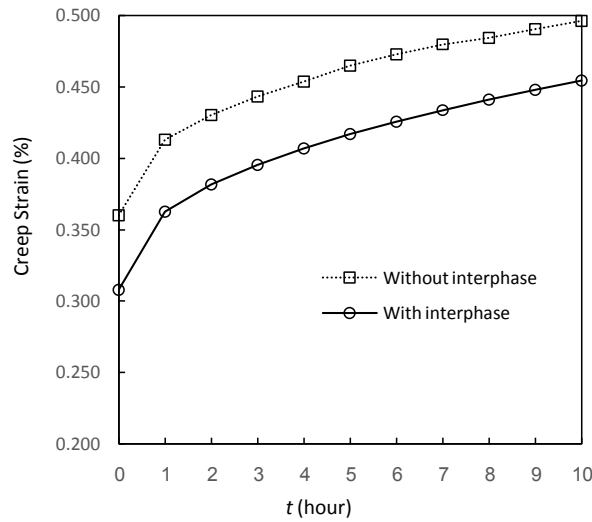


Fig. 9: Creep strain of silica nanoparticle/polyimide nanocomposite.

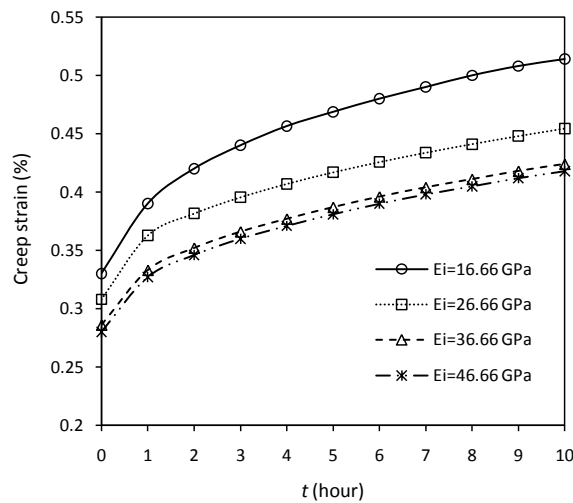


Fig. 10: Effect of interphase elastic modulus on the creep strain of the nanocomposite.

The effect of silica nanoparticle diameter on the creep strain of the nanocomposite is shown in Fig. 12. Four values including 25, 50, 100 and 200 nm are considered in order to investigate the influence of silica nanoparticle diameter on the viscoelastic behavior of silica nanoparticle/polyimide nanocomposite. The results reveal that silica nanoparticle diameter plays an important role in the viscoelastic behavior of the nanocomposites. According to the obtained results from Fig. 12 at a given time  $t$ , the creep strain of the nanocomposite decreases with the decrease of silica nanoparticle diameter. For example, when  $t=10$  hour, the creep strains for  $d=$

50 and 25 nm are 0.454 and 0.381%, respectively, which corresponds to a 16.1% reduction. Also, it is found that a peculiar value exists for the silica nanoparticle diameter about 100 nm beyond that the creep strain of nanocomposite is slightly affected by the increase of nanoparticle diameter. For example, when  $t=10$  hour, the creep strains for  $d= 100$  and 200 nm are 0.504 and 0.509%, respectively, which corresponds to a 0.99% increment. It may be due to the reduced influence of the interphase between silica nanoparticle and polyimide matrix. It is noted that at a specific nanoparticle volume fraction, as the nanoparticle diameter increases, the interphase volume

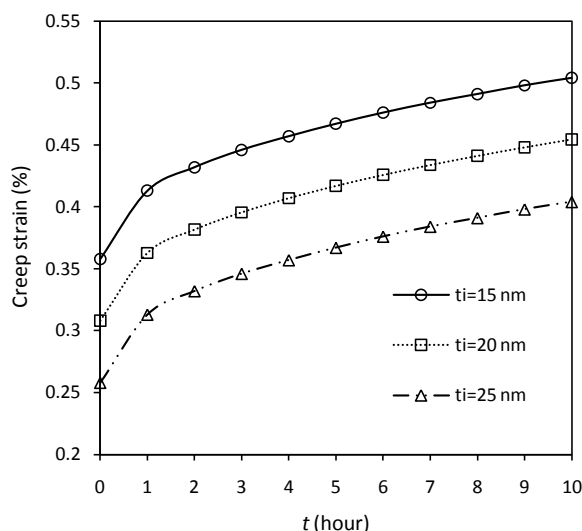


Fig. 11: Effect of interphase thickness on the creep strain of the nanocomposite.

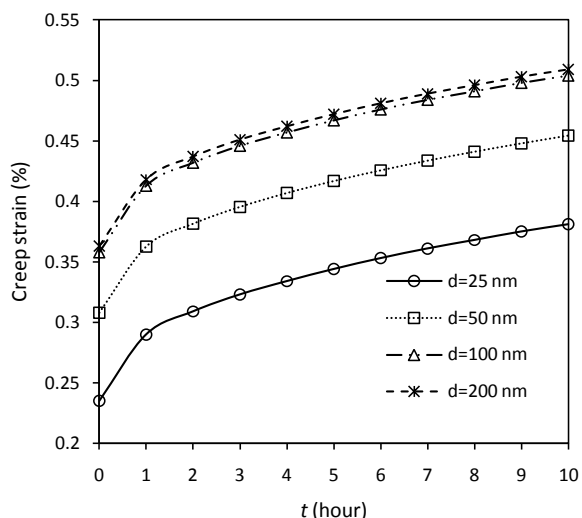


Fig. 12: Effect of silica nanoparticle diameter on the creep strain of the nanocomposite.



fraction decreases, as can be seen in Fig. 1. Thus, by increasing the nanoparticle diameter leading to a reduction in the interphase volume fraction, the effective properties of the nanocomposites become similar to those of the conventional composites reinforced with microscale particles in which the interphase does not affect their overall behavior.

Fig. 13 illustrates the effect of silica nanoparticle volume fraction on the viscoelastic behavior of the nanocomposite. Three different volume fractions

including 1, 3 and 5% are used for this analysis. As expected at a given time  $t$ , with increasing the volume fraction of silica nanoparticle, the creep strain of the nanocomposite decreases. It can be attributed to the fact that the elastic modulus of silica nanoparticle is significantly higher than that of polymer matrix. Consequently, increasing the silica nanoparticle volume fraction leads to an enhancement in the creep resistance of the silica nanoparticle-reinforced polymer nanocomposites. Also, note that by increasing nanoparticle volume

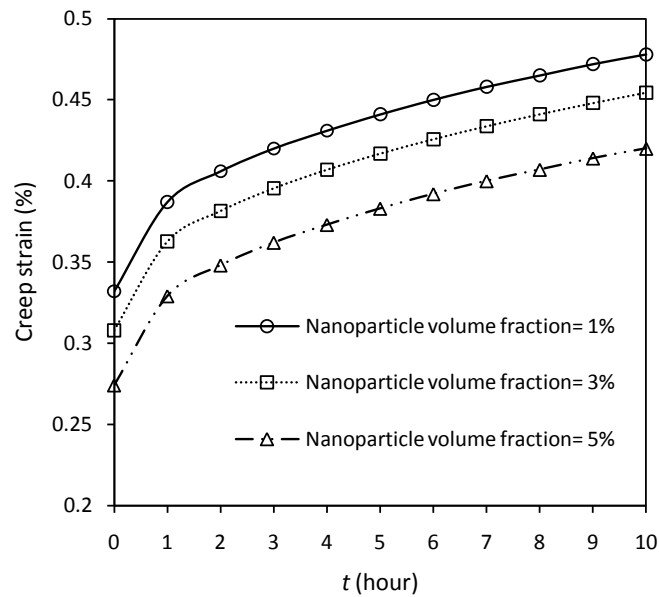


Fig. 13: Effect of Silica nanoparticle volume fraction on the creep strain of the nanocomposite.

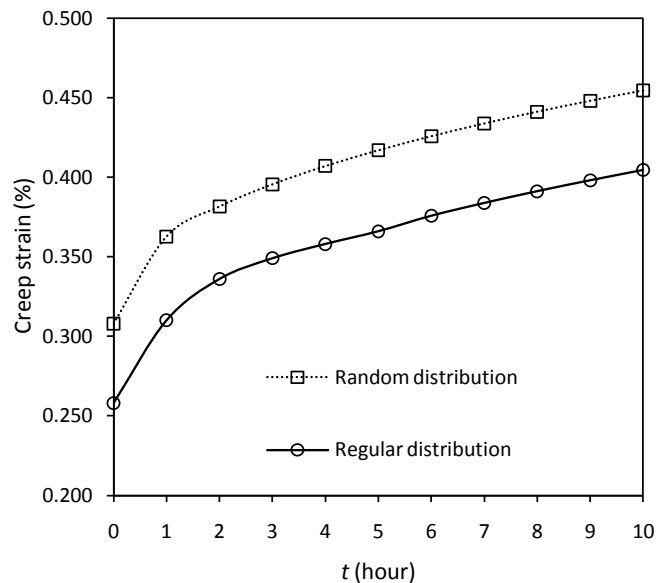


Fig. 14: Effect of type of distribution of silica nanoparticles on the creep strain of the nanocomposite.

fraction, the volume fraction of interphase surrounding the nanoparticles increases which is another reason for decreasing the creep strain of the nanocomposite. The effects of regular and random distributions of silica nanoparticles into the polyimide matrix on the viscoelastic behavior of the nanocomposite are studied in Fig. 14. When silica nanoparticles are regularly dispersed into the polyimide matrix, the creep strain of the nanocomposite considerably decreases. It can be concluded that regular distribution of silica nanoparticles provides more creep resistance. For example, when  $t=10$  hour, the creep strains for the nanocomposite with random and regular distributions are 0.454 and 0.404%, respectively, which corresponds to 11% reduction.

## CONCLUSIONS

In this paper, a three-dimensional FE modeling approach was developed to study the viscoelastic behavior of silica nanoparticle/polyimide nanocomposites. The interphase generated due to the interaction between the nanoparticle and polymer matrix in the nanocomposite was considered as an equivalent solid continuum. The effects of interphase elastic modulus and thickness, nanoparticle geometry and volume fraction as well as the type of distribution into the polymer matrix on the viscoelastic behavior of the nanocomposite were investigated in details. To obtain the viscoelastic behavior of the nanocomposite, it was assumed that the polymer matrix to be as a viscoelastic material. Also, silica nanoparticle and interphase were assumed to be elastic. The obtained results could be summarized as follows:

For silica nanoparticle/polyimide nanocomposites, the interphase presents a significant effect on the viscoelastic behavior. With increasing the interphase thickness or elastic modulus, the creep strain of the nanocomposite decreases. Furthermore, a critical elastic modulus was observed after which further increase of the interphase elastic modulus slightly affects the viscoelastic response.

With decreasing the silica nanoparticle diameter, the creep strain of the nanocomposite decreases. Moreover, a critical diameter was observed after which further increase of the nanoparticle diameter did not change the creep strain of the nanocomposite.

With increasing the nanoparticle volume fraction, the creep strain of the nanocomposite decreases.

For nanocomposite structures, the regular distribution of silica nanoparticles into the matrix presents minimum creep strain.

## CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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