Research Article

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Simple models for tensile modulus of shape memory polymer nanocomposites at ambient temperature

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Abstract: This article analyzes the tensile modulus of shape memory polymer nanocomposites (SMPNs) at ambient temperature. Several conventional models, such as rule of mixtures, Halpin–Tsai and Kerner–Nielsen, cannot practically estimate the modulus due to the absence of some main parameters for nanocomposites. Additionally, some parameters in Kerner–Nielsen and Sato–Furukawa models are useless and ineffective, due to the small concentration and high modulus of nanofillers in SMPNs. Therefore, Kerner–Nielsen and Sato–Furukawa models are simplified and modified to deliver the simple models for calculation of modulus in SMPNs. Various nanocomposite samples are provided to prove the validity of the suggested models. The results demonstrate that the predictions of the suggested models have a good match with the experimental results. The models also demonstrate high simplicity and good accuracy for the calculation of modulus in SMPNs at ambient temperature. Generally, the calculated results disclose that the modified Kerner–Nielsen model is preferable for approximation of modulus in SMPNs.

Keywords: shape memory polymer nanocomposites, tensile modulus, micromechanics models

1 Introduction

Shape memory polymers (SMPs) have a potential to store a temporary deformed shape and recover the original permanent shape [1–5]. The shape memory behavior is typically introduced by a change in temperature, stress, moisture, electric or magnetic fields, light or pH. All SMP structures include both hard and soft phases. The hard segments keep the permanent shape and do not melt or soften at the glass transition temperature (T_g) of the soft segments. Also, they can be cross-linked as rigid local structures or entanglements, which will not separate at the recovery temperature. On the other hand, the soft parts act as a switch to remember the original material shape. Figure 1 demonstrates the shape memory behavior in a SMP by temperature. A temporary shape is obtained by heating at temperature above T_g and applying force. The temporary shape is fixed by cooling to temperature below T_g and finally, the shape recovery occurs by heating to temperature over T_g .

SMPs demonstrate many advantages such as low density, large shape recovery, high recoverable strain, easy processing and low cost [6–9]. The recoverable strain in SMPs can reach 100%, while it reaches only to about 10 and 1% in shape memory metals and ceramics, respectively [10]. SMPs can be employed in many applications such as aerospace, automatics, electronics and biomedical materials [7,11–14]. However, there are various scientific and technological obstacles that prevent the extensive applications of SMPs. For example, SMPs have comparatively low shape recovery stress, which is usually 1–3 MPa compared to 0.5–1 GPa for shape memory metal alloys [15].

In recent years, it has been known that the addition of nanofillers to materials like polymers improves the physical, mechanical, thermal and barrier properties [16–32]. However, a competition between modulus enhancement and recoverable strain ratio is observed in SMPs, due to the significant effects of filler size and stiffness. SMPs

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Figure 1: A schematic for the shape memory behavior of SMPs.

reinforced with traditional micro-filler often show small shape memory effect, due to the high weight fraction of filler (20–30 wt%). However, by incorporation of small amounts of nanofiller, less effect on the macroscopic deformation of the composite is shown, due to much smaller nanofiller dimensions [21,33–37]. Therefore, shape memory polymer nanocomposites (SMPNs) can provide good mechanical and shape memory properties.

Although many researchers have focused on the experimental aspects of SMPNs, the theoretical analysis of SMPN properties was rarely studied in literature. The theoretical investigation can provide information that helps to achieve desired properties [38–41]. From the theoretical point of view, the stored energy of SMPs can be converted to force, but the rubbery modulus described as the elastic modulus above T_g should be increased [42]. The elastic modulus of an SMP increases by addition of nanofillers [43–48], but there is no efficient model to calculate the modulus of SMPNs. SMPNs store the mechanical energies at elevated temperatures and then release them in response to external stimulus. However, since the mechanical properties of SMPNs are different at various temperatures (below or above T_g), we focus on the modulus of samples at ambient temperature in this article. Actually, we assume the original status of shape memory nanocomposites to be below T_g , because the modulus of samples decreases at elevated temperatures after T_g . The modulus of SMPNs at ambient temperature can reveal the capability of samples for converting the stored energy to force.

In this article, the tensile modulus of SMPNs is analyzed at ambient temperature using the simple models for composites. Additionally, some models are proposed for prediction of tensile modulus in SMPNs. The modified models are evaluated by matching the predictions to experimental data of various samples containing dissimilar polymers and nanoparticles. This work provides a simple methodology for estimation of tensile modulus of SMPNs to guide researchers in this field.

2 Micromechanics models

The micromechanics models can estimate the mechanical properties of nanocomposites using the characteristics of each phase [49–51]. Here some micromechanics models are introduced, which can be simplified for SMPNs.

The simplest models for modulus of composites are parallel and series referred to rule of mixtures and inverse rule of mixtures, which assume equivalent strain and stress in both matrix and filler phases, respectively [52]. They can be given by:

$$E = E_m \phi_m + E_f \phi_f, \quad (1)$$

$$\frac{1}{E} = \frac{\phi_m}{E_m} + \frac{\phi_f}{E_f}, \quad (2)$$

where “ E_m ” and “ E_f ” are the Young’s moduli of matrix and nanoparticles, respectively. Also, “ ϕ_m ” and “ ϕ_f ” are the volume fractions of matrix and nanoparticles calculated by the weight percentage of nanofiller in the nanocomposite (m_f) as:

$$\phi_f = \frac{d_c m_f}{d_f}, \quad (3)$$

$$d_c = \frac{d_m d_f}{(1 - m_f) d_f + m_f d_m}, \quad (4)$$

$$\phi_m = 1 - \phi_f, \quad (5)$$

where “ d_c ”, “ d_f ” and “ d_m ” are the densities of nanocomposite, nanofiller and polymer matrix, respectively.

The rule of mixtures often predicts the high modulus for polymer nanocomposites in comparison to experimental results, due to the extremely high modulus of nanofillers. In contrast, the inverse rule of mixtures frequently underestimates the modulus of nanocomposites. Therefore, they have been modified by researchers.

Cox [53] introduced a reduction factor (λ) to the rule of mixtures:

$$E = E_m \phi_m + \lambda E_f \phi_f, \quad (6)$$

$$\lambda = 1 - \frac{\tanh(m)}{m}, \quad (7)$$

$$m = \alpha \sqrt{\frac{2G}{E_f \ln\left(\frac{r}{R}\right)}}, \quad (8)$$

where “ G ” is the shear modulus of the matrix. “ α ” is the aspect ratio of the nanofiller defined as $\alpha = l/t$; “ l ” and “ t ” are the length and thickness of the nanoparticles. “ R ” and “ r ” are the nanofiller radius and the center-to-center distance of the nanofiller, respectively.

Verbeek [54] also suggested a similar equation to Cox’s model (Eqs. (6) and (7)) in which “ m ” is expressed as:

$$m = \alpha \sqrt{\frac{(1-X)^3 G}{E_f} \frac{\phi_f}{1-\phi_f}}, \quad (9)$$

$$X = \frac{\Psi}{\phi_m(1-\Psi) + \Psi}, \quad (10)$$

$$\Psi = \frac{(1-\phi_m)^2 \phi_{\max}}{1-\phi_m \phi_{\max}}, \quad (11)$$

where “ X ” is the modified void content as the modified porosity relative to the polymer phase. “ Ψ ” is the voidage and “ $\phi_{\max}$ ” is the maximum volumetric packing fraction of the nanofiller as $\phi_{\max} = \text{true volume of the filler} / \text{apparent volume occupied by the particles}$.

Halpin and Pagano [55] proposed the most commonly used model as:

$$E = E_m \left(\frac{1 + \eta \chi \phi_f}{1 - \eta \phi_f} \right), \quad (12)$$

$$\eta = (E_f/E_m - 1)/(E_f/E_m + \chi), \quad (13)$$

$$\chi = 2\alpha. \quad (14)$$

Halpin–Tsai model also overpredicted the modulus of nanocomposites, due to the big levels of “ E_f ” and “ α ” in many nanocomposite samples.

Kerner and Nielsen [56] modified the Halpin–Tsai model as:

$$E = E_m \left(\frac{1 + AB\phi_f}{1 - PB\phi_f} \right), \quad (15)$$

$$A = (7 - 5\nu)/(8 - 10\nu), \quad (16)$$

$$B = (E_f/E_m - 1)/(E_f/E_m + A), \quad (17)$$

$$P = 1 + \phi_f^2[(1 - \phi_{\max})/\phi_{\max}], \quad (18)$$

where “ ν ” is the Poisson ratio of the polymer matrix.

Moreover, Sato and Furukawa [57] developed a model using an adhesion parameter (ζ) as:

$$E = E_m \left[\left(1 + \frac{0.5\phi_f^{2/3}}{1 - \phi_f^{1/3}} \right) (1 - \psi\zeta) - \frac{\phi_f^{2/3}\psi\zeta}{(1 - \phi_f^{1/3})\phi_f} \right], \quad (19)$$

$$\psi = \left(\frac{\phi_f}{3} \right) \left(\frac{1 + \phi_f^{1/3} - \phi_f^{2/3}}{1 - \phi_f^{1/3} + \phi_f^{2/3}} \right). \quad (20)$$

where a “ ζ ” value of 0 indicates good interfacial adhesion, while $\zeta = 1$ shows poor adhesion at the interface.

3 Results and discussion

In this section, the mentioned models are evaluated using the experimental results of many SMPNs containing different matrices and nanofillers from literature. Then, an attempt is made to simplify the models for modulus of SMPNs at ambient temperature. Finally, some evidence is provided to confirm the validity of the proposed models for modulus of SMPNs. The SMPN samples from literature and their phase characteristics are shown in Table 1.

Various types of SMPNs consisting of different matrices and nanofillers were chosen from valid literature. It is considered that the reported samples can provide a comprehensive study of the modulus. Therefore, no experimental work to prepare and characterize the SMPNs was performed. The predictions of rule of mixtures and inverse rule of mixtures models can be easily calculated using the densities, moduli and weight fractions of the components. Figure 2 compares the experimental results and the calculations for polystyrene (PS)/multi-walled carbon nanotube (MWCNT)/short carbon fiber (SCF) (No. 1) and polyurethane (PU)/CNT (No. 3) samples. As observed, the rule of mixtures and inverse rule of mixtures obviously over- and underestimate

Table 1: The characteristics of the studied SMPNs

No.	Samples	d_m (g/cm ³)	d_f (g/cm ³)	E_f (GPa)	Ref.
1	PS ¹ /MWCNT ² /SCF ³	0.92	1.9, 1.8	600, 240	[58]
2	PS/nanocarbon powder	0.92	1.85	450	[59]
3	PU ⁴ /CNT	0.94	1.90	240	[60]
4	PU/nanocellulose	1.00	1.53	150	[61]
5	PU/silicon carbide	0.94	3.22	450	[62]
6	PE ⁵ /nanoclay	0.92	1.77	178	[63]
7	PDMS ⁶ /MWCNT	0.97	2.10	600	[64]
8	PU/MWCNT	1.00	2.10	600	[65]

¹polystyrene; ²multi-walled carbon nanotubes; ³short carbon fiber;

⁴polyurethane; ⁵polyethylene; ⁶poly(dimethylsiloxane).

Figure 2: The calculations of modulus by rule of mixtures and inverse rule of mixtures models for (a) PS/MWCNT/SCF (No. 1) and (b) PU/CNT (No. 3) samples.

the modulus of samples, respectively. As a result, they are not appropriate for prediction of modulus of SMPNs at ambient temperature.

Cox and Verbeek models Eqs. (6)–(11) need precise characterization of several parameters such as “ α ”, “ R ”, “ r ” and “ $\phi_{\max}$ ” for modulus prediction. However, an accurate and practical method for determination of these parameters has not been presented in literature. Some authors determined them by fitting the models to experimental results [66,67], which is not acceptable. Others used the “ α ”, “ R ” and “ r ” values from the characteristics of pristine nanofillers [68,69], but the size of nanofillers change greatly due to high stress during the fabrication process of nanocomposites. Also, it was found that “ α ” and “ $\phi_{\max}$ ” may not have any important effect on the predicted modulus in nanocomposites, due to extremely high range of “ E_f ” [56]. Therefore, the modeling of modulus by Cox and Verbeek models may involve some errors in the results.

Figure 3 illustrates the measured modulus at ambient temperature and the calculations by modified rule of mixtures model Eq. (6) for four samples. Surprisingly, the predicted modulus shows good agreement with the experimental results by choosing an appropriate value of “ λ ”. Accordingly, “ λ ” can be determined by experimental measurement of modulus for one prepared SMPN sample. Obviously, it provides great simplicity and accuracy for modeling of modulus of SMPNs. The most suitable values of “ λ ” for all samples calculated by a fitting process are shown in Table 2. Many “ λ ” values are much less than 1, which indicate the high deviation of the modified model from the rule of mixtures Eq. (1).

According to Table 2, the highest “ λ ” value is found for the PS/MWCNT/SCF sample in which the modulus increased from 1.24 to 3.64 GPa by addition of only 2.5 wt% of MWCNT and 1.5 wt% of SCF [58]. The CNT particles could be well bonded with the polymer matrix due to the same typical size of CNT and polymer segments, which played a positive role in the shape memory behavior [58]. Moreover, the tensile shape recovery forces and bending shape recovery moments of PS/MWCNT/SCF sample increased with the addition of two fillers, which not only improved the mechanical properties, but also increased the actuating capability.

In addition, the Kerner–Nielsen model Eqs. (15)–(18) can be more simplified for SMPNs. Since the Poisson's ratio of all polymers (ν) changes from 0.33 to 0.5, the “ A ” parameter Eq. (16) is approximately constant for all polymer matrices. Moreover, the E_f/E_m term has a high level in SMPNs, and thus the “ B ” parameter Eq. (17) is roughly constant at different values of “ ν .” Also, the “ P ” Eq. (18) values do not vary significantly at different ranges of “ ϕ_f ” and “ $\phi_{\max}$ ”, which is attributed to the low values of “ ϕ_f^2 ” in the SMPNs. Table 3 shows the variation in “ P ” for poly(dimethylsiloxane) (PDMS)/MWCNT sample. As shown, “ P ” changes insignificantly at different values of “ ϕ_f ” and “ $\phi_{\max}$.”

Kerner–Nielsen model can be simply presented for SMPNs as:

$$E = E_m \left(\frac{1 + m\phi_f}{1 - \phi_f} \right), \quad (21)$$

where “ m ” is a constant parameter that can be determined by testing the modulus for only one sample. The

Figure 3: The obtained results by modified rule of mixtures and Kerner–Nielsen models for (a) PS/MWCNT/SCF (No. 1), (b) PS/nanocarbon powder (No. 2), (c) PU/silicon carbide (No. 5) and (d) PDMS/MWCNT (No. 7) samples.

calculation of modulus by the modified Kerner–Nielsen model is shown in Figure 3 and the best fitted values of “ m ” for all samples are shown in Table 2. Good agreement is shown between the theoretical predictions and the experimental results. As a result, the modified Kerner–

Nielsen model calculates the modulus by a simple and exact method, which leads to elimination of difficult and inaccurate characterizations of different parameters for prediction of modulus. Figure 3 also demonstrates that the modulus obtained by the modified Kerner–Nielsen

Table 2: The calculated parameters for reported SMPNs

No.	Samples	λ (Eq. 6)	m (Eq. 21)	ζ (Eq. 19)	k (Eq. 22)	Ref.
1	PS ¹ /MWCNT ² /SCF ³	0.270	93.0	−55	30.5	[58]
2	PS/nanocarbon powder	0.018	3.10	−0.75	1.01	[59]
3	PU ⁴ /CNT	0.090	89.0	−29	17.5	[60]
4	PU/nanocellulose	0.023	82.0	−22	12.0	[61]
5	PU/silicon carbide	6×10^{-4}	58.8	−35	23.5	[62]
6	PE ⁵ /nanoclay	0.008	34.0	−13	8.50	[63]
7	PDMS ⁶ /MWCNT	4×10^{-5}	33.0	−10	6.50	[64]
8	PU/MWCNT	0.006	13.8	−4.2	3.15	[65]

¹polystyrene; ²multi-walled carbon nanotubes; ³short carbon fiber; ⁴polyurethane; ⁵polyethylene; ⁶poly(dimethylsiloxane).

Table 3: The variation in “ P ” at different levels of “ ϕ_f ” and “ $\phi_{\max}$ ” for PDMS/MWCNT sample

No.	m_f (wt%)	ϕ_f (vol%)	$\phi_{\max}$ (vol%)	P Eq. (18)
1	1	0.0046	0.1	1.0002
2	2	0.0093	0.1	1.0008
3	3	0.0140	0.1	1.0018
4	4	0.0188	0.1	1.0032
5	7	0.0334	0.1	1.0101
6	1	0.0046	0.9	1.0000
7	2	0.0093	0.9	1.0000
8	3	0.0140	0.9	1.0000
9	4	0.0188	0.9	1.0000
10	7	0.0334	0.9	1.0001

model is much closer to the experimental data than those by the modified rule of mixtures. This means that the modified Kerner–Nielsen model is preferable for calculation of modulus of SMPNs.

Figure 4 shows good coherence between the calculated modulus by the Sato–Furukawa model Eqs. (19)

and (20) and the experimental data for PU/nanocellulose (No. 4), PDMS/MWCNT (No. 7) and PU/MWCNT (No. 8) SMPNs. The best fitted values of adhesion parameter (ζ) are shown in Table 2. Negative “ ζ ” values are obtained for all SMPNs, while “ ζ ” should vary from 0 to 1 in micro-composites. The extremely small ranges of “ ϕ_f ” in SMPNs result in the negative “ ζ ” values. However, the negative values of “ ζ ” express the strong interfacial adhesion between polymer and nanofiller phases in the SMPNs [70]. Although the predictions of the Sato–Furukawa model are well fitted to the experimental results, this model can be much simplified for estimation of modulus in SMPNs. By assuming $\zeta = 0$ as strong adhesion, this model is modified to:

$$E = E_m \left(1 + \frac{k\phi_f^{2/3}}{1 - \phi_f^{1/3}} \right), \quad (22)$$

where “ k ” is a constant parameter. The predictions of the simplified Sato–Furukawa model are shown in Figure 4 and the best fitted values of “ k ” are reported in Table 2.

Figure 4: The calculations of Sato–Furukawa and modified Sato–Furukawa models for (a) PU/nanocellulose (No. 4), (b) PDMS/MWCNT (No. 7) and (c) PU/MWCNT (No. 8) samples.

Clearly, the modified model can present good agreement with the experimental results. Furthermore, the most negative “ ζ ” and the highest “ k ” are found for the PS/MWCNT/SCF sample, as expected according to the earlier results.

4 Conclusion

The tensile modulus for SMPNs was investigated by simple models. The rule of mixtures and inverse rule of mixtures overestimated and underpredicted the modulus for SMPNs, respectively. However, the modified model Eq. (6) could calculate the modulus of SMPNs at ambient temperature. In addition, it was shown that “ A ”, “ B ” and “ P ” in Kerner–Nielsen model Eqs. (16)–(18) changed negligibly for different samples, owing to the small concentration and high modulus of the nanofiller. Therefore, some modifications were applied to the models to propose a simple equation for calculation of the modulus. Additionally, Sato–Furukawa model was simplified in the present study. Several samples from literature were provided to show the validity of the models. The predictions of the suggested models exhibited a good match with the experimentally measured modulus. “ m ” in modified Kerner–Nielsen model changes from 3.1 to 93 for the samples, while “ k ” in the modified Sato–Furukawa model varies from 1.01 to 30.5. The obtained results showed that the modified Kerner–Nielsen is preferable for calculation of modulus in SMPNs. It was found that the concentration of nanoparticles has the main effect on the modulus of samples. Also, the constant factors such as “ m ” and “ k ” have significant roles in the nanocomposite modulus.

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References

- [1] Li M-Q, Wu J-M, Song F, Li D-D, Wang X-L, Chen L, et al. Flexible and electro-induced shape memory Poly (Lactic Acid)-based material constructed by inserting a main-chain liquid

- crystalline and selective localization of carbon nanotubes. *Compos Sci Technol.* 2019;173:1–6.
- [2] Palza H, Zapata P, Sagredo C. Shape memory composites based on a thermoplastic elastomer polyethylene with carbon nanostructures stimulated by heat and solar radiation having piezoresistive behavior. *Polym Int.* 2018;67(8):1046–53.
- [3] Markad K, Lal A. Experimental investigation of shape memory polymer hybrid nanocomposites modified by carbon fiber reinforced multi-walled carbon nanotube (MWCNT). *Mater Res Exp.* 2021;8(10):105015.
- [4] Sliozberg YR, Kröger M, Henry TC, Datta S, Lawrence BD, Hall AJ, et al. Computational design of shape memory polymer nanocomposites. *Polymer.* 2021;217:123476.
- [5] Gopinath S, Adarsh NN, Nair PR, Mathew S. Shape-memory polymer nanocomposites of poly (ϵ -caprolactone) with the polystyrene-block-polybutadiene-block-polystyrene-tri-block copolymer encapsulated with metal oxides. *ACS Omega.* 2021;6(9):6261–73.
- [6] Lei M, Chen Z, Lu H, Yu K. Recent progress in shape memory polymer composites: methods, properties, applications and prospects. *Nanotechnol Rev.* 2019;8(1):327–51.
- [7] Wu G, Gu Y, Hou X, Li R, Ke H, Xiao X. Hybrid nanocomposites of cellulose/carbon-nanotubes/polyurethane with rapidly water sensitive shape memory effect and strain sensing performance. *Polymers.* 2019;11(10):1586.
- [8] Hassanzadeh-Aghdam MK, Ansari R, Mahmoodi MJ. Thermo-mechanical properties of shape memory polymer nanocomposites reinforced by carbon nanotubes. *Mech Mater.* 2019;129:80–98.
- [9] Hassanzadeh-Aghdam M, Ansari R. Thermal conductivity of shape memory polymer nanocomposites containing carbon nanotubes: a micromechanical approach. *Compos Part B: Eng.* 2019;162:167–77.
- [10] Cho JW, Lee SH. Influence of silica on shape memory effect and mechanical properties of polyurethane–silica hybrids. *Eur Polym J.* 2004;40(7):1343–8.
- [11] Quadrini F, Bellisario D, Santo L, Gaudio CD, Bianco A. Shape memory foams of microbial polyester for biomedical applications. *Polym Technol Eng.* 2013;52(6):599–602.
- [12] Herath M, Epaarachchi J, Islam M, Fang L, Leng J. Light activated shape memory polymers and composites: a review. *Eur Polym J.* 2020;136:109912.
- [13] Li F, Leng J, Liu Y, Remillat C, Scarpa F. Temperature dependence of elastic constants in unidirectional carbon fiber reinforced shape memory polymer composites. *Mech Mater.* 2020;148:103518.
- [14] Antony GJM, Raja S, Aruna S, Jarali CS. Effect of the addition of diurethane dimethacrylate on the chemical and mechanical properties of tBA-PEGDMA acrylate based shape memory polymer network. *J Mech Behav Biomed Mater.* 2020;110:103951.
- [15] Yan B, Gu S, Zhang Y. Polylactide-based thermoplastic shape memory polymer nanocomposites. *Eur Polym J.* 2012;49:366–78.
- [16] Zare Y, Rhee KY. Tensile modulus prediction of carbon nanotubes-reinforced nanocomposites by a combined model for dispersion and networking of nanoparticles. *J Mater Res Technol.* 2019;9:22–32.
- [17] Farzaneh A, Rostami A, Nazockdast H. Thermoplastic polyurethane/multiwalled carbon nanotubes nanocomposites: effect

- of nanoparticle content, shear, and thermal processing. *Polym Compos.* 2021;42:4804–13.
- [18] Farzaneh A, Rostami A, Nazockdast H. Mono-filler and bi-filler composites based on thermoplastic polyurethane, carbon fibers and carbon nanotubes with improved physicomechanical and engineering properties. *Polym Int.* 2022;71:232–42.
 - [19] Naghib SM, Behzad F, Rahmanian M, Zare Y, Rhee KY. A highly sensitive biosensor based on methacrylated graphene oxide-grafted polyaniline for ascorbic acid determination. *Nanotechnol Rev.* 2020;9(1):760–7.
 - [20] Bayat H, Fasihi M, Zare Y, Rhee KY. An experimental study on one-step and two-step foaming of natural rubber/silica nanocomposites. *Nanotechnol Rev.* 2020;9(1):427–35.
 - [21] Behdinan K, Moradi-Dastjerdi R, Safaei B, Qin Z, Chu F, Hui D. Graphene and CNT impact on heat transfer response of nanocomposite cylinders. *Nanotechnol Rev.* 2020;9(1):41–52.
 - [22] Cheng C, Song W, Zhao Q, Zhang H. Halloysite nanotubes in polymer science: Purification, characterization, modification and applications. *Nanotechnol Rev.* 2020;9(1):323–44.
 - [23] Gooneh-Farahani S, Naghib SM, Naimi-Jamal MR, Seyfoori A. A pH-sensitive nanocarrier based on BSA-stabilized graphene-chitosan nanocomposite for sustained and prolonged release of anticancer agents. *Sci Rep.* 2021;11(1):1–14.
 - [24] Haghgoo M, Ansari R, Hassanzadeh-Aghdam M. Synergic effect of graphene nanoplatelets and carbon nanotubes on the electrical resistivity and percolation threshold of polymer hybrid nanocomposites. *Eur Phys J Plus.* 2021;136(7):1–20.
 - [25] Jiang Q, Tallury SS, Qiu Y, Pasquinelli MA. Interfacial characteristics of a carbon nanotube-polyimide nanocomposite by molecular dynamics simulation. *Nanotechnol Rev.* 2020;9(1):136–45.
 - [26] Zhang Y-F, Du F-P, Chen L, Yeung K-W, Dong Y, Law W-C, et al. Supramolecular ionic polymer/carbon nanotube composite hydrogels with enhanced electromechanical performance. *Nanotechnol Rev.* 2020;9(1):478–88.
 - [27] Huang Z, Tsui GC-P, Deng Y, Tang C-Y. Two-photon polymerization nanolithography technology for fabrication of stimulus-responsive micro/nano-structures for biomedical applications. *Nanotechnol Rev.* 2020;9(1):1118–36.
 - [28] Huang Y, Zeng J. Recent development and applications of nanomaterials for cancer immunotherapy. *Nanotechnol Rev.* 2020;9(1):382–99.
 - [29] Rostami A, Moosavi MI. High-performance thermoplastic polyurethane nanocomposites induced by hybrid application of functionalized graphene and carbon nanotubes. *J Appl Polym Sci.* 2020;137(14):48520.
 - [30] Tajdari A, Babaei A, Goudarzi A, Partovi R, Rostami A. Hybridization as an efficient strategy for enhancing the performance of polymer nanocomposites. *Polym Compos.* 2021;42(12):6801–15.
 - [31] Moradi S, Yeganeh JK. Highly toughened poly (lactic acid)(PLA) prepared through melt blending with ethylene-co-vinyl acetate (EVA) copolymer and simultaneous addition of hydrophilic silica nanoparticles and block copolymer compatibilizer. *Polym Test.* 2020;91:106735.
 - [32] Sadeghi A, Moeini R, Yeganeh JK. Highly conductive PP/PET polymer blends with high electromagnetic interference shielding performances in the presence of thermally reduced graphene nanosheets prepared through melt compounding. *Polym Compos.* 2019;40(S2):E1461–9.
 - [33] Zare Y. “a” interfacial parameter in Nicolais–Narkis model for yield strength of polymer particulate nanocomposites as a function of material and interphase properties. *J Colloid Interface Sci.* 2016;470:245–9.
 - [34] Zare Y, Rhee KY. The effective conductivity of polymer carbon nanotubes (CNT) nanocomposites. *J Phys Chem Solids.* 2019;131:15–21.
 - [35] Zare Y, Rhee KY. Expansion of Takayanagi model by interphase characteristics and filler size to approximate the tensile modulus of halloysite-nanotube-filled system. *J Mater Res Technol.* 2022;16:1628–36.
 - [36] Wang W, Zhang B, Zhao L, Li M, Han Y, Wang L, et al. Fabrication and properties of PLA/nano-HA composite scaffolds with balanced mechanical properties and biological functions for bone tissue engineering application. *Nanotechnol Rev.* 2021;10(1):1359–73.
 - [37] Hu C, Wu L, Zhou C, Sun H, Gao P, Xu X, et al. Berberine/Ag nanoparticle embedded biomimetic calcium phosphate scaffolds for enhancing antibacterial function. *Nanotechnol Rev.* 2020;9(1):568–79.
 - [38] Zare Y, Rhee KY. A simulation work for the influences of aggregation/agglomeration of clay layers on the tensile properties of nanocomposites. *JOM.* 2019;3989–95.
 - [39] Zare Y, Rhee KY. Effects of interphase regions and filler networks on the viscosity of PLA/PEO/carbon nanotubes biosensor. *Polym Compos.* 2019;40:4135–41.
 - [40] Zare Y, Rhee KY. Expression of normal stress difference and relaxation modulus for ternary nanocomposites containing biodegradable polymers and carbon nanotubes by storage and loss modulus data. *Compos Part B: Eng.* 2019;158:162–8.
 - [41] Zare Y, Rhee KY. The strengthening efficacy of filler/interphase network in polymer halloysite nanotubes system after mechanical percolation. *J Mater Res Technol.* 2021;15:5343–52.
 - [42] Ohki T, Ni QQ, Ohsako N, Iwamoto M. Mechanical and shape memory behavior of composites with shape memory polymer. *Compos Part A: Appl Sci Manuf.* 2004;35(9):1065–73.
 - [43] Yang Y, He Q, Rao YN, Dai HL. Estimation of dynamic thermo viscoelastic moduli of short fiber-reinforced polymers based on a micromechanical model considering interphases/interfacial conditions. *Polym Compos.* 2020;41(2):788–803.
 - [44] Chen T, Chen S, Wang J, Lin T, Wu W, Yang W, et al. Preparation and characterization of poly (vinyl alcohol)/halloysite nanotubes composite sponges with improved mechanical properties. *Polym Compos.* 2021;42:3158–68.
 - [45] Zare Y, Rhee KY. Two-stage simulation of tensile modulus of carbon nanotube (CNT)-reinforced nanocomposites after percolation onset using the ouali approach. *JOM.* 2020;72:3943–51.
 - [46] Zare Y. A model for tensile strength of polymer/clay nanocomposites assuming complete and incomplete interfacial adhesion between the polymer matrix and nanoparticles by the average normal stress in clay platelets. *RSC Adv.* 2016;6(63):57969–76.
 - [47] Zare Y. Assumption of interphase properties in classical Christensen–Lo model for Young’s modulus of polymer nanocomposites reinforced with spherical nanoparticles. *RSC Adv.* 2015;5(116):95532–8.
 - [48] Zare Y, Rhee KY. Simulation of Young’s modulus for clay-reinforced nanocomposites assuming mechanical percolation, clay-interphase networks and interfacial linkage. *J Mater Res Technol.* 2020;9(6):12473–83.

- [49] Zare Y, Rhee K. Evaluation and development of expanded equations based on takayanagi model for tensile modulus of polymer nanocomposites assuming the formation of percolating networks. *Phys Mesomech.* 2018;21(4):351–7.
- [50] Zare Y, Rhee KY. Accounting the reinforcing efficiency and percolating role of interphase regions in the tensile modulus of polymer/CNT nanocomposites. *Eur Polym J.* 2017;87:389–97.
- [51] Zare Y, Rhee KY. Analysis of critical interfacial shear strength between polymer matrix and carbon nanotubes and its impact on the tensile strength of nanocomposites. *J Mater Res Technol.* 2020
- [52] Haghighat M, Zadhoush A, Khorasani SN. Physicomechanical properties of α -cellulose-filled styrene–butadiene rubber composites. *J Appl Polym Sci.* 2005;96(6):2203–11.
- [53] Cox H. The elasticity and strength of paper and other fibrous materials. *Br J Appl Phys.* 1952;3(3):72.
- [54] Verbeek C. The influence of interfacial adhesion, particle size and size distribution on the predicted mechanical properties of particulate thermoplastic composites. *Mater Lett.* 2003;57(13–14):1919–24.
- [55] Halpin J, Pagano N. The laminate approximation for randomly oriented fibrous composites. *J Composite Mater.* 1969;3(4):720–4.
- [56] Zare Y, Garmabi H. Analysis of tensile modulus of PP/nano-clay/ CaCO_3 ternary nanocomposite using composite theories. *J Appl Polym Sci.* 2012;123(4):2309–19.
- [57] Sato Y, Furukawa J. A molecular theory of filler reinforcement based upon the conception of internal deformation (A rough approximation of the internal deformation). *Rubber Chem Technol.* 1963;36:1081.
- [58] Yu K, Liu Y, Leng J. Conductive shape memory polymer composite incorporated with hybrid fillers: electrical, mechanical, and shape memory properties. *J Intell Mater Syst Struct.* 2011;22(4):369–79.
- [59] Leng J, Lan X, Liu Y, Du S. Electroactive thermoset shape memory polymer nanocomposite filled with nanocarbon powders. *Smart Mater Struct.* 2009;18(7):074003.
- [60] Ni QQ, Zhang CS, Fu Y, Dai G, Kimura T. Shape memory effect and mechanical properties of carbon nanotube/shape memory polymer nanocomposites. *Composite Struct.* 2007;81(2):176–84.
- [61] Auad ML, Contos VS, Nutt S, Aranguren MI, Marcovich NE. Characterization of nanocellulose-reinforced shape memory polyurethanes. *Polym Int.* 2008;57(4):651–9.
- [62] Guo Z, Kim TY, Lei K, Pereira T, Sugar JG, Hahn HT. Strengthening and thermal stabilization of polyurethane nanocomposites with silicon carbide nanoparticles by a surface-initiated-polymerization approach. *Compos Sci Technol.* 2008;68(1):164–70.
- [63] Rezanejad S, Kokabi M. Shape memory and mechanical properties of cross-linked polyethylene/clay nanocomposites. *Eur Polym J.* 2007;43(7):2856–65.
- [64] Ahir S, Squires A, Tajbakhsh A, Terentjev E. Infrared actuation in aligned polymer-nanotube composites. *Phys Rev B.* 2006;73(8):085420.
- [65] Mahapatra SS, Yadav SK, Yoo HJ, Ramasamy MS, Cho JW. Tailored and strong electro-responsive shape memory actuation in carbon nanotube-reinforced hyperbranched polyurethane composites. *Sens Actuators B: Chem.* 2014;193:384–90.
- [66] Ogasawara T, Ishida Y, Ishikawa T, Yokota R. Characterization of multi-walled carbon nanotube/phenylethynyl terminated polyimide composites. *Compos Part A: Appl Sci Manuf.* 2004;35(1):67–74.
- [67] Cadek M, Coleman J, Barron V, Hedicke K, Blau W. Morphological and mechanical properties of carbon-nanotube-reinforced semicrystalline and amorphous polymer composites. *Appl Phys Lett.* 2002;81(27):5123–5.
- [68] Safadi B, Andrews R, Grulke E. Multi-walled carbon nanotube polymer composites: synthesis and characterization of thin films. *J Appl Polym Sci.* 2002;84(14):2660–9.
- [69] Seyhan AT, Tanoğlu M, Schulte K. Tensile mechanical behavior and fracture toughness of MWCNT and DWCNT modified vinyl-ester/polyester hybrid nanocomposites produced by 3-roll milling. *Mater Sci Eng: A.* 2009;523(1):85–92.
- [70] Zare Y. Determination of polymer-nanoparticles interfacial adhesion and its role in shape memory behavior of shape memory polymer nanocomposites. *Int J Adhes Adhesives.* 2014;54:67–71.