CrossMark

Effects of Size and Aggregation/Agglomeration of Nanoparticles on the Interfacial/Interphase Properties and Tensile Strength of Polymer Nanocomposites

Muhammad Aqeel Ashraf[1,2], Wanxi Peng[1*], Yasser Zare[3] and Kyong Yop Rhee[4*]

Abstract

In this study, several simple equations are suggested to investigate the effects of size and density on the number, surface area, stiffening efficiency, and specific surface area of nanoparticles in polymer nanocomposites. In addition, the roles of nanoparticle size and interphase thickness in the interfacial/interphase properties and tensile strength of nanocomposites are explained by various equations. The aggregates/agglomerates of nanoparticles are also assumed as large particles in nanocomposites, and their influences on the nanoparticle characteristics, interface/interphase properties, and tensile strength are discussed. The small size advantageously affects the number, surface area, stiffening efficiency, and specific surface area of nanoparticles. Only 2 g of isolated and well-dispersed nanoparticles with radius of 10 nm ($R = 10$ nm) and density of 2 g/cm^3 produce the significant interfacial area of 250 m^2 with polymer matrix. Moreover, only a thick interphase cannot produce high interfacial/interphase parameters and significant mechanical properties in nanocomposites because the filler size and aggregates/agglomerates also control these terms. It is found that a thick interphase ($t = 25$ nm) surrounding the big nanoparticles ($R = 50$ nm) only improves the B interphase parameter to about 4, while $B = 13$ is obtained by the smallest nanoparticles and the thickest interphase.

Keywords: Polymer nanocomposites, Particle size, Aggregation/agglomeration, Interfacial/interphase properties

Background

The nanocomposites exhibit substantial properties by only small content of nanofiller [1–5]. The important properties of polymer nanocomposites cause a wide range of applications in various technologies such as advanced materials and goods, medicines, energy devices, and sensors [6]. The studies on different types of polymer nanocomposites aim to achieve high-performance products by an easy fabrication process and low cost.

The considerable properties of polymer nanocomposites are attributed to good interfacial properties between polymer matrix and nanoparticles such as interfacial area and interaction/adhesion at interface [7–13]. The high levels of interfacial properties lead to formation of another phase as interphase around the nanoparticles which is different from both polymer matrix and nanoparticles shows the advantage of nanocomposites compared to conventional micro-composites [14–18]. Many theoretical investigations on interfacial/interphase properties have given a large amount of information to attain the desirable properties. However, the high surface area of nanoparticles and the strong attractive interaction between particles result in the aggregation/agglomeration [19, 20]. The strong and dense collectives of nanoparticles denote the aggregation, but the loosely joint particles show the agglomeration which may be broken by mechanical stress [21].

The aggregation/agglomeration of nanoparticles reduces the potential enhancement of mechanical properties in nanocomposites, due to the restriction of interfacial area [22, 23]. Therefore, the main challenge in production of nanocomposites includes the achievement of small nanoparticles and good dispersion of nanoparticles. It is vital to

* Correspondence: pengwanxi@163.com; rheeky@khu.ac.kr
[1]School of Forestry, Henan Agricultural University, Zhengzhou 450002, China
[4]Department of Mechanical Engineering, College of Engineering, Kyung Hee University, Yongin 446-701, Republic of Korea
Full list of author information is available at the end of the article

Ashraf *et al. Nanoscale Research Letters* (2018) 13:214

Page 2 of 7

overcome the attractive forces between nanoparticles producing the aggregation/agglomeration, instead of disturbing the structure of nanoparticles. Surprisingly, Dorigato et al. [24] suggested a model which shows the primary filler aggregation reinforces the polymer nanocomposites, while the agglomerated nanoparticles commonly induce negative effects on the mechanical performances of polymer nanocomposites [21, 25]. Accordingly, the study on aggregation/agglomeration of nanoparticles is required to reveal its real effects on the properties of nanocomposites. Although the nanoparticle size is assumed as an attractive benefit in polymer nanocomposites, the effects of isolation or aggregation/agglomeration on the main properties of nanoparticles such as number, surface area, and specific surface area have not been studied in the literature. Moreover, the aggregation/agglomeration of nanoparticles has been assumed as a general term which qualitatively changes the behavior of nanocomposites. Also, the possible roles of nanoparticle and interphase dimensions on the interfacial/interphase properties have not been described in previous studies.

Methods

In this paper, the effects of filler size and density on the number, surface area, stiffening efficiency, and specific surface area of nanoparticles in polymer nanocomposites are explained by proper equations. Also, the aggregation/agglomeration of nanoparticles is assumed as large particles and their influences on various terms are revealed. Similarly, the possible roles of nanoparticle and interphase sizes in the interfacial/interphase parameters and tensile strength of nanocomposites are discussed. The main focus of this article is on the spherical nanoparticles, but other nanoparticle geometries can be studied by development of the suggested equations.

The number of spherically isolated nanoparticles in a nanocomposite can be calculated by the weight of nanoparticles (W_f) as:

$$N = \frac{W_f}{d_f \frac{4}{3}\pi R^3}.$$ (1)

where d_f and R are the density and radius of nanoparticles, respectively. In this condition, the total surface area of dispersed nanoparticles is given by:

$$A = N(4\pi R^2).$$ (2)

A can be considered as the interfacial area between polymer matrix and nanoparticles. Replacing of N from Eq. 1 into Eq. 2 leads to:

$$A = \frac{3W_f}{d_f R}.$$ (3)

which correlates the A with W_f, d_f, and R.

Each nanoparticle introduces a stiffening effect in polymer matrix by mechanical involvement of polymer chains. The level of stress sharing between polymer matrix and nanoparticles depends on the interfacial area and the stiffness of nanoparticles. As a result, a novel parameter as the stiffening efficiency of nanoparticles can be defined as:

$$SE = AE_f = \frac{3W_f}{d_f R}E_f.$$ (4)

where E_f is the Young's modulus of nanoparticles. The stiffening efficiency as a function of the properties of nanoparticles expresses the capability of nanoparticles for the stiffening of nanocomposites. Additionally, the specific surface area of particles is expressed as:

$$A_c = \frac{A}{m} = \frac{A}{d_f v} = \frac{4\pi R^2}{d_f \frac{4}{3}\pi R^3} = \frac{3}{d_f R}.$$ (5)

where m and v are total mass and volume of nanoparticles, respectively. This parameter expresses the surface area of 1 g particles and so, does not depend on the concentration of nanoparticles in nanocomposite.

Now, the tensile strength and interfacial/interphase properties are given by simple equations. Pukanszky [26] suggested a model for tensile strength of composites as a function of filler content and interfacial/interphase properties as:

$$\sigma = \sigma_m \frac{1-\phi_f}{1 + 2.5\phi_f} \exp\left(B\phi_f\right).$$ (6)

where σ_m shows the tensile strength of polymer matrix and ϕ_f is the volume fraction of nanofiller. This model was originally suggested for composites, but this model has shown good agreements with the experimental results of different polymer nanocomposites. A good agreement is obtained between the experimental data of tensile strength and the predictions of Pukanszky equation in many samples such as PP/SiO$_2$ [27], PEEK/SiO$_2$ [28], PVC/CaCO$_3$ [29], PP/CaCO$_3$ [30], and PVC/SiO$_2$ [31] calculating the B parameter as 4.12, 3.15, 3.07, 2.5, and 2.1, respectively. These examples validate the application of Pukanszky model for the tensile strength of polymer nanocomposites.

B is an interfacial parameter which shows the level of interfacial adhesion by:

Ashraf et al. Nanoscale Research Letters (2018) 13:214

Page 3 of 7

$$B = \left(1 + A_c d_f t\right) \ln\left(\frac{\sigma_i}{\sigma_m}\right). \quad (7)$$

where t and σ_i are the thickness and strength of interphase, respectively.

Replacing of A_c from Eq. 5 into the latter equation presents:

$$B = \left(1 + 3\frac{t}{R}\right) \ln\left(\frac{\sigma_i}{\sigma_m}\right). \quad (8)$$

Applying the above equation into Pukanszky model offers the relative strength (σ/σ_m) as:

$$\sigma_R = \frac{1-\phi_f}{1 + 2.5\phi_f} \exp\left[\left(1 + 3\frac{t}{R}\right) \ln\left(\frac{\sigma_i}{\sigma_m}\right)\phi_f\right]. \quad (9)$$

which explicitly links the tensile strength to filler and interphase properties. Also, we should indicate the size effects, which undoubtedly exist when modeling fracture [32–34].

The volume fraction of interphase (ϕ_i) for nanocomposites containing spherical nanoparticles can be considered [35] by:

$$\phi_i = \left[\left(\frac{R + t}{R}\right)^3 - 1\right]\phi_f. \quad (10)$$

in which $t = 0$ results in $\phi_i = 0$ indicating the absence of interphase in nanocomposite. The analytical models in this study may be applicable where other models such as cohesive zone describe the interphase regions. Some previous studies have considered the interphase by some models such as 2D finite element [36, 37].

In our previous work [38], a interphase parameter for polymer nanocomposites reinforced with spherical nanoparticles was defined as:

$$a = 10\left(\frac{t}{R}\right)\left(\frac{10E_i}{E_f} - 1\right). \quad (11)$$

where E_i is the modulus of interphase. This equation correlates the a to various effective parameters of nanofiller and interphase. a was calculated for some nanocomposites ranging from 0.8 to 19 [38]. It was reported that a higher level of a introduces a better modulus in nanocomposite.

Results and discussion

At the first part of this section, the effects of size and density on different properties of nanoparticles are plotted by contour plots and the results are discussed to clarify the influence of aggregation/agglomeration. At the next step, the roles of nanoparticle radius (including the aggregation/agglomeration) and interphase thickness

in the interfacial/interphase properties and nanocomposite performances are studied.

Figure 1 illustrates the aggregation/agglomeration of nanoparticles in a nanocomposite. When the isolated and dispersed nanoparticles are accumulated, it can be assumed that a large nanoparticle is formed. According to Fig. 1, if isolated nanoparticles with R radius aggregate/agglomerate, a big particle is produced with high radius. As a result, the aggregation/agglomeration of nanoparticles can be physically assumed by growth of particle size in nanocomposites. This occurrence affects the characteristics of nanoparticles and interphase which finally change the behavior of nanocomposites.

Figure 2 shows the roles of R and d_f in ln (N) and A levels at constant $W_f = 2$ g. According to Fig. 2a, low N is observed by the high values of R and d_f, but N increases when R and d_f decrease. So, the density and size of nanoparticles inversely affect the number of particles in polymer nanocomposites at a constant filler concentration. The small nanoparticles with low density produce a large number of nanoparticles in nanocomposites, while the big and dense nanoparticles make few particles. Accordingly, the aggregates/agglomerates significantly decrease the number of nanoparticles in nanocomposites at a constant filler concentration.

Figure 2b illustrates the effects of R and d_f parameters on the total surface area of nanoparticles (A in m^2) at $W_f = 2$ g. The surface area of nanoparticles is assumed as the interfacial area between polymer and nanoparticles transferring the stress from matrix to nanoparticles. The stress may be efficiently transported from polymer to nanoparticles to improve the mechanical properties, when the interfacial area is big enough [39, 40]. As observed in Fig. 2b, the largest interfacial area is achieved by the smallest ranges of R and d_f. It is also interesting that only 2 g of isolated and well-dispersed nanoparticles with $R = 10$ nm and $d_f = 2$ g/cm^3 produce about 250 m^2 interfacial area with polymer matrix. However, the interfacial area reduces by increasing the size and density of nanoparticles and A below 50 m^2 is obtained at $R >$ 40 nm and $d_f > 3$ g/cm^3. The significant difference between the interfacial areas at different particle sizes indicates that the nanoparticle size is an important parameter in nanocomposites. The large nanoparticles cause small interfacial area which deteriorates the significant
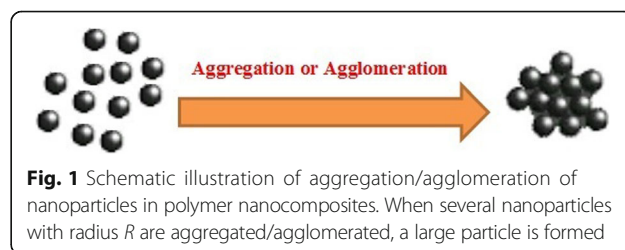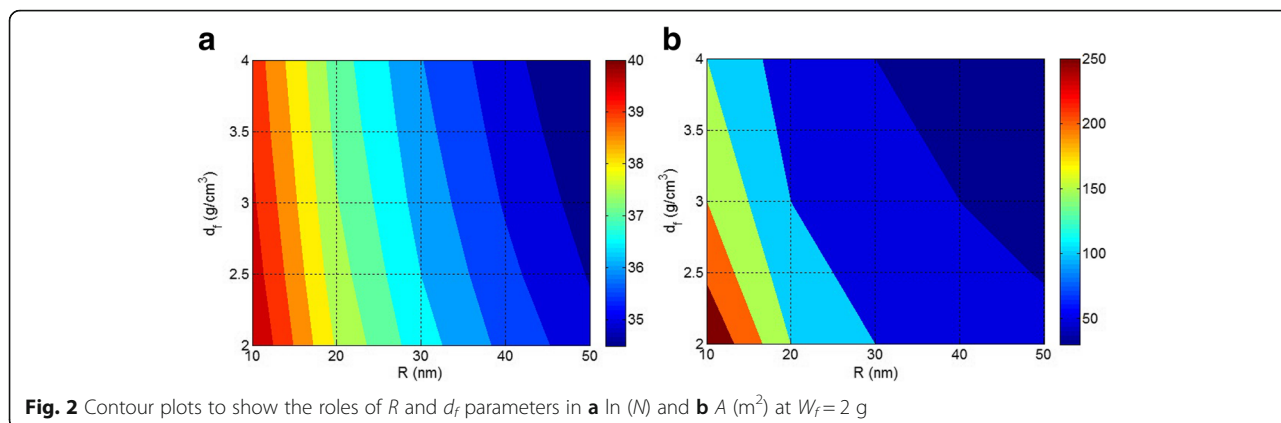

Fig. 1 Schematic illustration of aggregation/agglomeration of nanoparticles in polymer nanocomposites. When several nanoparticles with radius R are aggregated/agglomerated, a large particle is formed

Ashraf *et al. Nanoscale Research Letters* (2018) 13:214

Page 4 of 7

Fig. 2 Contour plots to show the roles of R and d_f parameters in **a** ln (N) and **b** A (m^2) at $W_f = 2$ g
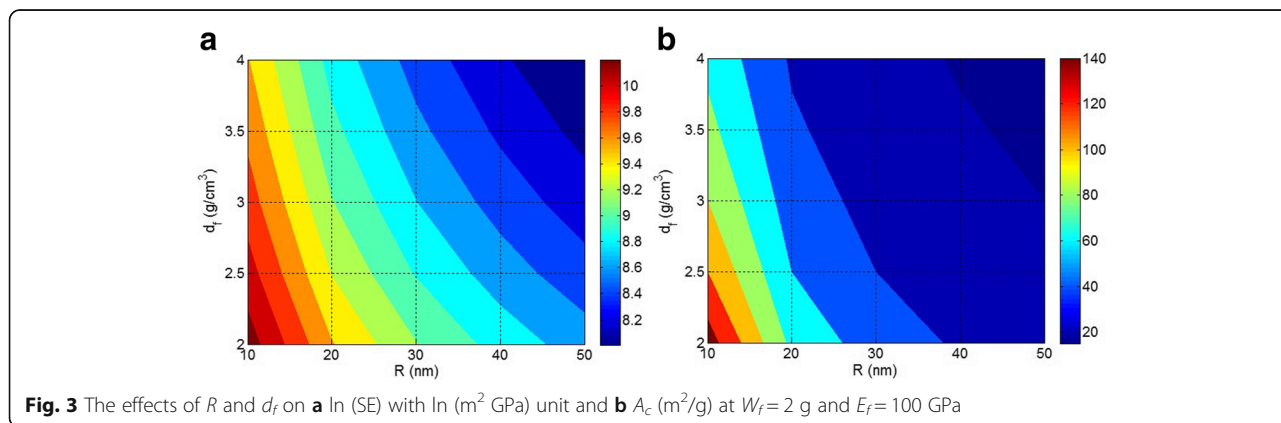
advantage of nanoparticles in nanocomposites. It should be noted that the filler concentration in nanocomposites may not be too high compared to micro-composites, but the extraordinary surface area of nanoparticles commonly results in the interaction between particles and aggregation/agglomeration. As a result, although the high contents of nanoparticles in nanocomposites strengthen the accumulation, the aggregation/agglomeration of nanoparticles generally occur in polymer nanocomposites at different filler concentrations which decrease the interfacial area and weaken the performance.

Figure 3a displays the contour plots of ln (SE) as a function of R and d_f at $W_f = 2$ g and $E_f = 100$ GPa. The stiffening efficiency of nanoparticles increases when small nanoparticles with low density are incorporated in polymer matrix, demonstrating that the nanoparticle size causes an effective role in the stiffening of nanoparticles in polymer nanocomposites. On the other hand, the aggregated/agglomerated nanoparticles deteriorate the performances of polymer nanocomposites by reduction of nanoparticle efficiency. The small nanoparticles with low density meaningfully increase the stiffness of nanocomposites through the great level of stress transferring between polymer chains and nanoparticles. A previous

study in this area has explained the physics of the influence of filler radius on the stress transfer from polymer matrix to fiber using the molecular dynamics simulations [41]. However, the large and dense particles cannot introduce the high stiffness of nanoparticles to polymer matrix suggesting a composite with poor stiffness. Therefore, the characteristics of nanoparticles significantly control the properties of nanocomposites.

Figure 3b also shows the levels of A_c parameter at different R and d_f values at $W_f = 2$ g and $E_f = 100$ GPa. It is observed that the best A_c is obtained by small and low-density nanoparticles, while the worst one is produced by large and dense particles. The A_c value of about 140 m^2/g is achieved by $R = 10$ nm and $d_f = 2$ g/cm^3, while A_c level of less than 20 m^2/g is shown by large particle size and high density. As a result, R and d_f parameters show negative effects on A_c in polymer nanocomposites. It is concluded that A_c parameter expressing the interfacial area of 1 g isolated nanoparticles gives the best levels by small nanoparticles. As a result, the large nanoparticles or aggregates/agglomerates cannot produce a considerable A_c which decreases the efficiency of nanoparticles in polymer nanocomposites. It is known that the performances of nanocomposites such

Fig. 3 The effects of R and d_f on **a** ln (SE) with ln (m^2 GPa) unit and **b** A_c (m^2/g) at $W_f = 2$ g and $E_f = 100$ GPa

Ashraf *et al. Nanoscale Research Letters* (2018) 13:214

Page 5 of 7

as mechanical, flame retardation, and barrier properties directly relate to the interfacial area between polymer and nanoparticles [10, 42]. A large A_c can produce acceptable levels for nanocomposite properties by little amount of nanoparticles, due to the high interfacial area between polymer matrix and nanoparticles. Accordingly, controlling the size and density of nanoparticles are challenging in nanocomposites to create the best properties.

Now, the effects of nanoparticle and interphase sizes on the interfacial/interphase properties and tensile strength of nanocomposites are explained by the proposed equations. Figure 4 illustrates the effects of R and t on B interfacial parameter and tensile strength by Pukanszky model (Eq. 6) at $\sigma_i/\sigma_m = 5$ and $\phi_f = 0.02$. Based on Fig. 4a, the B level of 13 is obtained by the smallest nanoparticles and the thickest interphase. Also, B decreases to below 3 when the size of nanoparticles grows to about 40 nm and the interphase thickness decreases to less than 10 nm. Therefore, the sizes of nanoparticles and interphase play dissimilar roles in $\underline{B}$ parameter. Also, it should be noted that the small nanoparticles without formation of a strong interphase cannot give a high B in polymer nanocomposites. On the other hand, a thick interphase ($t = 25$ nm) surrounding the big nanoparticles ($R = 50$ nm) only improves the B parameter to about 4. As a result, both nanoparticle and interphase dimensions are important to obtain a high level of B in nanocomposites. However, at a constant level of interphase thickness, the growth of nanoparticle size by aggregation/agglomeration decreases B parameter demonstrating the negative effects of aggregates/agglomerates on the interfacial/interphase properties.

Figure 4b also shows the effects of R and t parameters on the tensile strength of nanocomposites by Pukanszky model. It is observed that small nanoparticles and thick interphase improve the strength of nanocomposites. However, a poor strength is observed by big particles and thin interphase. Therefore, both R and t parameters affect the tensile strength of nanocomposites. Moreover,

it is found that the strength of nanocomposites reduces when the size of nanoparticles grows, due to aggregation/agglomeration. Accordingly, it is essential to isolate and disperse the nanoparticles in polymer matrix at small size to achieve the best performances. Since nanoparticles naturally tend to aggregation/agglomeration, modification of their surface or functionalization of polymer chains can prevent the accumulation [19, 43, 44].

Figure 5 depicts the dependences of interphase volume fraction (ϕ_i) and a interphase parameter on R and t parameters at $\phi_f = 0.02$, $E_f = 100$ GPa, and $E_i = 50$ GPa. According to Fig. 5a, the smallest nanoparticles and the thickest interphase give the highest level of ϕ_i as 0.8 which significantly reinforces the nanocomposite. This level of ϕ_i is more than ϕ_f demonstrating the effective roles of R and t parameters in the performances of nanocomposites. Furthermore, ϕ_i decreases to about 0 at $R > 30$ nm, i.e., a thick interphase ($t = 25$ nm) cannot make a high ϕ_i in polymer nanocomposites when large nanoparticles are incorporated in the polymer matrix. This occurrence shows the significant role of nanoparticle size in the formation of interphase regions. So, the size of nanoparticles considerably changes the interphase properties revealing that the aggregation/agglomeration of nanoparticles mostly decreases the interphase concentration which causes poor modulus and strength in nanocomposites [5, 45]. It should be mentioned that the interphase regions may overlap in the systems containing high filler concentration. Therefore, the expressed equation for ϕ_i (Eq. 10) is reasonable for normal nanocomposites containing low filler content.

Figure 5b also shows the effects of R and t levels on a interphase parameter. a increases by small nanoparticles and thick interphase, whereas it gives less values (less than 10) at $R > 40$ nm and $t < 10$ nm. This evidence reveals that a depends on both R and t parameters. Since a high a parameter improves the Young's modulus of nanocomposites [38], small nanoparticles and thick interphase are desirable for nanocomposites performances.
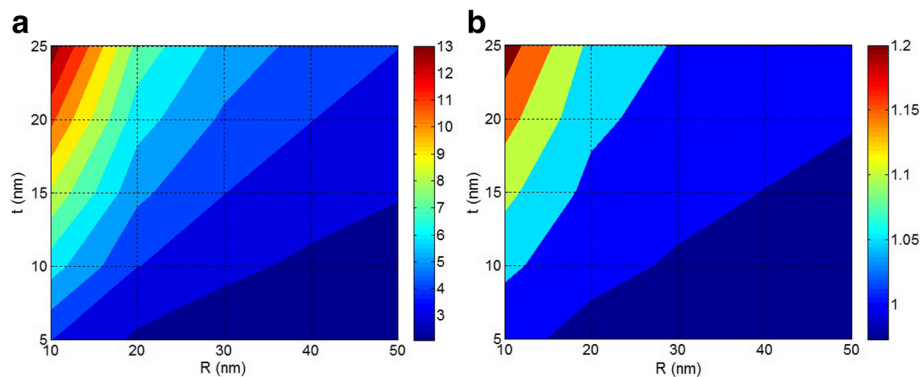

Fig. 4 a B interfacial parameter and **b** relative tensile strength by Pukanszky model at different ranges of R and d_f and constant $\sigma_i/\sigma_m = 5$ and $\phi_f = 0.02$
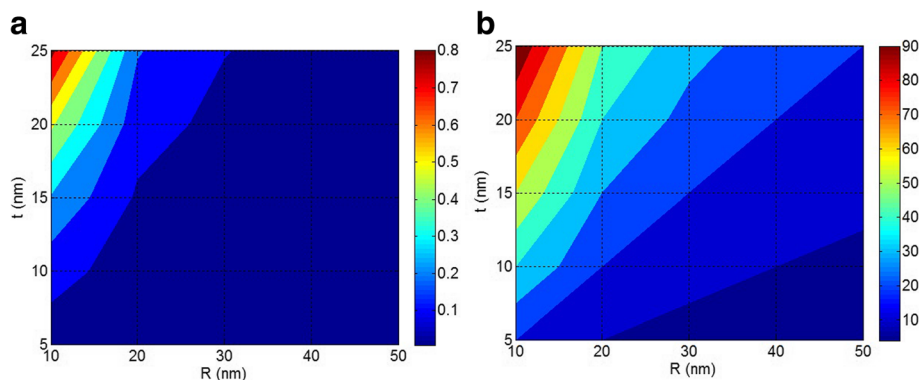
Ashraf *et al. Nanoscale Research Letters* (2018) 13:214

Page 6 of 7

Fig. 5 The correlation of **a** φ_i and **b** a interphase parameter to R and t parameters at $\varphi_f = 0.02$, $E_f = 100$ GPa, and $E_i = 50$ GPa

According to Fig. 5b, the aggregates/agglomerates of nanoparticles (high R) produce slight a event by thick interphase. This occurrence indicates that a strong interphase cannot give a great a or high modulus when the nanoparticles are aggregated/agglomerated in nanocomposites. As a result, the aggregates/agglomerates of nanoparticles cause negative effects on the properties of polymer nanocomposites. Based on the mentioned remarks, the aggregation/agglomeration weakens the benefits of nanoparticles and properties of interface/interphase; therefore, the nanoparticles cannot present a strong reinforcement in polymer nanocomposites.

Conclusions

The effects of filler size and density as well as interphase thickness on the characteristics of nanoparticles and the interface/interphase properties were studied by simple equations. Also, the aggregates/agglomerates of nanoparticles were assumed as large particles and their influences on the interphase parameters and the tensile strength of nanocomposites were discussed. The small size and low density cause significant levels for number, surface area, stiffening efficiency, and specific surface area of nanoparticles. Only 2 g of small and well-dispersed nanoparticles ($R = 10$ nm) with $d_f = 2$ g/cm^3 can produce about 250 m^2 interfacial area with polymer matrix. On the other hand, big size and aggregates/agglomerates weaken the positive attributes of nanoparticles in nanocomposites. Small nanoparticles and thick interphase present the high levels for B parameter, tensile strength, interphase volume fraction, and a interphase parameter. B decreases to below 3 when the size of nanoparticles grows to about 40 nm and the interphase thickness reduces to less than 10 nm. However, $B = 13$ is obtained by the smallest nanoparticles ($R = 10$ nm) and the thickest interphase ($t = 25$ nm). This occurrence confirms that the interfacial/interphase properties depend on the nanoparticle size beside the interfacial interaction/adhesion. Additionally, large nanoparticles produce low

interfacial/interphase properties and poor tensile strength even at high interphase thickness revealing the main role of particles size. The smallest nanoparticles and the thickest interphase give the highest level of ϕ_i, while ϕ_i decreases to about 0 at $R > 30$ nm. This evidence demonstrates that only a thick interphase ($t = 25$ nm) cannot make a high ϕ_i when large nanoparticles or aggregates/agglomerates are present in nanocomposites. Accordingly, the aggregated/agglomerated nanoparticles negatively affect the interfacial/interphase properties and tensile strength of polymer nanocomposites.

Author details
[1]School of Forestry, Henan Agricultural University, Zhengzhou 450002, China. [2]Faculty of Science, Department of Geology, University of Malaya, 50603 Kuala Lumpur, Malaysia. [3]Young Researchers and Elites Club, Science and Research Branch, Islamic Azad University, Tehran, Iran. [4]Department of Mechanical Engineering, College of Engineering, Kyung Hee University, Yongin 446-701, Republic of Korea.

References
1. Perets Y, Aleksandrovych L, Melnychenko M, Lazarenko O, Vovchenko L, Matzui L (2017) The electrical properties of hybrid composites based on multiwall carbon nanotubes with graphite nanoplatelets. Nanoscale Res Lett 12(1):406
2. Bershtein V, Fainleib A, Egorova L, Gusakova K, Grigoryeva O, Kirilenko D et al (2015) The impact of ultra-low amounts of amino-modified MMT on dynamics and properties of densely cross-linked cyanate ester resins. Nanoscale Res Lett 10(1):1
3. Zare Y (2016) Shear, bulk, and Young's moduli of clay/polymer nanocomposites containing the stacks of intercalated layers as pseudoparticles. Nanoscale Res Lett 11(1):479

Ashraf *et al. Nanoscale Research Letters* (2018) 13:214

Page 7 of 7

4. Sagalianov I, Vovchenko L, Matzui L, Lazarenko O (2017) Synergistic enhancement of the percolation threshold in hybrid polymeric nanocomposites based on carbon nanotubes and graphite nanoplatelets. Nanoscale Res Lett 12(1):140

5. Ma X, Zare Y, Rhee KY (2017) A two-step methodology to study the influence of aggregation/agglomeration of nanoparticles on Young's modulus of polymer nanocomposites. Nanoscale Res Lett 12(1):621

6. Kato M, Usuki A, Hasegawa N, Okamoto H, Kawasumi M (2011) Development and applications of polyolefin–and rubber–clay nanocomposites. Polym J 43(7):583–593

7. Zare Y, Rhee KY (2017) Development of Hashin-Shtrikman model to determine the roles and properties of interphases in clay/CaCO3/PP ternary nanocomposite. Appl Clay Sci 137:176–182

8. Zare Y, Rhee KY (2017) Development and modification of conventional Ouali model for tensile modulus of polymer/carbon nanotubes nanocomposites assuming the roles of dispersed and networked nanoparticles and surrounding interphases. J Colloid Interface Sci 506:283–290

9. Zare Y, Rhee KY, Park S-J (2017) Predictions of micromechanics models for interfacial/interphase parameters in polymer/metal nanocomposites. Int J Adhes Adhes 79:111–116

10. Chopra S, Deshmukh KA, Peshwe D (2017) Theoretical prediction of interfacial properties of PBT/CNT nanocomposites and its experimental evaluation. Mech Mater 109:11–17

11. Zare Y, Rhee KY (2017) Dependence of Z parameter for tensile strength of multi-layered interphase in polymer nanocomposites to material and interphase properties. Nanoscale Res Lett 12(1):42

12. Montazeri A, Naghdabadi R (2010) Investigation of the interphase effects on the mechanical behavior of carbon nanotube polymer composites by multiscale modeling. J Appl Polym Sci 117(1):361–367

13. Jahanmard P, Shojaei A (2015) Mechanical properties and structure of solvent processed novolac resin/layered silicate: development of interphase region. RSC Adv 5(98):80875–80883

14. Zare Y, Rhee KY (2017) Development of a conventional model to predict the electrical conductivity of polymer/carbon nanotubes nanocomposites by interphase, waviness and contact effects. Compos A: Appl Sci Manuf 100: 305–312

15. Zare Y, Rhee KY (2017) Development of a model for electrical conductivity of polymer graphene nanocomposites assuming interphase and tunneling regions in conductive networks. Ind Eng Chem Res 56(32):9107–9115

16. Zare Y, Rhee KY (2017) A simple methodology to predict the tunneling conductivity of polymer/CNT nanocomposites by the roles of tunneling distance, interphase and CNT waviness. RSC Adv 7(55):34912–34921

17. Rafiee R, Pourazizi R (2015) Influence of CNT functionalization on the interphase region between CNT and polymer. Comput Mater Sci 96:573–578

18. Herasati S, Zhang L, Ruan H (2014) A new method for characterizing the interphase regions of carbon nanotube composites. Int J Solids Struct 51(9): 1781–1791

19. Esbati A, Irani S (2018) Effect of functionalized process and CNTs aggregation on fracture mechanism and mechanical properties of polymer nanocomposite. Mech Mater 118:106–119

20. Jouault N, Vallat P, Dalmas F, Said S, Jestin J, Boué F (2009) Well-dispersed fractal aggregates as filler in polymer–silica nanocomposites: long-range effects in rheology. Macromolecules 42(6):2031–2040

21. Zare Y (2016) Study of nanoparticles aggregation/agglomeration in polymer particulate nanocomposites by mechanical properties. Compos A: Appl Sci Manuf 84:158–164

22. Chen J, Yu Y, Chen J, Li H, Ji J, Liu D (2015) Chemical modification of palygorskite with maleic anhydride modified polypropylene: mechanical properties, morphology, and crystal structure of palygorskite/polypropylene nanocomposites. Appl Clay Sci 115:230–237

23. Khan A, Shamsi MH, Choi T-S (2009) Correlating dynamical mechanical properties with temperature and clay composition of polymer-clay nanocomposites. Comput Mater Sci 45(2):257–265

24. Dorigato A, Dzenis Y, Pegoretti A (2013) Filler aggregation as a reinforcement mechanism in polymer nanocomposites. Mech Mater 61: 79–90

25. Zare Y (2016) The roles of nanoparticles accumulation and interphase properties in properties of polymer particulate nanocomposites by a multi-step methodology. Compos A: Appl Sci Manuf 91:127–132

26. Pukanszky B (1990) Influence of interface interaction on the ultimate tensile properties of polymer composites. Composites 21(3):255–262

27. Bikiaris DN, Papageorgiou GZ, Pavlidou E, Vouroutzis N, Palatzoglou P, Karayannidis GP (2006) Preparation by melt mixing and characterization of isotactic polypropylene/SiO2 nanocomposites containing untreated and surface-treated nanoparticles. J Appl Polym Sci 100(4):2684–2696

28. Zhang G, Schlarb A, Tria S, Elkedim O (2008) Tensile and tribological behaviors of PEEK/nano-SiO2 composites compounded using a ball milling technique. Compos Sci Technol 68(15):3073–3080

29. Xie X-L, Liu Q-X, Li RK-Y, Zhou X-P, Zhang Q-X, Yu Z-Z et al (2004) Rheological and mechanical properties of PVC/CaCO3 nanocomposites prepared by in situ polymerization. Polymer 45(19):6665–6673

30. Chen H, Wang M, Lin Y, Chan CM, Wu J (2007) Morphology and mechanical property of binary and ternary polypropylene nanocomposites with nanoclay and CaCo3 particles. J Appl Polym Sci 106(5):3409–3416

31. Chen G, Tian M, Guo S (2006) A study on the morphology and mechanical properties of PVC/nano-SiO2 composites. J Macromol Sci Part B 45(5):709–725

32. Talebi H, Silani M, Rabczuk T (2015) Concurrent multiscale modeling of three dimensional crack and dislocation propagation. Adv Eng Softw 80:82–92

33. Talebi H, Silani M, Bordas SP, Kerfriden P, Rabczuk T (2014) A computational library for multiscale modeling of material failure. Comput Mech 53(5):1047–1071

34. Budarapu PR, Gracie R, Yang S-W, Zhuang X, Rabczuk T (2014) Efficient coarse graining in multiscale modeling of fracture. Theor Appl Fract Mech 69:126–143

35. Ji XL, Jiao KJ, Jiang W, Jiang BZ (2002) Tensile modulus of polymer nanocomposites. Polym Eng Sci 42(5):983

36. Msekh MA, Cuong N, Zi G, Areias P, Zhuang X, Rabczuk T (2018) Fracture properties prediction of clay/epoxy nanocomposites with interphase zones using a phase field model. Eng Fract Mech 188:287–299

37. Hamdia KM, Silani M, Zhuang X, He P, Rabczuk T (2017) Stochastic analysis of the fracture toughness of polymeric nanoparticle composites using polynomial chaos expansions. Int J Fract 206(2):215–227

38. Zare Y (2015) Assumption of interphase properties in classical Christensen–Lo model for Young's modulus of polymer nanocomposites reinforced with spherical nanoparticles. RSC Adv 5(116):95532–95538

39. H-x L, Zare Y, Rhee KY (2018) The percolation threshold for tensile strength of polymer/CNT nanocomposites assuming filler network and interphase regions. Mater Chem Phys 207:76–83

40. Razavi R, Zare Y, Rhee KY (2018) A model for tensile strength of polymer/carbon nanotubes nanocomposites assuming the percolation of interphase regions. Colloids Surf A Physicochem Eng Asp 538:148–154

41. Vu-Bac N, Lahmer T, Zhang Y, Zhuang X, Rabczuk T (2014) Stochastic predictions of interfacial characteristic of polymeric nanocomposites (PNCs). Compos Part B 59:80–95

42. Xu Q-J, Wang S-B, Chen F-F, Cai T-C, Li X-H, Zhang Z-J (2016) Studies on the interfacial effect between nano-SiO2 and nylon 6 in nylon 6/SiO2 nanocomposites. Nanomaterials and Nanotechnology 6:31

43. Ferreira F, Pinheiro I, Gouveia R, Thim G, Lona L (2018) Functionalized cellulose nanocrystals as reinforcement in biodegradable polymer nanocomposites. Polym Compos 39(S1):E9

44. Brković DV, Pavlović VB, Pavlović VP, Obradović N, Mitrić M, Stevanović S et al (2017) Structural properties of the multiwall carbon nanotubes/poly (methyl methacrylate) nanocomposites: effect of the multiwall carbon nanotubes covalent functionalization. Polym Compos 38(S1):E472–E489

45. Zare Y, Rhee KY, Hui D (2017) Influences of nanoparticles aggregation/agglomeration on the interfacial/interphase and tensile properties of nanocomposites. Compos Part B 122:41–46