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Understanding the Effect of Interfacial Interphase on the Elastic Response of Hollow Glass Microsphere Reinforced Microcomposites

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ABSTRACT

The hollow glass microspheres (HGMS) has been recently used in the fabrication of low-density polymeric composites due to rather high stiffness nature of the fillers together with their lightweight that in turn results in the development of micro-composites of engineered properties with enhanced mechanical properties. Interfacial interactions at the filler/polymer interface control the load transfer and, thus, bulk properties of composites leading to unpredictable performance of composites embedded with inclusions. Nevertheless, useful analytical models are required to estimate the mechanical behavior of the HGMS based composites with the incorporation of the effect of interfacial interactions and possible agglomeration of fillers. No studies so far have reported the analytical modeling of HGMS reinforced thermosetting composites emphasizing the role of the interphase shaped at the vicinity of fillers. This study aims at the fabrication of 0-20 wt% HGMS/polyester micro-composites followed by micromechanical modeling of the fabricated parts whilst the effect of the interphase region is emphasized by models modification. The results indicated a strong correlation between the interphase characteristics and Young's modulus of the specimens revealing the dependency of the modulus on the thickness and modulus of the interphase as well as the level of agglomeration and interfacial debonding of the HGMSs. The results demonstrated that with considering no interphase, the models underestimate the modules of the parts, which suggests the presence of stiff interphase around the HGMS governed by changes in the interfacial cross-link density of the parent polymer as hypothesized supported by the mechanical response of the parts.

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NOMENCLATURE

A_i	Constants used on Eshelby's tensor	f	Fillers volume fraction (%)
d	Filler diameter (mm)	l	Filler length (mm)
E_{11}	Composite elastic modulus (GPa)	**Greek Symbols**	
E_C	Composite elastic modulus (GPa)	ζ	Shape factor in Halpin-Tsai relation, l/d
E_f	Filler tensile modulus (GPa)	η	Halpin-Tsai constant as a function of modulus and shape factor
E_i	Interphase tensile modulus (GPa)	ν_m	Matrix Poisson 's ratio
E_m	Matrix tensile modulus (GPa)	η	Halpin-Tsai constant as a function of modulus and shape factor

1. INTRODUCTION

The fabrication of lightweight high stiff composites has drawn increasing attention due to their extensive applications mainly due to the energy conservation aspects, the decrease in the processing costs and energy consumption during the processing. Therefore, the development of lightweight thermosetting composites with no or minimal decrease in their mechanical properties has drawn a great body of interest due to their broad applications in structures, automotive industries or energy sectors [1, 2]. However, due to the presence of multiple variables such as the interfacial interactions at the surface of filler, the agglomeration of fillers and the

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modified properties of the polymers at the surface of the fillers, the eventual mechanical properties of reinforced polymers cannot be easily predicted through analytical or computational tools. Interfacial interactions either repulsive or attractive can modify the physical properties of polymer chains at the interface of filler/polymer and, thus, result in either an imperfect or strong bonding at the contact of filler/polymer, respectively [3].

Most often, attractive interactions lead to the generation of a third stiff layer called "interphase" that is responsible for the quality of load transfer from the matrix to the filler. The interactions, moreover, may result in the formation of the agglomeration phase which suppresses the mechanical properties of the reinforced composites [4, 5]. One main challenge still existing is the unknown properties of the interphase for many composite systems as the thickness and mechanical peripeties of the interphase are remarkably material specific. In addition, the small size of the interphase requires sophisticated and advanced techniques for its accurate characterization [6]. The latter would lead to even more issues in the corporation of the interphase as a second mechanism into modeling tools [7].

The challenges arise when, for reliable characterization, the lack of any filler/polymer shear slippage, filler debonding and additional voids, micro-cracks and imperfections at the interface is assumed [8, 9]. Such phenomena significantly result in mechanical response exhibited by nano/micro-composites that cannot be easily predicted or engineered through even extensive experimental observations [10, 11]. Therefore, for a better understanding of the effect of interfacial interactions on mechanical performance of composites, although micromechanical models have been frequently employed, there is a need for approaches through which the effect of the interactions could be assessed.

Based on previous studies, numerous thermosetting-based composite systems have been designed and developed using micro size fillers from metal oxides, carbon-based materials to natural fibers [12, 13]. It could be consequently understood that the utilization of glass based reinforcements has been widely reported in the enhancement of mechanical properties of composites [14, 15]. Although this body of research reveals the dependence of mechanical and physical behavior thermosetting composites reinforced with fillers, the use of the hollow glass microspheres (HGMSs) as lightweight high stiff reinforcements has been rarely reported in respect of the elastic response of polyester-based composites filled with HGMS as the reinforcement [16]. In one study, Chen et al. [17] reported the use of HGMSs coated with silver particle aiming at the fabrication of electrically conductive microspheres to be employed as one of the reinforcement phase in epoxy-based liquid molded composites. The composites exhibited electromagnetic interference shielding properties whilst the HGMSs acted as materials to fill up the vacancies generated by the network of carbon fibers. Jiang et al. [18] have recently revealed their work on the use of HGMSs in reinforcing expanded polystyrene (EPS) and epoxy resin (EP) to develop compression molded foam balls of enhanced compressive strength. In another study, Altay et al. [19] incorporated HGMSs together with polystyrene (PS) microfiber membrane to examine the effect of HGMSs on the thermal insulation and sound absorption insulation behavior of glass fiber fabric-reinforced epoxy composites. Moreover, no research has been reported on the correlations amongst the elastic performance of the HGMS reinforced thermosetting micro-composites emphasizing the effect of interphase [20, 21]. To do so, this study aims at the analytical modeling of 0-20 wt% HGMS reinforced polyester micro-composites using the Halpin-Tsai and Tandon-Weng models accounting for the interphase thickness and elastic modulus for a better understanding of the role of the reinforcing mechanism, the interphase, in overall tensile behavior of the HHMSs based micro-composites.

To examine the interphase impact on the elastic modulus of the fabricated parts, tensile testing was performed and the moduli of the composites were compared with the models' predictions. Upon parametric changes in the interphase properties, best result fitting based on the experimental values was determined and possible links with the interfacial modification of the polyester matrix such as degree of cross-links at the surface of HGMS and the possible presence of agglomeration phase as an adverse clustered region at higher filler loading were discussed.

2. EXPERIMENTAL

2. 1. Material Industrial-grade HGMS was used as the micro-reinforcement with the density of 0.38 g/cm^3, the modulus of 1.99 GPa and an approximate diameter of ~30-40 μm. An industrial-grade polyester thermoset resin was used as the polymer matrix. The resin components of hardener and catalyst were mixed as described by the manufacturer. Figure 1 represents the SEM image of the fractured surface of polyester composites reinforced with HGMS illustrating the surface morphology and the average size of the HGMS fillers.

2. 2. Fabrication of HGMS/Polyester Micro-composites Micro-composites of polyester resin reinforced with 0 to 20 wt% of as-received HGMS were fabricated using direct mixing of the fillers within the resin utilizing a high shear mixer (HSM) consisting a rotor/stator mechanism at 3000 rpm for 30 minutes. The mixture of resin/hardener/catalyst was then mixed and cast into silicon molds per the ASTM standard required

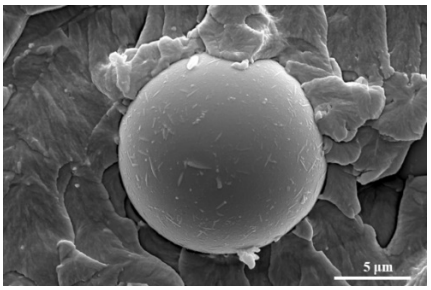


Figure 1. SEM image of a single HGSM particle exposed from the surface of HGMS/polyester composites

for each test near its gel time to avoid HGMS floating onto the surface of the resin due to its lightweight. To fabricate specimens, the curing process was performed at the ambient condition taking ~ 24 hours for the speciemens to be cured. No surface modification of HGMSs was used in the fabrication process of the current study [22]. The summary of the fabrication route and dispersion technique is given in Table 1.

2. 3. Characterization of HGMS/Polyester Composites

2. 3. 1. Tensile Properties To understand the effect of HGMSs on the tensile response of the composites including Young's modulus and tensile strength, tensile testing was performed per to the ASTM D638 test method using a universal tensile testing machine (Sanaf Co., Iran).

Three tensile test specimens were used in the case of each composite system and at least an average of two specimens were reported. The tensile values were obtained at the deformation rate (stroke speed) of 2.54 mm/min at the ambient temperature. The slop of stress-strain curve over the linear region was used to determine the modulus of the samples and the highest point on the curves were as the tensile strength of the composites.

2. 3. 2. Impact Measurement The Izod impact resistance of the composites was measured according to the ASTM D256 using an Impact machine (Sanaf Co., Iran). At least three specimens were used for the impact resistance performance of each composite system and the average and standard deviation were reported. The test was performed to better correlate the presence of stiffer interphase with the assumptions used in the incorporation of the interphase into the modeling techniques used.

TABLE 1. Fabrication process specification

Parameter	Value
HSM rotational speed	3000 (rpm)
HSM diameter	20 (mm)
Mixing time	30 (minutes)
Curing time	24 (hours)
Curing temperature	ambient
Mold cavity dimension	120×120×10 mm^3 3.2×12×70 mm^3

2. 3. 3. Drop Weight Testing HGMS reinforced composites of 120×120×10 mm^3 were fabricated to conduct the drop-weight analysis. A digital drop testing system based on the changes on the deflection of a single cantilever beam load-cell and electric out-put current of strain gages on a Whetstone Bridge was used for assessing the toughness of the parts. The system works in such a way that upon damage onto the specimens, a fraction of the potential energy applied is absorbed by the parts and the rest of the energy is to be transferred to the loadcell. The energy received by the cantilever leads to the deflection of the beams to be a measure of the absorbed energy by the composites in a differential manner.

2. 3. 4. Scanning Electron Microscopy (SEM) Studies The morphology, HGMS/polyester bonding conditions, dispersion quality and microstructure of the fractured surface of the composites were evaluated on a FE-SEM (QUANTA FEG 450-USA). The surface of the SEM composite samples cut from the fractured surface of the composites was gold sputtered before the analysis to eliminate electron charging effects created by the non-conductive nature of the polymer-based specimens.

3. MICROMECHANICAL APPROACH

To understand the effect of interactions on the elastic response of HGMSs reinforced composites, micromechanical models were utilized emphasizing the role of the interphase properties [23]. Assumptions including the perfect dispersion and distribution of HGMSs, linear elastic properties of the filler and matrix, isotropic properties of the constituents were made. The aspect ratio of HGMSs was assumed to be ~ 1 as the fillers with spherical geometry as clearly demonstrated in Figure 1. Moreover, it was assumed that perfect interfacial bonding exists at the interface of HGMSs/polyester as one main factor, although this assumption proved to be invalid at higher fillers content due to weakened interfacial strength. Therefore, it was assumed that any debonding conditions and micro-voids resulting in shear slippage of agglomerated phase are not generally present in the bulk micro-composites as confirmed by the SEM image represented in Figure 2. It is illustrated in this study upon the addition of further HGMS loadings, interfacial debonding is likely due to

the suppressed wettability of the polymer due to excess surface area of the fillers to be wetted by the polyester resin. In the current study, three analytical models were employed to examine the effect of the interphase.

The Halpin-Tsai (H-T) model and Tandon-Weng extracted from Eshelby's tensor components first introduced in the case of ellipsoidal particles into an infinite matrix were used as the modeling technique [24].

Required modifications were considered to incorporate the interphase modulus and thickness per to a parametric study using the core-shell concept as the interphase acts like the matrix for the filler. The properties of a single entity homogenized filler being a new filler previously consisting of a filler and surrounding matrix, thus, were approximated using the micromechanical models. In addition, as the models are normally utilized in the case of short-fibers and function more accurately for inclusions of considerable aspect ratios, the rule of mixtures (ROM) was also used for the case of comparison to understand the reinforcing effect of the filler/interphase entity embedded in the polyester. The Halpin-Tsai equations used for the estimation of Young's modulus of the composites are expressed as the following equation [25]:

$$E_c = E_m\left(\frac{1+\zeta\eta f}{1-\zeta f}\right) \quad (1)$$

where

$$\eta = \frac{\left(\frac{E_f}{E_m}\right)-1}{\left(\frac{E_f}{E_m}\right)+\zeta} \quad (2)$$

in which E_f, E_m and E_C are Young's modulus of the fillers, matrix and the composites, respectively and f the fillers volume fraction.

$\xi = 2\frac{l}{d}$ is to be ~2 in the case of spherical particles. ξ is a correction factor depending upon the shape and geometry of fillers reportedly exists in the literature [26]. Tandon-Weng (T-W) explicit model initially introduced based on the Mori-Tanaka method was used to compare the modeling results with those represented by the experimental observations and the H-T model. The Tandon-Weng model can be described as the equation below [26, 27]:

$$\frac{E_{11}}{E_m} = \frac{1}{1+f(A_1+2\nu_m A_2)/A}. \quad (3)$$

where E_{11} is the longitudinal Young's modulus in the direction of fibers/fillers, ν_m the Poisson's ratio of the polyester in this study and A constants were obtained using the Eshelby's tensor depending on the aspect ratio, Poisson's ratio of the matrix as well as the elastic Lame constants of the filler and matrix. The Young's modulus of the HGMSs was assumed to be 1.99 GPa based on the literature [28] and industrial-grade fillers. The modulus of 4.54 GPa for the neat polyester based on the experimental testing results, the density of 1.23 and 0.38 g/mm^3 based on the densitometry results and the manufacturer datasheet, respectively, and 0.23 and 0.40 as the Poisson's ratio of the HGMSs and the polyester-based on the literature values and manufacturer were used. The Lame constants were used based on the elasticity theory in respective constituents from literature as needed unless otherwise mentioned.

The hypothesis is that the vol% of the interphase region is significantly governed by the interphase thickness, the geometry of fillers and the agglomeration phase accordingly resulting in variations in the modulus of HGMS/polyester composites. To examine the effect of the ratio of interphase vol% to that of the filler, the interphase thickness was estimated to vary from 1 to 8 μm based on reasonable values reported in the case of micro-reinforced composites in literature [29]. The effect of interphase modulus, thus, was evaluated using the assumed values whilst the generation of a stiffer interphase compared to that of the matrix was hypothesized based on the experimental tensile moduli. In all cases, simultaneous variations in the interphase modulus as well as the changes in the interphase thickness were taken into account. It is pointed out that multiple variables exist to be considered in the

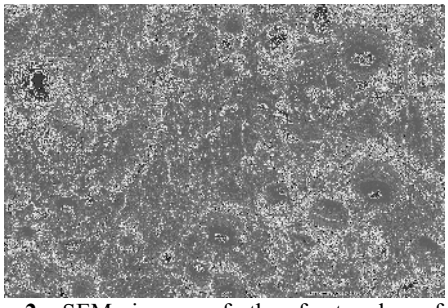


Figure 2. SEM image of the fractured surface of HGMS/polyester representing bonding conditions of HGMS/polyester

TABLE 2. Summary of the values used in the micromechanical modeling of the composites

Parameter	Value
HGMS diameter	30 (μm)
HGMS modulus	1.99 (GPa)
HGMS density	0.38 (g/mm^3)
HGMS wt. %	0-20
HGMS aspect ratio	~1
Interphase thickness	0-8 (μm)
Interphase modulus	2-15 (GPa)
Neat polyester modulus	4.54 (GPa)
Neat polyester density	1.23 (GPa)

incorporation of the interphase region into the analytical models. However, the thickness and modulus of this region are markedly affected by the interfacial strength and, thus, are taken into consideration as underlying parameters. Table 2 summarizes the parametric and experimental values employed in the micromechanical modeling of the HGMS/polyester composites.

4. RESULTS AND DISCUSSION

4. 1. Tensile Properties Figure 3 illustrates the tensile properties of polyester composites reinforced with 0-20 wt% of HGMS. As understood from the results, addition of HGMS results in a sharp decrease in the tensile strength whilst an increase in the modulus is observed. The decrease in the tensile strength as shown in Figure 3a is attributed to the highly porous nature of the fabricated parts with lower packing density and integrity of the specimens. It is hypothesized that an increase in the observed modulus as revealed in Figure 3b is correlated to the increased degree of interfacial cross- links generated at the filler surface and, thus, the enhanced stiffness of the composites. It is suggested that due to the lower Young's modulus of the HGMSs used compared to that of polyester, the second mechanism of reinforcement contributes to an increase in the modulus. However, upon the addition of HGMS up to the 12 wt% loadings, the modulus reaches a plateau suggesting the presence of opposing effects such as the formation of HGMSs agglomeration at higher loadings resulting in the decrease in the effective interfacial bonding and, thus, the interphase quality [5]. The latter is believed to decrease the shear resistance of the bulk composites as a result of weaker filler's wettability and, thus, suppressed load transfer at the interface of HGMSs and polyester matrix [30].

4. 2. Impact Behavior of the Composites The impact response of the specimens as a function of the HGMS wt% is demonstrated in Figure 4. As shown, the impact resistance of the composites experiences an overall increase with addition of filler content reaching an optimum value at 10 wt% of HGMSs. The observations could be ascribed to the generation of the greater volume of stiff interphase around the fillers as the HGMSs containing hollow structures cannot be essentially considered as a contributing phase to the enhanced impact performance of the composites. The hypothesis of stiffer interphase concerning the parent polymer matrix is interrelated to the polymer chains of cross-linking nature with significant growth in the degree of chains immobilization. The formation of constrained chains is thought to be occurred due to the presence of attractive interfacial forces, chains mechanical interlocking due to the fillers surface adsorption effect

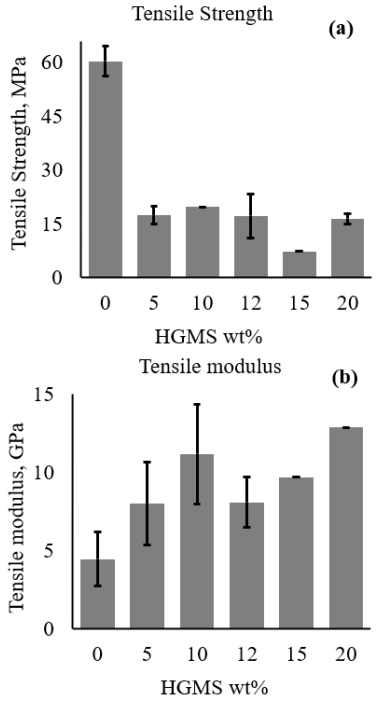


Figure 3. (a) Tensile strength and (b) tensile modulus of HGMS reinforced micro-composites as a function of HGMS loading

and interfacial van der Waals forces [31, 32]. The decrease in the results beyond this filler content is attributed to the increased agglomeration of the HGMS phase resulting from several competing factors. First, it is widely believed that at higher filler ratios, there is a lack of enough polymer available to wet the surface of fillers due to the extensive surface of fillers compared to that of polymer resin. Moreover, the agglomeration phase not only encourages crack propagation through the cluster phase with no polymer bonding, but also the agglomerated region leads to a decrease in effective interfacial interactions and, consequently, lowered interphase properties in terms of thickness and modulus [33].

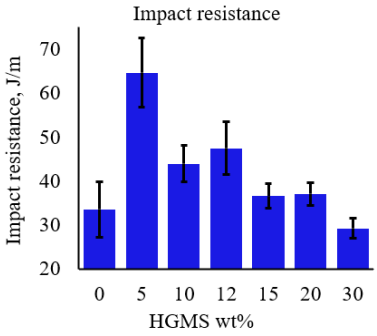


Figure 4. Impact resistance of HGMS filled micro-composites

4. 3. Toughness Performance of HGMS/Polyester Composites The HGMS/polyester composites energy consumption obtained from the drop weight impact as a measure of the toughness against the HGMS content is displayed in Figure 5. It is clearly understood from the figure that the addition of HGMSs into the polyester leads to an overall enhancement in the toughness exhibiting the greatest values in the case of composites at 12-15 wt% of HGMS loading. The findings support the creation of a stronger interfacial bonding and hence the formation of interphase with a stiffer nature compared to the neat polyester as earlier confirmed by the impact test results. As clearly understood, the results are in good agreement with the tensile response and impact performance of the parts where the presence of agglomeration phase at higher HGMSs loading adversely influences the toughness of the fabricated composites due to the factors discussed earlier.

4. 4. Micromechanical Predictions Figure 6 illustrates the micromechanical results predicted by the models compared to the experimental moduli. It is clearly shown that unmodified models underestimate the elastic modulus, which is ascribed to the lower modulus of the HGMS than that of the neat polyester. The presence of stiffer interphase is hypothesized. It is revealed by the findings as the aspect ratio of the fillers is ~1, the moduli are predicted the same regardless of the model used.

Several factors further contribute to the models underestimation including inadequate load transfer in the case of spherical particles, the assumption of uniform distribution of fillers in the matrix and perfect filler/polymer interfacial bonding.

Figures 7a to 7d depict the effect of incorporating the interphase of varying thickness while its modulus changes from 2 to 15 GPa. It is well defined that after the increase in the modulus beyond 12 GPa, the models start predicting more accurately concerning the experimental values and the increase is more intensified as the interphase thickness increases.

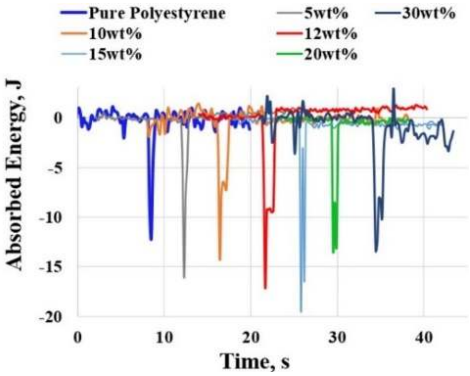


Figure 5. Drop weight energy absorption vs. time on based on absorbed energy difference. The downward peaks represent maximum energy absorbed upon damage by the specimens

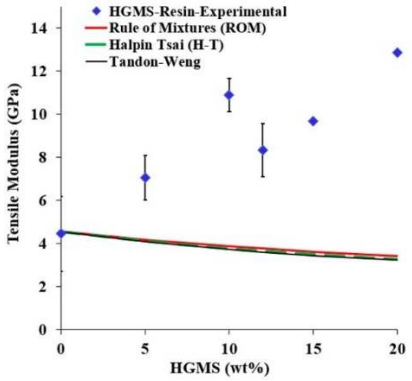
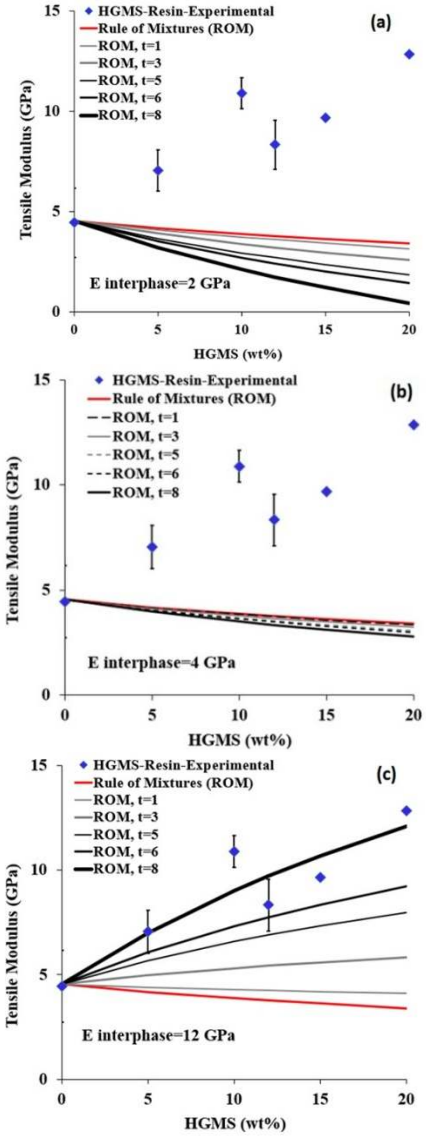


Figure 6. Modeling predictions as a function of HGMS wt%



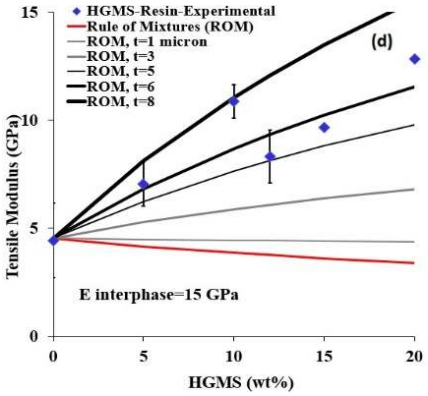


Figure 7. Modeling predictions as a function of HGMS wt% and interphase thickness in μm and modulus of: (a) 2, (b) 4, (c) 12, and (d) 15 GPa

The findings are correlated to and support the occurrence of a stiffer interfacial region as a result of an enhanced degree of cross-links at the vicinity of the HGMS surface forming stiff interphase. As mentioned earlier, a core-shell technique was employed to incorporate the interphase into the models. As expected with the addition of an interfacial phase surrounding the HGMSs, the overall elastic properties of the matrix changes resulting from the neat matrix replaced by either softer (at the interphase modulus of 2 GPa) or stiffer (at the interphase modulus of 15 GPa) material. Beside this phenomenon, as the interphase region of a given thickness occupies the neat matrix, the resulted shell material around the HGMS is expected to contribute to the overall modulus of the core-shell entity. Consequently, the modulus of the new filler (consisting of the interphase as the shell and the original HGMS as the core) is dramatically influenced by the thickness and the assumed modulus of the interphase.

As shown in Figure 7a, with the increase in the interphase thickness, the overall modulus of the composites is further decreased due to the addition of softer phase (modulus of 2 GPa) compared to the parent matrix (modulus of 4.54 GPa). The finding suggests that the interphase modulus is likely to be greater than 2 GPa based on the experimental value. As represented in Figure 7b, the presence of the interphase adversely contributes to the elastic response of composites. Nevertheless, the interphase thickness exhibits only a slight impact on the modulus as the interphase modulus and that of the neat polyester are comparable. Figures 7c and 7d clearly illustrate that the existence of a stiff interphase compared to the neat matrix not only leads to the enhancement in the modulus of the composites but also results in the sensitivity of the models to the interphase thickness. As discussed earlier, in this case, the interphase region of high modulus replaces the softer matrix, and, consequently, results in a marked improvement in the elastic response of the composites. As shown in Figure 7d, the impact of interphase thickness on the composites modulus is more intensified when the interphase gains greater degree of elastic properties.

Figures 8a and 8b in addition demonstrate the sensitivity of the models to the interphase thickness (6 and 8 μm represented) whilst the modulus varies. As understood from Figures 7 and 8, the composites modulus is more sensitive to both interphase thickness and modulus at lower HGMS wt%. It could be understood from the modulus curves represented in Figure 9 that there exists a trade-off between the interphase modulus and thickness suggesting that stiffer interphase of lower thickness may virtually lead to the same prediction as thick interphase of lower modulus. However, it is clearly understood from Figure 9 that the modulus predictions are more sensitive to the thickness of the interphase than the modulus resulting in a greater reinforcing efficiency [34]. The findings suggest that even though spherical particles, irrespective of their size, show an aspect ratio of 1, fillers of small diameters might contribute to the more effective interfacial load transfer than those with larger diameters. This effect could be thought of as the enhancement in the surface to volume ratio of fillers and, thus, an increase in overall volume fraction of the created interphase. The effect of the agglomeration phase on the overall elastic response of the fabricated part was also examined to give a better insight

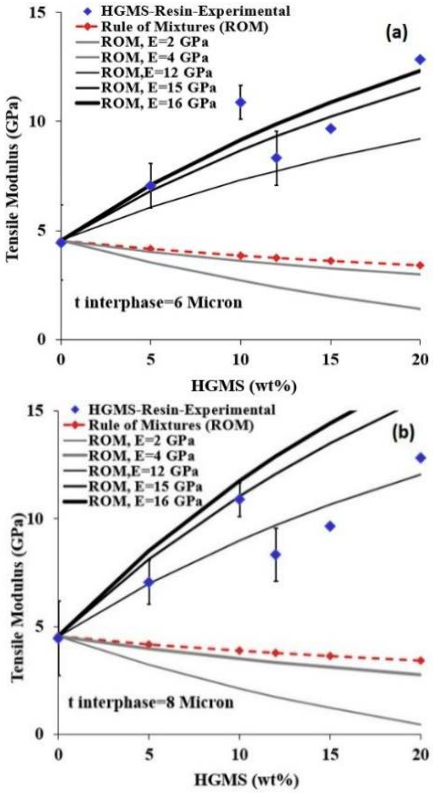


Figure 8. Modeling predictions as a function of HGMS wt% and modulus at the thickness of: (a) 6 and (b) 8 μm

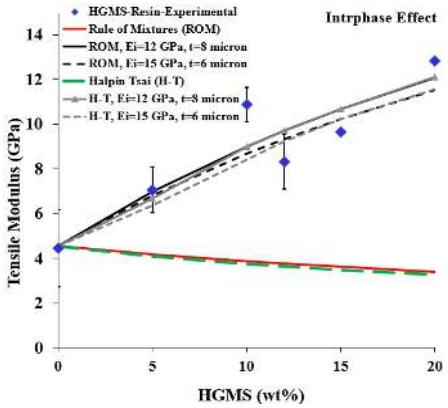


Figure 9. Optimized modeling predictions as a function of HGMS wt%

into the effect of interfacial interactions in particular at higher HGMSs loadings. As shown in Figure 10, the level of agglomeration phase was incorporated into the models considering the cluster size based on of a representative SEM image at high filler loading above 20 wt%. It is shown that with the incorporation of no agglomeration, the models somewhat overpredict the modulus depending on the models used (considering interphase); however, with the agglomeration involved, the modified models predict optimally closer to the experimental tensile modulus values. The agglomeration level/size was assumed using the SEM images of the fractured surface considering the equivalent particle size based on an average number of 4 HGMSs within each cluster as shown in Figure 11 (distinguished by circles) as discussed later in this work. It was found that each equivalent radius of HGMS is around 48-50 μm (v.s. 15-20 μm in the case of isolated HGMSs).

4. 5. Morphological Properties Figure 11 represents the SEM micrographs of HGMS reinforced polyester micro-composites filled with 3, 10, 20 and 30

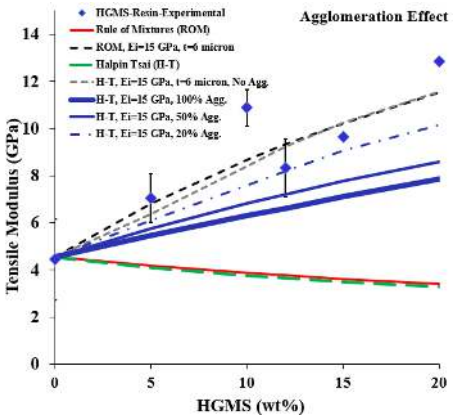


Figure 10. Modeling predictions as a function of HGMS wt% and agglomeration level from 0 to 100%

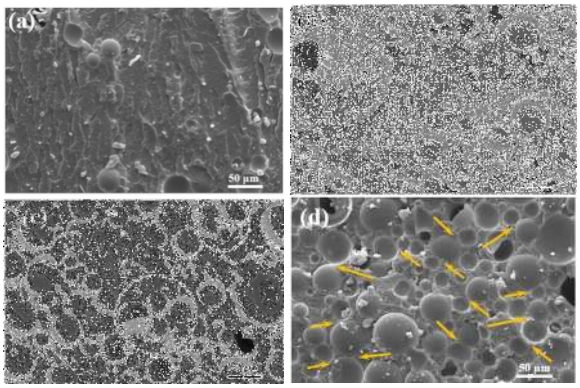


Figure 11. SEM fracture surfaced of HGMS/polyester composites filled with HGMS loading of (a) 3 wt% representing isolated single fillers dispersed within the matrix, (b) 10 wt% illustrating grown number density of fillers, (c) 20 wt% suggesting the presence of fillers clusters besides a dispersed phase and (d) 30 wt% as a high ratio filled composites displaying tangential interconnected spherical surface demonstrating lowered wettability of fillers due to their excess surface area compared to available polyester

wt% of the reinforcements. It is revealed that at low fillers loading, there exists uniform dispersion of HGMSs within the matrix illustrated in Figure 11a and composites where the models take into account high-level dispersion of fillers. Nevertheless, with an increase in the filler content, a higher number density of the fillers is observed, which leads to the possible presence of agglomerated phase as shown in Figures 11c and 11d. As reported frequently elsewhere, the existence of the agglomeration phase results in numerous mechanisms shown to be unfavorable to the interfacial load transfer at the filler/polymer matrix. Some factors adversely compete with the reinforcing mechanisms, which, accordingly, lead to the discrepancy between the model's prediction and the experimental elastic behavior of the composites as described in previous parts [35]. First, the agglomeration would result in the interparticle slippage against the shear forces upon loading because no bonding exists between the surface of fillers. Second, the lowered mechanical response of the fabricated part could be ascribed to the decrease in the effective surface area of the reinforcement phase due to the excess of fillers compared to the available polymer content and, thus, lower wettability of the HGMSs at the higher loadings [36, 37]. This observation could be better explained by the interconnected HGMSs surface where the fillers interspace is not enriched with polymer phase as clearly understood from Figures 11c and 11d (shown by arrows). Moreover, the higher number density of the fillers prohibits the formation of the interphase region with the assumed thickness due to the pinning effect of HGMSs sites [5, 38].

The exhibition confirms the agreement between the micro-mechanical model's prediction and the experimental elastic response resulting from the interfacial detachment at the interface of the HGMSs and polyester as understood from the SEM images.

5. CONCLUSION

Micro-composites of HGMS/polyester reinforced with 0-20 wt% of filler were fabricated and the effect of the presence of the interphase region and agglomeration on the overall elastic response of the composites was examined. The results indicated the existence of stiffer interphase as a result of perfect bonding at the contact of filler/polymer concerning the neat polyester leading to the increase in the modulus of the specimens upon the addition of fillers. It was further revealed that the elastic modulus of the composites is highly sensitive to the interphase modulus and thickness resulting in more accurate predictions with respect to the experimental values using the Halpin-Tsai, Tandon-Weng and the rule of mixtures when the interphase of a few microns up to 8 and modulus of 12-15 GPa are incorporated into the models. The findings confirmed the higher sensitivity of models to the interphase properties at lower filler content. It was shown through the morphological studies a perfect bonding of HGMS/polymer exists in the bulk specimens at loadings of lower filler ratio; however some levels of agglomeration leading to interconnected HGMSs and suppressed wettability of fillers were observed and accounted into the models to understand the effect of agglomerated phase on Young's modulus. The findings were linked to their severe number density of filler at higher HGMS content and, thus, models underestimation at such loadings. The study provided a methodology to give a better insight into the effect of interfacial interactions generated in thermosetting-based composites filled with HGMS reinforcements on the overall elastic response of the parts.

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Persian Abstract

چکیده

میکروگویچه های توخالی شیشه ای (HGMS) اخیراً در ساخت کامپوزیت های پلیمر با چگالی کم به دلیل سفتی بالای این تقویت کننده ها بهمراه وزن سبک آنها که به نوبه خود منجر به توسعه میکروکامپوزیت ها با خواص مهندسی شده و خواص مکانیکی بالا مورد استفاده قرار گرفته اند. برهم کنش های سطح فیلر/پلیمر مقدار انتقال نیرو و خواص بالک کامپوزیت ها را کنترل کرده که موجب خواص غیرقابل پیش بینی کامپوزیت ها با ذرات می گردد. با اینوجود، مدل های تحیلی مفید بمنظور تخمین خواص مکانیکی کامپوزیت های پایه HGMS با در نظر گرفتن اثر برهم کنش های سطح فیلر و آگلوموراسیون احتمالی مورد نیاز است. تاکنون مطالعاتی بر اساس مدل تحلیلی HGMS کامپوزیت های ترموست تقویت شده با این ذرات گزارش نشده در حالیکه بر نقش فاز میانی تشکیل شده در اطراف فیلر تاکید کرده باشد. این مطالعه بر ساخت میکرو کامپوزیت های پلی استر/HGMS تقویت شده با ۰ تا ۲۰ درصد ذرات تمرکز داشته که به دنبال آن مدل های میکرومکانیک صورت گرفته درحالیکه نقش ناحیه فاز میانی در اصلاح مدل ها تاکید می گردد. نتایج ارتباط قوی بین مشخصات فازمیانی و مدول یانگ در کامپوزیت ها را نشان داده که وابستگی ضخامت و مدول فاز میانی و سطح آگلوموراسیون و جدایش سطح مشترک ذرات HGMS را مشخص می کند. نتایج همچنین نشان داد که در نظر نگرفتن فاز میانی منجر به پیش بینی کمتر خواص شده که بیانگر وجود فاز میانی با سفتی بیشتر در اطراف HGMS بوده که مطابق با فرضیه تحقیق توسط تغییر در چگالی پیوندهای عرضی در پلیمر زمینه تعیین شده و توسط پاسخ مکانیکی نمونه ها تائید می گردد.