REVIEW

A REVIEW ON THE MECHANICAL PROPERTIES AND ENVIRONMENTAL IMPACT OF HOLLOW GLASS MICROSPHERE EPOXY COMPOSITES

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ISSN 0976-3104

ABSTRACT

The use of hollow glass microspheres (HGM) in composites is creating new opportunities in the composites industry. HGM consists of stiff glass hollow sphere filled with inert gas resulting in some specific properties like low weight, reduced dielectric constant, and reduced thermal conductivity. The typical wall thickness of HGMs is lying in between 0.5-2.0 μm and diameter 10-200 μm. On the basis of these properties, HGMs have been employed in the manufacturing of various composites for diverse applications. Therefore, the need of light-weight and high-strength materials for modern engineering applications may fulfill by the HGM composites. HGMs not only amending the properties but also ensuring the stability of molded articles by reducing viscosity and shrinkage of the composites. In this work, a comprehensive review on the properties of hollow glass microsphere (HGM) reinforced epoxy composites is presented. The summary of the paper shows the appropriateness of HGM-epoxy composites as an encouraging material for aerospace, automotive and cryogenic applications. The main focus area of the present study is the mechanical and environmental performance of the HGM composites.

THE IIOAB JOURNAL

KEY WORDS
Hollow glass microspheres, composite, epoxy, environment, mechanical properties

Received: 19 April 2018
Accepted: 26 May 2018
Published: 1 Oct 2018

INTRODUCTION

Composites are the materials produced employing two or more materials with different properties which on joining having assorted properties of both the constituents. The composite materials possess the indispensable properties borrowed from the best features of the two joined constituents. This material has the high strength to weight ratio and firm, fragile and other privative attributes of one material are concealed by the other one. In recent times, polymer composites are rapidly substituting the standard materials in various designing and product development in the engineering sector. Traditional materials and their modifications have a limited set of properties with a minor chance of improvements. On the other hand, composite materials can provide better quality control and required assortment of characteristics as per the application requirement. A proper blend of materials properties like specific modulus, a certain quality, low thickness etc. is doable in composites. The applications of composite materials are not limited to some particular areas. It also marked its presence in the space industry besides other common engineering industries owing to its sturdiness and potentials [1,2]. A thin walled sphere filled with inert gas or vacuum inside and a hard glass at the periphery are termed as hollow glass microspheres (HGMs). It is also named as hollow glass bead or glass bubbles or microballoons [3,4]. Syntactic foam is an example of HGM filled polymer composites [5]. The hollow glass microspheres have properties like feathery light, high specific area, inexpensive, low dielectric constant, flexible and noncorrosive. Additionally, the fragile microbeads are chemically strong, inflammable, unyielded and have excellent water resistance. The microspheres can be used in coatings, putty, artificial stones, emulsion explosives etc. [6–10]. It can also be used in oil and gas extraction industries as drilling fluid owing to its low density. It is also used to make superior optical glasses for visual resonators. Presently, the HGM reinforced composites especially syntactic foam are used to fabricate the gadgets employed for marine applications, deep sea exploration, ship bodies, helicopter and jet engine parts, detector instruments, noise reduction materials, sports merchandises. In the recent time, there is an increasing demand for the materials with low density and a high Young's modulus for various engineering applications. Therefore, HGM reinforced ploymer composites are gaining attention day by day. Foam glass owed Young's modulus and density of typically 1 GPa and 130 kgm^{-3}, respectively [11,12]. Moreover, it could be blown up to a full-size bubble of 2 mm diameter and frequently used to prepare HGM composite [12]. In the present article, the major focus of the study is the mechanical properties of the HGM reinforced composites reported in the various literatures. Additionally, the environmental and economic aspects of the same are also addressed in the article.

MECHANICAL PROPERTIES OF HGM COMPOSITES

In this paper, the mechanical properties of rarely reconnoitered HGM reinforced polymer composites are summarized. In the recent years, the structural properties of syntactic foams are studied by several researchers.

Compressive strength

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The compressive strength of HGMs filled epoxy composites are decreased with increasing volume fractions of the reinforcements [Fig. 1]. Additionally, higher density HGMs stimulates higher compressive strength of the composites. It can be marked from the plots [Fig. 1] that a stress plateau/valley exists in the plastic deformation region. The valley indicated the presence of broken HGMs owing to the compressive loading.

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ISSN: 0976-3104

The existence of stress plateau represents the high-energy absorbing capability of the composites. The main fracture mechanism of this type of composite is crushing and crumbling of HGMs.

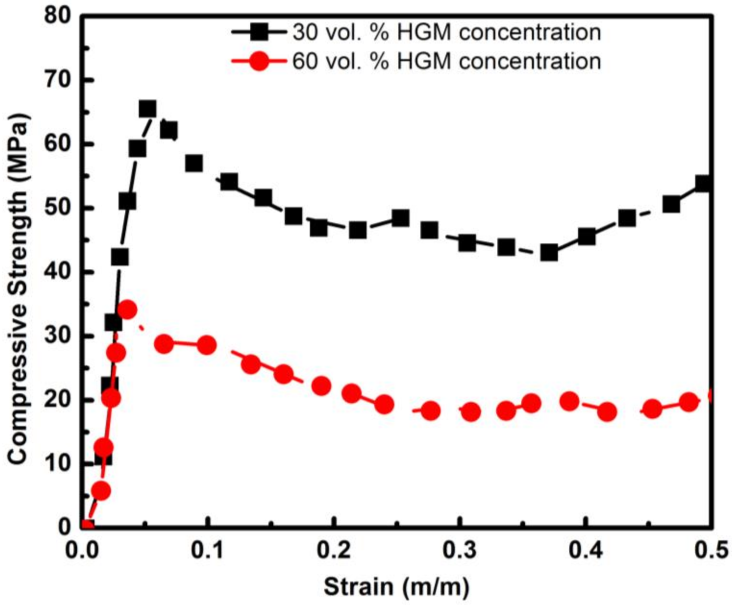

Fig. 1: Compressive stress-strain curve of epoxy matrix containing HGMs of 220 kg/m^3 in 60 and 30 vol% (Reconstructed from reference [13]).

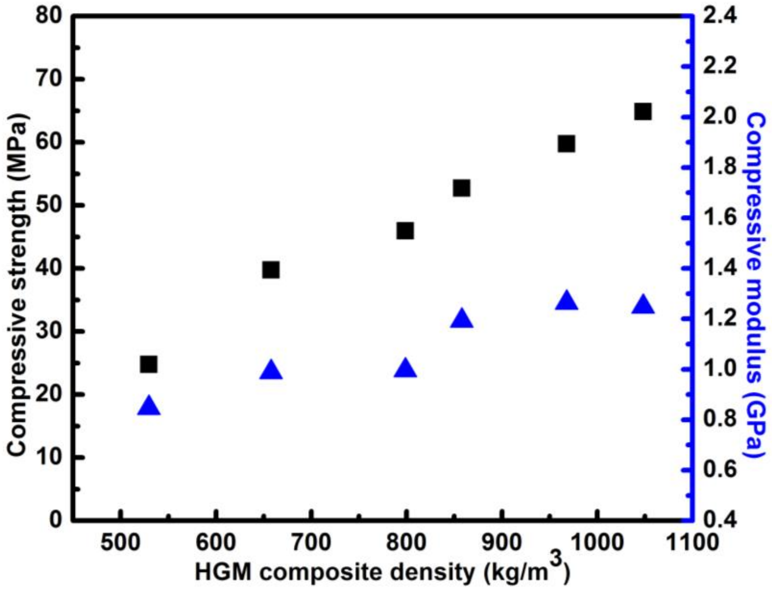

Fig. 2: Variation in compressive strength and modulus regarding the HGM composite density (Reconstructed from reference [13]).

The compressive strength/modulus versus density plots of HGM filled composites are recreated from [13] and shown in [Fig. 2]. This Figure is showing an increase in compressive strength with increasing density of reinforcements in composites. Other researchers have also reported the similar general trend [14–17]. Conversely, there is an insignificant change in compressive strength by changing the wall thickness and volume fraction of HGMs. Some papers depicted that broken HGM particles acted as solid fillers in the resin and improved the density of the composites. There is no specific trend reported in compressive modulus concerning density, and it was strongly dependent on the wall thickness of fillers. Thick walled HGMs increased the modulus whereas, thin-walled decreased the same [18]. [Table 1] depicts the maximum compressive strength of the various types of HGM polymer composites. When the HGM concentrations in the composites were less than or equal to 10%, the maximum compressive strength was reported. Higher HGM concentration might assist in improving Young's modulus in some composites. In

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some cases, it was observed that the addition of HGMs in polymer have no significant improvement in compressive strength or Young's modulus.

Table 1: The maximum compressive strengths of HGM based composites reported in literature

Matrix	Type and quantity of HGM	Young's modulus (MPa)	Compressive strength (MPa)	Ref.
Borosilicate glass	K46 (10%)	1388	19.0	[19]
Borosilicate glass	S60HS (10%)	1613	23.5	
Borosilicate glass	iM16K (10%)	1535	22.7	
Borosilicate glass	iM30K (10%)	1655	25.0	
Borosilicate glass	(383HS) FAC (10%)	1103	9.8	[20]
Araldite GY257	-	1648	104	[16]
Araldite GY257	K15 (10%)	1472	90.6	
Araldite GY257	S22 (10%)	1258	67.4	
Araldite GY257	S22 (20%)	1287	61.8	
Araldite GY257	K46 (10%)	1660	98.7	

Tensile strength

[Fig. 3] exhibits a representative stress-strain curve of HGM reinforced polymer composites under tensile loading. The plots were reconstructed from the data reported in reference [21]. The chosen composites contained 30 and 60 vol% HGMs of density 220 kg/m^3. The composites exhibited brittle type failure under tensile loading. Therefore, reduced tensile properties restricted the use of composite to the applications where loading conditions are the compressive type. It was reported that the strength might increase to some extent when HGMs-epoxy interfacial bonding is strong enough. The firm interfacial bonding assists in proper load sharing among the constituents. It can be examined from the [Fig. 4] that the tensile modulus of HGM composites improved with the incorporation of thick-walled HGMs. The high compressive strength of HGM composites is due to proper load distribution along with cracks closure ability during loaded conditions [22,23]. On the contrary, tensile load assisted in opening up the interfacial cracks resulted in poor strength. The tensile strength of the HGM composites is mainly dependent on polymer strength as well as fabrication defects. This problem might be solved by further reinforcing the HGM composites with glass fibers, carbon fibers, carbon nanotubes etc. [18, 24]. [Table 2] exhibits the maximum tensile strengths of HGM composites reported in the literature. It can be inferred from the table that low HGM concentration in the composites leads to better tensile strength. Apart from that untreated or medium size HGMs are exhibiting better strength than the other one. The yield strength of the composites did not follow any trend. In some cases, it was increased with the increment in HGM concentrations. On the other hand, the reverse is also true for some other instances.

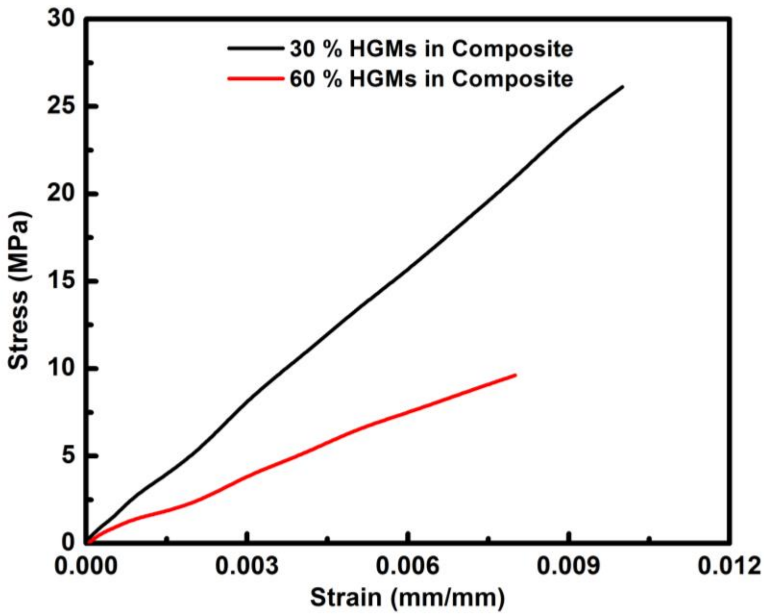

Fig. 3: Tensile stress-strain curves of HGM composites containing HGMs of 220 kg/m^3 density (Reconstructed from Reference [21]).

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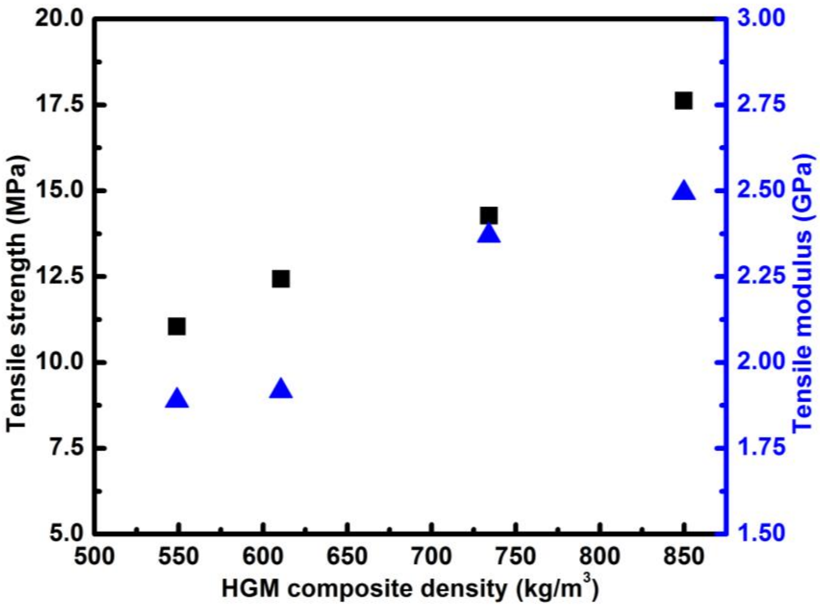

Fig. 4: Variation in tensile strength and modulus regarding the HGM composite density (Reconstructed from reference [21]).

Table 2: The maximum tensile strengths of various HGM based composites reported in literature

Matrix	Type and quantity of HGM	Young's modulus (MPa)	Tensile strength (MPa)	Ref.
Poly butylene succinate	-	330	34.7	[25]
Poly butylene succinate	T60 (05%)	355	32.2	
Poly butylene succinate	T60 (10%)	371	25.3	
Poly butylene succinate	T60 (15%)	439	23.7	
Poly butylene succinate	T60 (20%)	464	18.1	
Acrylonitrile Butadiene Styrene (ABS)	-	41	29.6	[26]
Acrylonitrile Butadiene Styrene (ABS)	TK70 (05%)	42	30.2	
Acrylonitrile Butadiene Styrene (ABS)	TK70 (10%)	41	30.7	
Acrylonitrile Butadiene Styrene (ABS)	TK70 (15%)	40	32.2	
Acrylonitrile Butadiene Styrene (ABS)	TK70 (20%)	38	32.3	
Polypropylene	-	33	29.3	[27]
Polypropylene	A-Glass 3000 (05%)	30	25.5	
Polypropylene	A-Glass 3000U (05%)	29	23.7	
Poly Vinyl Chloride (PVC)	-	48	33.8	[28]
Poly Vinyl Chloride (PVC)	HGMs-15µm (05%)	47	42.0	
Poly Vinyl Chloride (PVC)	HGMm-24µm (05%)	49	38.4	
Poly Vinyl Chloride (PVC)	HGMb-96µm (05%)	48	36.9	

Flexural properties

In flexural load-displacement curves, the load varies linearly with respect to displacement until failure occurs [29]. The flexural behavior of HGM filled composites were primarily determined by the tensile properties of the composites. The graphs of flexural strength/modulus versus density for thin and thick-walled HGMs were recreated from reference [15] and exhibited in [Fig. 5] and [Fig. 6]. The lower values of flexural strength and premature failure of HGM containing composites were articulated by the existence of matrix porosity and surface defects. Additionally, it was observed that the flexural modulus was slightly increased with increasing volume fraction of thin-walled HGMs. Conversely, incorporation of the higher amount of thick-walled HGMs reduced the flexural modulus of the composites significantly as shown in [Fig. 6]. Therefore it can be expounded that the surface roughness and air voids in the matrix hampered the flexural strength of the composites in a great extent and leads to premature failure in the HGM filled composites [18].

From [Table 3], it can be reiterated that in some instances when low-density HGM was present in relatively higher amount exhibited high flexural modulus and strength than its counterparts. Also, the improvement reported in the flexural strength is marginal in some composites, and there is deterioration in flexural strength than the matrix in some other composites. Therefore, the addition of HGMs imposed positive changes regarding mechanical properties in selective epoxies only.

IIOAB JOURNAL
ISSN: 0976-3104

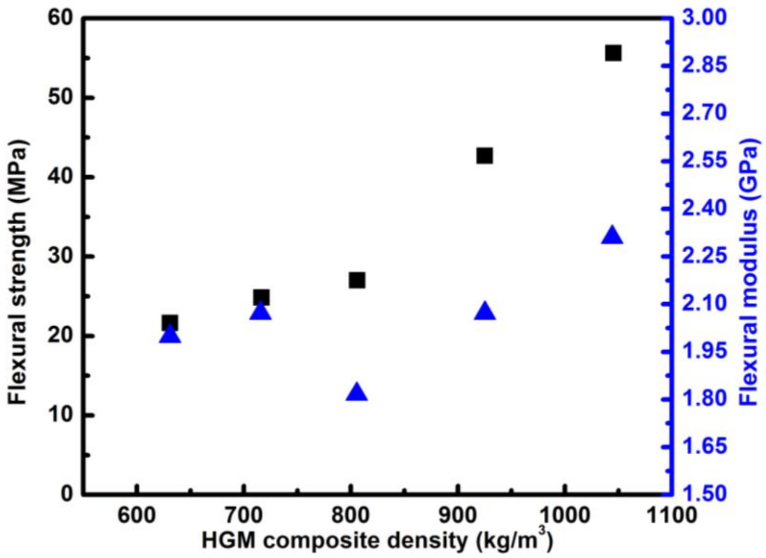

Fig. 5: Variation in flexural strength and modulus regarding the HGM composite density containing HGMs of 150 kg/m³ density (Reconstructed from Reference [15]).

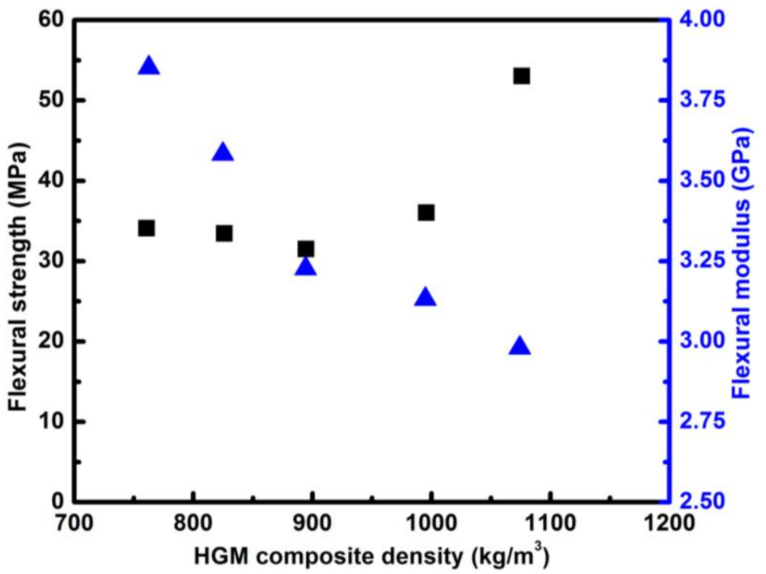

Fig. 6: Variation in flexural strength and modulus regarding the HGM composite density containing HGMs of 460 kg/m³ density (Reconstructed from Reference [15]).

Table 3: The maximum flexural strengths of various HGM based composites reported in literature

Matrix	Type and quantity of HGM	Flexural modulus (MPa)	Flexural Strength	Ref.
Vinyl ester	-	3229	103.5	[30]
Vinyl ester	HGM220 (50%)	3523	58.0	
Vinyl ester	HGM220 (60%)	3634	54.1	
Vinyl ester	HGM320 (30%)	2864	46.2	
Vinyl ester	HGM320 (60%)	3571	34.3	
Vinyl ester	HGM460 (30%)	2371	24.4	
Vinyl ester	HGM460 (60%)	3753	25.2	
Acrylonitrile Butadiene Styrene (ABS)	-	41	35.5	[26]
Acrylonitrile Butadiene Styrene (ABS)	TK70 (05%)	42	36.9	
Acrylonitrile Butadiene Styrene (ABS)	TK70 (20%)	38	39.0	

BRIEF OVERVIEW ON MECHANICAL PROPERTIES OF HGM COMPOSITES

Fu et al. [31] reiterated that the particle size, bonding capability and load are the deciding factors for stiffness, strength and toughness in particulate composites. Stiffness basically depends on load bearing capability of reinforcement rather than bonding whereas, toughness depends on bonding strength. Kinloch et al. [32] found that the mechanical and physical properties of thermosets or thermoplastic resins were

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ISSN: 0976-3104

significantly improved using HGMs as reinforcement. As per the investigation of Sahu and Broutman [33] the material with low melting viscosity or isotropy can be produced by associating glass beads into polyester and epoxy resins. They studied the mechanical behaviour along with fracture analysis of the same too. Mallick and Broutman [34] investigated the strength and fracture behavior of HGM filled epoxy composites. They did not observed better strength in HGM composite since critical crack size is not affected by the interparticle distance. Swetha and Kumar [16] prepared HGM composites of various densities using stir casting process and studied their mechanical properties. They found that the strength of the composite is high when the density of the HGM is high in the composite. Additionally, energy absorption capacity of the composite is increased till the addition of 40% HGM. The mechanical properties of HGM composites consisting HGMs of different diameter are investigated by d'Almeida [35]. He represented rupture of the microspheres as a single damage parameter. This investigation concluded that the ratio of wall thickness and mean diameter of the microspheres is the useful fabrication parameter. Yung et al. [36] studied the dielectric properties of HGM (0-51.3 vol.%) filled epoxy composites. They observed that the dielectric constant and dielectric losses decreased with increased HGM content. Apart from that the coefficient of thermal expansion (CTE) and the glass transition temperature (Tg) of the composites are enhanced significantly. Similar results were also observed by Park et al. [37] in HGM (0-2 wt.%) filled diglycidyl ether of bisphenol A (DGEBA) composites. They also found that the mechanical and interfacial properties of the HGM filled DGEBA are considerably higher than the neat DGEBA. Kim and Khamis [38] studied the fracture and impact behaviours of HGM (0-0.65 vol.%) filled epoxy resin composites. The addition slightly reduced the impact force and marginally increased the specific flexural modulus at the expense of specific fracture toughness and specific flexural strength. Hu et al. [39] studied the influence of broken HGMs on density, mechanical property and thermal conductivity in HGM filled silicon rubber composites. They found that these properties are enhanced even after the HGMs are broken inside the composites. Li et al. [25] investigated the morphology, melting/crystallization behavior, mechanical properties and thermal stability of HGM filled poly(butylene succinate) (PBS) composites. They observed that the crystallization rate and thermal stability of the composites were increased without hampering the crystalline nature. Additionally, the storage modulus of the composite is increased too. Huang and Li [40] developed a model for randomly distributed glass microspheres in epoxy using finite element modelling (FEM). They studied the elastic behavior and failure mechanism of different volume fractions of HGM filled composites. In this investigation they found that adjoining of adjacent microcracks along with voids due to broken HGMs are forming macro-cracks and propagating in a particular direction in the composite. Wang et al. [41] fabricated HGM-bisphenol A dicyanate ester (BADCy) composite using mechanical mixing followed by a stepped curing process. They found improved mechanical properties such as impact strength, flexural strength, flexural modulus and storage modulus without compromising its thermal properties. Im et al. [42] found that the HGM filled thermoplastic polyurethane composite have superior mechanical properties and a lesser amount of water adsorption properties for sonar encapsulating materials. Li et al. [43] studied the strain rates sensitivity of HGM containing epoxy syntactic foams in finite element stress analysis. As per the study, failures occurred in the composite due to crushing of glass microspheres and shear cracking of the epoxy matrix. Higher strain rate leads to the higher microsphere cracking and higher matrix debonding. Liang [26] studied the tensile and flexural properties of HGM filled ABS copolymer at room temperature. Increase in Young's modulus was observed till 5% volume fraction of HGMs. Till 15 vol.% HGM, ultimate tensile strength increased and remain constant afterwards. The tensile and impact strength of the HGM filled PVC composites were investigated by Liang [28]. He reported slightly lower yield strength with increasing HGM volume fraction in the PVC. On the other hand, somewhat better UTS of the composites was observed when volume fraction of HGMs lying in between 0–20%. Ferreira et al. [44] studied the mechanical behavior of hybrid composites prepared by HGMs and short fibre reinforcements. In terms of specific values, both flexural and compressive stiffness and impact absorbed energy increase with microsphere content.

ENVIRONMENTAL IMPACT OF HGM COMPOSITES

The composites fabricated using natural fibers are attracting attention among scientific and industrial communities owing to growing environmental concerns and more stringent norms [45]. These composites have several advantages over synthetic fiber composites such as low abrasion in processing, lower environmental impact, low cost, excellent specific properties and reasonable mechanical properties [46,47]. These fibers are generally employed in thermosets (e.g. polyester etc.) and thermoplastics (e.g. polypropylene, polyethylene etc.) composites. Among several thermoplastic polymers, Polypropylene (PP) is one of the commonly used semicrystalline thermoplastic. It has a set of attractive properties like high melting/service temperature, good chemical strength, higher stiffness, excellent durability, lightweight, easy to process and cost-effective [48,49]. The applications of neat PP are limited despite having many attractive properties [50]. The drawbacks might be subdued if vegetal, inorganic or mineral reinforcements are employed in PP. Likewise, other plastics whether it is thermoset or thermoplastic possess excellent properties but barely used in the unadulterated form for any structural applications. The reinforcement is always required to make it suitable for such type of applications. The composite development revolves around the natural or recyclable fillers are always encouraged for the betterment of human civilization. Hollow glass microspheres (HGM) are inorganic and spherical mineral fillers [39]. They are finely dispersed, free-flowing thin-walled powder particles with good thermal properties and low density [4]. As discussed in the previous section, the mechanical properties especially compressive strength of HGM containing composites are comparatively better than the neat epoxies in some instances. Additionally, HGMs are inorganic, recyclable and did not harm the environment. Therefore, HGM containing composites are considered as environmental friendly material, and regular attempts are going on for improving its properties worldwide.

CONCLUSION

HGM composites are having lower densities than the neat epoxies owing to the presence of hollow glass microbeads. The density of the composite will further go down if the thin-walled microbeads concentration is increased in it. The concentration of the filler contents is the deciding factors for the various properties of the composites, especially the mechanical one. The inferences drawn from the review indicates that lower concentration of the microspheres in most of the epoxy composites exhibited improved compressive strength, unlike yield strength. A similar trend was also reported for the tensile strength of HGM composites. However, the values of yield strength are more or less scattered and not following any specific pattern. It has a strong dependency on the epoxies as well as types of HGMs used. A relatively higher HGM concentration in the composites exhibited better flexural strength and modulus than its counterparts. But the improvement in the flexural strength is insignificant. In some instances, the mechanical properties are even weaker than the virgin matrix. Therefore, the choice of polymer and type of HGM should be as per the focused application area. The glass beads could be prepared from waste materials or as a byproduct of some critical industrial processes along with very high recyclability potential. Therefore, the use of glass microbead composite material is not imposing any potential harm to the ecosystem. It could be considered as a green material as per grown environmental concerns. It has a potential to replace many metals or composites in various industries. For this, extensive research efforts in multiple aspects are needed to scale the gap between HGM composites and conventional materials.

CONFLICT OF INTEREST

There is no conflict of interest

ACKNOWLEDGEMENTS

Authors wish to thank Dr. Anil from Accendere Knowledge Management Services, CL Educate, New Delhi for his contribution in manuscript preparation

FINANCIAL DISCLOSURE

None

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