

## Research Article

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# Processing of Hollow Glass Microspheres (HGM) filled Epoxy Syntactic Foam Composites with improved Structural Characteristics

<https://doi.org/10.1515/secm-2021-0011>

Received Aug 10, 2020; accepted Nov 30, 2020

**Abstract:** The objective of this work is to improve the structural characteristics of hollow glass microsphere (HGM) filled epoxy syntactic foam composites with little voids content and improved HGM dispersion in the composite.

A modified degassing technique has been introduced during resin casting process of the HGM filled syntactic foam composites. The effect of HGM content volume fractions (5-25%) on the degassing techniques was examined. The syntactic foam composites were characterized by analysing structural morphology using Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), and density measurements (theoretical and experimental).

Less than 5% void content has been achieved in this study. This resulted in improved tensile and dynamic mechanical properties (DMA).

**Keywords:** Hollow glass microsphere (HGM), syntactic foam, tensile properties, degassing, fracture mechanism

## 1 Introduction

Syntactic foam composites forms cellular structure and are entirely different from the commonly used foams such as polyvinyl chloride (PVC) and poly urethane (PU), geopolymer foam because of their isotropic nature and randomness of their microstructure [1, 2]. Thermoset resins such as epoxy are commonly used as binder or matrix and are widely used in structural applications and in adhesives. Hollow glass microspheres (HGM) are one of the common microballoons used for the development of syntactic foam.

This HGM is embedded in a polymer material to form syntactic foam composites. The hollow particles may sometimes be in the form of metal, carbon, ceramics, or polymer [1, 3]. Furthermore, thermoset resins such as epoxy are generally used as a binder for combining of microballoons, which produces composite material with improved properties. Composite material developed from this combination is widely used in structural applications due to its low cost, excellent specific strength, and ease of processing [4, 5].

There is a need to produce a lightweight material for the structural application that is economically friendly and fire retardant. Syntactic foams have been studied to fit into this aspect because they are known to be of good advantages for construction purposes due to their reduced weight thus making them applicable as core materials for structural and marine applications [6]. The use of HGM and their synergistic effect as filler in syntactic foam composites gives lightweight, resistances to bending, compressive stress and low thermal conductivity exceeding other foams [5–7].

Syntactic foam is a composite material which exhibits the same density before and after curing. Such low-density syntactic foam is employed in composite tooling for the manufacturing of aerospace structures [1, 5, 6]. Numerous studies have reported significant improvement in the mechanical properties of syntactic foam by changing the amount of filler particles. It was also noted that the properties of the resulting syntactic foam composites are enhanced by a strong interfacial bonding between the HGM filler and the polymer matrix. They are known to possess good impact behavior, low density, excellent compressive, flexural, hydrostatic, corrosive and tensile strength [5, 6, 8–16]. The attention of scholars has recently been drawn to its functionality and usage due to its low moisture absorption, good energy absorption, low weight and high specific strength which has made it applicable to most construction, transportation, machinery and aerospace industries [17–20]. Also, hollow glass microsphere syntactic foams are good material for electronic purposes because of their excellent dielectric constant [Dinesh Pinisetty], their excellent temperature resistance nature makes them suitable for

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buoyancy materials such as in the deep sea exploration or underwater resistance operation [21], they equally possess good heat-resistance ability when mixed with an inorganic adhesive binder matrix which is applicable for the nose cap of NASA/Marshall Space Flight [22], likewise, they can be suitable for high-temperature materials that are lightweight in nature for structural purposes [23].

One of the important parameters that are not properly studied in HGM filled syntactic foam composites is void content. Void content in HGM foam composites plays a very important role on the tensile and dynamic properties. Even at 2.5% increased in void content could reduce the tensile and DMA properties by 15-20%. Maximizing the void content in HGM syntactic foam composites is important as the HGM itself is a hollow void structure. Therefore the presence of void in composite further completes and induces and induce detrimental properties. The literature study also shows that HGM filled foam composites resulted in up to 10% void content, in addition of inherent HGM void. Most of the voids occurred during composite processing resulting in poor tensile strength. Therefore, a more efficient way of reducing the void content in the processing method of syntactic foam was adopted in this present work as the main area of interest. This study focusses on reducing the void content by a modified resin casting method. In this method, a degassing procedure is introduced as an intermediate processing step to eliminate voids before using of epoxy resin which resulted in good mechanical and thermal properties of the syntactic foam which is the novelty of the study. Additionally, the resultant composite was tested for density (experimental and theoretical), tensile and DMA properties and were discussed.

## 2 Experimental procedures

### 2.1 Materials

Epoxy resin LR 20 and hardener LR 281 were purchased from AMT composites, South Africa. The Hollow Glass Microballoons (HGM-T60) filler was procured from Anhui Elite Industrial Cop, Limited, Hong Kong Elite Industrial Group Limited, China (T60 trade name). The density of the HGM is  $0.6\text{g/cm}^3$  with 10-60 $\mu\text{m}$  particle sizes.

### 2.2 Processing

The processing for the fabrication of epoxy resin (EP) / HGM filled foam composites (EPT60) was carried out in two steps:

#### Step 1: Mixing and Degassing

Syntactic foam composites were fabricated by mixing (mechanically) epoxy resin and the desired volume fraction of hollow glass microspheres in a glass beaker. The mixture was thoroughly stirred for 30 min to obtain uniform slurry and a homogenous solution in order to reduce agglomeration. The viscosity of the matrix (epoxy resin) was lowered by heating it up to  $60^\circ\text{C}$  for 1 hour. Thereafter, hardener was gently added to it and mixed thoroughly. During mechanical mixing of composites, air bubbles were entrapped which led to open cell structure porosity called voids [24]. The epoxy-resin and hardener ratio were 10:3 and hollow glass microsphere volume fraction was added for each sample preparation. The volume fraction of hollow glass microspheres was varied from 5 to 25%.

#### Step II: Curing

This mixture was cast in the silicon mold made from mold max “30-part A” and mold max “30 STD catalyst part B” purchased from AMT composites as well. Silicon grease was used to coat the mold surfaces to ensure easy removal of syntactic foam slab after curing. The syntactic foam slab was cured at room temperature ( $27^\circ\text{C}$ ) for 24 hours and then post cured at  $80\pm 5^\circ\text{C}$  for 4 h. Tensile test specimens were cut from the cured slabs using a fixed bench blade tile saw.

### 2.3 SEM, TEM and DMA

Scanning Electron Microscopy (SEM) was used to investigate the fracture surfaces and morphology of the syntactic foam composites. The fractured tensile and flexural specimens were examined using a Zeiss EVO 1 HD 15 Oxford instrument X-max scanning electron microscope (SEM). The specimens were gold coated before the SEM was conducted because the syntactic foams are not conductive and there is need for the flow of electron for the fracture images to be seen properly, this was done by Quorum Q 150R ES machine for 6 minutes. The Transmission Electron Microscopy (TEM) was conducted using higher resolution (HR-TEM) Joel 2100, from Japan.

The Thermogravimetric Analysis TA (Model Q800 V20.6) instrument was used to perform the Dynamic Mechanical Analysis of the syntactic foam composites. The specimens used were measured according to ASTM D4065 standard 56mm x 12mm x 3mm in length, width and thickness respectively. The tests were run in a 3-point bending mode on a support span length of 50mm with heating temperature ranging from  $20^\circ\text{C}$  to  $200^\circ\text{C}$  at a frequency of 10Hz, rate of  $3^\circ\text{C/min}$  and amplitude of 20  $\mu\text{m}$ . Minimum tempera-

ture lag occurred between the sample and the furnace environment as a result of maintaining the parameters throughout the test.

## 2.4 Tensile Testing, and Density measurement

Specimens for tensile and flexural properties were prepared in accordance to ASTM D 3039 and ASTM D790-02 test standard specifications respectively. MTS 793 servo-hydraulic machine with a load cell of 100 KN and a test speed of 2 mm/min was used. The length, width and thickness for the tensile samples were 250mm × 25mm × 3mm respectively. Five specimens were tested from each syntactic foam sample and their mean values were used to calculate the final results.

## 3 Results and Discussion

### 3.1 Structures and Morphology

#### 3.1.1 Scanning Electron Microscopy (SEM)

Figure 1 (a and b) shows the nature of the HGM and structure under scanning electron microscopy (SEM) at different magnifications. Different sizes of the HGM were seen through the microscopy ranging from 10µm to 60µm. The image output was adjusted by using gold to sputter the surface before the scanning process. The effect of these sizes

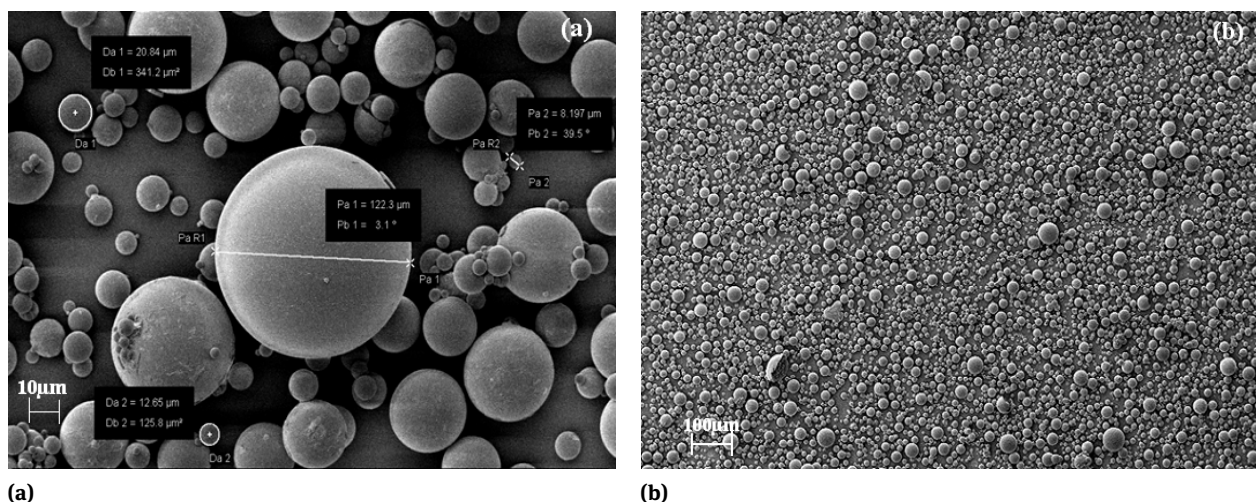
was seen during the mixture with matrix resin at various percentage concentrations (5%-25%) designated as (0.5%-2.5%) in the TEM and SEM images.

#### 3.1.2 Transmission Electron Microscopy (TEM)

The distribution of HGM sizes inside matrix resin was further investigated using transmission electron microscopy (TEM). The TEM images in Figure 2 (a-e) represent the distribution of HGM in the epoxy resin at different specifications. There was an uneven distribution of HGM filler in the matrix resin as the concentration increases from 5% to 25%, which affected the mechanical properties of the syntactic foam composites and contributed to an increase in the porosity level of the EPT60 syntactic foam composites. Figure 2 (a and b) shows little or no agglomeration of the HGM due to lower concentration. The agglomeration increased as the concentration increased as seen in Figure 2 (c-e) with much clustering and percolation of HGM at 25% (Figure 2e).

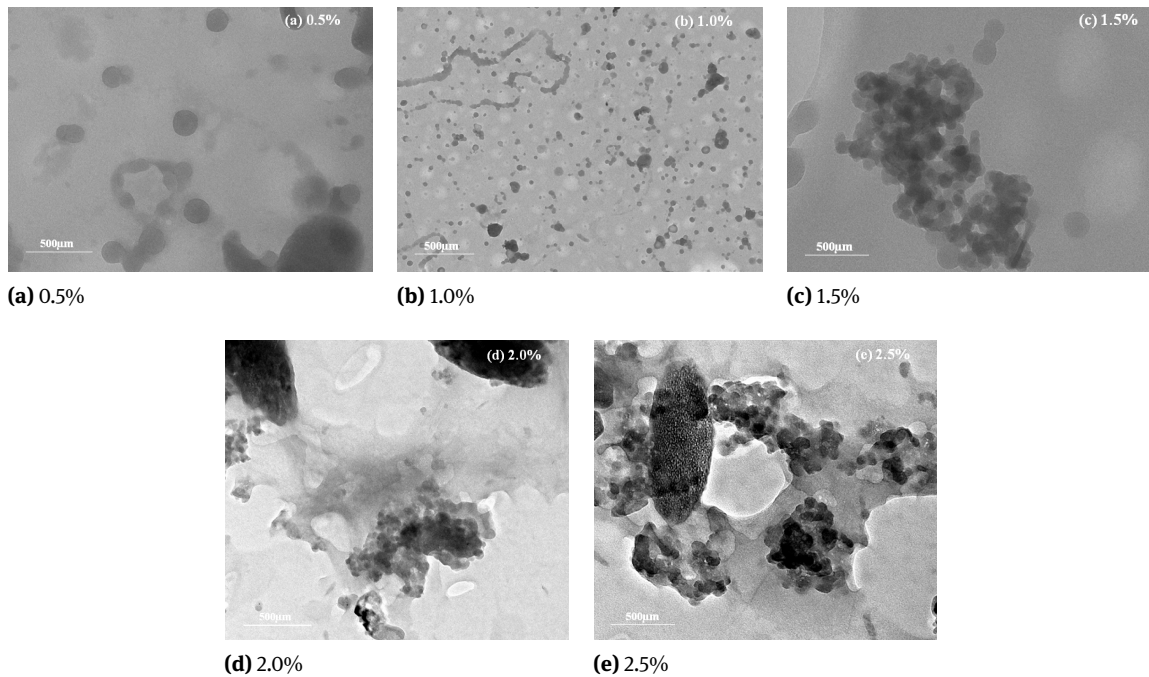
### 3.2 Density and Void Measurement of Syntactic Foam Composite

Based on the rule of mixture, the theoretical density values for HGM filled syntactic foam composites can be obtained. Theoretical density values are calculated using equation (1), while measured density was determined using ASTM C271-94, with specimen specification of  $25 \times 25 \times 12.5 \text{ mm}^3$ . Three specimens each were cut and measured accordingly with the mean value recorded. The value for the void volume



**Figure 1:** Microstructure of Hollow Glass Microsphere: (a) and (b) are Scanning Electron Microscopy (SEM) images showing different magnification of HGM at 10µm and 100 µm respectively.





**Figure 2:** Transmission electron microscopy (TEM) images showing the clustered particles of the hollow glass microspheres dispersed in the syntactic foam composite at each volume fraction.

fraction also known as matrix porosity was calculated by estimating the difference between the theoretical density ( $\rho_t$ ) and the measured density ( $\rho_m$ ) using equation (1).

$$\delta = \frac{\rho_t - \rho_m}{\rho_t} \cdot 100 \quad (1)$$

The theoretical density ( $\rho_t$ ), the measured density  $\rho_m$  and the void volume fraction ( $\delta$ ) are presented in Table 1. The density of the EPT60 syntactic foam composite increases compared with the HGM filler due to proper interaction between the constituent elements but shows decreasing values for both measured and theoretical densities as the HGM volume fraction increases. This corresponds to the report of Yingjie *et al.* [25] where the values of the measured and theoretical densities decrease with increasing HGM content. The measured densities were found to be lower

than the theoretical densities [25–28] for all the weight fractions of HGM. Void fraction and HGM porosities have been studied to be undesired and should be kept to the minimum level because its presence can lead to modulus and strength reduction in syntactic foams [29]. The void fraction which is higher than the measured and theoretical densities of the syntactic foams can also be responsible for an increase in moisture absorption of the syntactic foam composites [17, 30] which can cause diffusion in the foam specimen structure and get accumulated in the matrix porosity region.

Table 1 shows the values of the measured densities and theoretical densities as it relates to the void fraction in the syntactic foam filled with HGM composites. The void fraction increased with an increase in the filler content,

**Table 1:** The density and porosity (void) values of syntactic foam composites.

Specimen name	Volume fraction of HGM (%)	Density of the composite (g/cm <sup>3</sup> )		Void fraction of Composite (%)
		Theoretical	Measured	
EPT60-1	0	1.150	1.120	2.61
EPT60-2	5	1.145	1.122	2.01
EPT60-3	10	1.140	1.110	2.63
EPT60-4	15	1.135	1.105	2.64
EPT60-5	20	1.131	1.096	3.09
EPT60-6	25	1.126	1.075	4.53

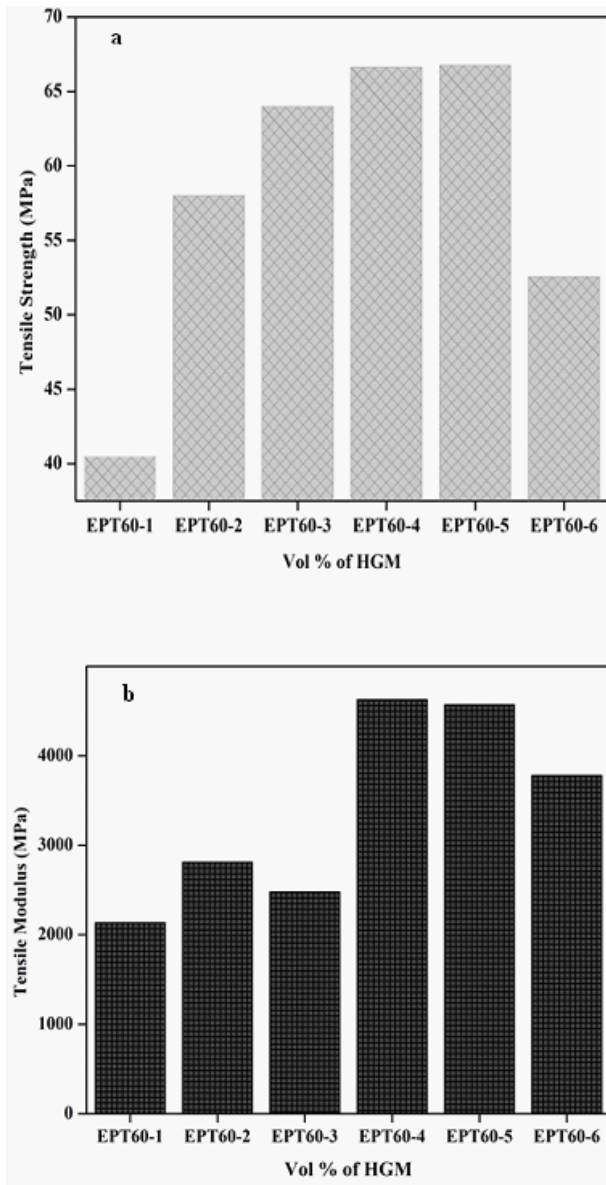
**Table 2:** Comparison of density and porosity (void) values of syntactic foam composites based on the present work and existing literatures.

S/no	Matrix type	Composition of HGM and type	Theoretical density ( $\rho_t$ ) (g/cm <sup>3</sup> )	Measured density ( $\rho_m$ ) (g/cm <sup>3</sup> )	Void fraction $\delta$ (%)	Reference
1	Epoxy resin LR20	—	1.150	1.120	2.61	Present work
2	Epoxy resin LR20	5% HGM-T60	1.145	1.122	2.01	
3	Epoxy resin LR20	10% HGM-T60	1.140	1.110	2.63	
4	Epoxy resin LR20	15% HGM-T60	1.135	1.105	2.64	
5	Epoxy resin LR20	20% HGM-T60	1.131	1.096	3.09	
6	Epoxy resin LR20	25% HGM-T60	1.126	1.075	4.53	
7	Vinyl ester VE	—	0.116	-	-	[33]
8	Vinyl ester VE	30% HGM-K46	0.951	0.905	4.8	
9	Vinyl ester VE	40% HGM-K46	0.881	0.798	9.3	
10	Vinyl ester VE	50% HGM-K46	0.811	0.811	0.0	[25]
11	Vinyl ester VE	60% HGM-K46	0.740	0.655	11.4	
12	Epoxy resin E51	47% HGM-S38	0.593	0.536	9.6	
13	Epoxy resin E51	49% HGM-S38	0.580	0.522	10.0	
14	Epoxy resin E51	51% HGM-S38	0.569	0.509	10.5	
15	Epoxy resin E51	53% HGM-S38	0.558	0.498	10.7	
16	Epoxy resin E51	55% HGM-S38	0.547	0.488	10.8	
17	Epoxy resin E51	57% HGM-S38	0.536	0.474	11.6	[34]
18	Epoxy resin DER 332	—	1.165	1.160	0.4	
19	Epoxy resin DER 332	30% HGM-S22	0.881	0.884	0	
20	Epoxy resin DER 332	50% HGM-S22	0.692	0.656	5.3	
21	Epoxy resin DER 332	30% HGM-K46	0.953	0.918	3.6	[31]
22	Epoxy resin DER 332	50% HGM-K46	0.812	0.823	0	
23	Epoxy resin Epon 8008 & 1031	10% HGM-S60	1.275	1.240	2.71	
24	Epoxy resin Epon 8008 & 1031	20% HGM-S60	1.216	1.180	2.96	
25	Epoxy resin Epon 8008 & 1031	30% HGM-S60	1.153	1.112	3.56	
26	Epoxy resin Epon 8008 & 1031	40% HGM-S60	1.087	1.056	2.85	
27	Epoxy resin Epon 8008 & 1031	50% HGM-S60	1.017	0.985	3.15	
28	Epoxy resin Epon 8008 & 1031	60% HGM-S60	0.943	0.885	6.15	

Figure 2(c-d). The more the concentration of the HGM content, the more the void volume fraction. The void volume fraction changes from 2.6% at EPT60-1 to 4.53% at EPT60-6 of the syntactic foam. This corresponds to an earlier report by Zhu *et al.* [31] where the void content also increased with an increasing volume fraction of HGM, which is as a result of surface infiltration between the matrix and the filler content. Also, it was an indication that there are heterogeneity and entrapment of air bubbles during the mechanical mixing of the constituent elements [32]. Comparison between

densities of the present work and some existing literature was also reported in Table 2.

Table 2 relates the void fraction of the present work with some existing literature, the void fraction value reduced compared to most previous works due to the degassing method used in its processing but increased with increasing HGM volume fraction [25, 31]. This is as a result of good dispersion and adhesion of HGM filler in the matrix resin and smaller volume fraction of HGM used.



**Figure 3:** a) Tensile strength relationship between the neat epoxy resin and EPT60 syntactic foam and b) Tensile modulus relationship between the neat epoxy resin and EPT60 syntactic foam.

### 3.3 Tensile properties

The tensile properties of the EPT60 syntactic foam composites are shown in Figure 3 and Table 3. Figures 3a and 3b, report the tensile strength and modulus relationship between the neat resin epoxy and the EPT60 syntactic foam respectively. With the addition of HGM, tensile strength and modulus increased at varying percentage volume fractions, highest tensile strength was at EPT60-5 (66.7 MPa), which is an increase of about 65% compared to the neat epoxy resin and highest tensile modulus at EPT60-4 (4.5 GPa). The tensile strength of syntactic foam increased consistently with an increase in the HGM content until EPT60-5 which correspond to the report by Nityanshu *et al.* and Rupan *et al.* where tensile strength of the Polypropylene increased upon the inclusion of HGM as a result of improved interfacial adhesion with the matrix [15, 16] before it declined at EPT60-6 due to increased void content caused by the agglomeration and reduced resin content resulting in low bonding and load transfer stability between the filler and the resin as shown in Figure 3a. The interfacial strength between the microsphere and the matrix is very important for the syntactic foam composites as it affects its overall tensile strength. Although, some previous studies [28, 32] reported a decline in the tensile strength by 60-80% as compared to neat epoxy. They stated that the increase in HGM volume fraction reduced the tensile strength because as the volume fraction of the epoxy resin in the material decreased, the strength of the composites also decreased due to higher range of microballoons in the composites structure. It was however observed that the tensile modulus at EPT60-3 decline as a result of high agglomeration (Figure 2c and 3b) between the filler and the matrix as a result of mixing which caused an early brittle failure at that point. The increase in modulus of EPT60 syntactic foam composites over that of neat epoxy resin confirms the report of Gupta *et al.* [35] that the syntactic foam has a higher Young's modulus in several compositions than the neat

**Table 3:** Tensile strength of syntactic foam composites.

Materials	Volume fraction	Tensile Modulus		Tensile Strength		Tensile Strain	
	HGM (%)	MPa	% increase	MPa	% increase	(mm/mm)	% increase
EPT60-1	0	2135	0	40.47	0	0.0297	0
EPT60-2	5	2813	31.76	57.97	43.24	0.0409	37.71
EPT60-3	10	2480	16.16	63.97	58.07	0.0277	-6.73
EPT60-4	15	4628	116.77	66.60	64.57	0.0219	-26.26
EPT60-5	20	4570	114.05	66.73	64.89	0.0256	-13.80
EPT60-6	25	3782	77.14	52.55	29.85	0.0217	-26.94

**Table 4:** Comparison of tensile properties of syntactic foam based on present work and existing literatures.

S/no	Matrix type	Composition of HGM and type	Tensile strength (MPa)	Tensile modulus (MPa)	Reference
1	Epoxy resin LR 20	—	40.47	2135	Present work
2	Epoxy resin LR 20	5% HGM-T60	57.97	2813	
3	Epoxy resin LR 20	10% HGM-T60	63.97	2480	
4	Epoxy resin LR 20	15% HGM-T60	66.60	4628	
5	Epoxy resin LR 20	20% HGM-T60	66.73	4570	
6	Epoxy resin LR 20	25% HGM-T60	52.55	3782	
7	Polybutylene succinate	—	34.7	330	[36]
8	Polybutylene succinate	5% HGM-T60	32.2	355	
9	Polybutylene succinate	10% HGM-T60	25.3	371	
10	Polybutylene succinate	15% HGM-T60	23.7	439	
11	Polybutylene succinate	20% HGM-T60	18.1	464	[12]
12	Epoxy resin DER 332	—	57.2	2752	
13	Epoxy resin DER 332	30% HGM- K46	25.1	3700	
14	Epoxy resin DER 332	40% HGM-K46	20.7	3641	
15	Epoxy resin DER 332	50% HGM-K46	15.6	3615	
16	Epoxy resin DER 332	60% HGM-K46	12.8	3491	
17	Epoxy resin DER 332	—	26.0	2700	[34]
18	Epoxy resin DER 332	30% HGM-S22	17.0	2200	
19	Epoxy resin DER 332	50% HGM-S22	16.5	2100	
20	Epoxy resin DER 332	30% HGM-K46	21.5	3200	
21	Epoxy resin DER 332	50% HGM-K46	23.0	4100	[37]
22	Acrylonitrile Butadiene Styrene (ABS)	—	29.5	1820	
23	Acrylonitrile Butadiene Styrene (ABS)	5% HGB-TK70	30.2	1840	
24	Acrylonitrile Butadiene Styrene (ABS)	10% HGB-TK70	30.8	1860	
25	Acrylonitrile Butadiene Styrene (ABS)	10% HGB-TK70	32.2	1875	
26	Acrylonitrile Butadiene Styrene (ABS)	20% HGB-TK70	32.2	1900	

The strain rate of the neat resin is substantially higher than that of the syntactic foam except for EPT60-2 with the lowest HGM volume fraction in the composites while that of other syntactic foam from EPT60-3 to 6 is lower than the neat epoxy. This resulted from their higher void content in the composites due to the larger percentage of HGM volume fraction.

Table 4 shows the compared tensile properties of the present work with some existing literature which observed that the tensile and modulus properties of the present work improved compared to the existing literature due to the degassing method of preparation and the small void fraction of the syntactic foam which resulted in good matrix/filler interaction. High volume fraction of HGM can result in ag-

glomeration leading to low tensile strength and modulus in the syntactic foam composites [36]. Also a poor interfacial bond between the matrix and the microballoons resulting from a high-volume fraction of HGM used can be responsible for the reduction in tensile strength of the syntactic foam [28]. However, good interfacial adhesion between the matrix and HGM volume fraction in the syntactic foam was responsible for the increase in tensile strength in the present work as shown in Table 3. Meanwhile, Ji-Zhao Liang *et al.*, Rupam *et al.*, and Naresh and Siva [14, 16, 37] reported a similar result to the present work, where tensile strength increases with increase in filler concentration. The effect of the good interfacial adhesion between the matrix and



the HGM was shown through the fracture mechanism in Figure 4.

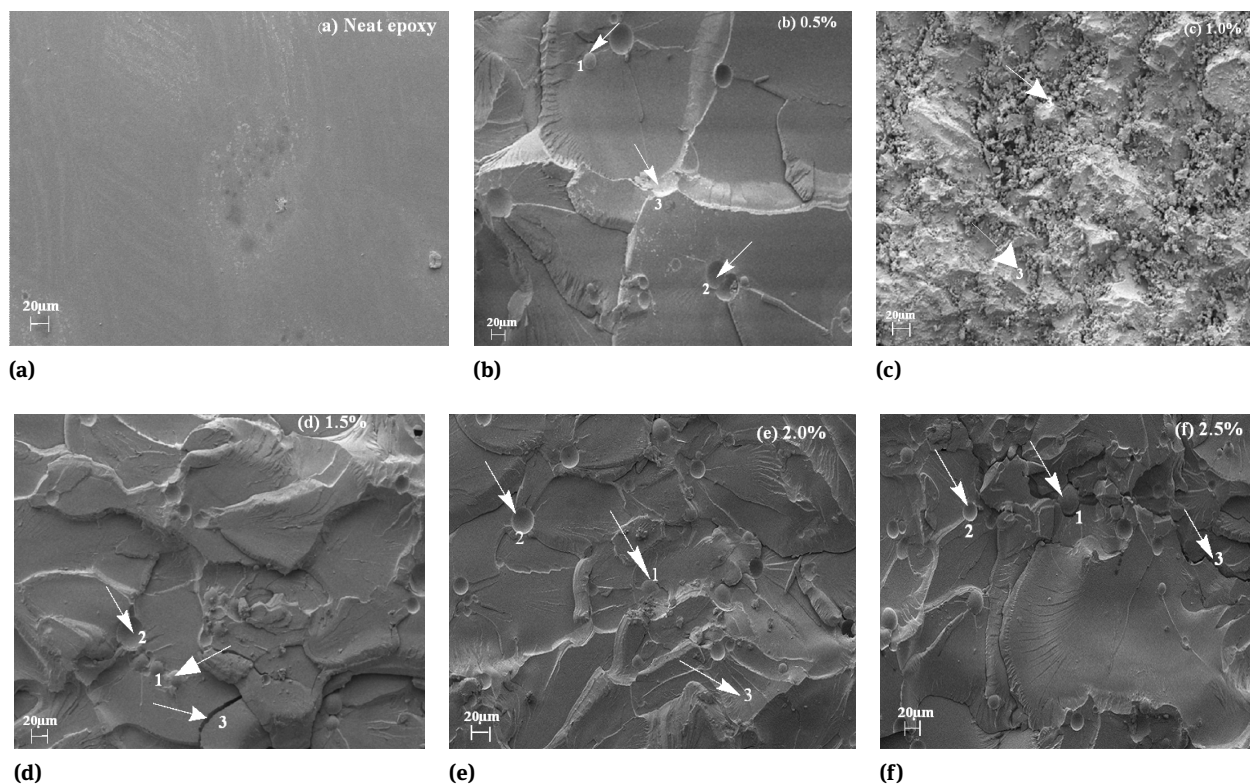
### 3.4 Fracture mechanism

The fracture mechanism for the tensile test conducted on the syntactic foam composites as observed under SEM are shown in Figure 4(a-f) for all the volume fraction compositions respectively. Figure 4(a) shows the cracked portion for the plain epoxy resin composites where the crack could be seen freely propagating around the surfaces which are responsible for their brittle failure [38]. The cracked surfaces become clearer with the introduction of HGM as shown in Figure 4(b-f). The marked regions identified as “1”- deboned microspheres, “2”- fractured microspheres and “3”- fractured surfaces on the syntactic foam. The fractured microsphere and deboned microspheres increased with an increasing volume fraction of HGM in the syntactic foam composites which are as a result of good bonding effect on interfaces between the matrix resin and can also be attributed to good interaction between the matrix and the filler [38], that the micrographs of HGM/epoxy composites show a good interaction of HGM with the epoxy matrix.

Figure 4b shows a little roughness on the fracture surface due to a reduced void volume fraction of the composites while much roughness could be seen from Figure 4(c-f) due to their higher void volume content in the syntactic foam which is also responsible for their low strain value compared to the neat resin, Table 3.

### 3.5 Dynamic mechanical analysis (DMA)

The dynamic mechanical analysis was conducted to study the viscoelastic properties of the syntactic foam composites. The storage, and loss modulus values for the EPT60 syntactic foam composites and neat epoxy matrix were taken from three different temperatures measurement 30°C, 50°C and 60°C as shown in Table 5. Figures 5(a-c) show the storage modulus, loss modulus and tan delta versus temperature graphs of the syntactic foam composites. At the temperature of 30°C, the storage modulus does not show a consistent trend but a decrease in loss modulus was observed as the HGM increases. At 50°C temperature, storage modulus increased while loss modulus decreased with respect to neat resin. At 60°C, both the storage and loss modulus increased at the addition of HGM with respect to neat resin. The maxi-



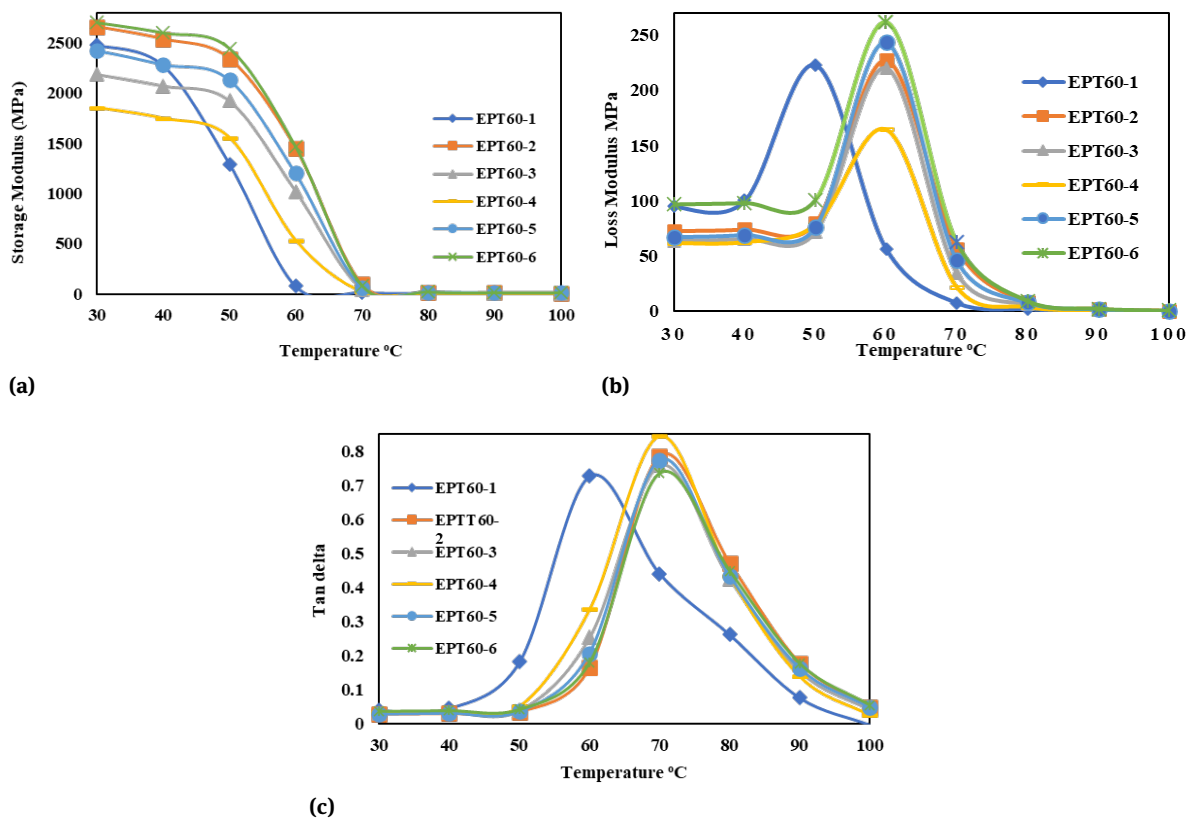
**Figure 4:** SEM images for tensile fracture surfaces for (a) neat resin and (b-f) percentage fraction of HGM 0.5-2.5% in syntactic foam at 500 magnifications showing the arrowed areas as 1: deboned microspheres, 2: fractured microspheres and 3: fractured surfaces.



imum storage modulus values were at EPT60-6 for the three selected temperatures which are 2705, 2447, and 1466 MPa respectively and the loss modulus was highest at 60°C of EPT60-6 (261.5 MPa). The improved storage and loss modulus values compared to neat resin can be attributed to good interaction and interfacial bonding between the microsphere and the matrix and also as a result of the lower density and void content. The storage modulus decreased with increased temperature due to the reduction in the void rate of the composites as a result of the degassing method in processing it which correlates with earlier reports from

Sankaran *et al.* [1] and Ghamsari *et al.* [3] while John B *et al.* [39] reported a sudden decrease in the storage modulus of cyanate ether syntactic foam composites with temperature.

The loss modulus shows the variation in values but increases mostly with the addition of HGM in the EPT60 syntactic foam. The maximum loss modulus for neat epoxy matrix resin was 223 MPa at 50°C and ~262 MPa at 60°C for EPT60-6. This shows that the loss modulus of syntactic foam was noticeably affected as the percentage of HGM



**Figure 5:** (a) Storage modulus, (b) loss modulus and (c) tan delta of HGMSF and neat epoxy matrix against temperature.

**Table 5:** Comparison of storage modulus (SMD) and loss modulus (LMD) values of HGMSF composites and neat epoxy matrix resin.

Specimen name	Volume fraction of HGM (%)	Temperature (°C)					
		30		50		60	
		SMD	LMD	SMD	LMD	SMD	LMD
EPT60-1	0	2478	94.78	1294	223.2	77.62	56.63
EPT60-2	5	2663	72.63	2343	79.9	1449	227.2
EPT60-3	10	2190	63.84	1924	71.76	1022	220.2
EPT60-4	15	1854	62.09	1555	78.63	535.4	164.6
EPT60-5	20	2426	67.48	2136	75.93	1203	243.5
EPT60-6	25	2705	96.74	2447	101	1466	261.5

**Table 6:** Comparison of  $\tan \delta$  values of HGMSF and neat epoxy resin at different temperature range.

Specimen name	Volume fraction of HGM (%)	Temperature		
		30°C	60°C	70°C
EPT60-1	0	0.03824	0.7259	0.4391
EPT60-2	5	0.02727	0.1637	0.7851
EPT60-3	10	0.02915	0.2518	0.7569
EPT60-4	15	0.03349	0.3349	0.8435
EPT60-5	20	0.02713	0.2055	0.7727
EPT60-6	25	0.03576	0.1793	0.7354

increases and it reflects either retention or a marginal increase till the maximum temperature was reached.

The storage modulus ( $E'$ ) of the EPT60 syntactic foam composite measures the stored energy in the elastic portion of the composite as shown in Figure 5a. However, the loss modulus ( $E''$ ) of the EPT60 syntactic foam composite measures the energy dissipated at heat per cycle under deformation of the material as shown in Figure 4b.

### 3.5.1 Damping Coefficient

The values for the damping coefficient or loss factor  $\tan \delta$  which is the measure of how efficient the syntactic foam loses energy to molecular rearrangement and internal friction as compared to the neat epoxy resin was varied at different temperatures “30°C, 60°C and 70°C” as reported in Table 6. The glass transition temperature ( $T_g$ ) was at 70°C which was the temperature at which the EPT60 syntactic foam reaches its maximum  $\tan \delta$  as shown in Figure 5c. The inclusion of HGM reduced the peak of epoxy resin  $\tan \delta$  which is an indication of good interfacial bonding. This shows that when the constituents material of the composites exhibit good interfacial bonding, it resulted in less energy dissipation resulting in a lower value of  $\tan \delta$  [3]. It also shows that epoxy resin exhibited a rubbery plateau before the EPT60 syntactic foam which was an indication of proper dispersion of HGM in the epoxy resin. In addition, the decrease in  $T_g$  with the addition of HGM resulted in the plasticization of the epoxy resin which can be linked to the reduced void fraction in the composites as a result of the degassing method [40]. Therefore, an increase in the interfacial interaction between the matrix and HGM leads to a decrease in the loss factor ( $\tan \delta$ ) of EPT60 syntactic foam composites and also improves the damping performance of the molecule motion in the foam composites [40, 41].

## 4 Conclusion

Syntactic foam composites prepared by degassing method with a varied volume fraction of HGM from 5-25% were prepared. The effect of varying compositions of HGM on the mechanical and dynamic mechanical properties was investigated. The tensile properties from the test revealed an improved strength as the volume fraction of HGM increased due to good interfacial adhesion between the filler and the matrix and the low void content in the composites due to degassing method of processing. The dynamic mechanical analysis of the syntactic foam composites showed a decrease in storage modulus and an increase in loss modulus with increasing temperature with the maximum value of storage modulus of 2705 MPa at 30°C and maximum loss modulus of ~262 at 60°C. The damping performance also reduces due to the reduction in the intensity of the  $\tan \delta$  peak due to good interfacial interaction between the molecules of the polymer and the HGM. The density of syntactic foam composites declined gradually as the volume fraction of HGM increased. In addition, this study shows that the properties of syntactic foam composite can be effectively controlled by reducing the void content when changing the volume fraction of HGM embedded in the matrix. The syntactic foam composite can be regarded as lightweight materials for structural application and in the context of the search for materials with better tensile properties for aerospace, marine, automobile and other design fields, this work contributes to our understanding of improving the properties of syntactic foam.

**Conflict of Interests:** The authors declare no conflict of interest regarding this manuscript.

**Acknowledgement:** The authors would like to appreciate the support gotten from the DST-CSIR U-295 grant and Durban University of Technology for the opportunity given to conduct this research work.

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