Journal of Material Science and Mechanical Engineering (JMSME)
p-ISSN: 2393-9095; e-ISSN: 2393-9109; Volume 6, Issue1; January-March, 2019 pp. 29-33

http://www.krishisanskriti.org/Publication.html

PET Waste as Reinforcement in Epoxy -Glass Microballoon Syntactic Foams

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Abstract—Epoxy based syntactic foams are formed by incorporation of hollow glass microballoons in an epoxy matrix. These classes of composites are characterized by high specific mechanical properties coupled with light weight features. In view of these interesting properties, epoxy syntactic foams are used in structural, automotive and in buoyant applications. In order to amplify the application range of these epoxy syntactic foams, reinforcements are added. Besides increasing the mechanical properties, this leads to an added concern of increasing the cost of such foams which prevents them from being used in certain applications where cost is a major constraint. In this study, waste PET fibres have been utilized as an alternative to the costly reinforcements, for the fabrication of epoxy syntactic foams. The sources of waste PET fibres are the empty beverage bottles collected from waste. When left undisputed, they are a major cause of concern to the environment. The PET bottles are collected, washed and dried prior to their shredding to form staple fibres.

PET fibres are added to the epoxy syntactic foams in varying amounts (1-5 % v/v). Quasi-static mechanical properties of the reinforced syntactic foams have been investigated. Inclusion of waste PET in the form of staple fibres leads to an increase in the properties of the neat foams without an appreciable increase in their density. About 8 % and 4% improvement in compressive strength and specific strength was obtained upon addition of PET fibres (3 % v/v). Similarly, improvements of the order of 30 percent and 26 percent were obtained for flexural strength and specific flexural strength respectively. These improved properties highlight the potential of waste PET as an effective reinforcement for epoxy based syntactic foam.

Keywords: *epoxy, hollowglassmicroalloons, syntactic foam, PET waste.*

1. INTRODUCTION

Plastics have become an integral part of human lives. The vast array of properties that are provided by the plastics materials has led to their usage in almost every field. However, with the increasing use of these plastics materials, their ill-effects cannot be ignored. One of the major concerns regarding the use of plastics is the lack of proper methods for waste disposal. Once discarded by the consumers, these plastics end up reaching dumping grounds and being incinerated with only a small fraction being recycled. Polyethylene terephthalate is one of the most widely used commodity plastic that is used for packaging of mineral and beverage bottles primarily due to its good mechanical properties[1], thermal properties[2], limited permeability to gases and water-vapour and transparency. In an attempt to exploit its useful properties and minimize accumulation of wastes, many research groups have investigated the use of waste PET in many structural applications.

There seem, however, limited studies when it comes to post-consumer usage of PET in reinforcements for polymer matrices. In this context, polymer syntactic foams are a new category of polymer composites and have emerged as strong contenders for a variety of applications ranging from undersea to aerospace. These foams have been used for the construction of less weight yet strong human operated vehicles and remotely operated vehicles for deep sea exploration[3]. The thermal insulation properties of these foams allow them to be used in the construction of spacecraft and fuel tanks insulation[4]. They are nowadays increasingly being used as core materials in sandwiched structures for blast mitigation applications[5]. Physical foams, as they are sometimes called, are made by incorporation of hollow microballoons of glass, polymer or flyash bounded by a polymer matrix. Glass microballoons are frequently being used as porogen in the fabrication of syntactic foams due to their availability in varying densities besides being chemical and corrosion resistant. For structural applications, epoxy is the preferred matrix material due to its superior mechanical properties. Reinforcements, such as carbon nanofibres[6, 7], graphene[8], polymer nanofibres[9], glass fibres[10, 11], etc. have been incorporated to enhance the strength of these foams. However, this leads to an obvious increase in the cost of manufacturing of such foams.

This work is an attempt to utilize waste PET as an effective and low cost reinforcement in the development of epoxy based syntactic foams for a variety of applications. We anticipate that the use of PET will not only enhance the mechanical properties but also reduce the cost of the composites and provide a solution for the effective utilization of this post-consumer feedstock.

2. EXPERIMENTAL

2.1. Materials and methods

Post-consumer plastic bottles were used as waste PET bottles. These were collected, washed and dried. Bottles were shredded manually into staple fibres (10 mm×2 mm). Diglycidyl ether of bisphenol A (DGEBA) based epoxy resin (LY 556), tri ethylene tetra amine hardener (HY 951) and Hollow glass microballoons (K46, 3M) were used for the preparation of syntactic foams. K46 has a density of 0.46 g $cc^{-1}$and an average particle size of 40 microns. The properties of hollow glass microballoons are presented in Table 1.

Table 1: Physical characteristics of hollow glass microballoons (K46)

HGM	Density (kgm^{-3})	Wall thickness (µm)	Radius ratio (η)	Microballoon size (µm) (mean diameter)	Isostatic crush strength (psi)
K46	460	1.29	0.9356	40	6000

2.2. Preparation of syntactic foams

Epoxy syntactic foams were fabricated by incorporation of hollow glass microballoons (40-60 % v/v) in epoxy matrix as per the procedure discussed in the literature[9]. Staple PET fibres (1, 3 and 5 % v/v) reinforced epoxy syntactic foams were prepared by incorporation of PET into epoxy resin. The contents were mixed manually followed by the introduction of calculated amounts of hollow glass microballoons and hardener. The system was degassed and transferred to aluminium moulds for curing at room temperature for 48 hours to prepare samples for mechanical testing. The amount of hollow microballoons and PET fibres was calculated as follows:

$$\frac{Mass\ of\ PET\ fibres}{Mass\ of\ composite} = \frac{\rho_{PET}\times\Phi_{PET}}{\rho_{PET}\times\Phi_{PET}+\rho_{K46}\times\Phi_{K46}+\rho_{matrix}\times\Phi_{matrix}}$$

Where ρ and Φ refer to the density and volume fraction of the constituent respectively, and the subscripts 'PET', K46 'matrix' refer to PET fibres, microballoons and epoxy respectively. For the purpose of calculation, the density of PET, K46 microballoons and epoxy have been assumed to be 1.38 g cc^{-1}[12], 0.46 g cc^{-1} and 1.17 g cc^{-1}[9] respectively. Reinforced syntactic foams were prepared maintaining a constant volume percentage of 40 for all the specimens. The sample designations and compositions are presented in Table 1.

TABLE 2: Different compositions and designations of neat and reinforced foams

Sample Code	Matrix (% v/v)	Microballoons (% v/v)	PET (% v/v)
S40	60	40	-
S50	50	50	-
S60	40	60	-
S40P1	60	39	1
S40P3	60	37	3
S40P5	60	35	5

For unreinforced syntactic foams, S refers to epoxy based syntactic foam followed by the volume percentages of hollow glass microballoons (40-60 % v/v) used for the fabrication of neat foams. For reinforced foams, S refers to epoxy based syntactic foams, two digits thereafter represents the volume fraction of total filler (hollow glass microballoons and PET fibre) content, P refers to waste PET fibres and 1, 3 and 5 refers to the volume percentage of its loading into epoxy.

2.3. Density Determination

The theoretical density was calculated using the standard rule of mixtures.

$$\rho_{th} = \rho_{PET} * \Phi_{PET} + \rho_{K46} * \Phi_{K46} + \rho_{matrix} * \Phi_{matrix}$$

Experimental density (ρ_{ex}) was determined by calculating the mass: volume ratio of five specimens as per ASTM D1622-98. The difference between the theoretical and experimental density was used to determine the air-void porosity trapped within the foam.

$$Void\ volume\ (\%) = \frac{\rho_{th}-\rho_{ex}}{\rho_{th}} \times 100$$

2.4. Mechanical Testing

Mechanical testing was carried out using Universal Testing Machine (Instron 3382) at ambient temperature. Compression testing was done using standard cylindrical specimens (12mm diameter, 6 mm thick) at cross head speed of 1.3 mm min^{-1}. Five specimens of each composition were tested and the load-displacement data obtained from the tests were used to obtain stress-strain curves for calculation of compressive strength.

Flexural testing of the samples was performed on specimens of standard dimensions (127 mm length x 12.5 mm width x 3.5 mm thickness) under three point bending mode as per ASTM D790 at a deformation rate of 2 mm min^{-1} and span length of 60 mm. The tensile properties were determined as per ASTM D638 using a Universal Testing System (Instron 3382) at ambient temperature. The dumb bell shaped specimens used for tensile testing were 165 mm long, 3 mm thick, and 13 mm wide along the centre of the casting for syntactic foams. The samples were subjected to a cross head speed of 10 mm min^{-1}.

2.5. Characterization

The thermal behaviour was investigated using Perkin Elmer (Pyris 1 TGA) under N_2 atmosphere in the temperature range 50-600°C. A heating rate of 20 °C/min and sample mass of 5.0 ± 0.5 mg was used for each experiment.The morphology of fractured surface was studied using a scanning electron microscope (JEOL- JCM6000PLUS) under an acceleration voltage of 1 kV. Samples were mounted on aluminium stubs

and sputter-coated with gold and palladium (10 nm) using a sputter coater (DII-29030SCTR Smart Coater) operating at 10-12 mA for 120 s.

3. RESULTS AND DISCUSSION

3.1. Density of Syntactic Foams

Increasing the microballoons concentrtion led to a proportional decrease in the density of syntactic foams. Entrapment of voidsin the specimen during processingsteered to a reduction in the experimental density and the same was observed in the specimens. The ratio of experimental and theoretical densities was therefore calculated to quantify the voidage in each sample, and the results are presented in Figure 1.

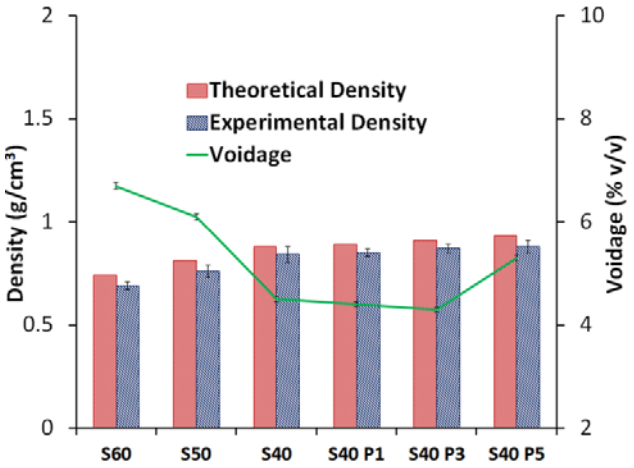

Figure 1: Theoretical, experimental densities and voidage of syntactic foams. Voidage is represented in the secondary axis.

3.2. Mechanical properties

The effect of incorporation of hollow glass microballoons (40-60 % v/v) on the mechanical properties of syntactic foams for unreinforced and reinforced specimens under different quasi-static modes of testing i.e., compressive, tensile and flexural is presented in Figure 2. Irrespective of the testing mode, an inclusion of weaker hollow glass microballoons (41 MPa) leads to a concomitant decrease in the strength of the specimens.In view of the improved specific properties for S40, the PET fibre has been added maintaining a constant loading of 40 (% v/v).Following are the observations from the mechanical testing of composites.

3.2.1. Compressive Testing

Quasi-static compressive stress-strain curves and corresponding mechanical properties of neat and PET fibre reinforced syntactic foams are presented in Figure 2 (a-b). All the samples exhibit a ductile mode of failure as evidenced by stress-strain curves. In general, a reduction in the compressive strength of unreinforced syntactic foams is observed as the volume percentage of glass microballoons increases. Increasing the microballoons loading leads to a reduction in the resin content which accounts for the increased shearing between the microballoons and their crushing which promotes their failure[5, 6]. In line with the previous studies[13-15], the stress-strain profile of syntactic foams is found to consist of three regions i.e. a linear region, also known as the Hookean region followed by a plateau region featuring a long strain at constant stress and a densification region, the end of which marks the failure of the foams. The linear region is representative of the elastic stretching of the cell walls. In the plateau region, the cells undergo crushing and thereby absorb the stress. The densification or the strain hardeningmarks the complete failure of the foams. Introduction of PET fibres increases the peak stress of the specimens. Maximum property enhancement is achieved at 3 % (v/v) of PET. This is attributed to the presence of a PET layer that absorbs the incoming stress leading to high values. Beyond 3 % (v/v), there is a precipitous drop in the compressive stress due to formation of agglomerates. Specific compressive strength, calculated by the ratio of compressive strength to density of the foams follows a similar trend. Improvement in compressive strength overcomes the minor increase in density upon introduction of PET fibres (3 % v/v) resulting in higher specific compressive strength. An 8 percent and 4 percent increase are obtained in the compressive and specific compressive strength of PET fibre reinforced syntactic foam. Our results are in correlation with the previous studies on syntactic foams wherein incorporation of filler leads to marginal improvements in the compressive properties of syntactic foams[16, 8]. In view of the improved properties due to the presence of PET fibres, the composite structures are assumed to take higher amounts of compressive loads and that demonstrates the capability of PET fibres as reinforcement in structural applications.

3.2.2. Flexural Testing

Flexural properties of neat and PET reinforced syntactic foams are presented in Figure 2 (c). It is to be noted that the failure of syntactic foams under flexure differs from that in compression. The failure under flexure is propagated by matrix failure with less dependency on the presence of hollow glass microballoons[17]. A decrease in the flexural strength is observed for neat syntactic foam specimens due to a reduction in the matrix content. The presence of PET fibres (3 % v/v) accounts for an increase in the load bearing components of the composite and is thus capable of resisting flexural stresses which leads to higher flexural strength compared to neat syntactic foams. The decrease in flexural strength beyond 3 % (v/v) PET fibres is primarily due to the agglomeration of fibres that act as stress concentration loci. An increase of 30 percent in flexural strength and 26 percent in specific flexural strength is observed at 3 % (v/v) addition of PET fibres (S40P3) compared to neat foam (S40).

3.2.3. Tensile Testing

The tensile properties of syntactic foam are presented in Figure 2 (d). The presence of varying volume fraction of microballoons does not have a major effect on the tensile properties of the syntactic foam. The failure of syntactic foams under tension depends upon matrix failure[7, 18]. A reduction in the epoxy content accounts for the reduced properties of neat syntactic foam.In the case of PET reinforced syntactic foams, there is an increase in the tensile properties compared to neat foam. Under tensile loading condition, the crack propagation continues uninterrupted leading to brittle failure of epoxy[19], however, the presence of a PET fibre serves to arrest this growing crack front by a mechanism known as crack pinning which is the primary reason for the improved properties of syntactic foams. Improvements of 13 and 9 percent in the tensile strength and specific tensile strength of PET fibre (3 % v/v) reinforced syntactic foams is therefore witnessed. The evidence of a crack pinning mechanism due to the presence of PET fibre is presented in Figure 3 by means of scanning electron microscopy image of the fractured surface of the specimen.

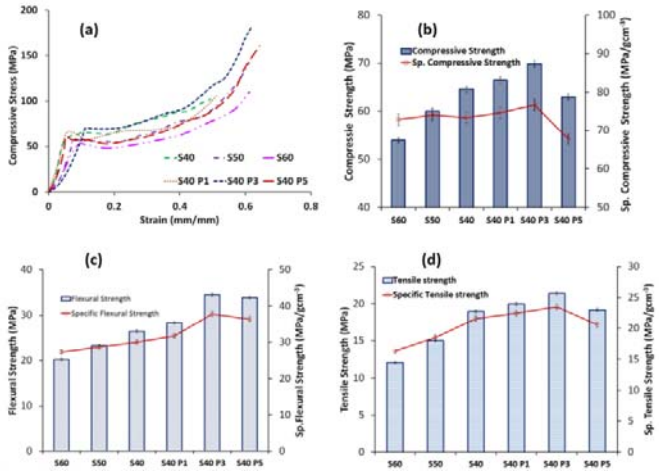

Figure 2: (a) Stress-strain curves (Compressive mode) and variation of b) compressive, (c) flexural and (d) tensile properties of syntactic foam

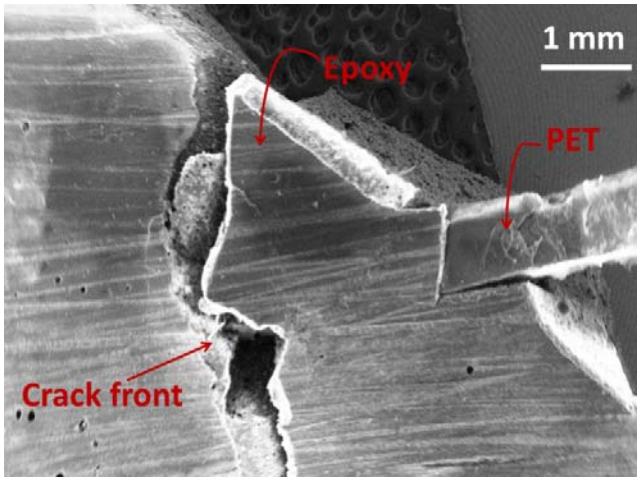

Figure 3: SEM micrograph of the fractured surface of epoxy syntactic foam

3.3. Thermal properties

Thermo gravimetric traces (under nitrogen atmosphere) of syntactic foams are presented in Figure 4. A single step degradation profile is observed for all specimens. However, there is a substantial increase in char percent due to the incorporation of hollow glass microballoons[13, 15]. Inclusion of PET staple fibres does not affect the service temperature of the syntactic foams. For the sake of brevity, only the TG trace of S40P1 is included in the graph as the thermal degradation profile remains unaffected (except for the minor decrease in char content) due to the presence of PET fibres.

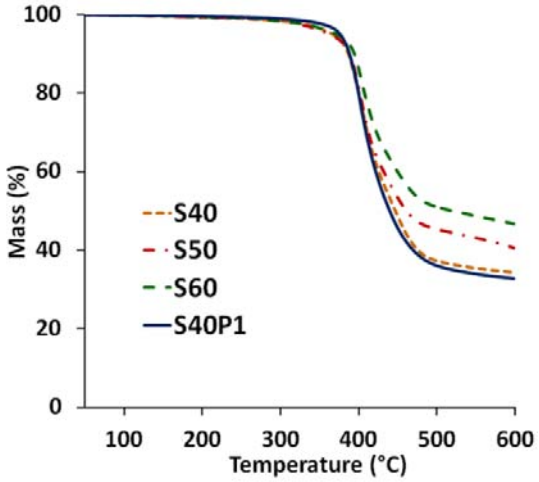

Figure 4:TG traces of syntactic foams with varying microballoon loading

4. CONCLUSION

Post-consumer usage, PET bottles were collected, washed, dried and shredded to obtain staple fibres. These were added as reinforcements (1-5 % v/v) into epoxy-glass microballoons formulations for the preparation of syntactic foams. PET containing syntactic foams reported improvements of the order of ~8 % and ~4 % in compressive and specific compressive strength respectively compared to neat foam.Interestingly, the extent of improvement, in terms of flexural strength was pronounced (~30 %) when PET was added as a load bearing component in the flexural testing of specimen. Importantly, the presence of PET fibres in epoxy matrix pins the crack and prevents it from propagating which resulted in improved tensile properties (~13 % and ~9 % improvement in tensile strength and specific tensile strength respectively). With such promising results, the reuse of this waste feedstock in syntactic foams not only foresees to address the issue of waste disposal in a cost-effective way but also seeks to explore the potential of PET as a utility in syntactic foams more often.

5 ACKNOWLEDGEMENT

The authors gratefully acknowledge CIPET:IPT Lucknow for providing necessary facilities to perform this work.

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