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Review Article

Enhancing the Strength, Electrical Properties and Versatility of Epoxy Polymer Composite Foam with Carbon Nanostructures

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Abstract

Epoxy resins (epoxy), also known as polyepoxides, are an extensive class of reactive polymers containing epoxide groups, and a very popular material is used in various applications such as marine, automotive, aircraft and aerospace architectures and electronic applications. However, epoxy is brittle and has high coefficient of thermal expansion, poor mechanical strength and electrical conductivity. Incorporating nano and micro filler in epoxy is an established approach to enhance all these properties. However, obviously also depends on the properties of the filler. In this mini review article, we will briefly describe how carbon nanomaterials, depending on their intrinsic structure and properties, can affect the epoxy composite foams, in terms of their fabrication methods, resulting mechanical, dielectric, and physical properties.

Keywords: Epoxy; Carbon nanostructures; Polymeric composites; Hollow glass microspheres; Strength

Introduction

A polymer composite, in simple description, can be considered as a sophisticated multi-phase material in which reinforcing fillers are intricately combined with a polymer matrix. This integration results in a synergistic enhancement of mechanical properties that cannot be attained by either component individually [1-3]. Such infusion of multiple component materials results in a final product with properties that surpass those of each individual constituent material. There are numerous compelling factors that can lead to the preference for new materials. Examples include materials that offer cost-efficiency, reduced weight, increased strength, enhanced durability [4-6] etc, compared to conventional materials. The distinct advantage of polymer matrix composites, in contrast to metals, lies in their manufacturing process that enables the production of intricately shaped components while offering lower density. This not only contributes to reduced fuel consumption in applications such as aviation and automotive industries but also facilitates higher speeds in competitive sports, longer ranges for missiles, and greater payload capacities in transportation [7,8].

In the majority of polymeric composite fabrication processes aimed at achieving desired material compositions, the primary challenges revolve around selecting the appropriate fibers, effectively distributing various phases, optimizing the aspect ratio of fibers, determining the spatial arrangements of continuous or short fibers, and devising an efficient process patterning strategy. In general, composite materials consist of one or more discontinuous phases dispersed within a continuous phase. In 'hybrid' composites, multiple discontinuous phases of varying natures are present [9,10]. The continuous phase is commonly referred to as the 'matrix,' while the discontinuous phase is termed as the 'reinforcement' material. By incorporating high-tensile strength reinforcements with exceptionally high modulus into a polymer matrix, it becomes feasible to enhance both the mechanical and thermal properties significantly. Additions of nano carbon materials [11,12] are trend to synthesize multifunctional composites. Also mechanical, electrical and thermal properties of these composites are fully customizable. Conducting fillers such as graphite-based nanostructures, carbon nanotubes, and carbon nanofibers are used as reinforcing agents for enabling the conducting behavior of epoxy resin.

Reinforcement Materials in Polymer Composites

Influencing properties of bulk materials by nano reinforcements are decade trend [13-16]. Hollow Glass Microspheres (HGMs) have low density and high strength with good thermal properties and thus can be employed in polymer matrix to form closed cell porous foams called syntactic foam. Unlike much other chemically synthesized polymeric foam, these syntactic foams have good mechanical, thermal and damping properties due to the presence of HGM into the polymer. These syntactic foams are used as buoyancy aid materials for marine applications [17]. These closed pore structures impart increased specific strength, reduced density, reduced coefficient of moisture absorptions and prevents thermal and electrical transports [18-22]. Syntactic foam properties are tailorable to good range and can be casted to any intricate shapes. Properties tailor ability is achieved either varying the volume fraction of HGM or by selecting HGM with suitable density (which depends on the shell thickness of HGM) [22,23]. Due to its reduced weight and improved specific properties attentions are given to use these composites for aerospace and marine structures as payload will be increased significantly [24,25]. The other applications of syntactic foams are discussed in [26,27]. Furthermore, previous works [28-30] have discussed the effects of properties on structural application of syntactic foams.

Nano filler reinforcement in the matrix enhances its cumulative properties which are attributed to commendable phase morphology and improved interfacial strength. Macro fillers forms finite interfaces with the matrix whereas, nano fillers have interfacial phases many folds increased [31-33]. Nano particulate syntactic foams are trending class of materials in which either HGM filler or epoxy matrix is modified by nano materials most commonly by nano carbon materials to achieve enhanced physical and engineering properties. When a small fraction of nano carbons is added to the composites it does not affect the density significantly and yields high performing multifunctional syntactic foams. Various nano carbon materials such as Carbon Nano Fibres (CNF), Carbon Nano Tubes (CNT) and Graphene (GP) evolve unique set of properties when reinforced with micro and bulk materials. High aspect ratio, smaller size and commendable mechanical properties makes carbon nano materials a highly preferable reinforcing material in syntactic foams [34-36]. Good dispersion of nano fillers in the matrix is crucial, as almost all the property enhancement in nano composites is attributed to increased interfacial phases, without which the adverse effect will be occurred in the composite. Entrapped voids during syntactic foam manufacturing are highly undesirable as it supports the initiation and propagation of the crack. Low density HGM floats on denser epoxy resin and higher viscosity of the resin-HGM are the major causes of voids entrapment. Reinforcing nano particles stabilizes voids in the system [37-39] which increases the risk of composite failure; hence effective processing methods are necessary.

Graphene platelets were proved to be potential two-dimensional filler in syntactic foams [40-44]. Large surface areas of graphene provide a good surface interaction with matrix and hence stress transfer [45-49]. Reinforcing graphene platelets to a maximum volume fraction of 0.5% in epoxy based syntactic foams shows insignificant increase in density [45]. However it's important to note that poor dispersion of graphene platelets causes its entanglement and wrapping which leads to voids in the composites. Its dispersion can be increased by functionalizing its surface to bear hydroxyl and amine groups. These groups make van der-wall interactions with the polymer and also surface roughness of GP increases which leads to wrinkled topology of GP and hence mechanical interlocking of polymer chain with GP takes place [42-45].

Synthesis and Growth Approaches

In the HGM/epoxy composite foam [50-56], the introduction of Nano Carbons (NCs) is carried out through two distinct approaches: (a) coating NCs onto the HGM surface [34,57], and (b) incorporating them within the matrix [42-45], as depicted in Figure 1. Chemical Vapor Deposition (CVD) stands out as the most commonly employed method for growing Carbon Nanotubes (CNT) on the surface of HGM, typically with a catalyst coating, where catalyst materials like nickel, cobalt, or ferrous are used. The CNT growth process involves the decomposition of carbon containing gases at elevated temperatures. For instance, Ephraim et al. successfully grew CNT on HGM with a cobalt coating, maintaining temperatures between 650 °C and 850 °C, using methane as the carbon source [34]. P. Bhat and colleagues achieved CNT growth on HGM coated with nickel at 600 °C, utilizing acetylene as the source gas [57]. These NCs, whether in their pure form or as NC-coated HGM, can be integrated into the matrix through various techniques, such as solvent evaporation or melt mixing methods.

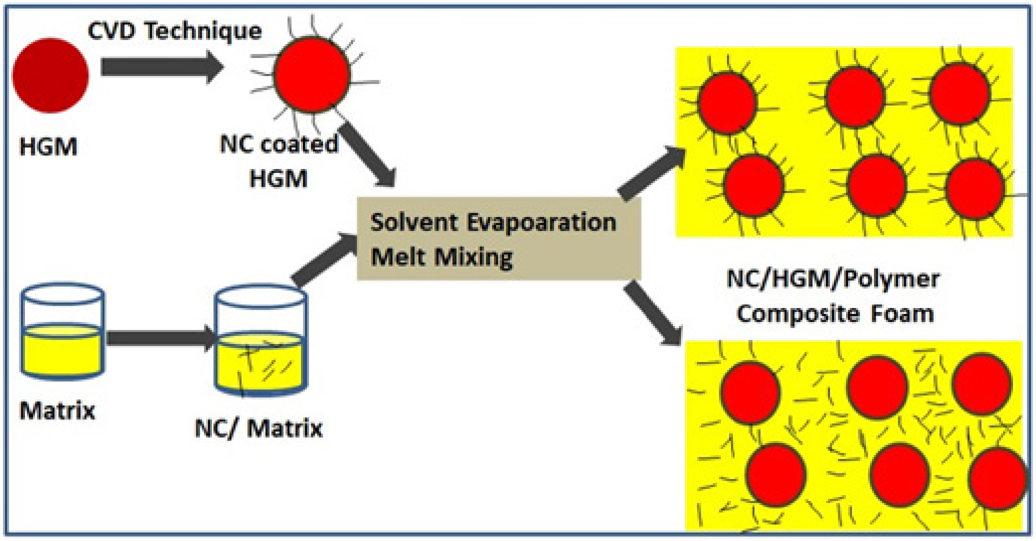

Figure 1: Schematic processing techniques of introducing carbon nanomaterials NC into epoxy polymer composite towards synthesis of efficient (NC/HGM/Polymer) composite foam.

Improved Properties of Polymer Composite Foam by Introducing NC

Mechanical properties

Enhancements of mechanical properties are very serious challenge for syntactic foams as these foams serve as core in aerospace and marine structures and failure of which may lead to property and life loss. Several research have been reported mechanical properties enhancement by matrix modification with fillers such as short fibers, micro sized fibers and particulates and nano scaled fillers such as nano clay, CNT, graphene and PEEKMOH [58]. Stable structural defects in nanostructures add to further versatility [59]. Macro fibers show tensile and shear strength enhancement at low volume fraction and higher volume of micro fiber reinforcement softens the matrix and also possesses poor interaction with the matrix and hence brings down the structural reliability of syntactic core. Reinforcement of nano clay in syntactic foams has been studied and found limited enhancement in tensile properties with increase in density significantly [60-65]. It is reported in [62,63,66,67] that a density increases of 17-26% has occurred in syntactic foams with nano clay inclusion of 2-4 vol% and hence limits its application as structural cores.

A very low volume fraction of graphene platelets has enhanced the tensile and fracture properties of syntactic foams [45], as shown in Table 1. Capacity to deflect crack propagation, high surface area and excellent mechanical properties of graphene platelets contribute the mechanical property enhancement of syntactic foams [58]. Also GP proves to have stronger interaction with the matrix which restricts polymer chain mobility and hence delaying crack initiation and growth. GP shows improvement in mechanical properties of syntactic foams when added in lower volume fraction (<3%) and higher volume fraction of GP shows negative performance which is clearly attributed to the dispersion challenge of GP when its volume increases. Poor dispersion of GP causes its agglomeration, wrapping and entanglement as they are 2D nano particles and hence restrict the strain transfer which results in poor performance.

Table 1: Effect of NC reinforcement on tensile and compressive properties of HGM/polymer composite foam.

Ref	Nano Carbon	ρ HGM kg/m^3	HGM vol %	NC vol%	Tensile Strength (% var)	Tensile Modulus (% var)	Compressive Strength (% var)	Compressive Modulus (% var)
[45]	GP	380	30	0.1	+15.9	+4.57	+3.6	+20
[45]	GP	380	30	0.3	+14.7	+14.7	+2.4	+26.6
[45]	GP	380	30	0.5	+10.5	+8.6	-11.1	+20
[61]	CNF	450	50	0.3	+27	+105	-	-
[61]	CNF	220	30	0.42	-7.8	-11.0	-	-
[61]	CNF	220	50	0.3	+29	+9.8	-	-
[61]	CNF	460	30	0.42	-14.4	-13	-	-
[61]	CNF	460	50	0.3	+46.6	+12.5	-	-

Syntactic foams show ductility under compression. Macro fiber reinforcement decreases the compression strength of syntactic foams. [62,63] reported that when 5 vol% of nano clay is added to epoxy based syntactic foams compressive strength increases with decrease in modulus significantly and with nano clay volume fraction less 5% the strength value decreases. Syntactic foams when loaded compressively it behaves elastically till the resilience level and then failure of microspheres occur corresponding to the plateau region in the stress strain curve and finally failure of matrix occurs. Syntactic foams are notably brittle when subjected to tensile loads. The tensile strength exhibits a decreasing trend with the rise in Hollow Glass Microspheres (HGM) volume fraction in syntactic foams, and failures occurring under tension are primarily influenced by the matrix. Tensile strength displays improvement when macro fibers are introduced, especially at volumes below 5%, but beyond that, it declines due to matrix softening with micro fiber reinforcements [66,67]. Interestingly, the addition of nano clay to epoxy-based syntactic foams, as reported in [45], increased tensile strength. However, these composites demonstrate poor damage tolerance and reduced load-bearing capacity. Furthermore, [45] reveals that the inclusion of graphene platelets in syntactic foams enhances both tensile modulus and tensile strength, particularly when the volume fraction of graphene platelets is at 0.3%, resulting in a remarkable 14.7% improvement compared to plain syntactic foams. However, when the volume of graphene platelets reinforcement exceeds 3%, adverse effects are observed [45]. Comparison of the resulting mechanical properties are in Table 1.

Electrical/dielectric properties

Conductivity of the composites increases with CNF reinforcement [68-71]. Electrical impedance increases with HGM volume in CNF/syntactic foams, which is because the porosity increases in the composites with HGM volume and hence the impedance. Denser HGM increases impedance of the composites. CNF decreases insulation and hence impedance decreases with CNF content in the composites [72]. Dielectric constant remains higher at low test frequencies, and it increases with the frequency. CNF volume in the syntactic foams increases dielectric constant and it reaches maximum at 10 wt% with 1 Hz test frequency. This increasing trend is because randomly connected CNF number increases with CNF volume in the composites and hence capacitance increases resulting higher dielectric constants [72-74]. Comparison of the resulting mechanical properties are in Table 2.

Table 2: Effect of NC reinforcement on electrical and dielectric properties of HGM/polymer composite foam.

Ref	NC	ρ HGM kg/m³	HGM vol %	NC wt%	Resistance		Capacitance (Fx10⁻¹¹)		Dielectric Constant	
					f=1 Hz (Ωx10⁸)	f=10⁵ Hz (Ωx10³)	f=1 Hz	f=10⁵ Hz	f=1 Hz	f=10⁵ Hz
[72]	CNF	-	-	1	9.4	2.2	4.2	2.6	19.9	12.4
[72]	CNF	-	-	2	7.3	2.8	50.3	3.1	222	13.8
[72]	CNF	-	-	5	7.7	1.9	5.2	3.1	26.4	15.8
[72]	CNF	-	-	10	7.7	1.6	5.1	3.1	26.6	16.1
[72]	CNF	220	15	1	9.8	3.6	2.9	1.8	12.6	7.9
[72]	CNF	220	15	2	21.9	3.3	13.6	2.2	62.9	9.8
[72]	CNF	220	15	5	7.0	1.5	5.5	3.4	25	15.5
[72]	CNF	220	15	1	4.1	2.4	$7.8 x10^3$	4.3	$3.7x10^4$	19.2
[72]	CNF	220	30	1	8.1	2.7	2.1	1.7	9.7	7.7
[72]	CNF	220	30	2	8.8	3.1	3	2	14.4	9.5
[72]	CNF	220	30	5	7	6.3	40.4	2.5	203.7	12.1
[72]	CNF	220	30	1	35.7	4.7	1	0.72	4.6	3.5
[72]	CNF	460	15	1	10.9	4.8	8.6	2	40.1	9.2
[72]	CNF	460	15	2	7.6	4.9	7.9	2.4	35.6	10.9
[72]	CNF	460	15	5	8.1	2.2	5.6	3	24.3	13
[72]	CNF	460	15	10	$1.9x10^{-3}$	7	$4x10^7$	8.6	$1.6x10^8$	41.1
[72]	CNF	460	30	1	23.1	1.5	1.8	1.5	8.3	7.1
[72]	CNF	460	30	2	9.6	2.9	3	1.9	13.7	8.4
[72]	CNF	460	30	5	4.6	3.6	$1.2x10^3$	3.3	$5.5x10^3$	14.1
[72]	CNF	460	30	1	37.5	3.9	2.8	1.1	13.2	5.4

Conclusion

In the materials industry, the imperative quest for weight reduction in designated applications takes center stage. This study underscores the remarkable accomplishments of Hollow Glass Microspheres (HGM) reinforcement, resulting in significant weight reduction and heightened structural rigidity. Furthermore, the paper delves into the documented contributions of nanocarbons, including graphene platelets, carbon nanotubes, and carbon nanofibers, in augmenting both mechanical and electrical characteristics. This concise review also underscores the ease with which nanocarbons can customize the electrical attributes of polymer composites, a versatile quality sought after in various applications.

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