Effect of Glass Spheres on Properties of E Glass/Cyanate Modified Epoxy Laminate

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ABSTRACT— ***High performance polymer composite laminates used in Aerospace and Electronics industries requires laminates that are structurally rigid besides exhibiting high stiffness and good di electrical properties. They are often required to be electromagnetically transparent in order to transmit the EM signals with nil transmission loss. Such materials could be a targeted with a focus on further enhancing their capabilities so that new material combinations with variety of fillers could be tried for different applications of the generation next, Electronics. Response of the laminates under tensile , flexural loading and a set of critical di electrical properties could by and large establish a potent material combination that can be used in signal processing divisions of aero electronics in particular viz Radome and similar such units. To augment this, thin laminates with cross ply configuration having 0/90/0/90 degrees as applicable to 4 plied, cross plied laminates were fabricated with cyanate ester modified epoxy resin and 1200GSM E glass unidirectional fiber. Solid glass spheres (9-13 micrometers diameter) as fillers were added to the laminate in 0%,10%,20% and 30%.Tensile strength & modulus, Flexural strength& modulus and di electric strength,di electric constant and dissipation factor /loss tangent at 1 MHz respectively were the properties chosen for the functionalization. It was found that best structural responses were from the balanced 4 plied laminate with 10% filler addition whereas the dielectric strength and loss tangent were found to be best for laminate with 10% filler addition. The dielectric constant was found to be the best for laminate with 30% filler addition. Thus, it was proven that filler addition has a vital role in influencing the properties of a laminate and a proper trade off between various parameters are required to be weighed before zeroing on a potential laminate for a specific application.***

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1. INTRODUCTION

The epoxy resin is the most widely used matrix material for many structural composites. It has many good properties like high stiffness, low shrinkage, good adhesion to glass/ carbon fiber, etc. Unfortunately, the very factor contributing to its high stiffness and heat resistance leads to its main drawback (lack of toughness), that is its highly cross-linked structure. Hence, among the different materials used for the modification of epoxy resins, cyanate esters are expected to be the best material to improve thermos mechanical properties. Cyanate esters also offer ease of handling and processing similar to that of epoxy resin systems. Thus, blending of epoxy with cyanate ester resin continues to attract research interest in order to impart improvements in fracture toughness without compromising other mechanical and thermal properties. Never a single choice of material could possibly possess both the properties at its best and hence proper compromises are often made with an intention of selecting the best combination with regard to a potential application. Since the late seventies, a variety of methodologies have been tried to improve the toughening of epoxy resin systems (1-3). Increasing fracture toughness in filled epoxies and the mechanism of crack pinning to enable this were also some important allied work carried parallel (4-9). Blending of Cyanate ester and Epoxy with the intention of toughening the latter, has gained lot of research potential (10-14) due to the advance structural requirements in the fields of aerospace and Electronics ,which essentially requires composites with high strength and good Di electric properties.In this scenerio, obviously the need for developing such materials has gained lot of momentum in the recent past. To prevent the transmission signals from getting blocked the investigational scope over here has been limited to thin laminate with simple 0/90/0/90 stacking orientations as applicable 4 plied laminates.Thus, cyanate ester modified epoxy resin as a matrix material and 1200 GSM, E Glass Uni directional fabric as an reinforcement are used here along with solid glass spheres as filler materials ,mainly to improve the di electrical properties of the laminate.

Here, 15% Cyanate ester loaded epoxy resin and 1200 GSM E Glass Unidirectional fiber were chosen as matrix and reinforcement elements respectively for fabricating cross plied laminates with 0/90/0/90 orientations as applicable to 4 plied, cross plied laminate. Lot of researchers have done wide variety of works to predict the influence composites filled with fillers of small aspect ratio, like that of glass.(15-22)Moreover in order to study the influence of glass microspheres as a filler on the properties of the laminates, filler percentages were varied in the laminates. Besides, with an intention of exploiting the vast potential in the field of aerospace and electronics, requiring structural composites with high stiffness and strength characteristics, this work focuses on the functionalization of the laminate in terms of blending of epoxy with cyanate and addtion of fillers.

Hence besides the mechanical properties of tensile and flexural strength and modulus the Di electrical properties Like dielectric strength, Di electric constant and dissipation factor were taken up with the focus on establishing best tradeoff between structural and dielectric performance

2. MATERIALS AND METHODS

2.1 Materials

Epoxy resin, Curing agent and Unidirectional fibers of 1200 GSM was procured from Sackthi fibers, Chennai, India and Bisphenol A Cyanate ester was imported from Shangai Righton, Shanghai, China. Glass spheres (9-13 micrometers diameter) were imported from Sigma-Aldrich, USA.

Figure 1 Basic materials for making Laminates

Figure 2 Different cured samples during Optimization

Trials

Curing agent (Tetraehyleneamine), was optimized with regard to Cyanate ester(BACY) modified Epoxy resin (Diglycidyl ether of bisphenol A/DGEBA). Optimization was done by varying the form, state and quantity of the curing agent, through a number of trials till the arrival of the defect free cured resin mixture. Before the optimization was achieved, defects like continuous foaming with sponginess in the cured blend, presence of glassy pinholes of varying sizes inside and open blow holes on the cured surface and presence of partially trapped and dissolved cyanate were observed visually. On the addition of the unoptimized curing agents and often, due to the added exothermic reaction, resulted in the accelerated gelation of the blend, thereby reducing the potable time of the resin mixture. Good potting period of around 15 to 30 minutes with almost zero defects was observed when 25.33%of the curing agent was mixed and later added to the blend of cyanate ester / Epoxy resin mix and allowed to cure at room temperature.

2.2 Methods and Testing

The composite laminates were fabricated from E-glass fiber (Unidirectional 1200 GSM) and cyanate modified epoxy resin by Hand layup technique. The addition of hardener (triethylene tetra amine) was based on optimized value as stated above through to a number of trials. The cyanate loading was fixed at 15% by weight of epoxy resin. The mixture of resin and cyanate ester was stirred for ensuring uniform dispersion of cynate ester, followed by the addition of tetraethylenetriamine (Hardener/curing agent) in 25.33% by weight of epoxy and stirred for around 10 minutes.Mylar sheet coated with stonewash polish as a release agent was further applied with a layer of resin using a brush. Then the first layer of the fiber (300×300mm) was placed on the resin, and consolidated using rollers. This process was repeated to consolidate five, 4 crosses plied laminates, namely L1 to L5.L1 was fabricated with neat epoxy. Necessary precautions were taken to keep the fabric well aligned. The fabricated laminated were then cured at room temperature for around 8 hours. Glass microspheres amounting to 10%, 20%,and 30% as fillers were added to the 4 plied laminates viz L3 and L5 respectively. The notations of the various cyanate modified epoxy glass fiber composites are presented in table 1.

Table 1. Laminate coding details

S. No	NAME	DESCRIPTION
1.	EP- L1	4Plies + Epoxy 100g + Hardener 25.33%+ Cyanate Ester 0%+ Filler 0%
2.	EPCY – L2	4Plies + Epoxy 100g + Hardener 25.33% + Cyanate Ester 15%+ Filler 0%
3.	EPCY 10F – L3	4Plies + Epoxy 100g + Hardener 25.33% + Cyanate Ester 15% + Filler 10%
4.	EPCY 20F– L4	4Plies + Epoxy 100g + Hardener 25.33% + Cyanate Ester 15% + Filler 20%
5.	EPCY 30 F- L5	4Plies + Epoxy 100g + Hardener 25.33% + Cyanate Ester 15% + Filler 30%

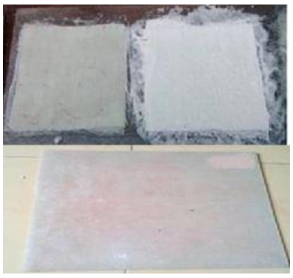

Figure 3 Processing of Laminate

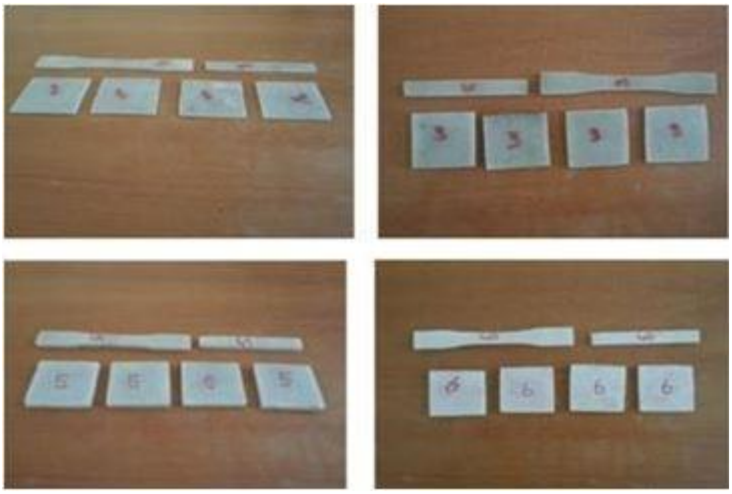

Figure 4 Few of the samples

A tensile specimen is a standardized sample cross-section as per ASTM: D638. It has two shoulders and a gauge (section) in between. The shoulders are large so they can be readily gripped, whereas the gauge section has a smaller cross-section so that the deformation and failure can occur in this area. For flexural strength specimens were prepared by ASTMD790, standard. The transverse bending test is most frequently employed, in which a specimen having a rectangular cross-section is bent until fracture or yielding using a three point flexural test technique. The flexural strength represents the highest stress experienced within the material at its moment of rupture. For electrical testing samples were cut using ASTM D149.

The dielectric strength is the maximum working voltage a material can withstand without breaking down, i.e., without experiencing failure of its insulating properties. At breakdown, the electric field frees bound electrons, turning the material into a conductor. The field strength at which breakdown occurs in a given case is dependent on the respective geometries of the dielectric (insulator) and the electrodes with which the electric field is applied, as well as the rate of increase at which the electric field is applied. Dielectric strength is usually expressed as a voltage gradient (such as volts per mm).

It is an important piece of information when designing capacitors and in other circumstances where a material might be expected to introduce capacitance into a circuit. The layers beneath etched conductors in printed circuit boards (PCBs) also act as dielectrics. Dielectrics, of course, are used in RF transmission lines. Dielectric constant is a dimensionless number.

The dielectric loss tangent can defined by the angle between capacitor's impedance vector and the negative reactive axis, as illustrated .it determines the loss line of the medium. Similar to dielectric constant, low loss tangent result in "fast" subtract while large loss tangent result in "slow" subtract. Beware that the exact values can vary greatly depending on the particular manufacture's processes, so we should seek out data from the manufacture for critical applications.

3. RESULTS AND DISCUSSIONS

The mechanical behavior of a composite is so complex that, the feasibility of a proper modeling is questionable, and the experimental approach may be the only acceptable solution. Much of our knowledge about the special nature of composite behavior has been derived from experimental observations. The measurement of the mechanical properties is also an important element of quality control and quality assurance process, associated with the manufacture of composite material.

1. The experimental values of tensile stress versus strain for composite specimen were determined during the tensile testing. In general, it is observed that the cyanate modified epoxy has better tensile strength invariably when compared to epoxy composite and this has been proved with respect to the experimental investigations.

The higher tensile properties are due to the rigid aromatic structure and rigid triazne ring formed as a result of trimeritative reaction.Also,this should be obviously the result of better bonding and adhesion characteristics between the various plies of the laminate and also should be contributed towards the higher GSM of the material.

It has also been observed that the addition of Fillers beyond 10% in the same laminates has brought down strength drastically. This should be attributed to the presence of fillers similar to the presence of voids and pores in isotropic materials which will act as stress risers during the loading processes. Under the influence of external loading the bonding in the vicinity of the fillers gets broken which in turn initiates the process of nucleation of micro cracks which tends to grow into macro cracks in the latter stages of loading, finally resulting in the failure under a relatively lesser load itself.

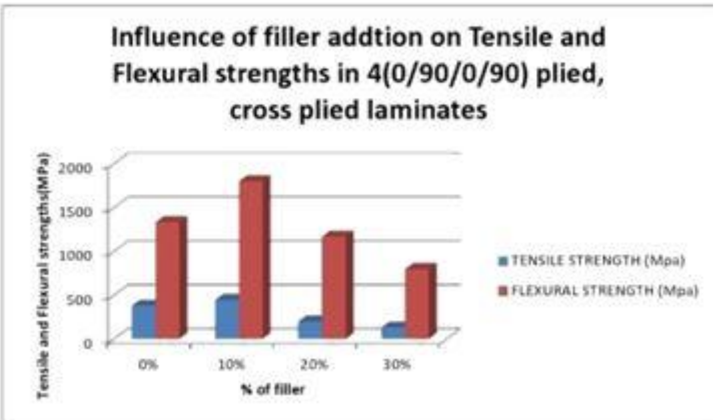

Figure 5 Influence of filler on Tensile and Flexural strength in 4 plied laminate

2. Similar trend was also observed in the values of flexural strength and the substantial increase in the flexural strength after cyanate loading is also observed and this due to the formation of a network structure between cyanate ester and the epoxy matrix. The marginal variations in the above properties might be attributed to other factors like the purity of the cyanate ester and also to the fabrication accuracy.

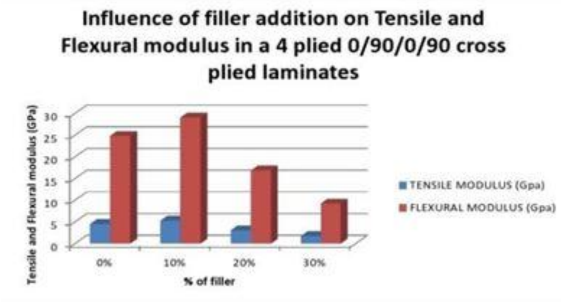

Figure 6 Filler influence on Tensile and Flexural modulus in 4 plied laminate

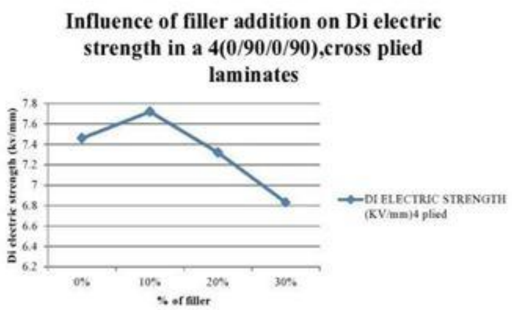

Figure 7 Filler influence on Di electric strength in 4 plied laminates

3. Electrical properties like Di electric Constant and Di electric strength by and large reflect ability of the laminate to transmit the RF signals through it with minimum attenuation or rather without any communication failure. Any noticeable loss here would mean communication failure between the Radar and the target. It is known that a higher Dielectric strength and lower Di electric constant and loss tangent are the requirement for the Radome wall. From the tests and results it could be observed that best value for Dielectric strength and Dissipation factor was observed for laminate with a 10 % filler addition. Dielectric constant was found to be good for the laminates with a 20 % filler added. This has proved the role of filler in influencing Dielectric properties in one way or the others. The quality of the ingredients could also have a strong say in the final outcome.

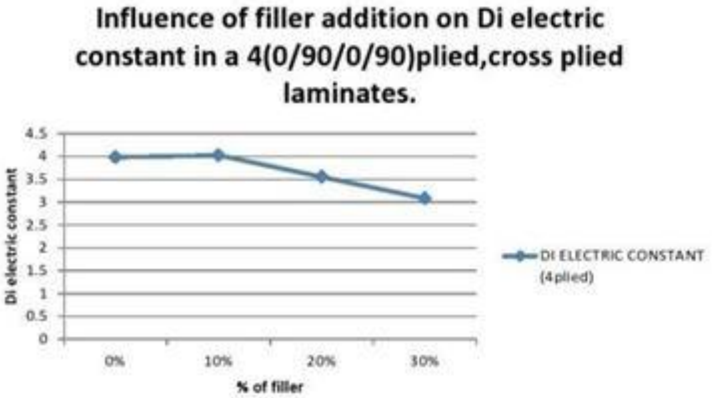

Figure 8 Filler influence on Di electric Constant in 4 plied laminate

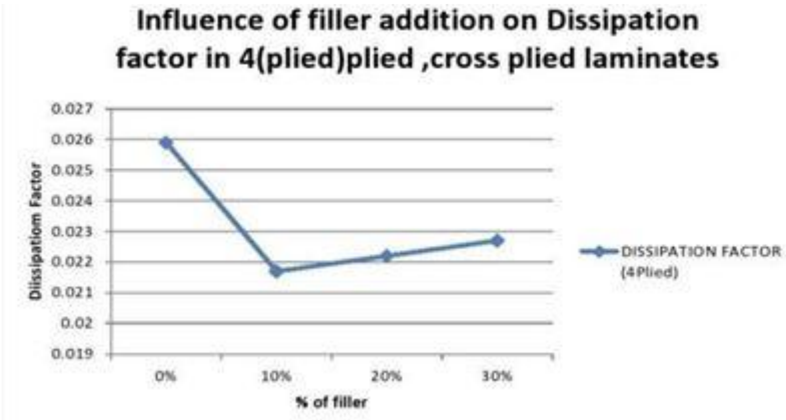

Figure 9 Filler influence on Dissipation Factor in 4 plied laminates

Table 2. Mechanical properties

TEST SPECIMEN	L1	L2	L3	L4	L5
TENSILE STRENGTH MPa	251	381	446	201	131
TENSILE MODULUS GPa	2.6	4.53	5.30	3.08	1.76
FLEXURAL STRENGTH MPa	333	1335	1800	1170	800
FLEXURAL MODULUS GPa	7.9	24.83	29.05	16.88	9.27

Table 3. Di Electrical properties

TESTSPECIMEN	L1	L2	L3	L4	L5
DIELECTRIC STRENGTH (kv/mm)	16	7.46	7.72	7.32	6.83
DIELECTRIC CONSTANT	8.1	3.98	4.02	3.55	3.08
DISSIPATION FACTOR	0.061	0.0259	0.0217	0.0222	0.0227

4. CONCLUSION

It has been found that the tensile and flexural strength with their respective modulus has shown an increasing trend in 4 plied, cross ply laminates loaded with 10% of glass microspheres as filler. The values show a decreasing trend beyond 10% filler addition. Though the glass fillers have been added here, mainly to enhance the electrical aspects due to its good dielectric properties, which it does here, the mechanical strengths viz., tensile and flexural increases first and then decreases latter with respect to 10% filler addition. This is due to the fact that ,fillers when initially added to the resin till 10% by volume , fills up the voids space effectively during the curing process thereby improving the resistance against the tensile and flexural deformations. Because of this, the toughness and hence the modulus values also reflects a similar increasing trend. But it has been found that after the optimum filling volume of 10%, further addition of fillers could not fetch a similar trend in the strength characteristics of the laminate. Besides the above fact, composite's better adhesion and hence better stiffness offered by the symmetrically balanced 4 plied laminate against the loading, cannot be over ruled here. But when it comes to Electrical properties higher Di electrical strength and loss tangent were found for 10% of filler and on the other hand, minimum Dielectric constant was recorded against the maximum filler addition of 20%. Thus, to conclude ,the role played by glass spheres as filler in influencing the structural and more importantly the dielectrically properties has been verified with the test results and laminates could be applied in the fields of aero electronics.

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