

ORIGINAL ARTICLE

Effect of layering pattern and fiber hybridization on viscoelastic properties of PALF/COIR hybrid epoxy composites

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ABSTRACT – To design and develop a hybrid biocomposite material for structural applications, it becomes necessary to determine the optimum fibers layering pattern. Therefore, in this research work, the different layered hybrid biocomposite boards i.e. bilayer pineapple/coir (P/C), trilayer (PCP, CPC), and intimately mixed (IM) were developed and characterized for viscoelastic properties. The composites were made by hand lay-up method, keeping the volume ratio of PALF and COIR 1:1 and the total fiber volume fraction is 0.40 volume of composite. Dynamic mechanical thermal analysis test was employed to characterize the viscoelastic behavior in terms of storage modulus, loss modulus, loss damping factor, and the glass transition temperature. Amongst all the different layered hybrid composites, the trilayer CPC has lowest value (0.635) of effectiveness coefficient with highest stiffness and activation energy (40.54 kJ/mole). It confirms the better fiber-matrix interaction at the interfacial region. The glass transition temperature of CF-EP and PF-EP was increased by 8.74% and 13.15% respectively by the synergistic hybridization of cellulosic fibers. The PCP layered composite possesses lowest value of phase transition energy (9.17 kJ/mole) and this was because of the poor fiber-matrix interfacial adhesion.

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INTRODUCTION

The increased awareness towards sustainability, product recyclability, and depletion of fossil fuels leads to the progressive utilization of green bio-based product and processes in various applications such as automotive, building and construction, railways, marine, and packaging. The lignocellulosic plant based fibers and their composites are active and feasible solution to replace non-renewable synthetic materials and it was due to their wide verities of profitable properties such as low cost, low density, high specific strength and stiffness, energy and ecological balance, and problem-free disposal at the end of use [1]. Amongst all the cellulosic strands, the pineapple leaf fiber (PALF) and coconut husk fiber (COIR) are enjoying a lot of attention for industrial and household applications and it was attributed to their superior physical, thermal, and mechanical properties. B.T. Mulyo et al. [2] have also proved the positive attributes of PALF based material as an alternative material for Indonesia National Standard (SNI) helmet. The COIR and PALF can be obtained from the fibrous husk (mesocarp) of coconut palm and the leaves of the pineapple fruit respectively using the biological and mechanical retting process. According to the report (2016) of Food and Agriculture Organization (FAO) entitled as "The State of Food and Agriculture", the total production of pineapple (Ananas-comosus) and coconut (Cocos-nucifera) in India is 1.9 and 11.1 million tonnes respectively.

PALF and COIR are multi-cellular fibers that consist of high cellulose (70-82%) and high lignin (40-45%) content respectively. Due to the presence of high cellulosic and low lignin content in PALF, it exhibits better tensile and flexural strength than the most commonly used natural fibers. As compared to PALF, the COIR is tougher, resilient, and highly durable in nature. Therefore, to combine their properties synergistically, it becomes necessary to incorporate them in the same polymeric matrix. The hybridization of stiffer fiber with the high strain to failure one leads to the improvement in impact energy absorbing capacity of material [3]. The combination of COIR and PALF may lead to the improved quality of performance. In most of the cases, the hybrid biocomposites have better or comparable performance than the glass fiber reinforced composites (GFRCs). The key benefits of hybrid composites over the single fiber reinforced ones are that they possess better impact strength, fatigue strength, and fracture toughness with significant amount of weight saving [4]–[12]. In order to make composites for various structural applications, it is desirable to test them in oscillating load, frequency, and dynamic thermal conditions.

Dynamic mechanical thermal analysis (DMTA) is a potent technique to interpret the structure/property relationship of a composite material. It can be used to analyze the effect of hybridization, fiber-resin volume fraction, fiber geometrical parameters and fiber orientation on the thermo-mechanical performance of a composite. The stiffness and damping behavior of a polymer and polymer based composites can be determined in terms of storage modulus (E' or G'), loss modulus (E''), and loss damping factor (tan δ). S.K Saw et al. [13] examined the effect of layering pattern on dynamic mechanical properties of Jute/Bagasse hybrid composites. C. Capela et al. [14] studied the effect of fiber length on the dynamic mechanical properties of carbon fiber reinforced composites. A. Chafidz et al. [15] investigated the effect of date palm fiber loading on the thermal and rheological properties of HDPE based composites. Q. Ahsan et al. [16] declared the positive impact of nano fibrillated kenaf fiber addition on the tensile properties of PLA based composites. Y. Nishitani et al. [17] investigated the viscoelastic properties of untreated and treated hemp fiber filled polyamide 1010 composites. W.S. Widodo et al. [18] examined the influence of ramie fiber addition on the sealing characteristics of silicon rubber gaskets and reported that the composite has better sealing performance than the pure silicon. J. Alexander et al. [19] reported that the storage modulus (E') of sisal/basalt/epoxy composite is higher than the sisal/glass/epoxy. S.S Chee et al. [20] evaluated the viscoelastic properties of kenaf/epoxy, bamboo/epoxy, and bamboo charcoal/epoxy composites and found that the kenaf/epoxy has higher E' and lower tan δ than the other composites. Y.S. Song et al. [21] examined the viscoelastic behavior of twill and plain woven Hemp/PLA composites and observed that the twill woven has higher stiffness and damping ability than the plain woven. S. Rwawiire et al. [22] examined the effect of bark cloth ply angle arrangement on viscoelastic properties of Bark/Epoxy composites. M. Idicula et al. [23] have examined the effect of layering pattern on the thermo-mechanical properties of Banana/Sisal/Polyester hybrid composites. R. Murugan et al. [24] reported that the Carbon/Glass/Carbon reinforced hybrid composite has higher storage and loss moduli than the single carbon, single glass, and hybrid Glass/Carbon/Glass-Epoxy composites. M. Jawaid et al. [25] analyzed the effect of layering pattern on dynamic mechanical characteristics of Jute/EFB filled epoxy based composites. They found that the Jute/EFB/Jute layered composite has higher stiffness than the EFB/Jute/EFB.

After reviewing the above mentioned work, it can be concluded that the relative arrangement of fibers significantly affect the overall performance of a hybrid composite. Therefore, this work considered the effect of different layering pattern on the viscoelastic properties of PALF/COIR hybrid composites. Moreover, the effect of fiber hybridization on the thermo-mechanical behavior of composites was also examined.

EXPERIMENTAL

Materials

The cellulosic fibers (PALF and COIR) were obtained from M/s Go Green Products, Chennai (India). Their physical properties and surface morphology are reported in Table 1 and Figure 1 respectively. The SEM micrographs revealed the network like structure that contains longitudinally oriented unit cells held together by lignin and fatty-waxy substances. The morphology of PALF is much smoother and continuous than the COIR and it is due to the presence of higher cellulose content and low fibril orientation angle (14^o). The epoxy resin [diglycidyl bisphenol-A (DGEBA)] and curing agent [tri-ethylene tetra-amine (TETA)] were obtained from M/s Sakshi Dies and Chemicals, Delhi (India). The typical properties of epoxy thermoset are mentioned in Table 2.



Figure 1. Surface morphology of (a) PALF and (b) COIR

roperties PALF C	OIR
(g/cm ³) 0.98	1.2
se (%) 70-82 32	2-43
lose (%) 18.8 0.1	5-0.25
u (%) 5-12.7 44	0-45
1.1	3-4
ontent (%) 11.8	8
se (%) 70-82 32 close (%) 18.8 0.11 a (%) 5-12.7 40 a (%) 1.1 2 pontent (%) 11.8 11.8	2- 5- 0- 3- 8

Table 1. Physical properties of PALF and COIR

Physical properties	PALF	COIR
Microfibrillar angle (deg)	14	30-49
Diameter (µm)	20-80	100-460
Tensile strength (MPa)	413-1627	131-220
Young's modulus (GPa)	34.5-82.5	4-6
Elongation at break (%)	1.6	15-40

Table 1. Physical properties of PALF and COIR (cont.)

Table 2. Typical properties o	f epoxy thermoset
he appearance of liquid resin	A clear pale yellow

The appearance of fiquid feshi	A clear pare yellow
Density (Kg/m ³)	1200-1250
Viscosity at 25°C (cps)	550
Tensile strength (MPa)	11-15
Tensile modulus (GPa)	0.45-0.52
Flexural strength (MPa)	23-28
Flexural modulus (GPa)	1.3-1.7
Max. Elongation (%)	4.2-5.6
Flashpoint	> 2000C

Preparation of Composites

The randomly oriented dried and chopped fibers of length 20 mm were impregnated with medium viscosity epoxy resin in a glass mold cavity of dimensions 70 mm X 10 mm X 2 mm. The different layered composite specimens [bilayer {pineapple/coir (P/C)}; trilayer {pineapple/coir/pineapple (PCP), coir/pineapple/coir (CPC)}; and intimately mixed (IM)] were made by hand lay-up technique, keeping the volume ratio of PALF and COIR 1:1 and V_f equal to 0.40 V_c. Figure 2 illustrates the various layering pattern for PALF/COIR hybrid composites. In all composite samples, the volume ratio of epoxy resin (DGEBA) with hardener (TETA) was fixed as 2:1(v/v). Before using the thermoset matrix for molding, ensure that the resin was completely mixed with hardener and no air bubbles were present. To accomplish this, the mixing was done in a necked round glass flask through mechanical stirrer at room temperature for 20 minutes. The curing of composite was performed at room temperature for 24 h under the dead weight of 35 kg. The DMA test specimens (Figure 3) of dimensions 35 X 10 X 2 mm³ were cut from the molded sheets and their designation are recorded in Table 3.

Table 3. Designation of DMTA test specimens

Designation	Details
EP	Neat Epoxy resin
CF-EP (1)	Coir fiber-Epoxy composite (frequency 1 Hz)
PF-EP (1)	Pineapple leaf fiber-Epoxy composite (frequency 1 Hz)
CPC (1)	COIR/PALF/COIR hybrid composite (frequency 1 Hz)
PCP (1)	PALF/COIR/PALF hybrid composite (frequency 1 Hz)
Bilayer (P/C) (1)	Bilayer (PALF/COIR) hybrid composite (frequency 1 Hz)
IM (1)	Intimately mixed hybrid composite (frequency 1 Hz)
CPC (5)	COIR/PALF/COIR hybrid composite (frequency 5 Hz)
PCP (5)	PALF/COIR/PALF hybrid composite (frequency 5 Hz)
Bilayer (P/C) (5)	Bilayer (PALF/COIR) hybrid composite (frequency 5 Hz)
IM (5)	Intimately mixed hybrid composite (frequency 5 Hz)



Figure 2. Different layering pattern of hybrid composites (a) PCP, (b) CPC, (c) Bilayer (P/C) and (d) IM



Figure 3. DMTA test specimens

Dynamic Mechanical Thermal Analysis (DMTA)

DMTA is a powerful technique in the development of composites aiming to establish a significant relationship between microscopic structure and thermo-mechanical properties. The comprehensive knowledge of viscoelastic properties such as stiffness, damping, thermal transition, melting & degradation, and crystallization is important for manufacturing a high-performance polymeric material. DMTA method has enormous potential to analyze the elastic and viscous response of polymer composites over a range of temperature, frequency, and time. The thermo-mechanical properties of a composite were determined using DMTA, Q800 instrument (Figure 4) which is supplied by Anamatrix Instrument Technologies Pvt. Ltd., Bangaluru (India). The specimens are of rectangular in shape having dimensions 35mm X 10mm X 2mm. They were used in double cantilever mode over a temperature range of 25-140°C with the heating rate of 3°C/min. The effect of frequency on storage modulus (E'), mechanical damping factor (tan δ), and glass transition temperature (T_g) has also investigated at two different frequencies, namely 1Hz and 5 Hz.



Figure 4. DMTA Q800 test instrument

RESULTS AND DISCUSSION

The thermo-mechanical behavior of a composite can be accurately assessed using the DMTA technique. DMTA is one of the most reliable and widely acceptable thermal analysis techniques that can record the viscoelastic response of polymer based composites under the sinusoidal load, temperature, and oscillation frequency. The test results which are reported in this section concerns the effect of layering pattern and fiber hybridization on the dynamic mechanical behavior of PALF/COIR hybrid composites in terms of storage modulus (E'), loss modulus (E''), and loss damping factor (tan δ).

Effect of layering pattern and fiber hybridization on the storage modulus (E')

The modulus curve provides the information about the viscoelastic rigidity and energy storage ability of a composite material. Figure 5 illustrates the effect of layering pattern and fiber hybridization on the storage modulus (E') of composites. The reinforced fibers and fillers play an active role to enhance the modulus of polymeric materials. The stiffness of thermoset resin was increased considerably by the incorporation of stronger and stiffer cellulosic fibers and this was attributed to the higher restriction of chain movements. The cross-linking density of epoxy polymer was increased after the loading of PALF and COIR. The E' curve approaches to lower values with the increase of temperature. Above the glass transition temperature (T_g), the E' of thermoset matrix was dropped sharply. The reinforcing fibers contribute much more for modulus at higher temperatures, above T_g than below it. It was due to the hydrodynamic mechanical action of reinforced fibers to impart constraints against the mobility, deformability, and relaxation of the macromolecular chain segments in viscoelastic medium. Irrespective of the fibers layering pattern, the stiffness of CF-EP was improved significantly by the incorporation of PALF. This was because of the synergistic compatibility between PALF and COIR. Among all the different layered hybrid composites, the bilayer (P/C) and trilayer CPC exhibits the higher E' below and above T_g (70^oC) respectively. The effectiveness of each fibers layering arrangement in regard to the increase of modulus can be determined by a coefficient 'C'.

$$C = \frac{\frac{E'_{G/E'_{R}}}{E'_{G/E'_{R}}}}{\frac{E'_{G/E'_{R}}}{E'_{G/E'_{R}}}}$$
(1)

where E'_G and E'_R are the storage modulus in the glassy and rubbery phase respectively.

The higher value of constant C signifies the poor effectiveness of fiber in regard to the increase of modulus. Usually, the filled systems have lower C value than the unfilled matrix polymer. Table 4 revealed that the CPC has lower 'C' value ($30-120^{\circ}$ C) as compared to the other hybridized composites. This was due to the higher wettability of coir fiber, less void content, and the formation of more covalent bond at the interface. The 'C' value of CF-EP (0.7048) was reduced after the loading of PALF and it confirms the synergistic compatibility between reinforcing fibers. It was observed that the bilayer (P/C) and intimately mixed (IM) composites have approximately same modulus at T_g. But at higher temperature (above T_g), the E' of bilayer composite was significantly greater than the intimately mixed one.



Figure 5. Effect of layering pattern and fiber hybridization with temperature on the storage modulus of fabricated composites at a frequency of 1 Hz

Effect of Layering Pattern and Fiber Hybridization on the Damping Loss Factor (tan δ)

The tan δ curve revealed the damping ability of a material. It provides indirect information about the stiffness, internal structure, interfacial characteristics, and the morphology of a composite. The damping ability of a composite depends on the orientation and distribution of fibers; fiber to matrix volume ratio; fiber/matrix interfacial interaction; and the concentration of void content. Figure 6 shows the tan δ curve of unhybridized and hybridized composites as a special reference of temperature. In all specimens, the tan δ curve goes on increasing and attains a peak value (phase transition region) followed by the reduction with the increase of temperature. The increment in tan δ peak corresponds to the greater mobility of small groups and chain segments. Table 4 revealed that the virgin epoxy occupies a higher tan δ_{max} value than the reinforced polymer composites, the trilayer PCP possesses shorter and wider tan δ peak. However, it occupies lower value of E' and T_g (62^oC) with higher coefficient 'C'. It confirms that the 'PCP' shows poor interfacial interaction with thermoset matrix and it was due to the presence more hydroxyl (-OH) group on the surface of PALF.



Figure 6. Effect of layering pattern and fiber hybridization with temperature on the loss damping factor (tan δ) of fabricated composites at a frequency of 1 Hz

The hybridization effect can be better understood in terms of T_g . The glass transition temperature (T_g) of CF-EP was increased from 64.9°C to 68.51°C and 70°C in CPC and bilayer (P/C) arrangements respectively. The CF-EP exhibits

higher T_g than the PF-EP and it was due to the better interaction between COIR and epoxy matrix at the interfacial region. In comparison to the CPC, the bilayer and intimately mixed have higher value of glass transition temperature (T_g). Furthermore, they (Bilayer and IM) possess higher peak width with lower peak height than the CPC. This was due to the better distribution of fibers and effective stress transfer from matrix to fibers without the matrix cracking. The shifting of T_g to a higher value revealed the reinforcement effectiveness of fiber. In this context, the intimately mixed (IM) has higher reinforcing ability than the others. The better performance of intimately mixed banana/sisal composite was also reported by M. Idicula et. al [23].

Effect of Layering Pattern and Fiber Hybridization on the Loss Modulus (E")

The loss modulus (E'') represents the ability of a material to dissipate mechanical energy as heat per cycle of sinusoidal viscoelastic deformation. Figure 7 shows the E'' curves of developed composites as a function of temperature. The E'' of epoxy thermoset was increased significantly by the incorporation of cellulosic fibers (PALF and COIR). It confirmed the prominent effect of fibers to increase the frictional force at the fiber-matrix interface. The peak value of E'' corresponds to the glass transition temperature (Tg). Beyond Tg, the loss modulus curve drop down to lower values and it was due to the free movement of polymer chain segments. Table 4 revealed that the Tg (obtained from E'' curve) of epoxy matrix was increased after the loading of reinforcing fibers. It was attributed to the inhibition in chain relaxation process, rigidity of polymer chain segments, and elevation in heterogeneity within the composite material. The phase transition temperature (Tg) of CF-EP and PF-EP was positively shifted to a higher value after the synergistic hybridization. The glass transition temperature obtained from E'' curves is lower as compared to the values resulted from tan δ curve. The intimately mixed composite possesses highest value of Tg, followed by bilayer, CPC, and PCP.

Table 4. Values of the effectiveness coefficient 'C', tan δ peak height and width, glass transition temperature (T _g),
E" peak height, E' and E" values at different temperature for virgin epoxy, pure coir fiber-epoxy, pineapple leaf fiber-
epoxy, and the different layered hybrid composites

Composites	С	$\begin{array}{c} T_{g} \\ from \\ Tan \\ \delta_{max} \\ (^{o}C) \end{array}$	Tan δ peak width	Tan δ peak height	Tg from E'' (^o C)	Log E'' Peak height (MPa)	Log E' (30 ^o C)	Log E' (50°C)	Log E' (70 ^o C)	Log E' (90°C)	Log E' (120 ^o C)	Log E'' (50°C)	Log E'' (70°C)
EP	-	61.2	23.66	0.83	44.3	8.79	10.98	9.6	5.83	4.93	4.72	8.78	5.29
CF-EP	0.70	64.9	20.39	0.76	55.88	9.14	11.33	10.99	7.45	6.79	6.91	8.71	7.02
PF-EP	0.61	62.9	19.08	0.73	52.17	9.21	11.12	9.9	7.95	7.79	7.85	9.16	7.02
CPC	0.64	68.51	17.12	0.63	61.56	9.21	11.45	11.32	8.61	7.73	7.75	8.12	8.12
PCP	0.67	62	27.52	0.43	49.24	9.14	11.42	10.46	7.81	7.33	7.32	8.99	6.71
Bilayer	0.66	70	18.64	0.63	62	9.13	11.51	11.33	8.68	7.54	7.54	8.46	8.22
IM	0.64	70	20.13	0.58	62.23	8.93	11.22	11.08	8.64	7.49	7.49	7.92	8.09



Figure 7. Effect of layering pattern and fiber hybridization with temperature on the loss modulus of fabricated composites at a frequency of 1 Hz

Effect of Frequency

The dynamic behavior of a material depends on the time, temperature, and frequency of oscillation. To characterize the viscoelastic behavior of polymer-based composites, it becomes necessary to analyze the effect of frequency on storage modulus (E'), loss modulus (E''), loss damping factor (tan δ), and the glass transition temperature (Tg). Figure 8 shows the variation in E' as a special reference of frequency and temperature. In all cases, the E' was increased with the increase of frequency of oscillation from 1 Hz to 5 Hz. It follows the same line of action as in the previous research works [Idicula et. al [23], Pothan et. al [26], Romanzini et. al [27], Mittal et al. [28]]. The static and dynamic moduli were found to be decreased over a long period of time and this may be due to the rearrangement of molecules in order to overcome the localized stresses. It was observed that the E' was decreased dramatically with the increase of temperature from 55^oC to 110^oC and it was due to the breakage of covalent bonding between the fibers and matrix resin.

The damping ratio (tan δ) is also influenced by the frequency of oscillation (Figure 9). At a higher frequency (5 Hz), the composites have higher value of tan δ_{max} and Tg. This behavior assured the higher degree of cross-linkage between polymeric chain segments. The tan δ peak and its corresponding temperature (Tg) is linked with the process of partial loosening of the polymeric structure and the cross-linkage density. Table 5 shows that the peak width of tan δ curve becomes broader by the increase of frequency (Figure 10 and Table 5). The response of the viscoelastic materials may vary with temperature and oscillation frequency. Therefore, it becomes significant to plot a three-dimensional thermogram (Figure 11) which represents the combined effect of frequency and temperature on the damping ratio of developed composites.

Table 5. Effect of frequency on viscoelastic properties of hybrid composites

Composite	T_g from tan δ_{max} (^O C)	Tan δ peak height	Tan δ peak width	T _g from E'' (^O C)	Log E'' peak height (MPa)
CPC (1)	68.51	0.63	17.12	61.56	9.206
CPC (5)	70.1	0.656	20.91	62.55	9.577
PCP (1)	62	0.43	27.52	49.24	9.138
PCP (5)	68.16	0.528	25.99	54.5	9.35
Bilayer (P/C) (1)	70	0.63	18.64	62	9.130
Bilayer (P/C) (5)	73.7	0.622	22.88	63.08	9.212
IM (1)	70	0.58	20.13	62.23	8.928
IM (5)	73.36	0.619	24.19	62.35	9.28



Figure 8. Effect of frequency with temperature on the storage modulus of fabricated hybrid composites



Figure 9. Effect of frequency with temperature on the damping ratio (tan δ) of fabricated hybrid composites



Figure 10. Effect of frequency with temperature on the loss modulus of fabricated hybrid composites



Figure 11. Three dimensional thermogram of different layered PALF/COIR hybrid epoxy composites

The Energy of Activation for Phase Transition

The energy required for phase transition from the glassy state to rubbery state can be easily and accurately determined using the Arrhenius equation:

$$f = f_0 \exp(-\frac{E}{RT})$$
(2)

where f is the measuring frequency, fo is the frequency when T approaches infinity, T is the glass transition temperature obtained from tan δ curve analysis, E is the activation energy, and R is the gas constant. The calculated activation energy for the different layered hybrid composites is mentioned in Table 6. It was observed that the CPC and PCP have highest and least value of activation energy. This outcome is in favor of the extent of reinforcement.

Hybrid composites	Activation energy (kJ/mole)
CPC	40.54
PCP	9.179
Bilayer (P/C)	18.65
IM	20.45

Table 6. The activation energy for the different layered hybrid composites

Cole-Cole Plots

The Cole-Cole plot is an effective and valuable tool to fully characterize the viscoelastic properties of polymer-based materials. It represents the relationship between loss modulus (E'') and the storage modulus (E'). It signifies the fibermatrix interaction and the heterogeneity or non-uniform dispersion of fibers in the matrix system. The Cole-Cole plot of a homogenous system is reported as a smooth semi-circular arc whereas the heterogeneous system displays irregular or imperfect semicircles. The dielectric relaxation and viscoelastic response can never be predicted by a single tan δ peak. Therefore, it is particularly important to understand the structure-property relationship using the Cole-Cole method. The Cole-Cole plot of hybrid composites at a frequency of 1 Hz and 5 Hz are reported in Figures 12-13. The Cole-Cole plot of EP was significantly changed and leads to a higher degree of imperfection by the addition of reinforcing fibers. The curve of PCP shows less heterogeneity and proposed the poor fiber-matrix interfacial adhesion. The shape of curves for IM, CPC, and bilayer was pointed towards the good fiber-matrix interfacial adhesion.



Figure 12. Cole-Cole plots of neat epoxy and hybrid composites at a frequency of 1 Hz



Figure 13. Cole-Cole plots of neat epoxy and hybrid composites at a frequency of 5 Hz

CONCLUSIONS

The viscoelastic properties of a composite material are significantly influenced by the hybridization and fibers layering pattern. In this regard, the different layered hybrid composites were made and characterized. Based on the experimental work, the following conclusions can be made:

- 1. The dynamic modulus and the glass transition temperature of single fiber reinforced composites (CF-EP and PF-EP) were increased by the synergistic hybridization of cellulosic fibers. After the incorporation of PALF and COIR in a single matrix resin, the shortcomings of each type of fiber was cracked which results in the formation of synergistic hybrid material.
- 2. Amongst all the different layered hybrid composites, the trilayer CPC possesses lowest value of effectiveness coefficient 'C' which results that it occupies maximum stiffness and higher activation energy for transition glassy phase to rubbery phase.
- 3. The bilayer (P/C) and intimately mixed hybrid composites have approximately same modulus (E') and the glass transition temperature. However, the intimately mixed one has lower value of coefficient 'C' and it was due to the effective and uniform stress transfer from matrix to fiber and fiber to fiber

- 4. The stiffness and phase transition temperature (T_g) of the epoxy resin were increased considerably by the loading of PALF and COIR.
- 5. The storage modulus (E'), glass transition temperature (T_g), and width of tan δ curve were increased with the increase of frequency from 1 Hz to 5 Hz. This behavior proved the better fiber-matrix interaction at a higher frequency (short time).

Overall we can conclude that the fibers layering patterns significantly affect the performance of a hybrid composite material. The incorporation of PALF and COIR in a single matrix medium result the high performing material having significant potential to be used in various engineering applications.

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