

Experimental Investigation of *Pterocarpus Marsupium* Resin/LY 556 Epoxy Blended Hybrid Polymer Material

PRASATH SRINIVASAN^{1*}, DHARMALINGAM SOMASUNDARAM²,
RAJENDIRAN SUBRAMANIAN², SANTHANAM VAJJIRAM³

¹Department of Mechanical Engineering, Suguna College of Engineering, Airport Road, Nehru Nagar West, Coimbatore, Tamilnadu, 641014, India

²Department of Mechanical Engineering, RVS Technical Campus Coimbatore, Kumaran Kottam Campus, Kannampalayam, Coimbatore, Tamilnadu, 641402, India

³Department of Mechatronics Engineering, Rajalakshmi Engineering College, Chennai, Tamilnadu, 602105, India

Abstract: *In this work an attempt had been made to hybridise the Epoxy resin by incorporating the Pterocarpus Marsupium natural resin powder derived from the Pterocarpus Marsupium tree. The mechanical, dynamic mechanical, biodegradability and thermal stability of the blended polymer was evaluated at different Pterocarpus Marsupium resin particulate loading (10, 15, 20, 25, 30 and 35 v/v %). The composite specimens were fabricated by using hand layup method. The mechanical properties such as tensile strength, flexural strength had shown significant improvement than the tensile modulus and flexural modulus due to blending, the experimental results indicated that the better properties of the blended polymer were obtained at 30% v/v Pterocarpus Marsupium resin incorporated Epoxy polymer. Soil burial test revealed that the incorporation of bio resin resulted in weight loss of the blended polymer over prolonged period of time.*

Keyword: *mechanical properties, epoxy resin, Pterocarpus marsupium, polymer fillers, DMA analysis*

1. Introduction

Because of its high strength-to-weight ratio, low cost, simplicity of availability, and ability to adjust the mechanical properties of polymer composites according to application needs, polymer composites have been widely used. Researchers [1-6] have been interested in using natural fibres and biopolymers as one of the constituent elements in composites for several decades, owing to environmental concerns and government laws. Synthetic polymers are combined with natural fibres and fillers to improve performance while reducing negative environmental impact. Fibers are used to improve the strength of the composite, while fillers are used to improve the moduli of the material and the thermal and biodegradability of the composite. The effect of combining epoxidised natural rubber (ENR) with Epoxy resin was examined by Chuayjuljit et al [7]. The epoxy resin's tensile and flexural strength were both reported to be reduced after the filler was added. The impact strength of the composite, on the other hand, increased up to 5% with the inclusion of rubber and reduced with the addition of rubber in epoxy. The thermal breakdown temperature and glass transition temperature of the hybrid polymer were found to be between 351°C and 364°C and 51°C and 58°C, respectively, according to the thermal and DMA analysis results, which are similar to the values for epoxy resin. Dinesh Kumar et al [8] modified the Epoxy resin by integrating natural rubber in various quantities. Due to inadequate compatibility between the rubber and epoxy polymer, the blended matrix's thermal and DMA properties did not considerably improve. The impact toughness of the blended epoxy was increased by 3% and then began to decline when more filler material was added. Alsagayar et al. [9] achieved a hybridization of Epoxy resin with bio-based epoxidised palm oil. The effect of blending on the mechanical properties of epoxy resin was investigated in this study. The integration of the bio filler resulted in a modest drop in the blend's tensile and flexural strength, according to the trials. However, the use of bio mix enhanced the impact strength and strain to failure. At 20 percent v/v of the bio filler, the maximum impact strength was 59.19 J/m².

*email: prasathvasan07@gmail.com

Ruamcharoen et al. [10] found that combining Epoxidised Natural Rubber (ENR) with epoxy resin did not result in a substantial increase in the blend's mechanical qualities. The glass transition temperature (T_g) was shifted to the lower side using differential scanning calorimetry (DSC). Pronob Gogoi et al. [11] produced bio composites by combining a *Jatropha curcas* oil-based alkyd and expanded graphite (EG) with epoxy resin. At 5% EG loading, the characteristics of the blended composite were reported to have improved significantly. The blended composite with 5% filler loading had a maximum tensile strength of 43MPa, while the elongation to break was lowered with the addition of the EG. TGA was used to determine the thermal stability of the composite, which was found to be 205°C without the addition of EG and 243°C with a 5 percent EG loading. At 18 percent filler loading, Hafiezal et al. [12] found that combining bio *jatropha* filler with epoxy resulted in enhanced dynamic and thermal performance of the blended composite. By integrating waste peanut shell powder in epoxy resin, Prabhakar et al. [13] produced bio composites, and studies were undertaken to determine the effect of filler content on the mechanical and thermal properties of the composite. The addition of the filler improved the mechanical properties of the blended composite, according to the test results. With 15 wt% filler loading, the composite achieved maximum thermal stability and tensile strength. Vinod Kumar et al. [14] extracted nanofiber from palm petiole and used it as a reinforcement in epoxy resin with various filler loadings. The results showed that the nano filler had a favourable effect at 3 wt%, and that the silane treatment of the nano fibre improved the composite's qualities when compared to the untreated nano fibre integrated composite. Vimalanathan et al. [15] created a Polyester composite by varying the quantities of *Shorea robusta* bio filler. The mechanical and thermal properties of the hybrid composite were assessed, and it was found that adding the bio filler improved the mechanical and thermal properties. The composite with 20% v/v filler material had better characteristics. Due to the addition of bio filler, a biodegradability test revealed that weight loss was expedited. Several authors [16 - 20] have attempted to blend bio-derived polymers with epoxy resin, with the majority of experimental results indicating a small increase in mechanical and thermal properties due to the addition of plant-based fillers to epoxy resin. As a bio filler in the epoxy matrix, a unique plant-based *Pterocarpus Marsupium* resin powder was used in this study. The impact of filler loading on the composite's mechanical, dynamic mechanical, and thermal properties was investigated.

2. Materials and methods

2.1. Materials

In this work, a natural filler material extracted from the *Pterocarpus Marsupium* tree bark had been used as filler in the epoxy composite. The resin was available in solid form and it was procured from P.V. enterprises, Kanchipuram. The resin was dried for two days under sunlight to remove any residual water present in it, then it was powdered into fine particles using ball milling process. The particle size was kept as 7-10 microns. The density of the filler material was estimated as 1.08 g/cm³. Epoxy resin was used as the matrix in this work. Epoxy composites were prepared by adding the bio filler at different volume fractions (10, 15, 20, 25, 30 and 35 v/v%) with the epoxy resin. A mechanical stirrer was used to thoroughly mix the epoxy and hardener with the bio filler to achieve uniform distribution of the constituent materials. Hand layup process was used to fabricate the composite samples followed by medium compression with 500N load for 24 h. The cured samples were then cut to the required dimensions as per the ASTM standards.

2.2. Testing methods

Tensile and flexural experiments were evaluated by using the Instron universal testing machine. The tensile and flexural experiments were carried out by following ASTM D638 and ASTM D790 standards respectively. The tensile test was conducted at a crosshead speed of 5 mm/min. Impact toughness of the composite sample was measured by Izod impact testing machine as per ASTM D256 standard. All the experiments were conducted for five samples each and the average value was tabulated for analysis.

Visco elastic properties of a material determine the extent to which they behave as solid and the transition temperature above which they exhibit viscous fluid properties. Dynamic mechanical Analysis (DMA) was used to assess the storage modulus (E''), loss modulus (E') and glass transition temperature of the composite material. SEIKODMAI-DMSC 6100 instrument was used in this work to determine the visco elastic properties of the composite material. The experiment was conducted in Nitrogen environment by increasing the temperature with 5°C increments from a temperature of 30°C to a final temperature of 180°C . DMA test was carried out in tensile mode under 1Hz, 5Hz and 10 HZ frequencies.

Thermal stability of the composite specimen was investigated by a TG/DTA 6200 SEIKO TGA analyzer over a temperature range of $0-800^{\circ}\text{C}$ with $20^{\circ}\text{C}/\text{min}$ increment. The powder was heated in nitrogen environment to prevent oxidation. Since the bio filler had been used as one of the constituent materials, bio degradability test was conducted with a simple soil burial test. The specimen is cut to a $10\text{mm} \times 10\text{mm} \times 3\text{mm}$ size and it was buried under wet soil to allow natural degradation. The sample was taken at regular intervals of 10 days to weigh the sample, the weight gain/loss was recorded and a plot was drawn to study the bio degradability of the composite.

Morphological studies were carried out using SEM to investigate the dispersion of filler material in the polymer matrix. It was performed in Hitachi S 3400 N scanning electron microscope. Composite samples were sputter-coated with gold to avoid charging.

3. Results and discussions

The present work aims at the use of *Pterocarpus Marsupium* as bio filler with epoxy resin to improve the properties of hybrid polymer material. Experiments were carried out to assess the impact of bio filler loading bio filler on the properties of epoxy resin.

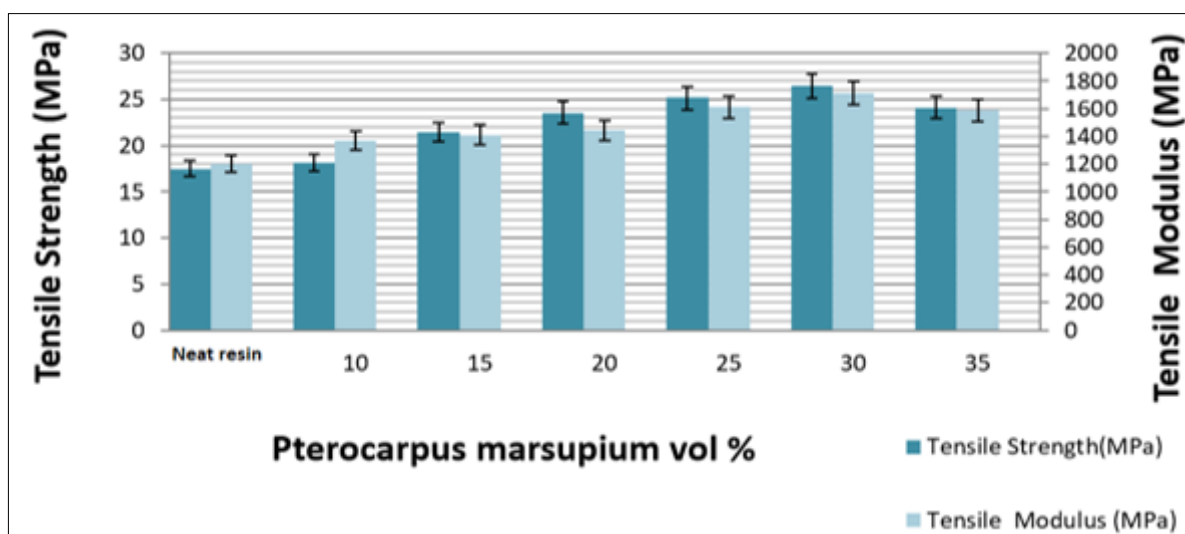


Figure 1. Tensile properties of *Pterocarpus marsupium* with epoxy blended hybrid material

3.1. Mechanical properties

The tensile strength and tensile modulus of the bio filler incorporated epoxy composite is presented in Figure 1. The figure shows that the incorporation of the bio filler considerably improves the tensile properties of the composite and a maximum value of tensile strength was observed for the specimen with 30 v/v% of bio filler. The tensile strength of neat resin was obtained as 17.5 MPa while the maximum value of the tensile strength was obtained as 26.48MPa. Also the increasing trend of the tensile strength showed better compatibility between the polymer and the bio filler, and also the uniform distribution of the bio filler in the matrix. The tensile modulus also had shown an improvement of 42.6% for the specimen with 30 v/v% bio filler from the neat epoxy resin. Further addition of bio filler in the epoxy resin resulted in a decrease of the values, which may be due to the coalescence of the bio filler particles

in the matrix. This leads to the poor wetting of the bio filler by the matrix and poor adhesion between the matrix and bio filler.

The Scanning Electron Micrographs (SEM) of 30 and 35 volume percent bio filler added hybrid polymer materials are presented in Figures 2, 3.

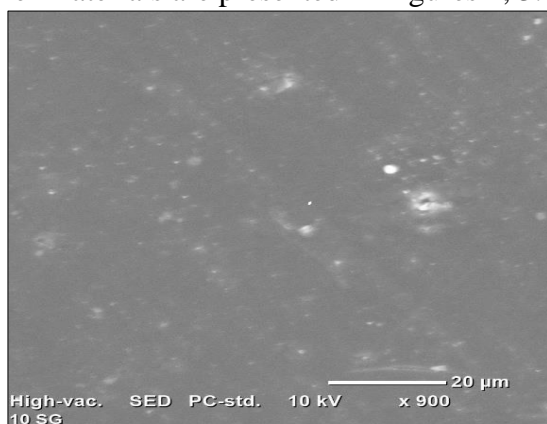


Figure 2. Tensile fractured specimen of Epoxy composite with 30 vol % *Pterocarpus Marsupium* bio filler

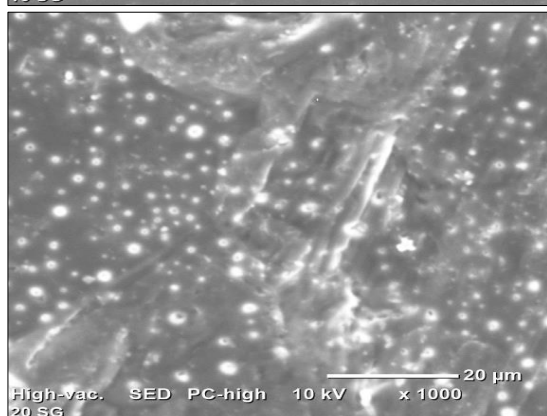


Figure 3. Tensile fractured specimen of Epoxy composite with 35 vol % *Pterocarpus Marsupium* bio filler

The SEM image in Figure 2 shows that the *Pterocarpus Marsupium* bio filler is uniformly dispersed in the epoxy matrix. The load carrying capacity of the composite was improved due to the even dispersion of the bio filler as this enhances the adhesion between the filler and epoxy. Further, Figure 3 shows the SEM image of the composite with 35 volume % of bio filler. It is evident from the SEM images that the addition of volume % of natural resin starts to agglomerates. Due to agglomeration the natural resin are unable to transfer the load evenly and hence the mechanical properties start to decline.

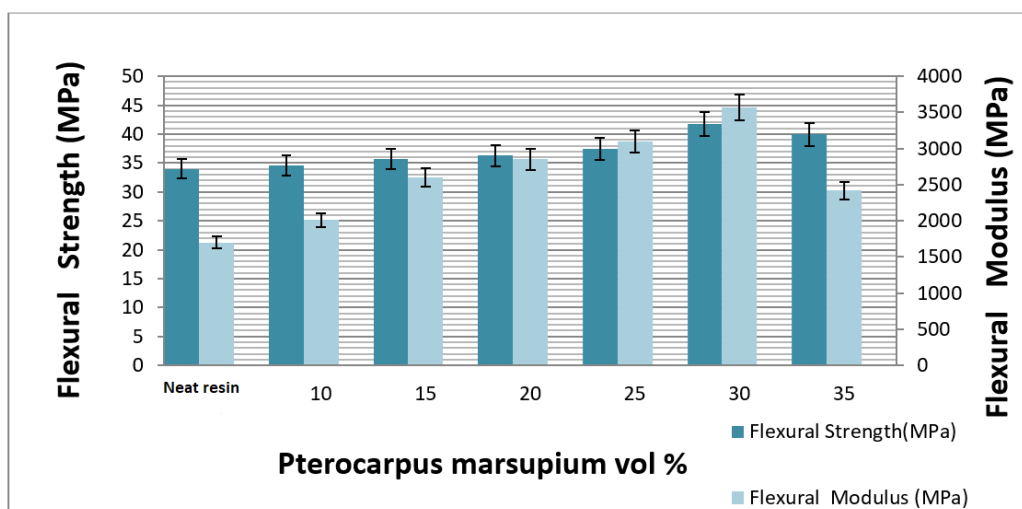


Figure 4. Flexural properties of *Pterocarpus marsupium* with Epoxy Blended Hybrid Material

Figure 4 shows the flexural strength and modulus of the neat epoxy and its composite. The flexural strength and modulus of the neat epoxy were obtained as 34 and 1700 MPa respectively. From the Figure 4 it is found that with the addition of *Pterocarpus marsupium* bio filler resulted in improvement in the properties similar to the tensile strength. An improvement of 22.7% was obtained in the flexural strength for the specimen incorporated with 30% v/v bio filler. Addition of bio filler beyond 30 v/v% resulted in reduction in the flexural properties similar to the tensile properties.

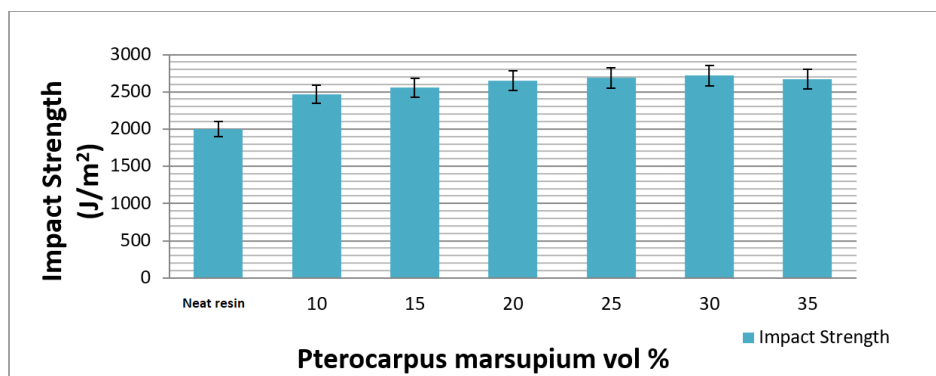


Figure 5. Impact properties of *Pterocarpus marsupium* with Epoxy Blended Hybrid Material

The Figure 5 shows the impact strength of the *Pterocarpus Marsupium* hybrid polymer material. From the figure it was observed that the impact strength for the neat epoxy is 2000 J/m², and for 10, 15, 20, 25, 30 and 35 volume % hybrid polymer materials the impact strength are 2,469, 2,557, 2,648, 2,687, 2718, 2,671 J/m² respectively. The maximum impact strength of 2,718 J/m² was observed for the hybrid polymer materials with 30 volume % of *Pterocarpus Marsupium*. The lower value of impact strength for the epoxy resin can be attributed due to its brittleness, whereas *Pterocarpus Marsupium* has better adhesion with the epoxy resin. This results in better ductility of the composite and impact strength was increased by 35.9%.

3.2. Dynamic mechanical analysis

In this study, the effect of *Pterocarpus Marsupium* filler loading on the storage modulus and damping factor of the epoxy hybrid polymer material was studied.

The effect of *Pterocarpus Marsupium* loading and the temperature on the storage modulus of epoxy composite at 10 HZ frequency is presented in Figure 6. The plots showed that the energy absorption by the epoxy composite improved due to the addition of the *Pterocarpus Marsupium* bio filler. Neat epoxy showed lower values of the storage modulus value, indicating lower stiffness value of the polymer. It is also observed that the glass transition temperature of the epoxy composite have not shown significant change due to the addition of the bio filler. Though the T_g value increased by 1-2°C. The maximum value of storage modulus was obtained for the epoxy composite with 25% of bio filler.

Another significant observation from Figure 6 is that the incorporation of *Pterocarpus Marsupium* natural resin improved the storage modulus (E'') in both glassy and rubbery region. Studies carried out showed that the storage modulus measured in shorter time (high frequency) results in higher values, whereas exposing over a longer time (low frequency) results in lower values. This is due to the molecular rearrangement of material in an attempt to minimize localized stresses.

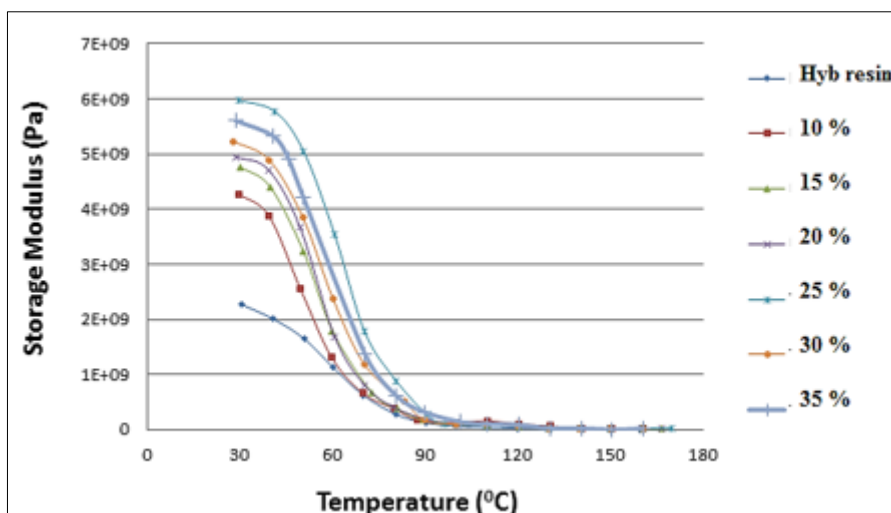


Figure 6. Storage modulus of *Pterocarpus Marsupium* with Epoxy Blender Hybrid at 10 Hz Frequency

Damping factor (Tan δ)

Damping factor (Tan δ) is the ratio between loss modulus (E'') and storage modulus (E') of the material which shows the degree of molecular movement in the polymer chain and indicates the energy dissipation of material during loading. Figure 7 shows the effect of bio filler addition with the epoxy matrix on the damping factor values.

Figures 7 show the damping factor of incorporation of *Pterocarpus marsupium* natural resin reported hybrid material. The results show that the addition of *Pterocarpus marsupium* natural resin up to 30 volume % increases the damping factor of the polymer material. This indicates that the polymer materials with lower volume % *Pterocarpus marsupium* natural resin loading have higher interaction between synthetic resin with natural resin. Because of better interaction, the energy dissipation of natural materials increases. However, the higher natural resin content (more than 30 volume %) reduces the energy dissipation, which is established as can be seen from Figures 7. The peak height of Tan δ (damping factor) for neat epoxy resin is lower compared to the peak height of *Pterocarpus marsupium* natural resin added hybrid polymer material. This indicates that higher loading of natural resin increases the stiffness of composite material.

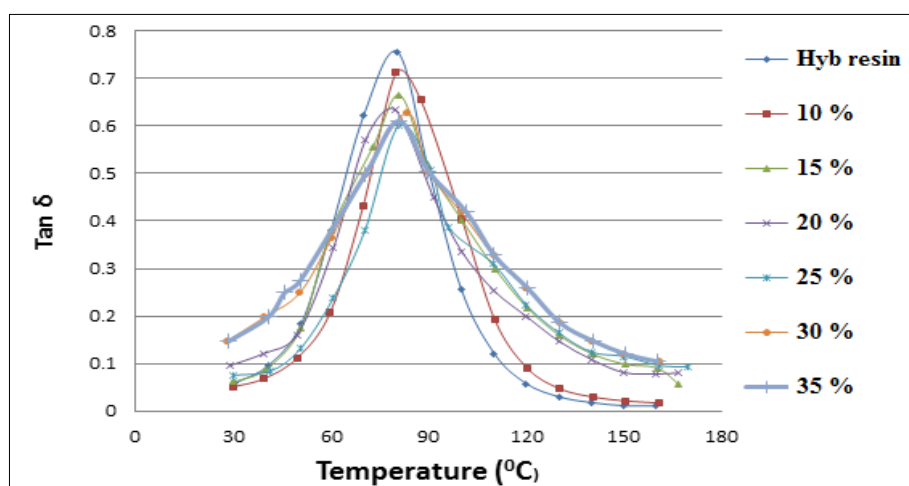


Figure 7. Tan δ of *Pterocarpus Marsupium* with Epoxy Blender Hybrid Material at 10 Hz Frequency

3.3. Cole- Cole plot

Linear viscoelastic behavior of the polymer composite within the glass transition temperature can be addressed using Cole-Cole plot. Cole-Cole plot is obtained by plotting loss modulus (E'') against storage modulus (E') at 10 HZ frequencies. This plot exposes the structural changes occurring in the cross-linked polymer due to the addition of natural *Pterocarpus marsupium* resin. It is represented by the following the nature of the curve indicates the homogeneity or heterogeneity nature of the material.

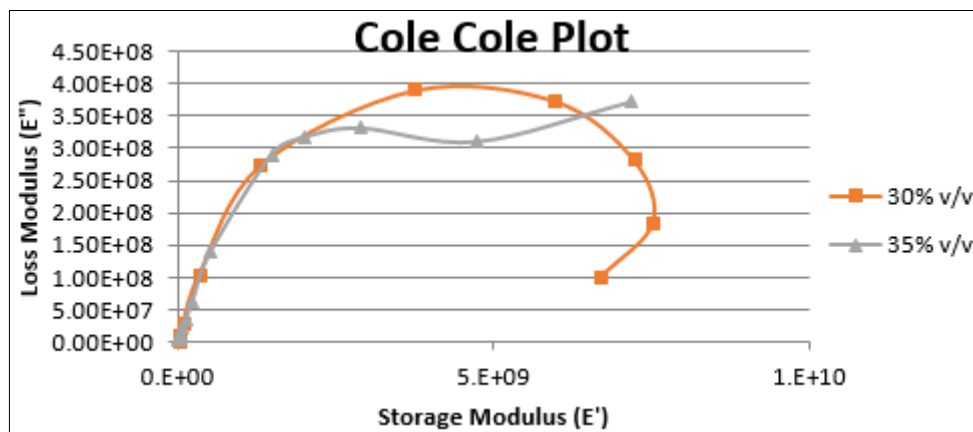


Figure 8. Cole- Cole Plot *Pterocarpus Marsupium* with Epoxy Blender Hybrid Material

Figure 8 presents the Cole-Cole plot, which indicates homogeneity in the properties of the hybrid polymer material. It is evident from the figure that the addition of 30 volume % *Pterocarpus marsupium* natural material results in homogenous material. Further addition of *Pterocarpus marsupium* natural resin (more than 30 volume %) changes the material behavior from homogeneity to heterogeneity. This is observed from the Cole-Cole plot wherein the curve is closer to semicircle for 30 Volume % and deviation from semicircular nature for 35% v/v indicates the composite becoming heterogeneous.

3.4. Thermogravimetric analysis

Thermogravimetric analysis (TGA) of neat epoxy resin and the hybrid polymer material are given in Table 1. The Initial Degradation Temperature (IDT), Mid Degradation Temperature (MDT) and Final Degradation Temperature (FDT) were tabulated based on the experimental results. The thermal stability of the epoxy with *Pterocarpus marsupium* resin incorporated hybrid polymer material was found to be slightly more than that of neat epoxy resin. The results indicate a favorable effect on the thermal stability due to incorporation of *Pterocarpus marsupium* hybrid material in the epoxy matrix. However, a slight improvement in the TGA values was observed due to the *Pterocarpus marsupium* natural resin materials. The first stage of weight loss was due to the evaporation of water from the sample. Hence, the addition of *Pterocarpus marsupium* natural resin in the epoxy matrix results in better thermal stability of the hybrid polymer material. Table 1 shows the stability in the epoxy resin up to 391°C. After this temperature, the resin degrades rapidly and its residue content is merely 0.2%. Whereas for *Pterocarpus marsupium* blended hybrid polymer, it is stable up to 401°C for 30% v/v content of bio filler with residue content of 19%. The initial degradation temperatures for the neat resin and hybrid polymer with 30% v/v bio filler are 302 and 313°C respectively

Table 1. Thermogravimetric analysis *Pterocarpus Marsupium* with Epoxy Blender Hybrid Material

Composition	Degradation temperature (°C)			Residue (%)
	IDT	MDT	FDT	
Net Resin	302	352	391	0.2
10% VF PM	305	373.4	397	9

15% VF PM	307	375.4	398	10
20% VF PM	308	375.6	398	15
25% VF PM	311	385.7	399	17
30% VF PM	313	386.4	401	19
35 % VF PM	316	391.3	407	18

3.5. Biodegradability test

Material sample was buried under a wet soil to evaluate its biodegradability. Influence of biodegradability of the *Pterocarpus marsupium* hybrid epoxy material is shown in Figure 9. Initial weight of the sample was noted before burying it under the soil. The sample was then taken at regular intervals, and its weight was measured to observe for any changes in its weight.

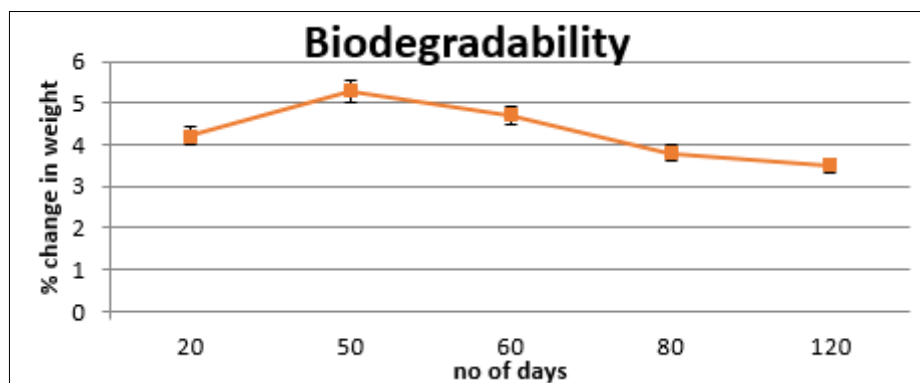


Figure 9. Biodegradable testing of *Pterocarpus marsupium* natural resin hybrid materials

The total weight loss of the sample was observed as 3.5% after 120 d. The weight of the sample was observed to increase at the initial stages due to moisture absorption, but the weight started reducing subsequently. This reduction in weight was attributed to the reaction of micro- and macro-organisms with the composite sample after prolonged duration. While comparing the results with those of other natural fiber- and filler-reinforced polymer composite, it was found that the material possessed better biodegradability. Hence, incorporation of *Pterocarpus marsupium* hybrid material results in the better environment friendly polymer material.

4. Conclusions

The influence of different volume % of *Pterocarpus marsupium* / LY 556 epoxy blended hybrid polymer material on mechanical, dynamic mechanical analyses and TGA with bio-degradability was analyzed. The mechanical, dynamic mechanical analysis, TGA and bio degradability test results revealed that the inclusion of 30 Vol % of *Pterocarpus marsupium* resin in the LY556 epoxy matrix resulted in better mechanical properties. Further addition of bio resin reduced the material properties due to non-uniform distribution. And filler agglomeration, which is evident from the SEM images.

References

1. MAHMUD, S., LONG, Y., TAHER, M. A., HU, H., ZHANG, R., ZHU, J., Fully Bio-based Microcellulose incorporated Poly (butylene 2, 5-furandicarboxylate) transparent composites: preparation and characterization. *Fibers and Polymers*, **21**(7), 2020, 1550-1559.
2. SANTHANAM, V., CHANDRASEKARAN, M., Effect of Surface Treatment on the Mechanical Properties of Banana-Glass Fibre Hybrid Composites. *Applied Mechanics and Materials* 591, 2014, 7-10.
3. VIMALANATHAN, P., SURESH, G., RAJESH, M., MANIKANDAN, R., KANNA, S. R., SANTHANAM, V., A Study on Mechanical and Morphological Analysis of Banana/Sisal Fiber Reinforced IPN Composites. *Fibers and Polymers*, 1-8, 2021



4. BHASKAR, K. B., SANTHANAM, V., DEVARAJU, A., Dielectric Strength Analysis of Acacia Nilotica with Chemically Treated Sisal Fiber Reinforced Polyester Composite. *Digest Journal of Nanomaterials and Biostructures*, **15**(1), 2020, 107-13.
5. KABIR, M. M., AL-HAIK, M. Y., ALDAJAH, S. H., LAU, K. T., WANG, H., Impact Properties of the Chemically Treated Hemp Fibre Reinforced Polyester Composites. *Fibers and Polymers*, **21**(9), 2020, 2098-2110.
6. MOCHANE, M. J., SEFADI, J. S., MOTSOENENG, T. S., MOKOENA, T. E., MOFOKENG, T. G., MOKHENA, T. C., The effect of filler localization on the properties of biopolymer blends, recent advances: A review. *Polymer Composites*, **41**(7), 2020, 2958-2979.
7. CHUAYJULJIT, S., N. SOATTHIYANON, P. POTIYARAJ., Polymer blends of epoxy resin and epoxidized natural rubber. *Journal of applied polymer science*, 102(1) 2006, 452-459.
8. KUMAR, K. DINESH, B. KOTHANDARAMAN, Modification of (DGEBA) epoxy resin with maleated depolymerised natural rubber. *Express Polym Lett*, 2, 2008, 302-311.
9. ALSAGAYAR, Z. S., RAHMAT, A. R., ARSAD, A., FAKHARI, A., Mechanical properties of epoxidized palm oil/epoxy resin blend. In *Applied Mechanics and Materials* 695, 2015, 655-658.
10. RUAMCHAROEN, P., UMAREE, S., & RUAMCHAROEN, J., Relationship between tensile properties and morphology of epoxy resin modified by epoxidised natural rubber. *Journal of Materials Science and Engineering*, **5**(5), 2011, 504-510.
11. GOGOI, P., BORUAH, M., BORA, C., DOLUI, S. K., Jatropha curcas oil based alkyd/epoxy resin/expanded graphite (EG) reinforced bio-composite: Evaluation of the thermal, mechanical and flame retardancy properties. *Progress in Organic Coatings*, **77**(1), 2014, 87-93.
12. HAFIEZAL, M. R. M., KHALINA, A., ZURINA, Z. A., AZAMAN, M. D. M., HANAFEE, Z. M., Thermal and flammability characteristics of blended jatropha bio-epoxy as matrix in carbon fiber-reinforced polymer. *Journal of Composites Science*, **3**(1), 2019, 6.
13. PRABHAKAR, M. N., SHAH, A. U. R., RAO, K. C., SONG, J. I., Mechanical and thermal properties of epoxy composites reinforced with waste peanut shell powder as a bio-filler, *fibers and polymers*, **16**(5), 2015, 1119-1124.
14. KUMAR, T. V., CHANDRASEKARAN, M., SANTHANAM, V., SUDHARSAN, V. D., Effect of coupling agent on mechanical properties of palm petiole nanofiber reinforced composite. In *IOP Conference Series: Materials Science and Engineering* 183(1), 2017, 012006.
15. VIMALANATHAN, P., VENKATESHWARAN, N., SANTHANAM, V., Mechanical, dynamic mechanical, and thermal analysis of Shorea robusta-dispersed polyester composite. *International Journal of Polymer Analysis and Characterization*, **21**(4), 2016, 314-326.
16. WANG, RONGPENG, THOMAS P. SCHUMAN., Vegetable oil-derived epoxy monomers and polymer blends: a comparative study with review. *Express Polym. Lett* **7**(3), 2013, 272-292.
17. SHARMA, S. 'Fabrication and Characterization of Polymer Blends and Composites Derived from Biopolymers', 2008
18. JIANG, L., ZHANG, J., Biodegradable polymers and polymer blends. *Handbook of Biopolymers and Biodegradable Plastics: Properties, Processing and Applications*, 2012, 109-128.
19. HU, B. Biopolymer-based lightweight materials for packaging applications. *Lightweight materials from biopolymers and biofibers*, 2014, 239-255.
20. AHMETLI, G., DEVECI, H., SOYDAL, U., GURLER, S. P., ALTUN, A., Epoxy resin/polymer blends: improvement of thermal and mechanical properties. *Journal of applied polymer science*, **125**(1), 2012, 38-45.

Manuscript received: 19.09.2021