**Influence of Graphite Filler in Hybrid Casuarina/Graphite Filled GFRP Composite**

Y. Shanti  
Research Scholar, Department of Mechanical Engineering, GITAM, Hyderabad, India.  
A. Satyadevi  
Professor, Department of Aerospace Engineering, GITAM, Hyderabad, India.

**Abstract** - The highest actual in advanced composites is lower weight and cost than improved mechanical characteristics. The probable promising of hybrid filler composites is that they exhibit a stable performance by overcoming the limitations of a single filler. The present work involves this attitude by taking hybridized natural and artificial filler. The mechanical and thermomechanical effects of a mixed filler gfrp composite containing equivalent wt%’s fillers, for example, casuarina wood residue and graphite, were researched. A significant increment in mechanical and thermomechanical properties is obtained for hybrid filler composite, compared to the published results of single filler composites. The addition of graphite is highly proposed for the wood-filled composite, which can assist in reducing voids in the composite, which improves the mechanical characteristics with less mechanical loss of the hybrid polymer composite.

**Index Terms** - Hybrid fillers; GFRP; Polyester resin; Mechanical and thermomechanical characterization.

**INTRODUCTION**

The research in polymer composites reinforced hybrid fillers made huge changes to make it superior to composites reinforced single filler [1]-[4]. Multiple fillers can be used to integrate the characteristics of all fillers in a single composite. By worrying about better diffusion of the hybrid fillers, good compatibility with the matrix can lead to better properties [5]-[7]. But still, the strength of multi-filled composites depends upon surface nature, distribution and content of parental filler. Masudul Hassan has studied the mechanical behavior of polypropylene (PP) composites hybridized with fillers such as betel nut/seaweed at varying filler loading. He observed that PP composite with bi fillers had enhanced properties than single filler, i.e., betelnut [8]. Mishra and Padhee have taken rice husk and fly ash as fillers to reinforce epoxy composite, which may be used in humid content applications such as washing machines, household gadgets, and packaging [9]. Nowadays, synthetic fillers usage is decreasing due to its cost, health, and environmental hazards through its mechanical performance is more[10],[11]. Similarly, natural fillers also have limitations in different applications due to moisture absorption, lesser density and low thermal stability, which results in debonding of composite constituents [12].

Wood is a renewable resource with less weight, low cost, durable, and easy availability among the many natural resources, attracting researchers' attention for so many years. For wood, behavioral attributes under mechanical loading include a known issue due to its natural unevenness, irregularity and anisotropy. Therefore comparative to other natural fillers like sisal, jute, palm, coconut coir, bamboo, hemp, wheat, kenaf, ramie, cotton etc., wood is being less strengthened to produce a usable end-product [13]. A couple of researchers have reported that after all chemical treatment of wood filler, composites’ mechanical and thermomechanical properties decrease because of weak interaction between the particle and Polymer [14]. [15].To improve the interaction between filler and matrix, many authors attempted by hybridizing wood fillers with the inorganic fillers, apart from chemical treatment of fillers, thereby getting enhanced mechanical and thermomechanical properties [16]-[18]. Zain has considered oil palm hybridized with wood flour to reinforce propylene to know its mechanical performance. He found a decrement in properties corresponding to the increased filler size and addition [19]. Organic wood fillers have low density, biodegradability and inorganic fillers have cost efficiency and abundance; mixing these fillers mostly attain appreciable strength of filler composites [20].

Few researchers reported hybridizations of wood and synthetic fillers, which is the motivation of the present work. The fundamental focus of the study is to compose a high geared partial green composite, which is of less cost. Therefore, to minimize damping and usage of synthetic fillers and further strengthen the wood filler composite, hybridization of casuarina wood (CW) with the graphite filler (GR) containing 5wt% each has been done in the current project within good accordance with the outcome. Compared mechanical and thermomechanical properties with the published results of single fillers, CW and GR[21].

**MATERIALS PREPARATION & EXPERIMENTAL PROCEDURE**

**I. Materials**

The investigation includes E-glass chopped strand mat 450 GSM (450 g/m²) density. Unsaturated Polyester Resin(UPR) was used as a matrix, with a viscosity of 450 and a specific gravity of 1.11. MEKP (Methyl Ethyl Ketone Peroxide) is taken as a hardener and mixed with the resin in the ratio of 33:100. The natural filler, i.e., Casuarina wood sawdust particles (CW) of size 60µm and graphite powder (GR) of size 80µm, were used in the fabrication of hybrid composite (CWGR) to enhance the mechanical properties.
II. Fabrication of Composite Laminate

Table 1 represents the details of CWGR composite laminate and the published details of single filler composites and the composite without filler coded as CW, GR and NR.

<table>
<thead>
<tr>
<th>Specimen Code</th>
<th>Matrix (wt%)</th>
<th>Glass fiber (wt%)</th>
<th>CW (wt%)</th>
<th>GR (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CWGR</td>
<td>65</td>
<td>25</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>CW           *</td>
<td>65</td>
<td>25</td>
<td>10</td>
<td>Nil</td>
</tr>
<tr>
<td>GR           *</td>
<td>65</td>
<td>25</td>
<td>Nil</td>
<td>10</td>
</tr>
<tr>
<td>NR           *</td>
<td>75</td>
<td>25</td>
<td>Nil</td>
<td>Nil</td>
</tr>
</tbody>
</table>

*From reference [21].

I treated a sieved CW sawdust filler with 10% NaOH solution. Post chemical treatment, the filler was washed with purified water and baked in the microwave for 24 h at 70˚C to remove the wetness of the wood filler, as shown in Figure 2. As per the proportions given in table 1, hybridized GR and CW fillers are stirred in the matrix by avoiding lumps. According to the hand layup method, three glass fiber(GF) sheets were placed above the other, with the matrix spread between the fibers to form a hybrid filler composite (CWGR). Properly moved each layer to assure uniform dispersion of resin over the glass mat. The whole assembly was permitted to cure full day under weather conditions.

III Mechanical Characterization

The specimen was tested for tensile and flexural strengths by a UTM of the 10-ton capacity of about 5mm/min to measure the elasticity and bending resistance according to ASTM D638 and 790. The specimens, prepared according to ASTM D256, were tested to find impact strength using an Izod testing machine of 25 Joule capacity. The energy absorbed in breaking the specimen was then recorded. Hardness was obtained with the Barcol impresser instrument as indicated by ASTM D2583. I determined each mechanical strength by taking the average value of 5 testing samples results. The testing sample for each test is displayed in Figure 3.
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IV Thermomechanical Characterization

The damping effects of the CWGR composite were assessed using a Dynamic Mechanical Analyzer (DMA) consisting of a 3-point bending fixture. It determines the viscoelastic character of the material. With this DMA test, determining properties such as storage modulus($E'$), loss modulus($E''$), tan delta(Tan δ) as a function of temperature, and also glass transition temperature, i.e., Tg for this composite. Here, the sample containing proportions as 50 mm × 10 mm × 3 mm is subjected to sinusoidal load at a working temperature span of 25°C to 130°C and a frequency of 1 Hz. Figure 4 represents the test sample for the DMA test.

RESULTS AND DISCUSSION

A detailed investigation was carried out on fractured test samples, as shown in Figure 5, to study the mechanical properties of hybrid CW and GR fillers reinforced grp composite.

Mechanical properties
Table 2 indicates the mechanical characteristics of hybrid filler composite and the published results of CW, GR and NR composites.

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Composite</th>
<th>Tensile Strength, (MPa)</th>
<th>Flexural Strength, (MPa)</th>
<th>Impact Strength, (MPa)</th>
<th>Hardness</th>
<th>Tensile Modulus, (MPa)</th>
<th>% Elongation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>CWGR</td>
<td>95.54</td>
<td>123</td>
<td>84.48</td>
<td>42.4</td>
<td>1045.7</td>
<td>14.3</td>
</tr>
<tr>
<td>2</td>
<td>CW*</td>
<td>63.54</td>
<td>64.86</td>
<td>69.01</td>
<td>42.2</td>
<td>699.9</td>
<td>13.88</td>
</tr>
<tr>
<td>3</td>
<td>GR*</td>
<td>86.77</td>
<td>95.38</td>
<td>67.54</td>
<td>48.2</td>
<td>1053.6</td>
<td>11.5</td>
</tr>
<tr>
<td>4</td>
<td>NR*</td>
<td>81.75</td>
<td>121</td>
<td>82</td>
<td>41</td>
<td>905.67</td>
<td>12</td>
</tr>
</tbody>
</table>

*From reference [21].

The CW composite has higher ductility and can restrain the deformation of the matrix before fracture. From the stress-strain behavior of single CW and GR fillers, one can conclude that deformation at break decreased compared to the hybrid CW/GR fillers as shown in Figure 6A. The reason for this decrement is due to the poor interface between the filler and the polyester resin. The other feasible cause is the high viscosity of CW filler, which results in a nonhomogeneous mixture of resin and CW filler during the preparation of composites. This may result in weak boundaries between the particles and bubbles trapped during the preparation of CW composite.

![Figure 6](image)

(A) STRESS-STRAIN CURVE, (B) MECHANICAL PROPERTIES OF COMPOSITES

The tensile test was conducted for the CWGR composite and observed increased strength and modulus compared to CW, GR and NR as shown in Figure 6(B). This strength reaches the highest value of 95.54 MPa at 10 wt% of hybrid fillers, as shown in table 2. This improvement indicates that the hybrid fillers are properly dispersed in the matrix, resulting in increased interfacial area and interaction between the fillers and the matrix. Therefore, some authors reported a reduction in the volume of clusters and voids and effective stress transfer compared to single filler composites [22]-[24]. The current study showed that CWGR had increased flexural strength because of the improved resistance to the bending of the composite. It may achieve greater adhesion of filler particles with the matrix, which indicates a good chemical balance of the filler/matrix, thereby obtaining effective stress transfer.

The extent of impact energy for the composite relies on elements, the essential characteristics of the composite and components connection. Figure 6(B) shows the improvement in impact strength for the CWGR composite compared to the single filler composites and even NR. The fracture mechanisms noticed in polymer composite include matrix splitting and fiber withdrawal during Izod testing. CW and GR may emerge filler particle agglomeration around stress concentration that requires low strength for a crack extension. The hardness of an FRP composite system is a direct result of the composite, how it is cured. Also, natural filler polymer matrix composite cures slightly slower than synthetic filler matrix composite because of some moisture absorption present in the parental particles. The stiffer and harder the filler particles and resin, the higher the hardness level obtained for GR composite according to the values given in Table 2. As CW particles are soft, CW composite has less hardness value. The presence of hard graphite particles in CWGR may increase the hardness compared to CW and NR.

**II Thermomechanical Properties**

In the DMA study, E1, E11, Tan δ and Tg concerning temperature were observed and recorded in Table 3.
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TABLE 3
THERMOMECHANICAL PROPERTIES OF COMPOSITES

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Composite</th>
<th>E₁, (MPa)</th>
<th>E₁₁, (MPa)</th>
<th>Tan delta</th>
<th>T₉ for E₁ (°C)</th>
<th>T₉ for E₁₁ (°C)</th>
<th>T₉ for Tan delta (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>CWGR</td>
<td>2430</td>
<td>255</td>
<td>0.32</td>
<td>61.9</td>
<td>74.5</td>
<td>82</td>
</tr>
<tr>
<td>2</td>
<td>CW*</td>
<td>1410</td>
<td>176</td>
<td>0.35</td>
<td>62</td>
<td>70.2</td>
<td>87.2</td>
</tr>
<tr>
<td>3</td>
<td>GR*</td>
<td>3090</td>
<td>313</td>
<td>0.35</td>
<td>68.6</td>
<td>77</td>
<td>95.9</td>
</tr>
<tr>
<td>4</td>
<td>NR*</td>
<td>2410</td>
<td>250</td>
<td>0.33</td>
<td>62</td>
<td>71.5</td>
<td>90</td>
</tr>
</tbody>
</table>

*From reference [21].

Figure 7 shows the storage modulus of gfrp composites with and without fillers go through the raised temperature (0 - 130°C). CWGR composite has an increase in E₁ upto 30°C, compared to CW and NR. The Storage modulus of single filler composites such as GR and CW is 3090 MPa and 1410 MPa, respectively. In incorporating, CW filler leads to a noticeable reduction in composite strength attributable to the cellulose deficit during chemical treatment. CWGR composite got E₁ as 2430 MPa with better potential to sustain load with recoverable viscoelastic distortion apart from single filled and even unfilled composite. Also, in the glassy state, hybrid fillers have increased stiffness of the composite with dense structure obtaining less molecular mobility than a single CW filler, which is in better conscience with the static tensile test outcome. Obviously, from figure 8, a fall in E₁ has been observed over the transition period of 30°C to 130°C on the glass transition phenomena of composites. Once the T₉ point has crossed, the Polymer becomes rubbery, and molecules easily progress with reduced friction.

In Table 3, changes in loss modulus values in the transition zone, i.e., 30°C to 130°C. Figure 8 represents variation in heat loss, which reaches heights after that decreases with the temperature increment. Also, it shows decrement in E₁₁ with the CW/GR fillers compared to single GR filler and composite without filler. Because the molecular mobility of hybrid CW/GR fillers has increased with moderate stress transfer with less dense structure than graphite filler, by attributing positive effects of hybridization.
Therefore GR offered more molecular friction with maximum energy loss as 313 MPa at $T_g$ equal to $77^\circ$ C, and it is degraded to 255 MPa in CWGR at $T_g$ equal to $74.5^\circ$ C. Against NR, except CW, the whole composites Manifesta $T_g$ rise on filler reinforcement results. The CW composite has decreased strength due to the pores occupancy at the coupling of filled resin, resulting in weak adhesion to transfer the stress.

The fraction of $E_1$ and $E_1^{\prime}$ is called $\tan \delta$ (mechanical loss factor). The inclusion of hybrid fillers reduces the $\tan \delta$ value for CWGR composite compared to GR and CW, as shown in Figure 9. Lesser $\tan \delta$ value of CWGR composite indicates a strong interfacial bonding between composite constituents with fewer voids. Hence, it will undertake less energy loss besides diminished damping than GR. We can observe from Table 3 that the glass transition temperature decreased to $82^\circ$ C from GR to CWGR composite. Clustering of CW filler in the CWGR composite may also cause a decrease in its damping ratio compared to CW. However, at 10% of filler loading, the $T_g$ value shows a negative shift from single filler composites to hybrid filler composite, indicating more free space between the molecules of filler matrices and changing the crystalline nature filler materials. Therefore, $T_g$ may reduce at lower filler loading.

**CONCLUSIONS**

The hybridization of natural filler with synthetic filler has produced encouraging results because of fruitful interaction and increased surface area. The hybridization of casuarina wood and graphite fillers has improved mechanical properties with less mechanical loss than GR. CWGR composite results improved the storage and loss modulus values and the energy dissipating interface than CW and NR.
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