Analysis Different Properties Of Reinforced With Cardanol Resin Coconut Shell Biocomposites

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Abstract: *To assess the possibility of using alkaline-treated coconut shell (CS) particles reinforced with cardanol resin (CR) as a new material in engineering applications, it is necessary to evaluate mechanical, thermal, and morphological properties of them.composites based on cardanol resin containing different amounts of alkaline-treated coconut shell (CS) particles (up to 40 wt.%) were prepared from CR matrix. In this study, we investigate the effects of CS particles on the mechanical properties of the composites by UTM, morphological properties by scanning electron microscopy (SEM) and the thermal properties by DSC.A maximum of 10 wt% impact strength of the composites is obtained. It is observed that impact strength of the composites decreases with addition of CS particles. The DSC analysis helps to study the thermal properties of virgin CR and the treated CS/CR composites. From the DSC analysis, it is reported that glass transition temperature (T_g), melting temperature (T_m), and crystallization temperature (T_c) of virgin CR, and the treated CS/CR composites increased up to 30% but decreased by increasing CS particles further. The CS particles added to the CR matrix increased the percentage of water absorption. This is evident that CS particles can be used to improve the properties of CR biocomposites that are used in eco-buildings.*
Key Word*: coconut shell, cardanol resin, impact strength, scanning electron microscope, biocomposite, differential scanning calorimeter (DSC)*

I. Introduction

In earlier days, the production of biocomposites involved incorporation of natural filler materials into conventional polymer. Non-degradable and non-renewable petroleum products are used for modern polymer composites. Polymer composites are generally very difficult to recycle or acquire substantial cost for disposal. This leads to increasing interest in production and use of polymer from renewable resources, which resolve this issue. This conscientious situation provides biocomposites possessing great properties, which has never been found in conventional composites. Thus, bio-based biodegradable and renewable composites have become the next generation composite materials. The environmental protection is a permanent alarm and, in this regard, several studies are being investigated with an aim to transforming natural wastages into useful materials, which could be disposed off with no harm to the environment, or even better, into recyclable goods.

A number of useful phenolic derivatives with meta-substituted saturated/unsaturated hydrocarbon long chain are extracted from cashew nut shell liquid (CNSL), which is an agricultural waste of cashew nut tree. Figure 1 shows the structure of Cardanol, which is one of the key components of CNSL constituents identified as a potential raw material for synthesizing various chemicals such as epoxy, alkyds, phenolics, polyols, and so on [1].

The usage of natural fibers as load-bearing constituents in composite materials has been investigated by many researchers for the last several years, and due to the low cost and the recycle capability of such fibers, the amount of usage has been increased drastically. In India, coconut shells are the important natural fillers. Alternatively, in the recent years, many researchers have been committed to use other natural fillers. However, coconut shell is considered as a potential filler because of their inexpensive, high strength, and renewable [2, 3]. This paper is to study the effects of alkaline treatment on tensile, impact, and thermal and morphological properties of CS/CR biocomposites.

Figure 1 Structure of Cardanol.

II. Material And Methods

Cashew nut shell liquid (CNSL), has been obtained from Golden Cashew Products Pvt-Ltd, Pondicherry, The epoxy LY 556 used in this study is diglycidyl ether of biphenol A (DGEB-A), and tri-ethylene tetra-amine (TETA) and sodium hydroxide (NaOH) have been purchased from Merck Life Science Pvt. Ltd, Mumbai, India. The average size of coconut shell and ground should be in 25 μm.

2.1. Preparation of Alkaline Treated CS

In this analysis, the CS is added to 5% sodium hydroxide solution for 5 hrs with intermittent stirring, rinsed, and washed with water until the water became neutral. Then, the water is filtered and the CS particle is dried for further use. The photographic Figures 2 and 3 show the schematic representation of the preparation of coconut shell particles and compression moulding machine used for biocomposite, respectively.
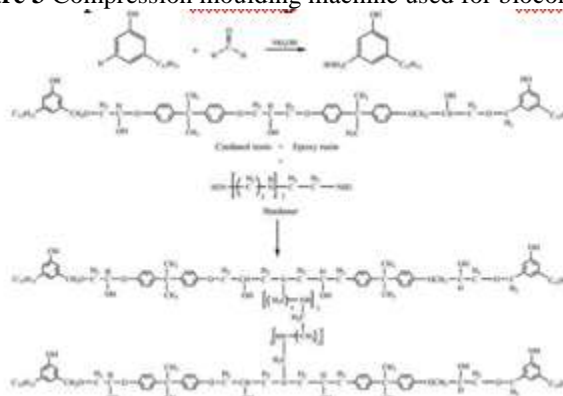
2.2. Preparation of the Coconut Shell Particles

Figure 2 Preparation of coconut shell particles.

Figure 3 Compression moulding machine used for biocomposite.

**Figure 4** Schematic representation of the fabrication of CS/Cardanol-epoxy biocomposite.

2.3. Fabrication of bio-Composites

The compression moulding method is used to prepare biocomposites by loading various CS particles (10–40 wt %). The 25-µm size CS particles are mixed with the Cardanol resin, epoxy resin and hardener matrix in a ratio of 10:1. However, due to its good adhesive nature and greater flexibility, diglycidyl ether of bisphenol-A is added as a curing agent in the Cardanol resin to prevent inherent release of water to porous materials. Adding hardener has improved the thermal properties of the composites. The mixture is then shifted to a mould with a dimension of $290 \times 290 \times 3$ mm and is fabricated. And, the material is next taken into the compression moulding machine. Using this compression moulding process, under the temperature 60°C and pressure 100kgf/cm^2, the composite is formed. The composites are left to post-cure treatment at a temperature ranging from 60 to 70°C for 24 hours in hot air oven. Figure 4 shows the schematic representation of the fabrication of CS/CR biocomposites.

III. Characterization

3.1. Tensile and Impact Test

Tensile test of ten samples taken from different types of composites are used with respect to ASTM D-638 method. The average values had been reported. The tensile test has been used on universal testing machine UTM -model-UNITEK 94100 at a crosshead speed of 5–250 mm/min by using a 100 kN load cell. The notched Charpy-impact testing is carried out by using an EMIC pendulum machine with respect to ASTM D-256, with a hammer weight of 0.6 kg and a falling height of 0.125 m. The notched specimens are mounted on the span of 20 and subjected to normal impact testing. Figures 5 and 6 show the schematic diagram and photographs of tensile and impact specimens subjected for the test, respectively. The fractured surfaces in the impact specimens are characterized by scanning electron microscopy (SEM-JEOL JSM 6610 LV).
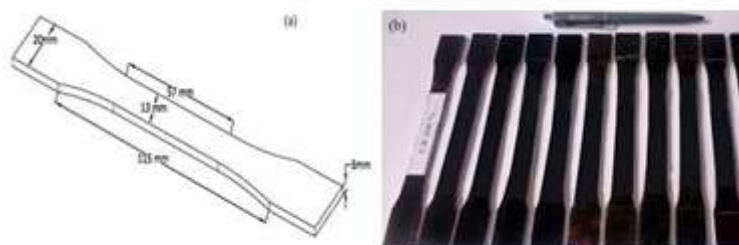
Figure 5 (a) Schematic diagram for CR and CS/CR tensile specimens and (b) photographic view of CR and CS/CR biocomposites specimens for tensile test before fracture.

3.2. Thermal Properties

The differential scanning calorimeter is used to characterize the thermal properties of the biocomposites and record the glass transition temperature (T_g), melting temperature (T_m), and crystallization temperature (T_c). Samples are heated from 30°C to 600°C with heating rate of 10°C/min under flow of nitrogen to prevent oxidation.

3.3. Water Absorption

The water absorption test on CS/CR biocomposites is carried according to ASTM D570 [4]; they are oven-dried and then weighed. The samples of virgin CR and treated CS/CR biocomposites are immersed in distilled water at room temperature for 40 days with daily change of water. The weight of water absorbed divided by dry weight of the specimen determine the water adsorption. Using the following equation, the percentage of water absorption, W_A, has been calculated.
$$W_A (\%) = [(W_2 - W_1)/W_1] \times 100$$
where W_2 and W_1 are the final weight after an immersion period and initial weight of sample before immersion, respectively.

IV. Result & Discussion

4.1. Tensile Strength

Figure 7 shows the tensile strength and tensile modulus of alkaline-treated CS/CR biocomposites. The tensile strength of biocomposites is increased by adding CS particles up to 30 wt%, which indicates that the filler is properly dispersed in the matrix structure.

However, the deformation at break decreases as the CS particles concentration increases after 30 wt% CS particle biocomposites. The reason for this decrease is due to the CS particles aggregated in the matrix,

which caused a poor interface between the particles and CR. Several other possible reasons are noted. One possibility is that the flaw existing in the biocomposites may have weak boundaries between particles and the bubbles trapped during the sample preparation. Another possible reason is non-homogeneous network density of the samples. Similarly, tensile modulus is also increased by adding CS particles up to 40 wt%, which promotes stiffness of regenerated CR biocomposites. The improvement is attributed to strong interaction between hydroxyl groups of CR that improves CS dispersion and adhesion with regenerated CR matrix.
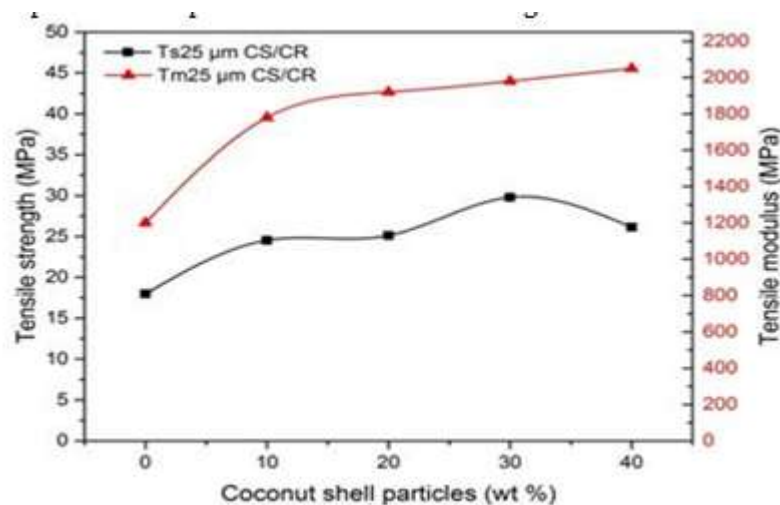

Figure 7 Tensile strength and tensile modulus of virgin CR treated CS/CR biocomposites at different filler loadings.

4.2. Impact Test

The impact behavior of CS-reinforced CR biocomposites is experimentally investigated using various notched Charpy-impact test specimens. The nature of composites, geometry, filler arrangement, and filler matrix interface are the primary factors for impact energy level. The matrix de-bonding and filler pull out are the failure modes observed in the filler composites due to impact loading. The load is transferred through shear and when the shear force exceeds the filler matrix interaction force, the filler matrix de-bonding takes place. The filler fracture will be predominating when the stress level exceeds the filler stress, and then the fractured filler are pulled out of the matrix. The fracture energy reflects the integrative force of the sample over the range of sample deflection recorded in an impact test.

Figure 8 shows the impact strength of alkaline-treated CS biocomposites. It is evident that impact strength of alkaline-treated CS 10 wt% biocomposites (1.56 J) is lesser than the virgin CR composite (1.64 J) particle pull out was the deciding factor, and the failure mechanism may be due to the cracks being easily propagated through void regions of composites in Figure 9(c), Figures 9(d) and (e). The absence of such voids Figure 9(a) was the main reason, for virgin CR matrix to have offered higher impact strength than treated composites. As the particles agglomerate, it leads to poor stress transfer between matrix and particle [5]. It has also been clearly observed that the weight of the particle also affects the impact strength. The CR matrix and the composite with 10 wt% CS particles have the highest impact energy. By increasing weight in composites, the ability of the composites to absorb impact energy decreases. The strength of the CR matrix is greater than that of the composites, which is observed from the impact test [6].
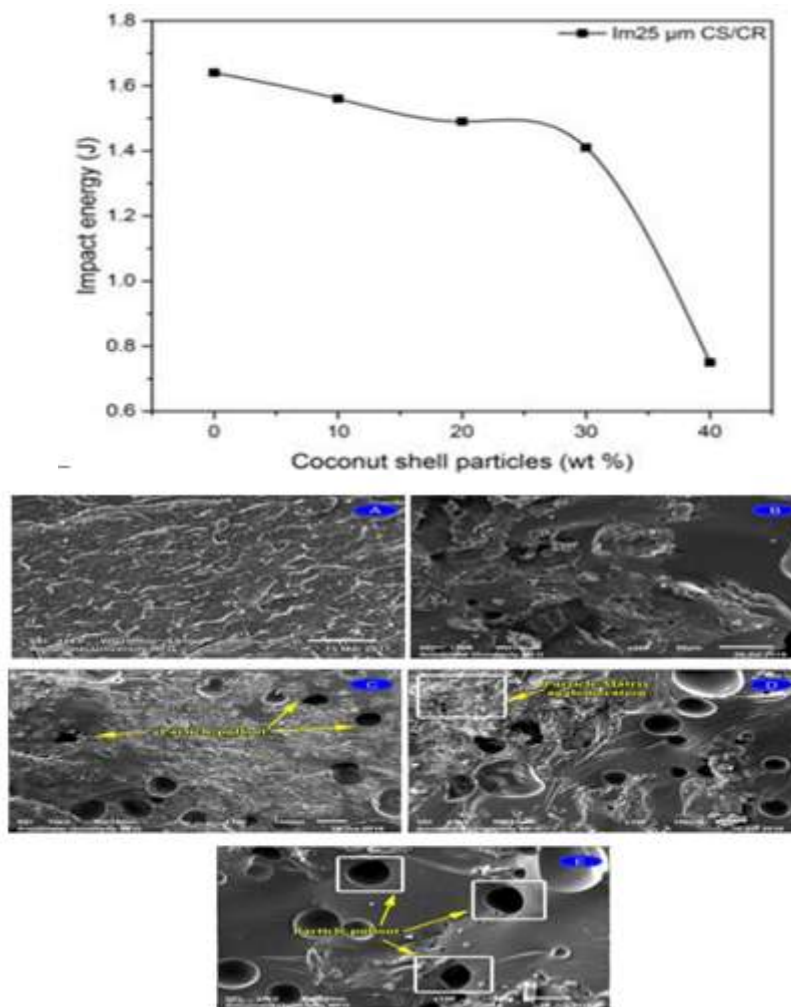
Figure 9 SEM fractograph of impact specimen of (a) virgin CR, (b) 10% CS/CR, (c) 20% CS/CR, (d), 30 % CS/CR and (e) 40 % CS/CR biocomposite.

4.3. Thermal Properties

The varying thermal properties of the CS/CR biocomposites are determined by using the DSC analysis. The glass transition (T_g), melting temperature (T_m), and crystallization temperature (T_c) of the biocomposites are shown in Figure 10. The DSC curve for alkaline-treated CS biocomposites of 10, 20, 30, and 40% with 25-µm size are also shown in Figure 10. The diagram delineates the DSC of the crude state and after soluble base treatment filler. The DSC curves display endothermic and exothermic procedures.

It is observed that the T_g values of biocomposites increased in the range of 60–110°C, compared to T_g (63°C) of virgin CR. A change in degree of plasticization is noticed after adding alkaline-treated composites that help to increase T_g values. This interaction had, however, been enhanced by treatment of CS particles, which in turn caused an increased value of T_g for biocomposites. This substantiates biocomposites further reduces the macromolecular mobility through strong adhesion between CR and CS particles.

It is observed that the crystallization temperature (T_c) of neat composite is 152°C and melting temperature is 174°C. The addition of CS affects the crystallization behavior of the biocomposites. The crystallization temperature of CS composites is in the range of 160– 175°C .This result showed that the crystallization exothermic and melting endothermic peaks indicate that the inclusion of CS in virgin CR did change the melting temperature significantly. T_m of the biocomposite shifted to a higher temperature, indicating its effective heat transfer. This may be caused by the superior bonding between CR matrix and treated CS [7–8].

The CS biocomposites have an impact on crystallization temperature; the particle size accelerates the crystallization and depends on the weight percentage of CS reinforcement. Figure 10 shows that a maximum crystallization temperature is observed in 30 wt% CS biocomposites, beyond that T_c decreased for 40 wt% specimens. The DSC can measure the thermo systems such as CR based on identifying energy changes associated with a change in the relationship between crystalline and amorphous in the resin.

The melting temperature of the CS Particles biocomposites is in the range of 180–200°C as shown in Figure 10. Melting temperature of the CS particles biocomposites is improved compared to neat CR composites. The maximum melting temperature is observed in 30 wt% CS particle biocomposites. So, as per the melting point broadening the crystal formed in the CS particles increases the crystallization and melting temperatures.

Due to the moisture content in the sample, it is observed that there are a few changes noted in thermal behavior in the biocomposites than in neat composites. The crystallization rate is affected after adding CS particles. So, increasing CS particles lead to lower crystallization peak shifts in the thermogram, and their reinforcement alters the overall melting behavior of the biocomposites. As per the melting point broadening, the crystals formed in the CS particles are more heterogeneous than those in the neat composites. The temperature increases with a proportion ranging from 10 to 30% and then it slowly decreases with respect to specimens. In this DSC strategy, the difference in the measure of heat required to expand the temperature of a sample, and references are measured as an element of temperature. Glass transition temperature (T_g), crystallization temperature (T_c), and melting temperature (T_m) are deliberate. This procedure is either exothermic or endothermic that relies on the heat flow to the samples. For instance, as a strong specimen melts to a fluid it will require more heat flowing to the sample to build its temperature at an indistinguishable rate from the reference [9]. These transitions show up as a stage in the pattern of the recorded

DSC signal. Table 1 lists the thermal properties, such as glass transition temperature T_g (°C), crystallization temperature T_c (°C), and melting temperature T_m (°C), of virgin CR and treated CS/CR biocomposites obtained from the DSC scans.

4.4. Water Absorption

The water absorption depends on several factors such as the type of reinforcement, surface protection, interfacial adhesion, voids, and its particle loading [10–11]. Figure 11 shows the time dependence of water absorption in composites. As shown in Figure 11, it is found that all samples projected the same tendency, and the rate of water absorption is higher in the initial stage and then progressively slowed to reach saturation [12]. The neat CR shows total water absorption of 1.08% after 40 days. This is due to the existence of void in the specimen while the manufacturing process is done. The water absorption of the 40% CS-particle composite is 3.42% after 40 days of immersion time. This could be attributed to the poor interface between particle and matrices. As for the 10 wt% CS-particle composite, the growth trends of water absorption are significantly weakened, and the water absorption eventually decreased to constant value of 1.80 % at 40 days. Removing non-cellulosic components such as hemicelluloses and lignin lead to decrease in water adsorption. So, in sum, the removal of hemicelluloses has decreased the accessible hydroxyl groups of CS particles and diminished the total amount of the free OH groups. The voids in the composite should be as minimal as possible, depending on the intended application for the material, in order for the lower rate of water adsorption since larger voids promote diffusion [13] and higher rate of water absorption The filler treatment will rise to lower rate of water absorption and better mechanical properties [15]. This assumption is supported by SEM.
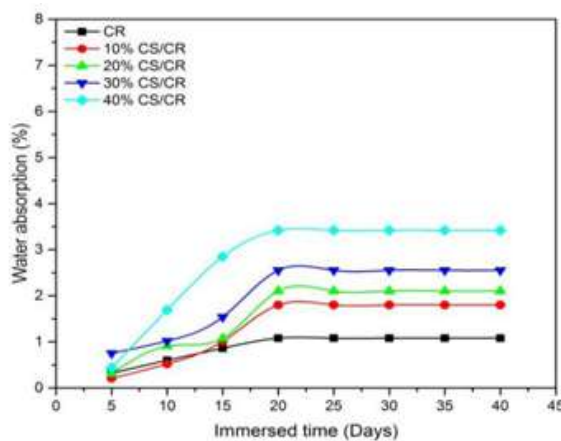
Table 1 DSC data for CR and biocomposites

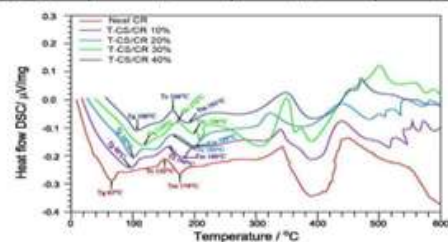| Samples	Glass transition temperature (T_g)	Crystallization temperature (T_c)	Melting temperature (T_m)
Pure CR	63	152	174
10% CS/CR	98	160	180
20% CS/CR	102	163	189
30% CS/CR	110	172	198
40% CS/CR	106	166	191

Figure 10 DSC scans for virgin CR and treated CS/CR biocomposites.

Figure 11 Water absorption behaviors of CR and CS/CR biocomposites at various particles loading

V. Conclusion

In this current work, the mechanical and thermal properties of pure CR and various formulations of CS/CR biocomposites have been investigated. On the basis of the results and discussion, the following conclusions are made:

☐ Adding CS up to 30 wt% leads to increase in tensile strength. This proves that the filler is properly dispersed in the matrix structure and decreased at higher CS particles.

☐ The impact strength of CS/CR biocomposites decreased with increasing filler content.

☐ Adding CS particles helps to increase the glass transition (T_g), melting temperature (T_m), and crystallization temperature of the biocomposites (T_c).

☐ The CR matrix increased the percentage of water absorption by adding CS particles.

☐ The SEM micrographs show the poor interfacial interaction between CS and CR matrix.

☐ Finally, we conclude that in order to realize the application of CS particle in various fields of engineering, the performance of CS particle as reinforcing filler for CR biocomposites can be improved by subjecting the CS particle to chemical treatment.

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