1 Introduction

Renewable materials are being sought after due to the fact that they are sustainable and environmentally friendly. Government policy on reducing the emission of greenhouse gases is the main drive towards sustainability. Many researchers are working on bio-based materials in order to improve the mechanical properties and to possibly discover a wider range of applications.

There have been many reports on the reinforcement of biodegradable thermoplastics with natural/plant-based fibers. Preparation and characterization of biocomposite materials from natural fibers and natural matrices has been reported by Takahashi et al.

Textile-reinforced composites based on natural fibers have been studied by many research groups in recent years due to their good mechanical performance, excellent drape ability, easy handling, excellent integrity, conformability for advanced structural applications, and reduced manufacturing cost. Woven fabrics have been found to be better than non-woven fibers as reinforcements, because the weave architectures of woven fabrics affect the permeability, and the mechanical and fracture properties of the composite. On the other hand, non-woven mats with aligned fibers are of interest since they have no crimp, and are of low cost. Textile structural composites are finding use in various high-performance applications. Various treatments can be done to these natural fibers to improve their wettability and consequently improve the fiber-matrix adhesion in the resulting composite. Many authors have done extensive work on natural fiber treatment.

Lyocell is a regenerated cellulose fiber derived from bleached wood pulp. Lyocell is obtained by a solvent spinning technique, using N-methylmorpholine N-oxide as the solvent. The spinning process is simpler and more environmentally sound than the Viscose spinning process, since it uses a solvent that is less toxic than the carbon disulfide used in the Viscose process, and since it can also be recycled in the manufacturing process. The regenerated cellulose fibers are of interest in structural composites, as they represent chemically pure cellulose fibers with an even quality and performance that cannot be achieved with mechanically treated natural fibers such as flax and hemp.

A hybrid bio-based composite is a combination of the individual characteristics of at least two different types of natural fiber reinforcements in a single renewable matrix. The properties of hybrid composites are a weighed sum of the individual components, but there may be a more favorable balance between the inherent advantages and disadvantages. This means that the attributes of one type of fiber can complement ones lacking in the other. As a result, a balance in cost and performance can be achieved through proper material design.
In this study, woven jute fabrics and carded Lyocell fiber mat were used as reinforcements in a methacrylic anhydride-modified soybean oil thermoset and the properties of the composites were analyzed by tensile and flexural testing, testing of impact resistance, and dynamic mechanical thermal analysis.

2 Experimental

2.1 Materials
Methacrylic anhydride modified soybean oil (MMSO) was used as matrix in the composite preparation. The synthetic pathway for chemical modification of the MMSO is shown in Figure 1. The matrix resin was synthesized according to the method published earlier. Three different types of jute reinforcements and carded Lyocell reinforcement were used in the composite preparation.

2.2 Composite preparation
The jute fibers were washed with 4% sodium hydroxide solution and dried over night and post treated by heating.

The MMSO was blended with 2 wt% tert-butylperoxy benzoate as free radical initiator.

Composites and hybrid composites were made for the purpose of comparison. Composite laminates were made by stacking sheets of fiber mats as a preform and resin impregnation done by hand spray. The prepreg was then inserted in a mould and compression moulded at 160 °C for 5 minutes using a pressure of 40 bar. The hybrid composites were made by sandwiching plies of Lyocell fiber in between the jute fibers, the jute-Lyocell ratio was maintained at approximately 60:40 by weight.

W1, W2 and W3 denoted composites reinforced with woven jute fabrics having surface weights of 240, 300 and 100g/m² respectively. Lyocell reinforced composite was denoted with L. The hybrid composites were denoted with W1L, W2L and W3L.

2.3 Characterizations
The tensile testing was performed according to ISO 527 standard test method for fiber reinforced plastic composites with a universal H10KT testing machine (maximum capacity 10 kN).

The flexural testing was performed according to ISO 14125, with the same testing machine. At least 7 specimens were tested for every material.

Impact testing was done on the composite laminates to determine the Charpy impact strength of the un-notched specimens. This was evaluated in accordance with ISO 179 using a Zwick test instrument. Ten specimens in total were tested to determine the mean impact resistance. The samples were tested edgewise.

The time-temperature dependency of the mechanical properties was determined by dynamic mechanical thermal analysis (DMTA), using a Q-series TA instrument dual cantilever supplied by Waters LLC, Newcastle, DE, USA. The temperature range was from 30°C to 150°C and the frequency was 1 Hz.

3 Results and discussions
The tensile strength of the Lyocell reinforced composite is higher than other composites reinforced with jute fabrics (Fig. 2). The hybridization with Lyocell fiber resulted in increase in both tensile strength and tensile modulus (Figs. 2 and 3). The effect of Lyocell hybridization was negligible on the flexural properties of the composites (Figs. 4), but the Lyocell reinforced composite had the highest flexural properties.

Fig. 5, represents the Charpy impact resistance (energy absorbed/cross-sectional area). The jute composites (W1, W2, and W3) showed relatively low impact resistance between 11 and 13 kJ/m², which could be attributed to good fiber-matrix adhesion. Higher fiber-matrix adhesion resulted in shorter average pull-out lengths, and therefore caused lower impact resistance or strength. The results from the flexural tests showed higher flexural strengths for the jute composites of between 120 and 137 MPa, and flexural moduli of between 5 and 8.5 GPa.
The matrix used in the preparation of the composites (MMSO) has a high cross-linking density due to a high number of reactive double bonds in the molecular structure. The neat MMSO resin is also very brittle due to the high cross-linking density.

The Lyocell composite (L) had the highest impact resistance (36 kJ/m²; Fig. 5), which indicated a longer fiber pull-out length, and this could be due to the structural and morphological nature of the Lyocell fiber (regenerated cellulose fiber). The hybridization of the jute fibers with Lyocell fiber increased the impact resistance of the composites slightly, to between 14 and 15 kJ/m² (see composites W1L, W2L, and W3L in Fig. 5).

Storage modulus is a measure of the elastic response of a material, and in this study the Lyocell composite L showed the highest storage modulus (Fig. 6). This indicates that it has better elastic properties than the other composites. Loss modulus is a measure of the viscous response of a material. The Lyocell composite L had the highest loss modulus (Fig. 7). The results indicate that Lyocell-reinforced composite L had the best viscoelastic properties of all the composites and hybrid composites manufactured. Viscoelasticity is the ability of a material to exhibit both elastic and viscous behavior. The better properties of the Lyocell composite L could be attributed to its reinforcing effects, which were also supported by the other mechanical analyses.

Hybridization with Lyocell fiber reduced the storage modulus of all jute composites (Fig. 6), and this could be due to delamination during constant heating and deformation for about 1 h in the equipment and the possibility of mismatch in the hybrid composite structure. Lyocell fiber and jute fabrics were combined in this case, and a micro-structural analysis of a transverse section of the specimen might give a better explanation.

The glass transition temperature can be determined by the tan δ curves; (see Fig. 8). The tan δ peak for the Lyocell composite corresponds to the temperature at 146°C, while the other jute composites (W1, W2, and W3) have their Tg at about 130°C. The effect of hybridization reduced the glass transition temperature of all the jute composites, which might be due to the explanation given in the previous paragraph.

The Tg values obtained in the loss modulus curves (Fig. 7) for all the composites were about 120°C, while they were between 90°C and 100°C for the hybrid composites. It can be concluded that the Tg values obtained from the loss modulus curve are lower than those obtained from the tan δ curve, which confirms the findings of many authors that the values of Tg from the tan δ curve are always exaggerated, whereas those from the loss modulus curve are more reliable. Increase in storage and loss modulus indicates better fiber-matrix adhesion. The ratio of E" to E' (loss modulus to storage modulus) gives the tangent of the phase angle δ; tan δ is known as the damping and is a measure of energy dissipation. Such parameters provide quantitative information about the behavior of a material. The storage and loss modulus and the glass transition temperature can be increased by blending the matrix with styrene, but in this study neat resin was used as a matrix.

![Fig. 1. The synthetic pathway of chemical modification of MMSO.](image-url)
Fig. 2. Comparison of the tensile strengths between the jute and Lyocell reinforced composites.

Fig. 3. Comparison of the tensile moduli of the jute and Lyocell reinforced composites.

Fig. 4. Flexural strength of the hybrid composites compared with Lyocell reinforced composite.

Fig. 5. Impact resistance of both the composites and the hybrid composite.

Fig. 6. Storage modulus of the various composite and hybrid composite samples obtained from the DMTA.

Fig. 7. Loss modulus for individual samples obtained from the DMTA.
Fig. 8. Tan δ peak of the various composite and hybrid composite samples obtained from the DMTA

Conclusions
There were some variations in the mechanical properties, which were due to the different types of reinforcements. In this study, the weave architectures were the same for the woven fabrics but the difference was the distance between adjacent roving wefts and warps, which contributed greatly to the surface weight of the fabric. Although one would have expected composite W2 to have better mechanical properties than the other woven jute composites because the fiber was compact, due to the short distance between the two adjacent roving wefts (about 0.1 cm), this was not the case.

The other woven jute fabrics had 0.2 and 0.5 cm between the adjacent wefts and warps, which led to lower surface weight and therefore a higher ply number in the composite. The mechanical properties of composites W1 and W3 were superior to that of W2.

The composites and the hybrid composites had very good mechanical properties. Hybridization with Lyocell fiber increased the overall mechanical properties of the composites but reduced their viscoelastic properties. Although Lyocell offers better performance at low cost, Lyocell fiber cannot replace woven fabric but it can be used as hybrid to complement the properties that are lacking in woven fabrics. Although the weight ratio of the jute-Lyocell fiber was 60:40, the tensile and flexural properties might be improved if the ratio of the Lyocell fiber is increased. Percentage elongation generally improved with inclusion of the Lyocell fiber. In order to increase toughness in a composite, Lyocell fiber should be used as hybrid.

References