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ELASTIC PROPERTIES OF CNT MIXED/CNT COATED SISAL FRP COMPOSITES USING EXPERIMENTAL AND FE METHODS

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Abstract. In this work, natural sisal fiber reinforced composite elastic properties are estimated by using Carbon Nano Tubes (CNT) as filler. The composite specimens are prepared with sisal and nano-CNT-reinforced polymer composites and tested for tensile modulus. Further, using micromechanics, hybrid sisal fiber composites are analyzed for elastic properties and interfacial stresses. Sisal fibers coated by CNTs and sisal fiber reinforced in homogenized CNT mixed polymer composites are analyzed and compared in terms of their elastic properties and interfacial stresses. From the results, it is found that sisal fiber coated with CNTs gives more longitudinal modulus (E_1) whereas sisal fiber mixed with CNT polymer composite shows good transverse modulus (E_2). Interfacial stresses are higher at the matrix interface of the CNT mixed composite than in the CNT coated composite.

Keywords: *Carbon Nanotubes, Sisal fiber, Micromechanics, Interfacial stresses*.

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1. Introduction

Nano reinforcement in pure polymer or fiber-reinforced polymer composites is increasing in the current decade due to the benefits associated with nano size. Owing to the superior strength-to-weight ratio and stiffness-to-weight ratio, carbon-based nanoparticles are becoming good candidates for the preparation of composite materials, which find applications in the aerospace, automotive, railway, and power sectors. (Swolfs *et al.*, 2018; He *et al.*, 2019; 2020; Chowdhury *et al.*, 2018; Li *et al.*, 2017; Kong *et al.*, 2018). The use of carbon nanotubes in regular fiber-reinforced composites made of both synthetic and natural fiber-reinforced composites is improving. Aramid fiber-reinforced composites are studied by using carbon nanotube mixed matrix material. These aramid and carbon nanotube reinforced hybrid composites showed a good load transfer mechanism that results in good tensile properties (Sharma *et al.*, 2020).

Few attempts have been made by researchers to use carbon nanotubes as coating materials. A char layer coated with carbon nanotubes improved its compression resistance and thermal insulation. (Xi *et al.*, 2021). Using the Multiscale modelling approach, the interfacial properties of carbon and carbon nanotube mixed and coated composites are reported and highlight the modelling techniques with the finite element method (Malekimoghadam & Icardi, 2019). The benefit of adding carbon nanotubes as fillers in the elastomers is that they improve their performance in the automotive and aviation sectors (Valentini *et al.*, 2018).

Using molecular dynamics and continuum approaches, nanoscale materials can be analyzed. Both methods can be justified by their outcomes. Using a micromechanics approach with a finite element process, the CNT reinforced composite Young's modulus is being estimated and compared with the Rule of Mixtures (ROM) (Gupta & Harsha, 2016). Along with the static, fatigue load-bearing capacity is also verified for carbon nanotube-based composites (Davis *et al.*, 2011). Maximum performance can be achieved with carbon nanotubes by promoting a good interface. This effect is proven for Young's modulus and fracture toughness (Saha & Bal, 2017; Cha *et al.*, 2017; 2016; Rafiee & Mahdavi, 2015; 2016).

The Micromechanics methodology and finite element method have been used by many researchers to explore the behaviour of reinforced matrix materials. This methodology is applied to the prediction of mechanical and thermal properties of carbon nanotube reinforced composites (Hassanzadeh-Aghdam *et al.*, 2018; Wang *et al.*, 2019; Ahmadi *et al.*, 2019; Shi *et al.*, 2019). Few authors investigate the elasto-plastic behaviour of carbon nanotube reinforced composites. Using the Micromechanics approach (Karimi *et al.*, 2018), Because of the high modulus (1800 GPa) and Poisson's ratio (0.3), carbon nanotubes are often selected as a coating material. (Tümer *et al.*, 2020). A multiscale approach has been developed by Mahmoodi *et al.*, 2020; Najafi *et al.*, 2021; Zaccardi *et al.*, 2021) to characterize the electro-thermo mechanical behaviour of CNT-based composites by selecting one unit cell. Homogenization techniques are applied to CNT-reinforced composites to quantify their bulk mechanical properties. (Cheng *et al.*, 2019; Alam *et al.*, 2020). Compared to silicon, germanium, tin nanotubes, and carbon tubes, they have given a better performance in elastic modulus (Dastmard *et al.*, 2021).

The continuum approach can be used by selecting RVE to assess the volume fraction, orientation, distribution, aspect ratio, and effects of the CNT on the resulting properties (Greco, 2020; Hassanzadeh-Aghdam *et al.*, 2018). Various modelling techniques used in CNT reinforced composites are highlighted by (Pal & Kumar, 2016). Carbon nanotubes, graphene, and Buckminster fullerene are allotropes of carbon. The finite element method is used to identify the Buckminster fullerene reinforcing effect on Young's modulus with and without defect (Prasanthi , 2015; 2014).The distribution of stresses along with the material properties are estimated as a function of interphase modulus and thickness by using the finite element method (Banerjee *et al.*, 2016). The relationship between the effective length of CNT and load transfer efficiency is explored (Haque & Ramasetty, 2005). Researchers estimate the buckling properties of carbon fibre and carbon nanotube reinforced composites (Ahmadi *et al.*, 2020). From the theoretical computations, it is indicated that a small amount of CNT is also making a difference in the mechanical properties. (Chwał & Muc, 2021). Damage initiation of carbon nanotube reinforced polymer composites has also been reported. (Kada *et al.*, 2018). Modeling and analysis of coiled carbon nanotube reinforced polymer composites is presented (Khani *et al.*, 2016).

In this work, the focus has been given to carbon nanotubes' influence on Young's Modulus of polymer matrix, conducting experimental studies. These studies are performed by varying the weight fraction of carbon nanotubes. Further, using the continuum approach, carbon nanotube- reinforced composite elastic properties are addressed using the Micromechanics method and the finite element method. The differences in the carbon nanotube infusion and coating of nanotubes on the sisal fiber-reinforced composite are presented from the perspective of elastic properties such as longitudinal, transverse, shear modulus, major and minor Poisson's ratio. In both methods, the stresses at the interface are identified using homogenization techniques.

2. Experimental Procedure

Commercially available SWCNT is in fine powder form with an apurity of > 99.9% with grade LF-HFC and is procured from Go Green Products, Chennai. The outer diameter is 7.5 nm, the inner diameter is 5 nm, and the length of the CNT is 100 nm with a Young's modulus of 1 TPa and a density of 1.9 g/cm3. The SWCNT material is considered a fibre material for the preparation of nano-composites.

The Epoxy Resin (Aradite 556) along with the Hardener (H3 95) is procured from Bindhu Agencies, Vijayawada, and Andhra Pradesh, India. The resin with 1.4g/cm^3 density and Young's modulus of 5.171GPa is used as a matrix material or the hosting agent for the composite. Sisal is a plant-based fibre and is attracting many researchers due to its structural, thermal, and mechanical properties (Chand & Fahim, 2021). These plant-based fibre reinforced composite elastic properties are further enhanced by the addition of carbon-based nano materials. The method to be adopted for the enhancement of the elastic properties plays an important role in their percentage. In this work, the coating and infusion effects on the elastic properties are presented.

2.1. Nano Composite Preparations and Testing

The fabrication of the CNT-reinforced composite was done by the traditional hand lay-up technique. At first, the fine powdered SWCNT is mixed with the epoxy resin with the help of ultra sonicator equipment at Prasad V Potluri Siddhartha Institute of Technology, Vijayawada, Andhra Pradesh, India. CNT with a weight of 0.5 to 3% by weight fraction is collected by using a Conteck precision balance. After collecting the measured CNT's, the specimen is prepared by mixing the CNT's into the epoxy with a compatible hardener. An ultrasonicator is used to mix or ensure the uniform mixing of CNTs into the matrix materials. The sonication process is carried out for 30 minutes. After sonication, the agglomeration in the resin disappeared. And this process has been widely used to produce a homogeneous mixture (Hatami & Panah, 2017) Fig. 1.

After sonication, the mixture is poured into the mould and the specimens are cured for 24 hours. For each weight fraction of the CNT, four specimens are prepared. After curing, the specimens are removed from the mould and tested for Young's modulus on a digital universal testing machine at the P.V.P. Siddhartha Institute of Technology, Vijayawada. The Nano composite specimens were prepared by varying the SWCNT weight percentage from 0.5 to 3% with an interval of 0.5. The ASTM D638 standard specimens are tested for tensile and elongation by using a UTM 5000kgf computerised machine. Young's modulus is obtained from the Universal Tensile Machine.

2.2. Sisal Hybrid composite preparation and testing

In this section, carbon nanotube mixed epoxy matrix is used as a matrix material, and sisal fibers are reinforced in the same matrix. The sisal fiber weight fraction is maintained at 50%. To prepare the specimen for tensile testing, the same hand lay-up techniques are used. The specimen is made of a sisal fiber-reinforced carbon nanotube mixed epoxy matrix. The Young's modulus of sisal fiber-reinforced in the homogenized

CNT mixed matrix is identified by conducting a tensile test as mentioned in subsection 2.1.

The sisal fiber-reinforced CNT mixed epoxy composite specimen is presented in Fig.1.

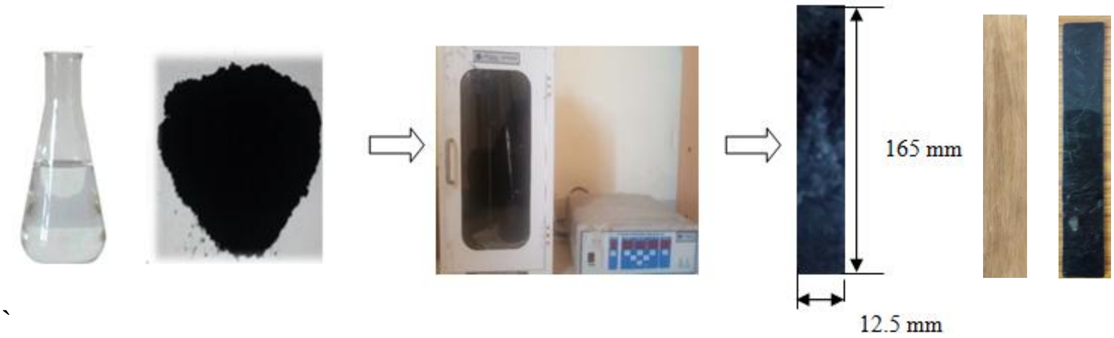

Figure 1. Nano composite specimen preparation and nano, sisal and hybrid composite

2.3. Simulation of carbon nanotube reinforced composite Elastic modulus

In this section, the procedure being adopted for the evaluation of Young's modulus and Poisson's ratio of single-wall carbon nanotube reinforced epoxy composites is presented using numerical simulations. Using the micromechanics approach, carbon nanotube (CNT's) reinforced epoxy lamina has been analysed. In that lamina, the CNTs are assumed to be distributed regularly in the epoxy matrix. The theory behind micromechanics involves the idealization of the CNT in the epoxy following a specific distribution, let us say square or hexagonal, and picking one from the array of the total lamina is the represents the entire lamina, ensuring proper boundary conditions. The selected cell for the analysis is called a Representative Volume Element (RVE) (Koutsawa *et al*., 2017).

For this problem, the square unit cell is considered, and it is presented in Fig.2. This selected model is analyzed for Young's modulus and Poisson's ratio by using finite element method based software ANSYS. As symmetry is observed in one unit cell from the perspective of the geometry, loading, and surrounded boundary conditions, the one-eighth portion is opted for the analysis. The volume of the CNT in the unit cell is governed by a CNT volume fraction, which is the ratio of the volume of the CNT to the total volume of the unit cell. A finite element method is implemented to study the response of the unit cell under uniaxial loading.

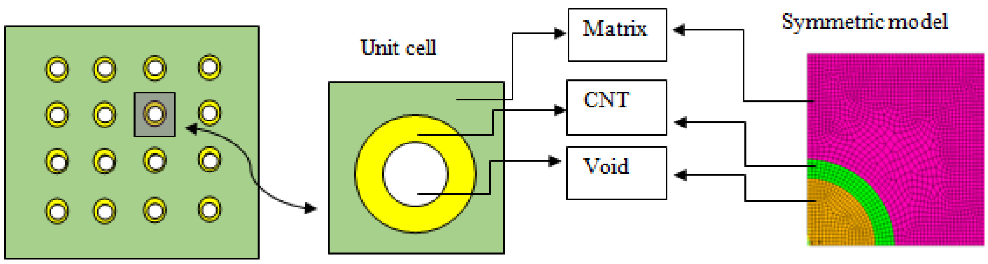

Figure 2. CNT distributed in epoxy matrix and selected unit cell

The experimentation was done by using weight fractions which are to be converted into volume fractions for Finite Element Analysis of CNT reinforced composites. The weight percentages of the SWCNT which are taken for the analysis are

0.5, 1, 1.5, 2, 2.5, and 3. The weight percentages are converted into volume fractions by using the density of the SWCNT and epoxy matrix. The element used for the present analysis is SOLID95 from ANSYS, which is a higher-order 3-D 20-node solid element that exhibits quadratic displacement behaviour.

The element is defined by 20 nodes having three degrees of freedom at each node translation in the nodal x, y, and z directions. The material properties provided by the supplier of CNTs and epoxy are used for the present analysis. The continuum approach has been used for the CNTS. In the continuum approach, the carbon nanotubes are assumed to be hollow cylinders. (Koutsawa *et al.*, 2017). The hollowness is treated as a void. The CNT modulus is considered to be 1 TPa and Poisson's ratio is taken as 0.3, for the epoxy matrix. The modulus is 5.171 GPa and the Poisson's ratio of 0.35 is considered for the analysis. For the void, 1e-17 GPa and Poisson's ratio of 0.4 are assigned. The cylindrical tube with an inner radius of 5 nm and an outside diameter of 7.5 nm is considered, and the size of the RVE is calculated from the volume fraction of the CNT. The FE model is presented in Fig.2. The modulus is obtained by applying load in the direction of CNT, i.e., 1 MPa, solving the finite element model by applying periodic boundary conditions and applying uniform pressure load, the displacement of the finite element model is obtained. From the displacement in the loading direction, the strain, Young's modulus is calculated using Hook's law. The finite element models are validated by comparing the Young's modulus calculated based on Hook's law to the rule of mixture (Liu & Chen, 2003) and comparisons between the results are presented in the results in the discussion section.

2.4. Simulation of sisal fiber and CNT mixed polymer composite and CNT coated sisal fiber reinforced in Polymer matrix

Further, one of the objectives of the present work is to compare two different methods, such as mixing nanoparticles before reinforcing the fibers into a matrix or coating the fiber with nanoparticles before reinforcing it into a matrix. The difference between these concepts is illustrated in Fig.3. In this paper, two different hybrid composites are studied, named H1 and H2. H1 means sisal fiber-reinforced in a homogenized CNT mixed epoxy matrix, whereas the H2 mode represents the sisal fibers which are coated with CNT and reinforced in a pure epoxy matrix. In the H1 model, the CNTs are assumed to be aligned in the sisal fiber direction, where as in the H2 model, the CNTs are coated along the length of the CNT. In both models, the properties of the constituents are the same.

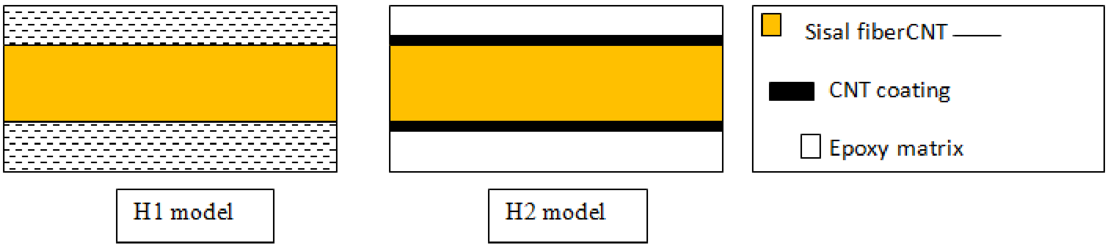

Figure 3. Representation of sisal/ CNT mixed matrix and CNT coated sisal epoxy composite

Sisal is a natural fiber. To enhance the properties of these fibers reinforced composite applications, matrix modification techniques and coating methods have been studied and compared in terms of their resulting elastic properties and interface stresses. In this section, sisal fibre is reinforced with homogenised CNT mixed polymer mixed composite (H1) and sisal fibre coated with CNT, which is then reinforced in a pure epoxy matrix (H2). In either case, the volume fraction of carbon nanotubes and sisal fiber is the same.

To achieve this objective, the micromechanics approach has been implemented. As it is covered in section 1, many researchers are using micromechanics to evaluate the elastic properties and interfacial stresses of composite materials, connecting nano to microscale reinforcements (Liu & Chen, 2003) . These studies can be performed with the support of the finite element method as it gives a piecewise approximation, which could be a better option for this problem. (Fig.4)

While applying micromechanics to composite materials, the first step is to idealise the reinforcement distribution in the hosting medium, such as matrix material. In this study, two types of reinforcement phases, such as sisal fiber and carbon nanotube, and two types of matrix materials, pure epoxy and homogenised carbon nanotube epoxy, are considered. For these composites, longitudinal modulus (E_1), transverse modulus (E_2), major and minor Poisson's ratios, and homogenised interfacial stresses

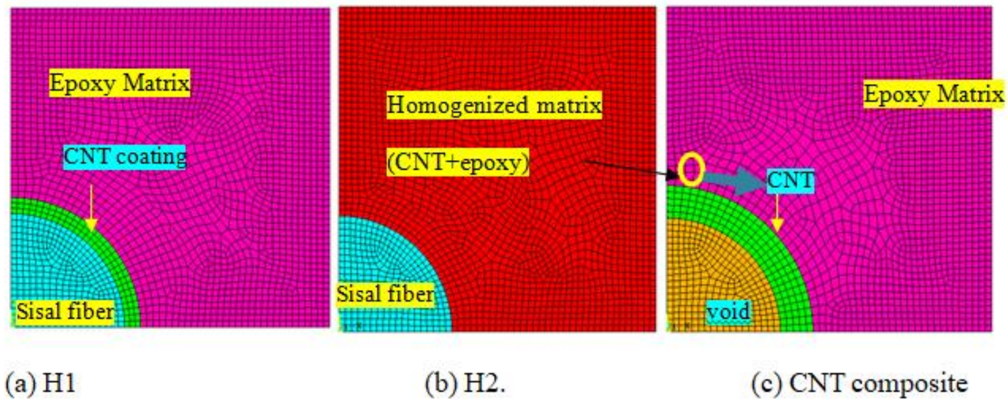

Figure 4. Finite element models of hybrid composite, (a). CNT coated sisal fiber epoxy composite (H2), (b). sisal fiber reinforced in homogenized CNT mixed epoxy matrix (H1). c. CNT reinforced in epoxy matrix

3. Results and Discussions

Fig.5. shows the variation of Young's modulus, which is increased with the rise in the weight percentage of the CNT reinforcement in the epoxy matrix, since the modulus depends upon the particle loading and that too small particle contribution leads to a high effect on the modulus. From Fig.6, as the percentage of SWCNT increases, the % elongation decreases. This is because there is a loss of ductility in the composite with an increase in the weight fraction of the SWCNT. The ductility of the composite has been suppressed by the presence of single-walled carbon nanotubes. The decrease in elongation is more prominent at higher particle loading. It is a fact that the deformation of the reinforcement is commonly much less than that of the epoxy resin. The reinforcement forces the matrix to deform less than its original form as the SWCNT locks the deformation and enhances the stiffness of the resulting material.

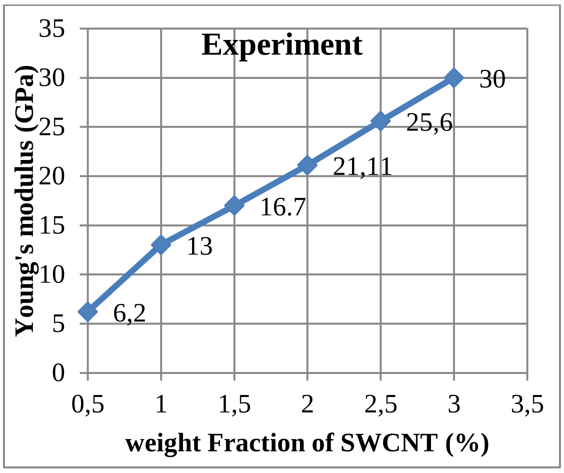

Figure 5. E with swcnt epoxy composite

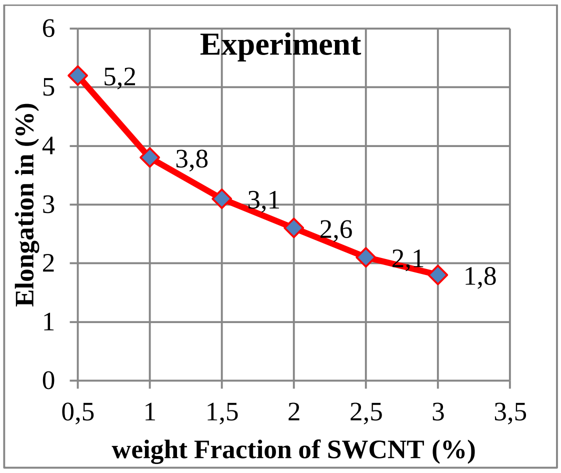

Figure 6. dl of swcnt epoxy composite

Fig.7. shows the modulus of the CNT-reinforced epoxy composite obtained from experiments and the finite element method. The percentage error between the two findings is also presented in the same figure. Close agreement is found between the experimental and finite element results, particularly at lower volume fractions and at higher weight fractions, where deviations are observed between the two results. The deviations are due to the assumptions considered while characterizing the elastic modulus in the finite element method.

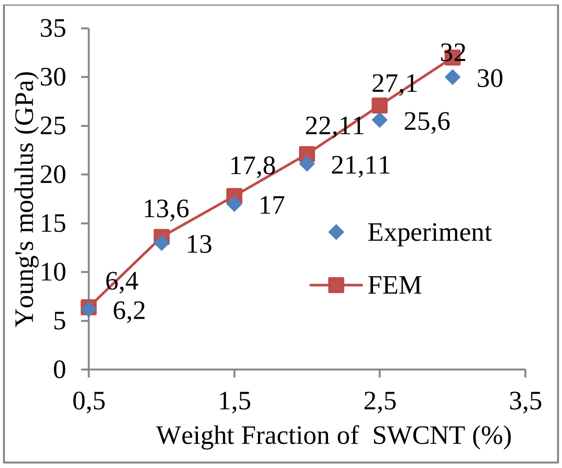

Figure 7. E with weight fraction of CNT

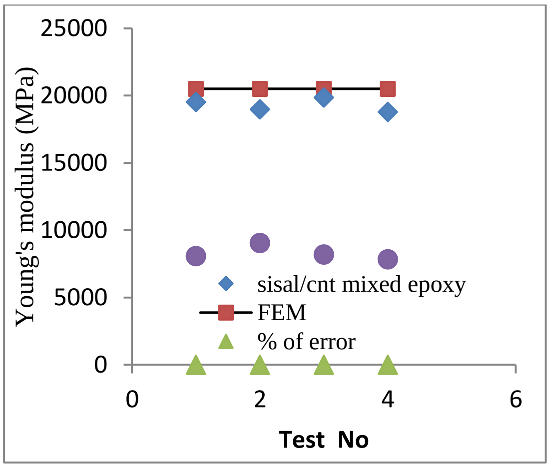

Figure 8. Sisal fiber and CNT mixed epoxy composite at 50% W_f of sisal fiber

Fig.8. shows the Young's modulus of sisal/epoxy and hybrid sisal composites, i.e obtained by reinforcing sisal fiber in a CNT mixed epoxy matrix (H1composite). The results are obtained from both experimental and simulation studies (Finite element method). Compared to the sisal fiber/epoxy composite, the sisal fiber/CNT mixed epoxy composite showed higher modulus. This study is performed by maintaining the weight fraction of sisal fiber at 50%. The percentage of error between the experimental and numerical methods is also presented.

The longitudinal modulus of two hybrid composites, i.e sisal fiber-reinforced in homogenised CNT mixed epoxy matrix (H1) and CNT coated sisal fiber reinforced epoxy composite (H2) is identified from finite element results and validated with the Rule of Mixture. Two different behaviours are noticed in two-hybrid composites. The longitudinal modulus is decreasing with increasing the volume fraction of sisal fiber in the sisal/CNT infused epoxy hybrid composite, whereas in the second composite (H2), which is CNT coated sisal fibre and pure epoxy composite, the longitudinal modulus is increasing with sisal fiber volume fraction. Fig.9. The magnitude is higher for CNT coated sisal fiber-reinforced composite than for CNT mixed sisal fiber-reinforced composite. In a two-hybrid composite, the sisal fiber and CNT volume fraction are the same. The reason for this behaviour is understood in the following way.

While applying load parallel to sisal fiber, the epoxy material transfers the load to sisal fiber. The CNT in the epoxy material in the first hybrid composite (H1) will also take the load as these are the stiffer materials than the matrix, and in other ways, these fillers also protect the fiber by receiving the load. As the sisal fiber, the contribution of these CNT's will be decreased in the polymer matrix. As a result, the modulus is declining.

In the second hybrid case, i.e. CNT coated sisal fiber-reinforced pure epoxy composite, the load applied in the fiber direction will be passed into the fiber, as these fibers are already coated with strong CNT in the same fiber direction and used to offer more resistance to the load than sisal fiber. That means the fiber is fully protected by the CNT coating, and these coatings are very effective up to 75% of the sisal fiber volume fraction. As a result, a direct relation is observed between the sisal fiber volume fraction and longitudinal modulus.

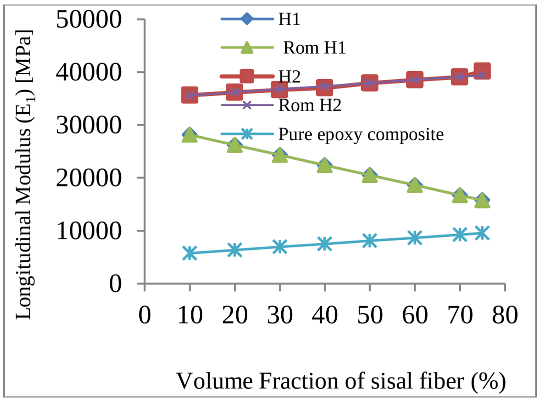

Figure 9. Longitudinal modulus

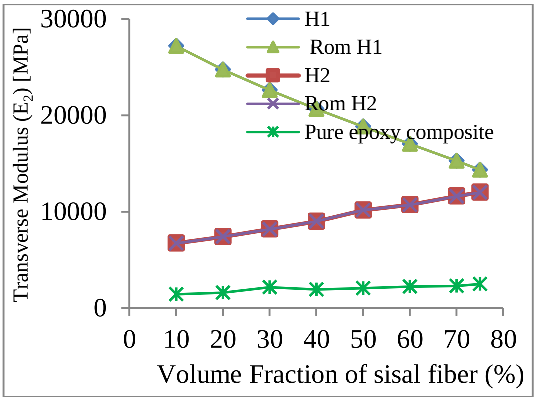

Figure 10. Transverse modulus

From these results, it is understood that the mismatch between the modulus of sisal fiber and the matrix material is very high. Enhancing the longitudinal modulus coating with the CNT will give more benefits than mixing the same CNTs in the epoxy composite. In the same figure, the modulus without any CNT is also presented. Compared to pure sisal reinforced epoxy composite, H1 and H2 composites enhanced the longitudinal modulus in an effective way.

Fig. 10 shows the transverse modulus of two hybrid composites with respect to the volume fraction of sisal fiber. The response of transverse modulus in the two-hybrid composite is the same with respect to sisal fiber volume fraction as that of longitudinal modulus. However, one important deviation is observed in the transverse modulus. The composite with CNT coated sisal fiber composite (H2) showed less magnitude than the sisal fiber with CNT infused epoxy composite (H1). In the composite transverse direction, which is also called the negative direction, the matrix is the key load-taking member in the transverse direction. Most of the composite shows high strength and stiffness in the fiber direction, and most of the failure of the composite starts in the transverse direction.

Applying the load perpendicular to the sisal fiber i.e transverse direction, the matrix used to receive the load and, due to the CNTs in the matrix material, the load will be able to handle the load effectively. Decreased matrix percentage also decreases the load-bearing capacity of the composite. As a result, increasing the sisal fiber volume fraction means decreasing the matrix percentage. As a result, the stiffening members' contribution (CNT) also decreases.

In the CNT-coated sisal fiber composite, the transverse modulus increases with an increase in the sisal fiber volume fraction (H2). In this case, the load perpendicular to the sisal fiber is received by the CNT coating, which also protects the fiber from the load. The coating is active in every volume fraction of sisal fiber.

Fig. 11-12 shows the major and minor Poisson’s ratio of hybrid composites. The major Poisson’s ratio is decreasing in two-hybrid composites with an increase in the volume fraction of sisal fiber. The decrement in the property is more in the sisal fiber CNT mixed epoxy composite (H1) than in the CNT coated sisal fiber reinforced epoxy composite (H2). Increasing the sisal fiber and CNT coating and CNT mixed epoxy decreases the lateral strain, and lateral strain directly impacts the magnitude of the major Poisson’s ratio. No differences in the magnitude of major Poisson’s ratio are observed between the two composites, i.e., H1 and H2 composite

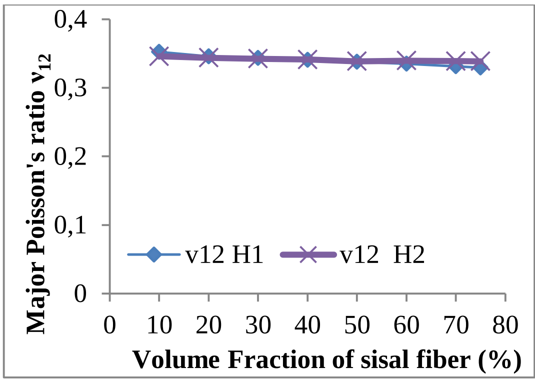

Figure 11. ν_{12} hybrid composites

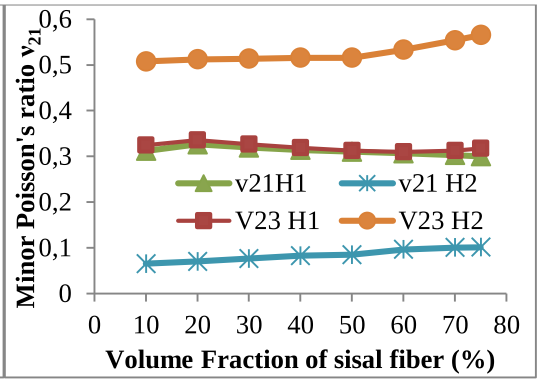

Figure 12. ν_{21} of hybrid composites

The variation of minor Poisson’s ratio is presented in Fig.12. V_{21} is high for H1 composite and v_{23} is high for H2 composite due to their high in-plane transverse modulus and out-of-plane transverse modulus, respectively. In-plane and out-of-plane shear modulus is presented in Figs. 13–14, and the response of these properties is the same as that of transverse modulus. The differences in the stress contours of H1 and H2 models are presented in Fig. 15–16. These contours clearly indicate the response of the material under given loading conditions.

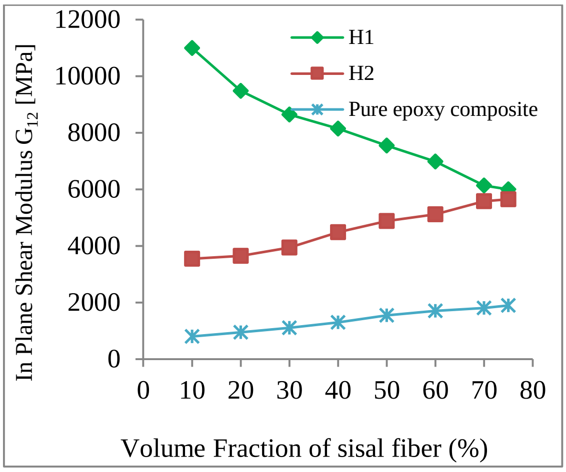

Figure 13. G_{12} hybrid composites

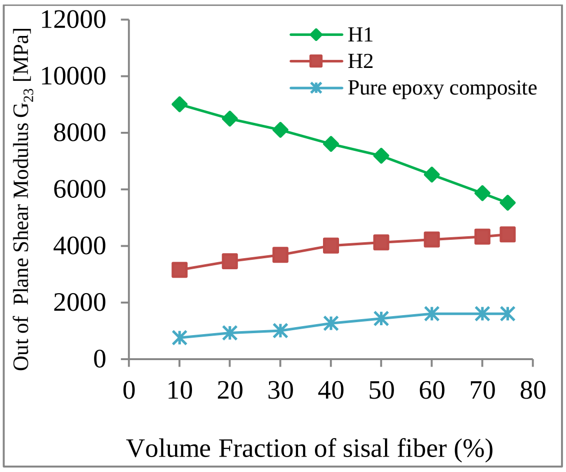

Figure 14. G_{21} hybrid composites

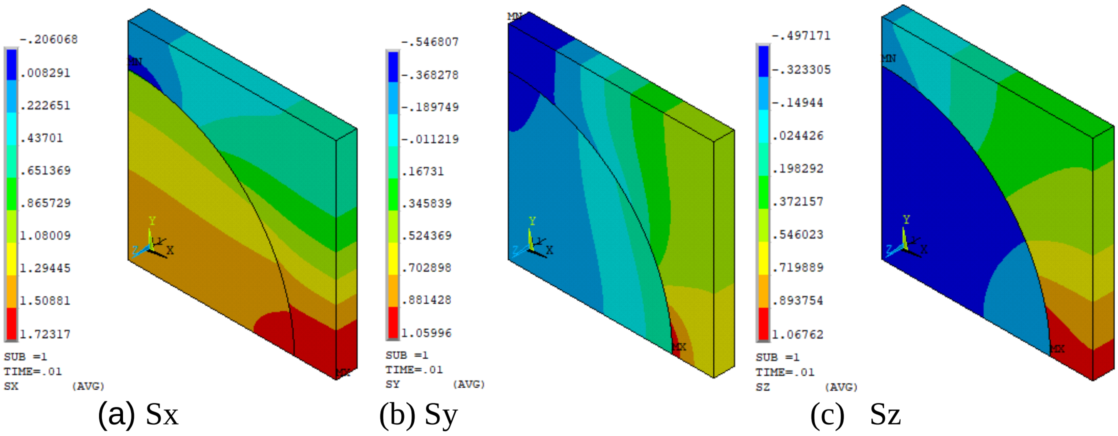

Figure 15. Stresses contours of H1 Model at 50% sisal fiber and 3% CNT under transverse load

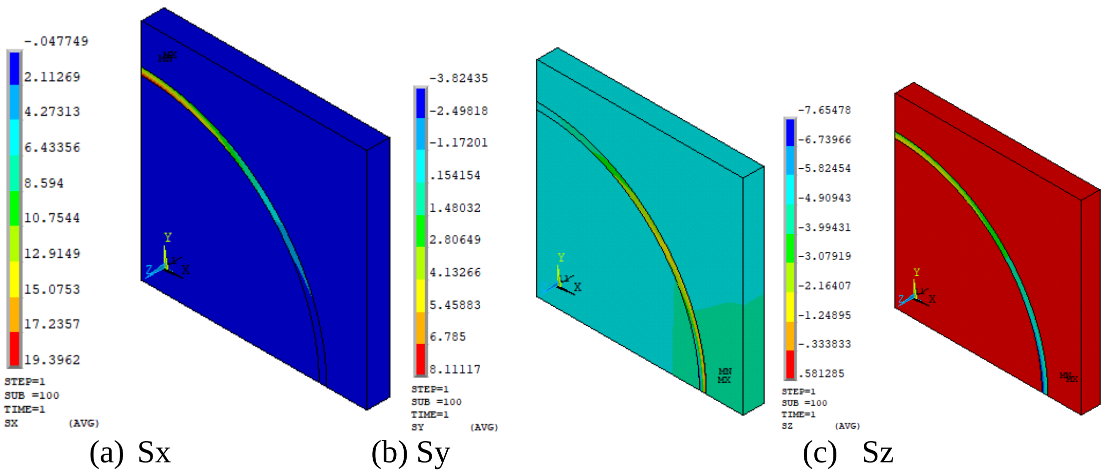

Figure 16. Stresses contours of H2Model at 50% sisal fiber and 3% CNT under transverse load

Interfacial stresses

Along with the elastic properties, the identification of interfacial stresses is very vital while addressing composite material behaviour, and through micromechanics and finite element methods, it is possible to estimate the interfacial stresses. In this section, the interfacial stresses of H1 and H2 composites are identified under longitudinal and transverse loading directions. The normal and shear stresses are plotted at every volume fraction of fiber at both the fiber and matrix interface, though the stresses are not the same at this junction of fiber and matrix. Due to the involvement of CNT reinforcement in 2 different ways, the interface of the sisal fiber and epoxy matrix experiences different stresses under longitudinal and transverse loading. Fig.17-18 show the interface stresses in fiber and matrix composite under longitudinal loading. The stress generated in the fiber direction (z) is greater in the H1 model, particularly in the matrix.

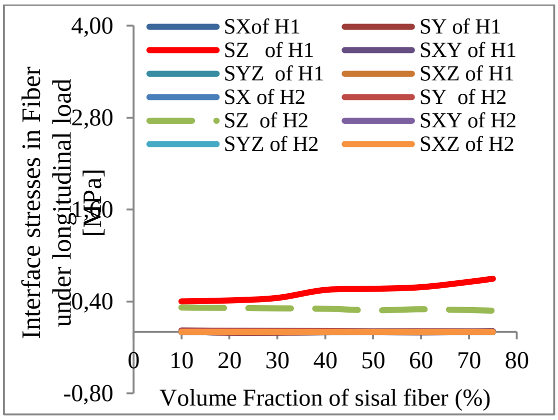

Figure 17. Interface stresses of fiber

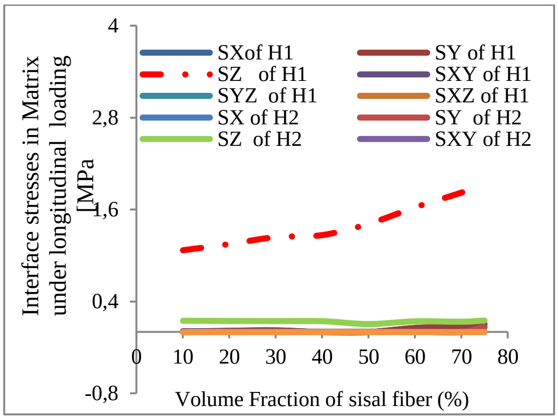

Figure 18. Interface stresses of matrix

In the transverse loading, the Sx is maximum in H2 model in both fiber and matrix. The coating stresses are presented in Figs. 19 and 20. Very high stresses are generated in the coating as these coatings, used to offer more resistance. As a result, they experienced more stress and obstructed the passage of these stresses to fiber as a protecting medium. Fig.21-22 shows the variation of interfacial stresses of the sisal fibers coated with CNT. In this case, the interfacial stresses are very high compared to the H1 model.

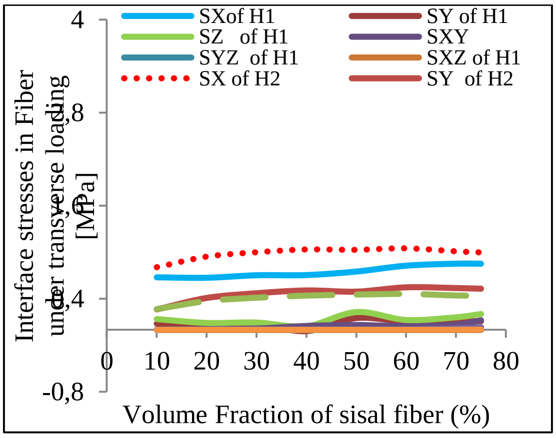

Figure 19. Interface stresses of fiber

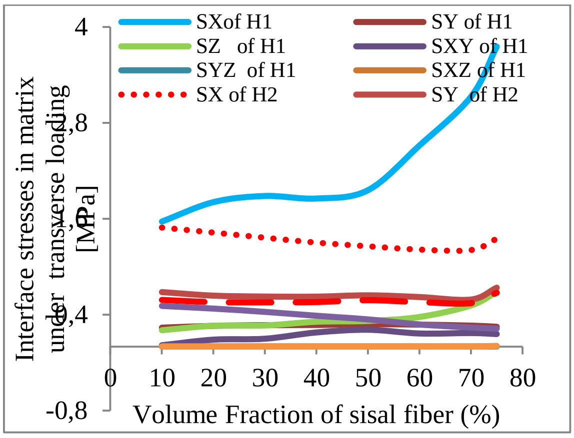

Figure 20. Interface stresses of matrix

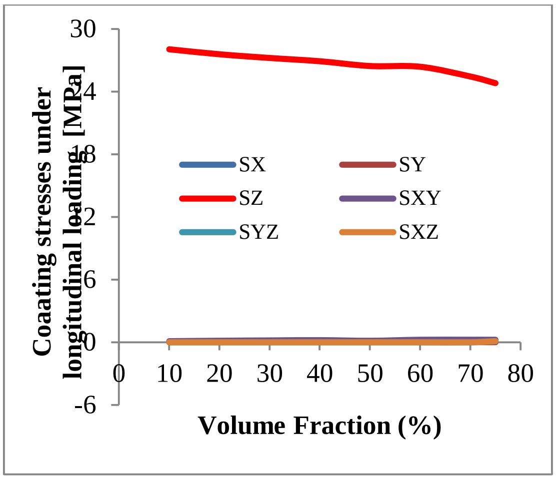

Figure 21. Stresses in coating of matrix

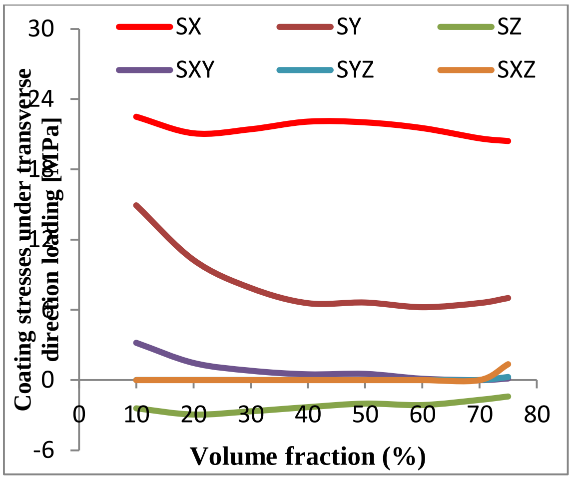

Figure 22. Stresses in coating of matrix

4. Conclusion

CNT reinforced epoxy and CNT coated sisal fiber-reinforced composite elastic properties and interface stresses are evaluated by experimental and finite element methods, respectively. The following conclusions are obtained from the present work:

- The longitudinal modulus (E_1) is higher for the CNT coated sisal fiber composite (H1) than for sisal fiber reinforced with homogenized CNT mixed epoxy composite. To enhance the longitudinal modulus of natural fiber, CNT coating is more beneficial than CNT mixing in the matrix material.
- The transverse modulus of sisal fiber is higher for CNT mixed composite than for CNT coated composite.
- The major Poisson's ratio is the same for H1 and H2 composites, and in-plane and out of plane stresses are greater for the H1 model.
- The interfacial stresses are greater for CNT coated composites than for CNT mixed composites as per the conditions considered for the study.
- From this study, it is found that natural fiber like sisal modulus in longitudinal and transverse directions will be enhanced greatly with CNT and CNT mixing in the polymer matrix is better than CNT coated sisal composite in terms of their transverse, in-plane and out-of-plane shear stresses, whereas the longitudinal modulus will be greatly enhanced by CNT mixed epoxy composite.

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