Formulation of nanocellulosic fibres and particle fillers, and their mono and hybrid reinforced polymer composites

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Abstract
Finite element analysis was used to formulate novel models of both mono and hybrid nanocomposites, and, also to predict their flexural properties. This study also formulated the optimal combination mix ratio of fibres and matrix of the flexural properties for use in different applications. The predicted properties were verified using the experimental method. This study found that the flexural properties of all the composites depicted an increase with fibre additions up to a certain optimum point, beyond which they began to fall gradually with further fibre additions. There was also evidence of transverse matrix fracture in rice husk nanoparticle-reinforced epoxy resin composites' flexural stiffness graphs. Moreover, it was found that the increases in flexural properties of mono composites were about two times lower in magnitude than hybrid composites. With regards to the formulation of optimal combination mix ratio of fibres and matrices for different applications, it was noted that the optimal combination mix ratio of fibres to the matrix for applications where all the flexural properties were maximized was 4:1. Moreover, the optimal combination mix ratio of fibres to the matrix for applications where the flexural properties were minimized was 1:2. Furthermore, the optimal combination mix ratio of fibres to the matrix for applications with a target of 10 MPa flexural strength properties and 10 GPa for flexural stiffness properties was 2:3.

1. Introduction

Nanoscience and nanotechnology have opened a completely new way in the development of fibre-reinforced composites. Nanotechnology is a very promising field for improving the properties of fibre-reinforced composites with nanoscale fillers [1].

In Material Engineering, a composite is a material that contains one or more discontinuous phases within a continuous phase. The discontinuous phase also referred to as the reinforcing phase, is usually harder and stronger than the continuous phase. This continuous phase is also called the matrix material [2]. The purpose of the reinforcing phase is to improve the mechanical properties of the composites, while that of the matrix phase is to transmit power to the reinforcing phase. The matrix material also serves to resist heat and chemical agents [2].

Most industries are willing to use nanocellulosic fibres because these fibres are considered renewable resources. Furthermore, they have lower prices, are lighter and have less wear on production equipment compared to synthetic fillers [1, 3].

Nanocellulose materials are also important from an environmental perspective and have numerous advantages, including the following: These materials create very little waste and can be re-used. Furthermore, the wastes produced from nanocellulose materials are not hazardous [3].

There are two main types of nanocellulose materials, namely, nanofibrillated cellulose (NFC), which is sometimes also referred to as microfibrillated cellulose; and cellulose nanocrystals (CNC), which are sometimes also referred to as nanocellulose whiskers [4–6].

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Many researchers have mentioned that nanocellulose materials could be used in medicines, bio-composites, pharmaceuticals, batteries, tissue engineering, bio-sensors, aircraft, paper coatings and many other applications [5].

This study used sisal fibres and rice husks to extract the nanofibres and nanoparticles, respectively, due to their ease of availability. A review of literature is illustrated hereunder:

Roszowska-Jarosz et al [1] studied the effect of addition of a small amount of nanocellulose on epoxy resin to form nanocomposites whereby the mechanical properties were investigated. The fabricated nanocomposites were subjected to mechanical tests such as three-point flexural tests and impact tests. The study concluded that nanocomposites with nanocellulose loading had improved flexural stress at break of about 15% when compared to the unmodified epoxy resin. Thermal testing also indicated a fairly good resistance of the composite at high temperature.

Johar et al [3] used alkali and bleaching method to extract cellulose from rice husk. The authors further investigated the production of nanocrystals from natural fibres. The study concluded that the chemical treatment led to an increase in cellulose content from 31% to 96%. The crystallinity index also increased to 59.0% from 46.8%.

Kargarzadeh et al [4] carried out an excellent review on the existing extraction methods for various kinds of nanocellulose. Specific conditions for extraction of nanocellulose from various natural sources were discussed in detail. The effects of the extraction methods, pretreatments, and conditions on the structure, morphology and properties of nanocellulose were described.

Farooq et al [5], did an excellent review of Cellulose from sources to nanocellulose and an overview of synthesis and properties of nanocellulose/Zinc oxide nanocomposite materials.

Wang et al [7] carried out an excellent review on the preparation of nanocellulose and its potential on reinforced composites. Several methods such as acid hydrolysis, oxidative methods, physical methods, ionic methods among others were reported.

Jaafar et al [8], investigated the effects of rubber toughened epoxy/silica/kenaf composites on mechanical properties and microstructure. The authors noted that the addition of 1 part per hundred of resin (phr) of LMG30 in epoxy/silica/kenaf composite exhibited the highest impact and flexural strength, which were 13.83 kJ m⁻² and 62.2 MPa, respectively. Furthermore, SEM analysis proved that the addition of LMG30 helped in lowering the stress transfer and resulted in optimum mechanical properties.

Collazo-Bigliardi et al [9] did a comparison of the cellulose fibres and the cellulose nanocrystals which had been extracted from both coffee husk and rice husk. The authors used the acid hydrolysis procedure to obtain the cellulose nanocrystals. The researchers further studied the thermal properties, x-ray diffraction among other characterization methods. The study reported that there was similarity in the cellulose contents of rice and coffee husk which were in the range of 34%–55%, making them good sources of cellulosic material for different uses in the industry.

Pham et al [10] used nanosilica obtained from rice husk as a filler material in epoxy resins. The study showed that there was an improvement in fracture toughness by 16.3% upon the addition of nanosilica fillers to the epoxy resins.

Deepa et al [11] reported on the use of acid hydrolysis to extract nanocellulose from sisal fibres. The authors, thereafter characterized the extracted nanofillers by means of microscopy analysis.

Mamat Razali et al [12] extracted cellulose nanofibres from rice straws using both mechanical and chemical treatments. Morphology studies were carried out using Field Emmission Scanning Microscopy (FESEM) and/or transmission electron microscopy (TEM). The fourier-transform infrared spectroscopy was used to perform structural analysis. The study reported that the mechanical properties of Kevlar were improved upon the addition of CNC epoxy laminates.

Suhot et al [13] carried out an excellent review on the recent progress of rice husk reinforced polymer composites between 2017 and 2021. The study recommended that there was need to use optimization techniques in order to reduce the costs of experimental set ups. In addition, the researchers recommended that there was dire need to develop the finite element method in order to compare the mechanical properties of rice husk (RH) composites from experimental works. Ayswarya et al [14] studied the reinforcement effect of rice husk ash (RHA), modified rice husk ash (MRHA) and nanosilica (NS) on epoxy resins. The authors evaluated the mechanical, thermal and dynamic mechanical properties of the composites. The researchers noted that the modulus of the composites increased with fibre additions.

Moosa et al [15] studied the preparation of silica nanoparticles from rice husk ash, which was to be used as nanofiller in epoxy/SiO₂ nanocomposites. Fourier transformation infrared spectroscopy (FTIR) showed that the broadband appeared at about 3429.43 cm⁻¹ which represented the presence of hydroxyl groups on the SiO₂ NPs surface and no further functionalization was needed.

Conradi et al [16] synthesized composite epoxy coatings containing silica particles on austenitic stainless steel. The authors compared the surface morphology and mechanical properties of these coatings. The study
noted that the silica particles significantly improved the microstructure of the coating matrix, which was reflected in an increased hardness, increased surface roughness and induced hydrophobicity.

Jumahat et al. [17] investigated the effect of nanosilica on the tensile stress-strain response of Epikote 828 epoxy polymer. A 40 wt\% nanosilica/epoxy masterbatch was used to prepare a series of nanocomposites with 5–25 wt\% nanosilica content. Static uniaxial tensile tests were conducted to investigate the tensile stress-strain response and tensile properties of unmodified and nanomodified epoxy polymers. The authors noted that the addition of nanosilica improved the tensile properties.

Xue et al. [18] studied a novel type of high strength regenerated cellulose composite fibre reinforced with cellulose nanofibrils and nanosilica. The authors noted that there was an improvement of the tensile strength of the composite cellulose by 47.46\%. Moreover, the authors noted that the surface of the regenerated fibre exhibited a scaly structure with pores, which could be reduced by adding cellulose nanofibrils (CNF) and nanosilica, resulting in the enhancement of physical strength of regenerated fibre.

Nguyen et al. [19] synthesized nanosilica from rice husk. The authors characterized the structural features using energy dispersive x-ray, field emission scanning electron microscopy and Fourier transformation infrared. The authors carried out tests to determine the beneficial effects of nanosilica on seed germination. The seeds were analysed with regard to germination percentage, mean germination time, seed germination index and seed vigour index in order to investigate the efficiency of nanosilica. The authors noted that the samples which added nanosilica had higher average length of shoot and root than the blank samples.

Lanna et al. [20] reported on the performance of a new engineered material made from epoxy resins with modified epoxidized natural rubber and nanofillers. Characterization was done using FTIR, dynamic light scattering among other methods. The study found that the greatest improvement in mechanical properties was shown by the nanofiller loading in epoxy composites. Mor’an et al. [21] carried out a study on the extraction of cellulose and preparation of nanocellulose from sial fibres. The researchers used chemical procedures such as bleaching, alkaline extraction, chlorination and acid hydrolysis. Characterization was done using thermogravimetric analysis (TGA), x-ray diffraction (XRD), Differential Scanning Calorimetry (DSC) and Scanning Electron Microscopy (SEM). The researchers were finally able to extract nanocellulose using acid hydrolysis.

Nia et al. [22], investigated, a new type of silica-cellulose hybrid aerogel that was synthesized through a green and facile chemical cross-linking process. The developed aerogel contained both positively and negatively charged functional groups and was a highly efficient substrate for dye adsorption from water, for both cationic and anionic organic dyes. Moreover, the aerogels were found to have an outstanding adsorption capacity towards methylene blue (MB) as a cationic dye and methyl orange (MO) as an anionic dye.

Ismail et al. [23], used a slow evaporation method to fabricate organic-inorganic hybrid films from cellulose nanofibrils (CNF) and nanosilica (5\%–30\% wt) embedded in a chitosan (Ch) biopolymer matrix. The self-standing films exhibited high strength and modulus up to 120 \pm 5 \text{ MPa} and 7.5 \pm 0.4 \text{ GPa}, respectively, which were remarkably high values for biopolymer/chitosan hybrids.

Bay et al. [24], investigated the thermal and mechanical properties of the nanocomposite made of polyvinyl alcohol (PVA) and nano fiber celluloses (NFC)/-nano silicon dioxide. Samples of the nanocomposite were prepared by the casting method with different nano cellulose and nano silicon dioxide loadings. The authors noted that the strength and modulus of the nano composite were significantly improved with the increase of cellulose nanofibrils loading.

El-Feky et al. [25], evaluated the effects of different contents of nano silica particles (0, 1, 1.5, and 2\% by weight of cement) on the mechanical properties, and microstructure of cement composites reinforced with various contents of nano cellulose fibres (0, 0.35, 0.55, and 0.75\% by weight of cement). The authors revealed, that the addition of the NCell particles separately, helped in improving the strength by about twice the value of the control mix when appropriate content of NCell was incorporated (0.35\%).

Despite the marked growth in the use of nanocellulosic fibres and particles, there is still a dire need to reposition this material system if it is to enjoy progressive, diverse economic feasibility and acceptability in various industrial sectors. This study opened up a new line of thought in the challenge of repositioning the material from an economic, industrial and environmental perspective. It explored the novel idea of the possibility of using new processing technology for modelling composites through the use of Finite Element Analysis (FEA). Finite Element Analysis (FEA) was used to develop novel models of mono and hybrid nanocomposites and to determine their mechanical properties. Moreover, this study sought to formulate an optimization technique using the Minitab software version 2017. This novelty coincides with the recommendations of Suhot et al. [13] in their review paper covering research between 2017 and 2021, whereby they recommended the development of finite element method to compare the mechanical properties of rice husks from experimental works. Furthermore, they also recommended the formulation of an optimization technique. The relevance of the present study is to increase the database of knowledge on the production, properties and use of nanocellulosic fibres and particles. This study used both the numerical simulations and the
nanocomposites were studied. The percentage results arising from both FEA and experimental approaches. This paper has 5 sections. Section 1 is an overview of the background study, rationale, hypothesis and relevance of the study, section 2 outlines both the experimental and numerical procedures followed in this study. Section 3 gives the results and analysis of the work carried out in this study. Section 4 gives the discussion of the results, whereas section 5 gives the conclusions and recommendations for future study.

2. Experimental procedure

The full factorial experimental design was adopted for this study, and was determined using the minitab software. The flexural strength and flexural stiffness mechanical properties for mono and hybrid nanocomposites were studied. The percentage fibre volume fractions (v/%) were varied at an increasing rate until the optimum fibre volume fractions were achieved. Sisal and rice husk fibres were found to be suitable for this study because they could easily be found. This study first extracted sisal nanocellulose fibres and rice husk nanoparticles. These extracted nanocellulose fibres and nanoparticles were then combined separately with epoxy resin to form monocomposites. They were then subsequently, combined together with epoxy resins in order to form hybrid composites. ASTM standards were applied to specimens which were used in this study. A total of 100 samples were prepared and used for testing purposes.

2.1. Materials

Epoxy resins (LR 20), which are commercially found, and their corresponding hardener (LH-281) were used as the binder in this study. The epoxy resin was deemed suitable for use in this study due to the benefits noted by Pham et al. The nanocellulosic fibres were obtained from sisal fibres, while the nanoparticles were obtained from rice husk.

2.2. Extraction of nanocellulosic fibres and nanoparticles

The nanocellulose fibres were obtained from sisal fibres using the standard acid hydrolysis procedure. This procedure of acid hydrolysis was deemed convenient since the acids which are needed for this purpose could be found easily. The sisal fibres were washed in distilled water several times in order to remove any solid particles in them. They were then dried in an oven at 80 °C for 24 h. The fibres were then cut to 5 mm length and dewaxed in a mixture of toluene/ethanol. Following onto this, filtration was done. Subsequently, the fibres were mercerized, treated with hydrogen peroxide and thereafter, immersed in 60 wt% sulphuric acid. The fibres were subsequently washed with deionized water and dried in an oven at 45 °C for 24 h.

Standard procedures were also employed in extraction of nanoparticles from rice husks. The type of nanoparticles which were obtained from rice husks were nanosilica. Nanosilica particles from rice husks for use in this study were obtained using the standard method. Rice husks were mixed with 5% sodium hydroxide and stirred for 1 h. They were then filtered, and thereafter washed with distilled water in order to remove the sodium hydroxide from them. Following onto this, they were immersed in 1% acetic acid to remove any remaining sodium hydroxide. Subsequently, they were immersed into 67.5% solution of hydrochloric acid for 5 h. Following onto this, they were washed with de-ionized water and dried in an oven at 45 °C for 24 h in order to ensure that they were dry. Subsequently, they were calcinated in a furnace at 500 °C for 4 h, then crushed in a ball mill in order to obtain nanosilica particles.

2.3. Composite fabrication

Sisal nanocellulosic fibres and rice husk nanoparticles were weighed separately using an electronic balance with an accuracy of ±0.5 g. Following onto this, masses which had the same fibre volume fractions were grouped together, separately for sisal nanofibres and rice husk nanoparticles. A thin layer of wax was smeared on a rectangular mould measuring 50 cm by 100 cm, so as to ensure that the fabricated composites could be easily removed after curing. In order to fabricate the monocomposites, predetermined nanofibres or nanoparticles corresponding to different fibre volume fractions were spread separately on the mould in a longitudinal direction. In the case of hybrid composites, predetermined fibre volume fractions of combined nanofibres and nanoparticles were spread on the mould in the same way as for the monocomposites. The matrix and its
hardener in their appropriate ratios were also measured by the electronic balance. Subsequently, the matrix and
the hardener were mixed together using a spatula. In order to remove air from the matrix, a sharp needle was
used to puncture the air bubbles. Thereafter, the matrix was spread to the nanofibres and nanoparticles
separately in order to fabricate monocomposites or spread to the combined nanofibres and nanoparticles, in
order, to fabricate hybrid composites. The nanocomposites were subsequently, cured for 24 h.

2.4. Thermal analyses
The following thermal analysis was conducted:

2.4.1. Thermo-gravimetric analysis (TGA)
The thermal stability of the nanocomposite samples was investigated with a thermo gravimetric analyser
called discovery from TA instruments (USA). The heating temperature ranged from 20 °C to 600 °C at a
controlled heating rate of 10 °C min⁻¹. The change in weight of the sample was measured in a controlled
atmosphere, varying temperature as a function of time in a systematic way. The flow rate of high purity
nitrogen was 100 ml min⁻¹.

2.4.2. Differential scanning calorimetry (DSC)
The glass transition temperature (Tg) and melting temperature (Tm) of the samples were determined by DSC
measurements, using a differential scanning calorimeter. The nanocomposite samples were scanned at a rate of
10 °C min⁻¹ from 20 °C to 600 °C, under nitrogen atmosphere and a flow rate of 100 ml min⁻¹.

2.5. Scanning electron microscopy (SEM)
SEM was performed to investigate the morphology of the nanocomposite specimens. SEM micrographs of the
surfaces of the specimens were taken using SEM equipment, model EVO HD 15, with an acceleration voltage of
30 KV. The specimens were coated with gold so as to avoid electrical charging [4]

2.6. Experimental approach to flexural testing
ASTM D 790–02 test standard specifications were adopted during the 3-point bending test done in this study.
Load deflection data for each sample was collected, and the maximum flexural strength and stiffness were
calculated as follows using equations (1) and (2), respectively.

\[ \sigma_f = \frac{3PL}{2bh^2} \] (1)
\[ E_f = \frac{\sigma_f^2 - \sigma_f1}{\varepsilon_f^2 - \varepsilon_f1} \] (2)

Where;
\( \sigma_f \) = Stress at the centre of the specimen (MPa)
\( P \) = Load (N)
\( L \) = Support span of the specimen (mm)
\( b \) = Specimen width (mm)
\( h \) = Specimen thickness (mm)
\( E_f \) = Flexural modulus (MPa)
\( \varepsilon_f \) = Strain at the centre of the specimens

2.7. Composite density and void volume fraction
Specimens which had initially been dried in the oven were dipped in tap water that had a temperature of 22 °C
for a duration of 48 h. Following onto this, the specimens were dried using a towel and subsequently, weighed
using an electronic balance in order to give the saturated mass of the specimens \( W_s \). The saturated specimens
were then dried in an electric oven at a temperature of 100 °C for 24 h. After removing the specimens from the
oven, they were allowed to cool at room temperature conditions and then weighed again. This gave the mass of
the dry specimen \( W_d \). The average amount of water absorbed at each fibre content was calculated. The
percentage water absorption by weight \( W_w \) was obtained from the equation (3) as follows

\[ W_w = \frac{W_s - W_d}{W_d} \times 100 \] (3)

This was followed by the determination of the percentage void volume fraction in accordance with ASTM C
220-75 as given in equation (4).
Table 1. Material definition for composite constituents (Source: experimental data).

| Property/material                     | Sisal nanofibre | Rice husk nanoparticle | Epoxy resin |
|---------------------------------------|-----------------|------------------------|-------------|
| Axial Young Modulus ($E_1$) (MPa)     | 18000           | 15000                  | 3800        |
| In-plane Youngs Modulus ($E_2$) (MPa)| 17500           | 14500                  | 3700        |
| Poisson’s ratio                       | 0.1             | 0.3                    | 0.4         |
| In-plane Shear Modulus ($G_{12}$) (MPa)| 17000          | 14000                  | 3500        |
| Transverse Shear Modulus ($G_{13}$) (MPa)| 17000       | 14000                  | 3500        |
| Transverse Shear Modulus ($G_{23}$) (MPa)| 17000       | 14000                  | 3500        |

\[ V_v = \rho \ V_w \] (4) 

Where, $V_v$ is the void volume fraction and $\rho$ is the density of the specimen. Composite density and void volume values were obtained as an average of 5 specimens at each fibre volume fraction. Finally, the variations of $\rho$ and $V_v$ with reinforcement volume fractions were plotted.

2.8. Finite element analysis of flexural properties of composites

A finite element analysis was formulated for modeling the mono and hybrid nanocomposites, and also to obtain their flexural properties using ABAQUS software. The quad-dominated element shape was employed as it not only uses quadrilateral elements but also allows triangles in transition regions. ABAQUS software was preferred as it allows the computation of accurate eigenvalues and eigenvectors at the nodes.

The values shown in table 1, were obtained experimentally, and were used to create the material definition for sisal nanofibre, rice husk nanoparticle and epoxy resin.

2.9. Validation of results

The Finite Element Analysis (FEA) results generated using ABAQUS/CAE were compared with their corresponding results which were generated experimentally. The experimental solution was selected as the reference method in this study and used as a basis for calculation of error percentages. The results of interest were whether the FEA results generated by Abaqus/CAE models matched with the experimental results. The acceptable error margins were to be within ±5% [26]. Therefore, this study used this standard procedure to determine the degree of correlation between both numerical models and experimental results.

Furthermore, the analysis of variance (ANOVA) was used for validation purposes. In the analysis of variance, the degree of freedom (df), the mean square (ms) and the F statistic are defined and are associated with the normality test under the basic statistics option. The degree of freedom is the difference between the number of observations and the number of groups. The mean square is the average variation either between or within groups. It is a ratio between the sum of squares and the degree of freedom. F statistic is the test statistic. It is a ratio between a mean square (between groups) and a mean square (within groups). If F statistic > F critical, the test is significant, and there is a difference between the means of the group. Moreover, if P value < 0.05, the test is significant. If the two conditions mentioned above are fulfilled, the test is significant, and there is a difference between the means of the groups.

2.10. Formulation of optimal combination mix ratio of fibre and matrix for different applications

The formulation of the combination mix ratio of fibre and matrix that would give optimal results for the properties of a nanocellulosic composite in both mono and hybrid form was done using Minitab software version 2017. The experimental design to identify the best option in fibre and matrix, which upon combination would yield an optimal design for mono and hybrid composites, was formulated using the Minitab software version 2017. The fibre and matrix were held as the independent variables whereas, flexural strength and flexural stiffness were held as the dependent variables. Therefore, the use of the Minitab software allowed the combination of these variables to yield an optimal solution.

The Minitab software was used to determine the optimal combination mix of the fibre and the matrix for the following different applications:

- Where all the dependent variables (flexural properties) of mono and hybrid composites were maximized
- Where all the flexural properties of mono and hybrid composites were minimized.
- Where a target of 10 MPa for flexural strengths, and 10 GPa and flexural stiffness of mono and hybrid composites were considered.
3. Results

3.1. Thermal analyses

Both Thermal Gravimetric Analysis (TGA) and the Dynamic Scanning Calorimetry (DSC) for sisal nanofibre reinforced epoxy resin composites, rice husk nanoparticle reinforced epoxy resin composites, and sisal/rice husk hybrid reinforced epoxy resin nanocomposites were investigated. The Thermal Gravimetric Analysis curves and the Dynamic Scanning Calorimetry curves for all the composite samples tested were plotted on the same graph as shown in figure 1.

From figure 1, it can be observed that the Thermal Gravimetric Analysis curves exhibit 3 sections: an initial flat section, then a section with a constant slope and finally a flat section. At the initial flat section, there was a weight loss of 5%, 10% and 10% for the sisal/rice husk hybrid reinforced epoxy resin nanocomposite, rice husk nanoparticle reinforced epoxy resin composites and, sisal nanofibre reinforced epoxy resin nanocomposite samples, respectively. At the constant slope section, there is a weight loss of 80%, 75% and 75% for sisal/rice husk hybrid reinforced epoxy resin nanocomposite, rice husk nanoparticle reinforced epoxy resin composites, and sisal nanofibre reinforced epoxy resin composite samples, respectively. Finally, all the mass degraded and left behind a black carbonaceous residue known as char, as is clearly depicted in the last flat sections.

The initial weight loss of all samples was attributed to the volatilization of water adsorbed in the composites before 100 °C [9, 22], and the second stage began at temperatures between 250 °C and 300 °C. This was caused by the absorbed moisture and the degradation of the lignocellulotic components. All the samples of sisal/rice husk hybrid reinforced epoxy resin nanocomposite, rice husk nanoparticle reinforced epoxy resin composites and, sisal nanofibre reinforced epoxy resin nanocomposite samples fully degraded at temperatures of between 400 °C and 475 °C. The complete degradation in the fibre composites arose when all the cellulose had fully degraded. The full degradation can be evidenced by the sudden levelling of the curve. It is also evident that the hybrid nanocomposites showed good thermal endurance compared to those of the mono composites. This observation is in agreement with the works of Jumahat et al [27].

Dynamic Scanning Calorimetry is performed to study the glass transition (Tg) temperatures and melting temperatures (Tm) of the nanocomposites. The DSC curves of all formulated nanocomposites are shown in figure 1. In the Dynamic Scanning Calorimetry (DSC) graphs, all the samples exhibited a glass transition temperature of between 50 °C and 75 °C. Further, all the samples exhibited a melting temperature of between 350 °C and 400 °C.

The first peak of the DSC curve of sisal/rice husk hybrid reinforced epoxy resin nanocomposite, rice husk nanoparticle reinforced epoxy resin composites and, sisal nanofibre reinforced epoxy resin composite samples in figure 1 corresponds to a heat flow of 0.3 W g⁻¹, 0.6 W g⁻¹ and 0.7 W g⁻¹, respectively, which results in the
loss of moisture. The remaining material seemed to be thermally stable up to a temperature of 325 °C, 350 °C and 350 °C for sisal/rice husk hybrid reinforced epoxy resin nanocomposite, rice husk nanoparticle reinforced epoxy resin composites, and sisal nanofibre reinforced epoxy resin composite samples, respectively. Thereafter, the pyrolysis rate reduced until it reached the maximum at 400 °C, 380 °C and 390 °C for sisal/rice husk hybrid reinforced epoxy resin nanocomposite, rice husk nanoparticle reinforced epoxy resin composites and sisal nanofibre reinforced epoxy resin composite samples, respectively. From 400 °C to 600 °C, pyrolysis continued at a very low rate progressively.

3.2. Composite density and void volume variation

Figure 2 shows the variation of the composite density with fibre content in the mono and hybrid composites fabricated. A general trend of decreasing density was seen with increasing reinforcement volume fractions. A nearly constant rate of decrease was seen initially, but at higher reinforcement levels, the composite showed a sudden decrease.

A plot of the void volume value against the reinforcement volume fraction is shown in figure 3. The void volume value was seen to increase with increasing fibre contents. A sudden rate of increase in the number of voids was seen up to around 3.000 v% This was followed by a region of gentle increase up to about 6.5 v%. Finally, a sudden rate of increase was seen again until the maximum fibre volume fraction was attained.
The density and void volume fraction in a material are interrelated because both depend on the number of voids in the material. The fibre content in the material was found to influence the composite unit weight. Unit weight varies at every fibre volume fraction ($\nu_f$) with the composite becoming lighter as fibre content increases. The composite density was therefore seen to decrease, as shown in figure 2. This can be attributed to an increase in the number of voids due to poor workability at high reinforcement levels [28]. The present results also showed an increase in void volume with increasing fibre contents as shown in figure 3 which may probably be due to agglomeration of the nanofillers and inhomogeneity caused to the epoxy matrix [23]. The agglomeration phenomenon was probably due to the decrease in distance between the nanofillers resulting in an increase in the
van der Waals attraction (aggregation factors) and a reduction in the electrostatic repulsion (dispersion factors), which in turn resulted in an irreversible agglomeration and led to an increase in void volume [23, 29].

The present results are in agreement to those of Nia et al [22]. The authors noted similar observations when they determined porosity measurements on hybrid aerogel nanocomposite of dendritic colloidal silica and hairy nanocellulose.

3.3. Scanning electron microscopy

SEM results of the sisal nanocellulosic fibres and the rice husk nanoparticles are illustrated in figures 4 and 5.

It is evident from figure 4 that the hemicellulose, lignin, pectin and wax were effectively removed using the acid hydrolysis procedure of nanocellulose extraction since the sizes of the nanofibres were 5 μm as evidenced from the SEM image. It can also be observed from figure 5, that the diameters of the nanoparticle were 10 μm, which implied that the chemicals were very effective in removing the non-cellulosic materials inorder for the nanocellulosic materials to remain. Different fibre sizes have been reported by different authors depending on the plant fibres used. As an example, water hyacinth fibre was 25–50 μm in size [30].

The acid hydrolysis procedure is considered instrumental in removing the amorphous regions so that the cellulose regions remain behind. Moreover, the acid hydrolysis procedure reduce these cellulotic regions to nanometric sizes [31]. The aspect ratios of the nanofibres were found to be 35.5. It has also been reported by Kallel et al [32] that the aspect ratio usually vary from 10 to 70 for cellulose nanocrystals [CNC] extracted from different materials. Despite this finding by Kallel et al [32], higher values have been obtained from garlic straw (L/d = 80) [9, 32]. It has also been reported by Silv‘ério et al [33] that for cellulose nanocrystals to be considered as a good reinforcing material, their aspect ratios must exceeds a value of 10. In this study, majority of the sisal nanofibres and rice husk nanoparticles had aspect ratios which exceeded 10, implying that they were good reinforcing material.

3.4. Results of flexural strength for sisal nanofibre reinforced epoxy resin composites

The flexural strength results for sisal nanofibre reinforced epoxy resin composites, which were obtained using both the experimental and FEA methods are shown in table 2.

| Fibre volume fraction (v%) | FEA results (MPa) | Experimental results (MPa) |
|---------------------------|-------------------|---------------------------|
| 0.00                      | 2.543             | 2.646 ± 0.251             |
| 1.672                     | 4.405             | 3.924 ± 0.455             |
| 2.691                     | 5.299             | 4.962 ± 0.549             |
| 3.355                     | 6.210             | 5.346 ± 0.532             |
| 4.711                     | 7.696             | 6.444 ± 0.613             |
| 5.754                     | 8.046             | 7.189 ± 0.522             |
| 6.750                     | 8.345             | 7.524 ± 0.550             |
| 7.783                     | 8.549             | 8.285 ± 0.525             |
| 8.516                     | 9.702             | 8.494 ± 0.714             |
| 9.305                     | 9.017             | 8.610 ± 0.988             |
| 11.200                    | 7.991             | 7.810 ± 0.540             |

The standard deviations recorded in table 2 at all fibre fractions for the experimental results were found to be very low, ranging from 0.251 to 0.714. However, the variations between specimens at a given fibre volume fraction was found to be larger for fibre reinforced specimens than in unreinforced test pieces. This can be attributed to the difficulty in achieving the same uniform distribution and orientation of fibres.
3.4.1. Comparison of FEA and experimental results of flexural strength for sisal nano fibre reinforced epoxy resin composites

Figure 6 shows a comparison of the results obtained from both the finite element analysis (FEA) and experimental approaches.

It can be seen from figure 6 that for the experimental results, the rate of increase of flexural strength with fibre volume fraction is fairly rapid initially up to fibre content of about 6.8 $\nu_f$%, thereafter, the rate of increase reduces with fibre additions up to the maximum strength, corresponding to 9.5 $\nu_f$% fibre content. For the FEA results, a steady increase in flexural strength was observed up to the maximum point. The FEA results gave the maximum flexural strength at around 9 $\nu_f$% fibre volume fraction, corresponding to around 9.8 MPa. The coefficient of correlation shown in the graphs (0.9825 and 0.9966) for both the FEA and experimental results were found to be closer to 1, implying a perfect fit of the data set.

3.4.2. Validation of FEA results of flexural strength for sisal nanofibre reinforced epoxy resin composites

The FEA results of sisal nanofibre reinforced epoxy resin composites were validated by computing the percentage errors between them and the experimental results, as shown in table 3.

It can be seen from table 3 that the deviation between experimental and FEA results is insignificant and in close agreement for 36.36% of the results since the margin of error is within the acceptable engineering percentage error limit of ±5%.

![Figure 6: A graph of fibre volume fraction versus flexural strength for sisal nanofibre-reinforced epoxy resin composite.](image)

Table 3. Error margin of experimental and FEA results.

| Fibre volume fraction ($\nu_f$) | FEA (MPa) | Experimental (MPa) | % Error |
|-------------------------------|-----------|--------------------|---------|
| 0.000                         | 2.543     | 2.646              | -3.893  |
| 1.672                         | 4.405     | 3.924              | 12.258  |
| 2.691                         | 5.299     | 4.962              | 6.792   |
| 3.355                         | 6.210     | 5.346              | 16.162  |
| 4.711                         | 7.696     | 6.444              | 19.429  |
| 5.754                         | 8.046     | 7.189              | 11.921  |
| 6.750                         | 8.345     | 7.524              | 10.912  |
| 7.783                         | 8.549     | 8.285              | 3.186   |
| 8.516                         | 9.702     | 8.494              | 14.222  |
| 9.505                         | 9.017     | 8.610              | 4.727   |
| 11.200                        | 7.991     | 7.810              | 2.318   |

Mater. Res. Express 9 (2022) 035404 W Webo et al
3.5. Results of flexural strength for rice husk nanoparticle reinforced epoxy resin composites

The results of flexural strength for rice husk nanoparticles reinforced epoxy resin composite specimens are shown in Table 4.

A maximum value of 4.923 MPa was obtained for the flexural strength at the fibre content of 8.516 μf%. The flexural strength of unreinforced epoxy specimens was obtained as 2.646 MPa. Furthermore, results from Table 4 show that the increments of experimental flexural strength values for rice husk nanoparticle reinforced-epoxy resin composites over the values obtained for the unreinforced specimens were: 11.07%, 5.83%, 4.25% and 0.51% at fibre contents of 1.672 μf%, 4.711 μf%, 6.750 μf%, and 8.516 μf%, respectively. Standard deviations for all fibre volume fractions were found to be very low, with the lowest value being recorded at 2.691 μf%. This low standard deviation implies that there was no much scatter in the data set. However, the variations between specimens at a given fibre volume fraction was found to be larger for fibre reinforced specimens than in unreinforced test pieces. This can be attributed to the difficulty in achieving the same uniform distribution and orientation of fibres.

3.5.1. Comparison of FEA and experimental results of flexural strength for rice husk nanoparticle reinforced epoxy resin composites

Figure 7 shows the results obtained from both the experimental and finite element analysis approach for rice husk nanoparticle reinforced epoxy resin nanocomposites. It can be seen from Figure 7 that for the experimental results, the rate of increase of flexural strength with fibre volume fraction is fairly rapid initially up to fibre content of about 7.8 μf%, then the rate of increase reduces with fibre additions. For the FEA results, a steady increase in flexural strength was observed up to fibre content of about 3.5 μf%, beyond which there was a reduction in flexural strength. It is also evident that the FEA plot and

![Figure 7](attachment:image.png)

**Table 4.** Flexural strength values for rice husk nanoparticle reinforced epoxy resin composites.

| Fibre volume fraction (μf%) | FEA (MPa) | Experimental (MPa) |
|-----------------------------|-----------|--------------------|
| 0.000                       | 2.543     | 2.646 ± 0.251      |
| 1.672                       | 3.145     | 2.939 ± 0.247      |
| 2.691                       | 3.955     | 3.849 ± 0.141      |
| 3.355                       | 4.226     | 4.079 ± 0.212      |
| 4.711                       | 4.106     | 4.317 ± 0.190      |
| 5.754                       | 3.894     | 4.568 ± 0.232      |
| 6.750                       | 3.799     | 4.762 ± 0.222      |
| 7.783                       | 3.689     | 4.898 ± 0.210      |
| 8.516                       | 3.581     | 4.923 ± 0.155      |
| 9.505                       | 3.436     | 4.156 ± 0.225      |
| 11.200                      | 3.391     | 3.572 ± 0.241      |
the experimental plot intersect at around 3.6 \( \nu_f \)%, with the corresponding flexural strength being 4 MPa. The coefficients of correlation shown in the graphs (0.9815 and 0.9414) are closer to 1, implying a perfect data set fit.

3.5.2. Validation of FEA results of flexural strength for rice husk nanoparticle reinforced epoxy resin composites

The experimental results of rice husk nanoparticle reinforced epoxy resin composites were used as the reference in order to validate the FEA results. This validation was done by computing the percentage errors between them and the FEA results, as shown in table 5.

It is evident from table 5 that the deviation between experimental and FEA results are in close agreement for 45.45% of the values recorded since their error values are within the acceptable engineering percentage of errors of \( \pm 5\% \). The other 54.55% of the experimental results have errors which are significant since their margin of error is outside the acceptable engineering percentage of error of \( \pm 5\% \). These deviations can be attributed to fibre bunching in some instances. Similar observations were noted by Bisanda [33].

3.6. Results of flexural strength for sisal/rice husk hybrid reinforced epoxy resin nanocomposites

Table 6 shows flexural strength results which were obtained from sisal/rice husk hybrid reinforced epoxy resin nanocomposites.

A Maximum value of 14.769 MPa was obtained for the flexural strength at 8.516 \( \nu_f \)% . It was also evident that in the FEA results, the increase in flexural strength over the values for the unreinforced specimens were 369.13%, 493.39%, 404.92% and 396.26% at 2.691 \( \nu_f \)% , 5.754 \( \nu_f \)% , 9.505 \( \nu_f \)% and 11.200 \( \nu_f \)% , respectively. The corresponding increments for the experimental results at the same fibre volume fractions were 292.74%, 417.91%, 371.20% and 304.99%, respectively.

3.6.1. Comparison of experimental and FEA results of flexural strength for sisal/rice husk hybrid reinforced nanocomposites

A graph of flexural strength versus fibre volume fraction of sisal/rice husk hybrid reinforced epoxy resin nanocomposites showing both experimental and finite element analysis (FEA) results is presented in figure 8 hereunder.
A general trend was seen of flexural strengths increasing with fibre additions for both the experimental and FEA approaches. In the experimental approach, it is evident that the optimum flexural strength of sisal/rice husk hybrid reinforced epoxy resin nanocomposites was realized at around 7\(\%\) fibre. Beyond these optimum values of reinforcement volume fraction, the flexural strength began to fall gradually with fibre additions. The rate of increase of flexural strength with fibre contents decreased at higher fibre volume fractions, as can be depicted in figure 8.

In the FEA approach, it is evident that there is a steady increase in the flexural strength up to the optimum point. The optimum point is at around 5.8 \(\%\) fibre. Beyond this optimum point, the flexural strength begins to reduce with an increase in fibre additions.

It is also evident that both the FEA and the experimental plots intersect at around 6.5 \(\%\) fibre and 9.6 \(\%\) fibre. The coefficient correlation of both the FEA and the experimental plot (0.9726 and 0.9657), as seen in figure 8, is closer to 1, implying a perfect fit of the data set.

### 3.6.2. Validation of FEA results of flexural strength for sisal/rice husk hybrid reinforced epoxy resin nanocomposites

The FEA results of sisal/rice husk hybrid reinforced epoxy resin nanocomposites were validated by computing the percentage errors between them and the experimental results, as shown in table 7.

| Fibre volume fraction \((\%f)\) | FEA \((\text{MPa})\) | Experimental \((\text{MPa})\) | % Error |
|-------------------------------|----------------|-----------------|---------|
| 0.000                         | 2.543          | 2.646           | -3.893  |
| 1.672                         | 10.790         | 9.603           | 12.361  |
| 2.691                         | 11.930         | 10.392          | 14.800  |
| 3.355                         | 12.610         | 12.237          | 3.048   |
| 4.711                         | 13.490         | 12.951          | 4.162   |
| 5.754                         | 15.090         | 13.704          | 10.114  |
| 6.750                         | 14.620         | 14.286          | 2.338   |
| 7.783                         | 14.080         | 14.694          | -4.179  |
| 8.516                         | 13.550         | 14.769          | -8.254  |
| 9.505                         | 12.840         | 12.468          | 2.984   |
| 11.200                        | 12.620         | 10.716          | 17.768  |

It can be seen from table 7 that the deviation between FEA and experimental results is insignificant and are in close agreement for 54.55\% of the results since the margin of error is within the acceptable engineering percentage of error of ±5\%.

### 3.7. Comparison of flexural strength of mono and hybrid nanocomposites

Figure 9 compares the flexural strength of sisal nanofibre reinforced epoxy resin composites, rice husk nanoparticle reinforced epoxy resin composites and sisal/rice husk hybrid reinforced epoxy resin nanocomposite.
From the graph in figure 9, it is clear that sisal/rice husk hybrid reinforced epoxy resin nanocomposite had the highest values of flexural strength. This was followed by sisal nanofibre reinforced epoxy resin composites and, finally, rice husk nanoparticle reinforced epoxy resin composites. It is also evident that the Flexural strength of sisal/rice husk hybrid reinforced epoxy resin nanocomposite is almost twice as much as the flexural strength of sisal nanofibre reinforced epoxy resin composites and rice husk nanoparticle reinforced epoxy resin composites. The better performance of hybrid composites than the monocomposites can be attributed to a bigger increase in the fibre loading in the hybrid composites than in the monocomposites.

### 3.8. Analysis of variance (ANOVA) for flexural strength

Table 8 shows a summary of variances of the mono and hybrid composites under experimental and the FEA approaches, whereas table 9 shows the analysis of variance between and within groups.

**Figure 9.** Comparison of flexural strength of mono and hybrid composition.

**Table 8.** Summary of variances within the mono and hybrid groups.

| Summary Groups                                              | Count | Sum    | Average | Variance |
|--------------------------------------------------------------|-------|--------|---------|----------|
| Sisal nanofibre reinforced epoxy resin composites(Experimental)) | 11    | 77.803 | 7.073   | 4.820    |
| Sisal nanoparticle reinforced epoxy resin composites (FEA)    | 11    | 71.234 | 6.476   | 4.004    |
| Rice husk nanoparticle reinforced epoxy resin composites(Experimental) | 11    | 39.765 | 3.615   | 0.231    |
| Rice husk nanoparticle reinforced epoxy resin composites (FEA) | 11    | 44.709 | 4.064   | 0.581    |
| Hybrid(Experimental)                                         | 11    | 134.163| 12.197  | 11.730   |
| Hybrid(FEA)                                                  | 11    | 128.466| 11.679  | 12.058   |

**Table 9.** Analysis of variance between groups and within groups.

| Source of variation | SS      | df | MS     | F      | P-value | F crit |
|---------------------|---------|----|--------|--------|---------|--------|
| Between Groups      | 744.104 | 5  | 148.820| 26.7164| 4.22E-14| 2.368  |
| Within Groups       | 334.223 | 60 | 5.571  |        |         |        |
| Total               | 1078.327| 65 | 5.371  |        |         |        |
It is evident from the results presented in table 8, that the sisal/rice husk hybrid reinforced epoxy resin nanocomposite done using the FEA approach had the highest variance of 12.058 while the rice husk nanoparticle reinforced epoxy resin composites done using the experimental approach had the lowest value of the variance of 0.231. Furthermore, it is evident from table 9 that the variability between the groups was 744.104, while the variability within the groups was 334.223. Moreover, The F statistic test and p statistic test indicate that these tests are significant. Since the conditions stated in section 2.9 of this paper are fulfilled, it means that the difference between the means of the six groups is deemed significant.

3.9. Results of flexural stiffness for sisal nanofibre reinforced-epoxy resin composites

The FEA and experimental results of flexural stiffness for sisal nanofibre reinforced epoxy resin composites at varying reinforcement volume fractions ($\nu_f$) are shown in table 10.

From table 10, it is evident that the highest flexural stiffness values were obtained at 6.750 $\nu_f$% and 7.783 $\nu_f$% for FEA and experimental approaches, respectively. The corresponding values of flexural stiffness at these points were 11.570 GPa and 13.670 GPa, for FEA and experimental approaches, respectively.

3.9.1. Comparison of FEA and experimental results of flexural stiffness for sisal nanofibre reinforced epoxy resin composites

A graph of flexural stiffness versus fibre volume fraction of sisal nanofibre reinforced epoxy resin composites showing both experimental and FEA results is presented in figure 10 hereunder.

From figure 10, it is evident that at around 5 $\nu_f$%, both the FEA and the experimental approaches gave the same value of flexural stiffness corresponding to around 9 GPa. Furthermore, it can be observed that the critical/optimum reinforcing fibre volume fraction of flexural stiffness under the FEA and the experimental approaches were at around 7.8 $\nu_f$% and 8.2 $\nu_f$%, respectively. The corresponding flexural stiffness values at these critical/optimum points were around 9.8 GPa and 12.2 GPa. Beyond these optimum values of reinforcement volume fractions, the flexural stiffness began to reduce gradually with fibre additions in both the FEA and the experimental approaches. This reduction of flexural stiffness beyond the critical values can be attributed to instances of fibre bunching at higher fibre volume fractions [28].

3.9.2. Validation of FEA results of flexural stiffness for sisal nanofibre reinforced epoxy resin composites

The approach stated in section 2.9 of this thesis was used to validate the FEA results, whereby the experimental results were used as the reference. The percentage errors of the results obtained through the FEA approach in relation to those obtained from the experimental approach have been computed in table 11.

It can be seen from table 11 that 36.36% of the results have margins of error that are within the acceptable engineering error limit percentage of ±5%. It is also evident that for 63.64% of the results, the margins of error
are outside the acceptable engineering error limits of ±5%. These deviations can be attributed to the difference in the accuracy levels of both the FEA and the experimental approaches.

### Table 10. Flexural stiffness values for sisal nanofibre reinforced-epoxy resin composites.

| Fibre content (υ%) | FEA (GPa) | Experimental (GPa) |
|--------------------|-----------|--------------------|
| 0.000              | 4.200     | 3.800 ± 0.250      |
| 1.672              | 4.581     | 4.400 ± 0.150      |
| 2.691              | 7.261     | 6.200 ± 0.225      |
| 3.355              | 8.009     | 7.500 ± 0.350      |
| 4.711              | 10.490    | 9.250 ± 0.200      |
| 5.754              | 9.832     | 10.570 ± 0.215     |
| 6.750              | 11.570    | 12.644 ± 0.265     |
| 7.783              | 10.450    | 13.670 ± 0.215     |
| 8.516              | 9.250     | 11.350 ± 0.245     |
| 9.505              | 8.860     | 10.250 ± 0.230     |
| 11.200             | 7.780     | 9.250 ± 0.200      |

### Table 11. Error margin of FEA and experimental results.

| Fibre content (%) | FEA (MPa) | Experimental (MPa) | % Error |
|-------------------|-----------|--------------------|--------|
| 0.000             | 4.200     | 4.300              | −2.326 |
| 1.672             | 4.581     | 4.400              | 4.114  |
| 2.691             | 7.261     | 6.200              | 17.113 |
| 3.355             | 8.009     | 7.700              | 4.013  |
| 4.711             | 10.490    | 9.250              | 13.405 |
| 5.754             | 9.832     | 10.270             | −4.265 |
| 6.750             | 11.570    | 12.644             | −8.494 |
| 7.783             | 10.450    | 13.670             | −23.555|
| 8.516             | 9.250     | 11.350             | −18.502|
| 9.505             | 8.860     | 10.250             | −13.561|
| 11.200            | 7.780     | 9.250              | −15.892|

### Table 12. Flexural modulus values for rice husk nanoparticle reinforced-epoxy resin composites.

| Fibre content (%) | FEA (GPa) | Experimental (GPa) |
|-------------------|-----------|--------------------|
| 0.000             | 4.200     | 4.300 ± 0.250      |
| 1.672             | 3.963     | 3.870 ± 0.350      |
| 2.691             | 4.227     | 3.910 ± 0.200      |
| 3.355             | 3.894     | 4.050 ± 0.150      |
| 4.711             | 3.681     | 4.630 ± 0.200      |
| 5.754             | 3.585     | 4.510 ± 0.250      |
| 6.750             | 6.154     | 5.870 ± 0.150      |
| 7.783             | 6.365     | 5.910 ± 0.200      |
| 8.516             | 6.654     | 6.290 ± 0.130      |
| 9.505             | 7.200     | 6.630 ± 0.200      |
| 11.200            | 6.800     | 6.510 ± 0.250      |

### 3.10. Results of flexural stiffness for rice husk nanoparticle reinforced-epoxy resin composites

The FEA and experimental results of flexural stiffness for rice husk nanoparticle reinforced epoxy resin composites, which were done using the FEA and experimental methods are shown in table 12.

From table 12, it is evident that the highest flexural stiffness for both the FEA and experimental approaches were recorded at 9.505 υ%. The corresponding flexural stiffness values at 9.505 υ% for both the FEA and experimental approaches were 7.200 GPa and 6.630 GPa, respectively. The increments in flexural stiffness values over the values recorded for the unfilled matrix in the FEA approach were −7.286%, 46.52% and 61.90% at
3.355 $\nu_f$, 6.750 $\nu_f$ and 11.200 $\nu_f$, respectively. The increments in the same fibre volume fractions as the FEA approach were $-5.814\%$, $36.51\%$, and $51.40\%$ in the experimental approach.

3.10.1. Comparison of FEA and experimental results of flexural stiffness for rice husk nanoparticle reinforced epoxy resin composites

A graph of flexural stiffness versus fibre volume fraction of rice husk nanoparticle reinforced epoxy resin composites showing both experimental and FEA results is presented in figure 11.

From figure 11, it can be observed that the minimum flexural stiffness reinforcing fibre volume fraction in the FEA and experimental approaches were around 2.1 $\nu_f$ and 1.8 $\nu_f$, respectively. The corresponding stiffness values at these minimum flexural stiffness reinforcing fibre volume fractions in both the FEA and experimental approach were around 3.5 GPa and 3.8 GPa, respectively. It is also evident that the curves to the right have two different gradients in both the FEA and experimental approaches, which is a sign of transverse matrix fracture. The advent of the transverse matrix fracture begins at around 8 $\nu_f$.

3.10.2. Validation of FEA results of flexural stiffness for rice husk nanoparticle reinforced epoxy resin composites

The percentage errors of the results obtained using the FEA approach in relation to the results obtained using the experimental approach have been computed in table 13.

From table 13, it can be observed that the deviations between the FEA and experimental results are insignificant and in close agreement for 45.45% of the results since the margin of error is within the acceptable engineering percentage error limit of $\pm 5\%$. It is also evident that the errors obtained from the other 54.55% of
the results are outside the acceptable engineering percentage of error of ±5%. These deviations arise from the fact that both FEA and experimental approaches have different levels of accuracy.

3.10.3. Results of flexural stiffness for sisal/rice husk hybrid reinforced-epoxy resin nanocomposites

The results for sisal/rice husk nanoparticle reinforced epoxy resin composites obtained using both the FEA and experimental methods are shown in table 14 hereunder.

| Fibre volume fraction (v%) | FEA (GPa) | Experimental (GPa) |
|---------------------------|-----------|--------------------|
| 0.000                     | 4.200     | 4.300 ± 0.250      |
| 1.672                     | 8.345     | 8.254 ± 0.220      |
| 2.691                     | 9.859     | 9.956 ± 0.380      |
| 3.355                     | 12.880    | 13.060 ± 0.250     |
| 4.711                     | 14.010    | 14.840 ± 0.230     |
| 5.754                     | 15.100    | 16.400 ± 0.350     |
| 6.750                     | 14.525    | 14.875 ± 0.235     |
| 7.785                     | 14.215    | 14.300 ± 0.150     |
| 8.516                     | 14.040    | 17.920 ± 0.225     |
| 9.505                     | 13.570    | 15.440 ± 0.315     |
| 11.200                    | 10.615    | 10.453 ± 0.265     |

3.10.4. Comparison of results of flexural stiffness for sisal/rice husk hybrid reinforced epoxy resin nanocomposites

A graph of flexural stiffness versus fibre volume fraction of sisal/rice husk hybrid reinforced epoxy resin nanocomposites showing both experimental and FEA results are presented in figure 12 hereunder.

The graph in figure 12 shows a steady increase in flexural stiffness up to the optimum point for both the experimental and FEA approaches, after which the increase in flexural stiffness with fibre additions reduces gradually. The optimum flexural stiffness corresponds to around 6.7 v% for the FEA method and a range of about 7.6 v% to 7.8 v% for the experimental approach. Beyond these optimal values, the flexural stiffness begins to reduce. This reduction in flexural stiffness beyond the optimum level could be attributed to fibre bunching at higher fibre contents [28].
3.10.5. Validation of FEA results of flexural stiffness for sisal/rice husk hybrid reinforced epoxy resin nanocomposites

The percentage errors of the results obtained through the FEA approach in relation to those obtained using the experimental approach have been computed in Table 15.

It can be seen from Table 15 that the deviation between FEA and experimental results are insignificant and in close agreement for 63.64% of the results since the margin of error is within the acceptable engineering percentage of error of ±5%. For 36.36% of the results, the margin of error falls outside the acceptable engineering percentage of error of ±5%. This deviation could be attributed to the difference in accuracy levels between the FEA and the experimental results.

3.10.6. Comparison of flexural stiffness of mono and hybrid nanocomposites

Figure 13 shows a graph comparing the flexural stiffness of sisal nanofibre reinforced epoxy resin composites, rice husk nanoparticle reinforced epoxy resin composites and sisal/rice husk hybrid reinforced epoxy resin nanocomposites. A total of 6 graphs have been plotted, indicating both the results of both the FEA and the experimental approaches.

From figure 13, it is evident that sisal/rice husk hybrid reinforced epoxy resin nanocomposite had the highest values of flexural stiffness. This peak magnitude was followed by sisal nanofibre reinforced epoxy resin composites and, finally, rice husk nanoparticle reinforced epoxy resin composites. Furthermore, both the mono and hybrid composites have steady increases in their flexural stiffness values with increasing fibre addition to the optimum level.
This increment in the flexural stiffness property up to the optimal level of both the mono and hybrid composites could be attributed to an increase in adhesion between the fibres and the matrix.

### 3.10.7. Analysis of variance (ANOVA) for flexural stiffness

Table 16 shows a summary of variances of the mono and hybrid composites under FEA and the experimental approaches, whereas table 17 shows the analysis of variance between and within groups.

| SUMMARY Groups                                      | Count | Sum   | Average | Variance |
|-----------------------------------------------------|-------|-------|---------|----------|
| Sisal nanofibre composite (Experimental)             | 11    | 92.283| 8.389   | 5.566    |
| Sisal nanofibre composite (FEA)                      | 11    | 99.284| 9.026   | 9.668    |
| Rice husk nanoparticle composite (Experimental)      | 11    | 56.723| 5.157   | 2.102    |
| Rice husk nanoparticle composite (FEA)               | 11    | 56.480| 5.135   | 1.222    |
| Hybrid (Experimental)                               | 11    | 131.359| 11.942 | 11.300   |
| Hybrid (FEA)                                        | 11    | 139.798| 12.709 | 16.335   |

Table 17. Analysis of variance between groups and within groups.

| Anova Source of variation | SS     | df | MS     | F       | P-value | F crit |
|---------------------------|--------|----|--------|---------|---------|--------|
| Between Groups            | 572.512| 5  | 114.503| 14.873  | 1.76E-09| 2.368  |
| Within Groups             | 461.933| 60 | 7.699  |         |         |        |
| Total                     | 1034.445| 65 |        |         |         |        |

This increment in the flexural stiffness property up to the optimal level of both the mono and hybrid composites could be attributed to an increase in adhesion between the fibres and the matrix.

### 3.11. Formulation of optimal combination mix ratio of fibre and matrix for different applications

The minitab software was used to formulate the optimal mix ratio of fibres and matrices which were validated by the experimental approach. For applications that require the flexural properties to maximized, the ratio was found to be 4:1. When all the flexural properties were minimized, the ratio was found to be 1:2. For applications that required a target value of flexural strength of 10 MPa and flexural stiffness of 10 GPa, the ratio was found to be 2:3.

### 4. Discussions

#### 4.1. Fibre volume fraction effects on flexural strength

The general trend in both the FEA and experimental approach is shown in figures 6–8, which shows that there is a steady increase in the rate of flexural strength between 0 % f/v and the optimal points. After the optimal points, there was a reduction in the rate of increase of flexural strength with fibre additions. This increase in the flexural strength up to the optimal point is due to the high specific strength, homogeneity and self assembly of the nanofibres and the nanoparticles [23, 34]. Moreover, this increase in strength may be due to the inherent high flexural strength of nanofibres and nanoparticles which acted as a suitable reinforcer and was able to form a good bond with the polymer [24].
When sisal nanofibres, rice husk nanoparticles and sisal/rice husk hybrid were used as reinforcement, it was found that there was a limit beyond which fibre additions did not improve the strength of the composites. This limit occurred at 9.5 v/%, and 8.5 v/% in the experimental and FEA approaches, respectively, in sisal nanofibres. In rice husk nanoparticles, this limit occurred at around 7.8 v/%, and 3.4 v/% in experimental and FEA approaches, respectively. In sisal/rice husk hybrid reinforced epoxy resin nanocomposites, these limits occurred at around 7 v/% in both the experimental and FEA approaches. This apparent loss in strength beyond a certain volume fraction can be ascribed to a number of reasons namely:

(a) Poor compaction at high fibre volume fractions is a result of an increase in the number of voids in the composites, especially as the mixture becomes more and more unworkable, which subsequently, results in lowering of the fibre/matrix adhesion and, consequently, the composite strength [28].

(b) In the case of hybrid composites, agglomeration, balling-up and curling of fibres increased with an increase in fibre contents resulting in inhomogeneity of the nanofillers within the matrix [23]. The agglomeration phenomenon was probably due to the decrease in distance between the nanofillers resulting in an increase in the van der waals attraction (aggregation factors) and a reduction in the electrostatic repulsion (dispersion factors), which in turn resulted in an irreversible agglomeration and led to a decrease in flexural strength [23, 29].

These two phenomena could have resulted in the formation of more voids and eventually decreased the fibre/matrix adhesion. Furthermore, these phenomena could be correlated to the reduction in the stiffness of the epoxy matrix [8]. This might be due to the reduction in cross linking density of the polymer composites [8]. El-Feky et al [25] stated that the aforementioned phenomena could be attributed to the fact that nanomaterials got highly agglomerated when being used in relatively high contents due to the van der waals forces which result in the fibres to get entangled and create weak void spots within the matrix, and consequently reducing the strength in such cases.

It was observed that the hybrid nanocomposites produced higher flexural strengths than the monocomposite specimens in both the experimental and FEA approaches. The higher flexural strength of hybrid composites can be attributed to the fact that the hybrid composite (sisal/rice husk hybrid reinforced-epoxy resin nanocomposite) had higher aspect ratios than the monocomposites (sisal nanofibre reinforced epoxy resin composites and rice husk nanoparticle reinforced the epoxy resin composite). Bay et al [24] stated that this increase in the flexural strength in hybrid composites is due to the addition of the nanocellulose content and the restriction of the mobility of the nanocomposites due to the addition of nano particles. This finding was also consistent with the present study and those of Roohani et al [35].

Upon comparing the FEA results with experimental flexural results (see tables 3, 5 and 7), it was established that the margins of errors of 36.36% of the results obtained from the sisal nanofibre reinforced epoxy resin composites were within the acceptable engineering percentage of error of ±5%. For the rice husk nanoparticle reinforced epoxy resin composites, 45.45% of the results were within the acceptable engineering error of ±5%. For the sisal/rice husk hybrid reinforced epoxy resin nanocomposites, 54.55% of the results were within the acceptable margin of error of ±5%. The deviations noted in the results could be attributed to the differences in the accuracy levels of the two methods. The experimental method has uncertainties that usually arise during procedural setups, such as the inherent differences between the presence and orientation of fibres. The FEA method gives approximate results since it considers the effects of non-linearities common in the fabrication of composites.

The results in the present study are in agreement to those of Ismail et al [23] who carried out a research on hybrid films of cellulose nanofibrils, chitosan and nanosilica. The researchers noted that the mechanical properties increased up to some point, beyond which they began to reduce.

The present results are also in agreement to those of Bay et al [24], who determined the mechanical properties of nanocomposite films made of polyvinyl alcohol/nanofiber cellulose and nanosilicon dioxide using ultrasonic method. The authors stated that the increase in the strength properties up to some point is due to the intermolecular forces that keep the strength of the fibrils intact resulting in an enhancement of the strength properties of the nanocomposites.

Comparable results to the present study were also recorded by Jaafar et al [8], who determined the effect of the liquid natural rubber on mechanical properties and microstructure of epoxy/silica/kenaf hybrid composite for potential automotive applications.

El-Feky et al [25] also noted similar findings to the present study when they determined the effect of nano silica addition on enhancing the performance of cement composites reinforced with nanocellulose fibers.
4.2. Fibre volume fraction effects on flexural stiffness

A general trend of flexural stiffness increases with fibre additions for the sisal nanoﬁbre-reinforced-epoxy resin composites, rice husk nanoparticles reinforced-epoxy resin composites and sisal/rice husk hybrid reinforced-epoxy resin nanocomposites were identiﬁed (see ﬁgures 10–12). This increase in the ﬂexural stiffness with ﬁbre additions is due to the high speciﬁc stiffness, homogeneity and self assembly of the nanomaterials and the nanoparticles [23, 34]. Moreover, this increase in stiffness may be due to the inherent high ﬂexural stiffness of nanofibres and nanoparticles which acted as a suitable reinforcer and was able to form a good bond with the polymer [24].

The optimum ﬂexural stiffness of sisal nanoﬁbre reinforced epoxy resin composites were realised in the range of 8.00 $\pm$ 0.03 to 8.20 $\pm$ 0.04 % by volume fraction using the experimental approach and 7.80 $\pm$ 0.16 % by volume fraction using the FEA approach (see ﬁgure 10). For the rice-husk nanoparticle reinforced epoxy resin composites, the optimum ﬂexural stiffness was realized in the range of 9.50 $\pm$ 0.05 to 9.80 $\pm$ 0.05 % by using both the experimental and the FEA approaches (see ﬁgure 11). For the sisal/ rice husk hybrid reinforced-epoxy resin nanocomposites, the optimal value was realized in the range of 7.40 $\pm$ 0.02 to 7.80 $\pm$ 0.03 % using the experimental approach, and 6.5 $\pm$ 0.02 % to 7.0 $\pm$ 0.02 % using the FEA approach (see ﬁgure 12). Beyond these optimum values of reinforcement volume fractions, the ﬂexural stiffness began to fall gradually with ﬁbre additions. This apparent loss in stiffness beyond a certain volume fraction is probably due to the formation of aggregates or agglomerates which caused phase inversion and ﬂexibilization of the matrix to take place [8]. This ﬁnding was supported by Khairulazfar et al [36] in their study that the mechanical properties of ﬂexural stiffness showed a reduction with increasing ﬁbre loading.

It was also evident that the ﬂexural stiffness graphs for the rice husk nanoparticle reinforced epoxy resin composites, in both the FEA and experimental approaches (see ﬁgure 11), exhibited two gradients on the right-hand side. This was a sign of a transverse matrix fracture. The advent of transverse matrix fracture began at around 7.8 $\pm$ 0.03 % in both the FEA and experimental approaches. The corresponding value of ﬂexural stiffness at this point of transverse matrix fracture was in the range of 5.8 GPa to 6 GPa in both the FEA and experimental approaches.

It was also observed that the hybrid composites produced higher ﬂexural stiffness values than the monocomposites (i.e. sisal nanofibre reinforced-epoxy resin composites and rice husk nanoparticle reinforced epoxy resin composites). The higher ﬂexural stiffness of hybrid composites can be attributed to the fact that the hybrid composite (sisal/rice husk hybrid reinforced-epoxy resin nanocomposite) had higher aspect ratios than the monocomposites (sisal nanofibre reinforced epoxy resin composites and rice husk nanoparticle reinforced the epoxy resin composite). Bay et al [24] stated that this increase in the ﬂexural stiffness in hybrid composites is due to the addition of the nanocellulose content and the restriction of the mobility of the nanocomposites due to the addition of nano particles. This finding was also consistent with the present study and those of Roohani et al [35].

Upon comparing the FEA ﬂexural stiffness results with the experimental ﬂexural stiffness results, it was established that the margin of error of 36.36% of the FEA results obtained from sisal nanofibre reinforced epoxy resin composites, was within the acceptable engineering percentage error limit of ±5% (see table 11). For the rice husk nanofibre reinforced epoxy resin composites, 45.45% of the FEA results were within the acceptable engineering error limit of ±5% (see table 13). For the sisal/rice husk hybrid reinforced epoxy resin nanocomposites, 63.64% of the FEA results were within acceptable error margins of error of ±5% (see table 15). The deviations between FEA and the experimental results could have resulted from the differences in the accuracy levels of the two methods. The experimental method usually involves a lot of uncertainties during experimental procedures, such as the inherent differences which exist between the orientation of ﬁbres. On the other hand, the FEA method considers the effects of non-linearities that are common in the fabrication of composites.

The ﬁndings in this study are consistent with those of Jaafar et al [8] who determined the effects of liquid natural rubber on mechanical properties and microstructure of epoxy/silica/kenaf hybrid composite for potential automotive applications.

The results in the present study are also in agreement to those of Ismail et al [23] who carried out a research on hybrid films of cellulose nanofibrils, chitosan and nanosilica. The researchers noted that the mechanical properties increased up to some point, beyond which they began to reduce.

The present results also compare well to those of Bay et al [24], who determined the mechanical properties of nanocomposite ﬁlms made of polyvinyl alcohol/nanofiber cellulose and nanosilicon dioxide using ultrasonic method. The authors stated that the increase in the strength properties up to some point is due to the intermolecular forces that keep the strength of the ﬁbrils intact resulting in an enhancement of the strength properties of the nanocomposites.
Table 18. Comparison of the current study DSC results with other authors.

| Parameter               | Panaitescu et al [38] | Bay et al [24] | Current study |
|-------------------------|------------------------|----------------|--------------|
| Glass transition temperatures | 50°C–57.2°C           | 70°C–100°C      | 50°C–75°C    |
| Melting temperatures    | 300°C–500°C            | 280°C–350°C     | 350°C–400°C  |

4.3. Thermal analyses

Thermal Gravimetric Analysis (TGA) was carried out to monitor changes in the sisal nanofibre reinforced epoxy resin composites, rice husk nanoparticle reinforced epoxy resin composites, and sisal/rice husk hybrid reinforced epoxy resin nanocomposites, as a function of exposure to heating.

From figure 1, it can be observed that the Thermal Gravimetric Analysis curves exhibit 3 sections: an initial flat section, then a section with a constant slope and finally a flat section. At the initial flat section, there was a weight loss of 5%, 10% and 10% for the sisal/rice husk hybrid reinforced epoxy resin nanocomposite, rice husk nanoparticle reinforced epoxy resin composites, and sisal nanofibre reinforced epoxy resin composite samples, respectively. At the constant slope section, there was a weight loss of 80%, 75% and 75% for sisal/rice husk hybrid reinforced epoxy resin nanocomposite, rice husk nanoparticle reinforced epoxy resin composites, and sisal nanofibre reinforced epoxy resin composite samples, respectively. Finally, all the mass degraded and left behind a black carbonaceous residue known as char, as is clearly depicted in the last flat sections.

The initial weight loss of all samples was attributed to the volatilization of water adsorbed in the composites before 100°C [9, 22], and the second stage began at temperatures between 250°C and 300°C. This was caused by the absorbed moisture and the degradation of the lignocellulosic components. All the samples of sisal/rice husk hybrid reinforced epoxy resin nanocomposite, rice husk nanoparticle reinforced epoxy resin composites, and sisal nanofibre reinforced epoxy resin composite samples fully degraded at temperatures of between 400°C and 475°C. The complete degradation in the fibre composites arose when all the cellulose had fully degraded. The full degradation can be evidenced by the sudden levelling of the curve. It is also evident that the hybrid nanocomposites showed good thermal endurance compared to those of the mono composites. This observation is in agreement with the works of Jumahat et al [27].

The degradation temperatures in the current study are in agreement with the works of Ismail et al [23], who carried out TGA tests on nanocomposites made of hybrid films of cellulose nanofibrils, chitosan and nanosilica. The authors reported the first degradation temperature at at temperatures below 100°C, and the second degradation temperature was reported at around 200°C. Ireana et al [37], on the other hand, reported degradation temperatures of between 264°C and 300°C. Furthermore, the results in the present study compare well to those of Bay et al [24] who performed TGA tests on nanocomposite films made of polyvinyl alcohol/nanofibre cellulose and nanosilicon dioxide using ultrasonic method. The authors reported that the initial degradation began at temperatures below 100°C, and the second degradation temperature started at about 250°C, while the third degradation began at about 350°C to 400°C.

In the Dynamic Scanning Calorimetry (DSC) graphs, all the samples exhibited a glass transition temperature of between 50°C and 75°C. Further, all the samples exhibited a melting temperature of between 350°C and 400°C.

The first peak in the DSC graphs is attributed to the pyrolysis of hemicelluloses, while the second peak in the DSC graphs is attributed to the pyrolysis of cellulose and lignin [38]. The behaviour in the Dynamic Scanning Calorimetry samples can be explained by a decrease in the cohesive energy in the samples due to the rupture of the hydrogen bonds as a result of heating [38].

The values found in this study compare closely to those of Panaitescu et al [38], who found glass transition temperatures of between 53°C and 57.2°C and melting temperatures of between 300°C and 500°C for PVA composites. The findings in the present study are also in agreement to those of Bay et al [24] who determined the glass transition temperatures in the range 70°C–100°C and the melting temperatures in the range 280°C–350°C. A comparison of the current result with those of Panaitescu et al [38] and Bay et al [24] are shown in table 18.

4.4. Formulation of optimal mix ratio for different applications

The Minitab software version 2017 was used to perform the optimization procedure in order to formulate the optimal mix ratio of fibres and matrices for different applications. These optimal values obtained using the Minitab software were validated using the experimental values. For applications that require the flexural properties to maximized, the ratio was found to be 4:1. When all the flexural properties were minimized, the ratio was found to be 1:2. For applications that required a target value of flexural strength of 10 MPa and flexural stiffness of 10 GPa, the ratio was found to be 2:3. The study found that there was agreement in the optimal values obtained using the Minitab software and the experimental values. The formulation of the optimal combination
mix ratio was deemed necessary to ensure the predictability of results and repeatability in the different mechanical engineering applications.

Similar ratios to the present findings were recorded by Das et al [39], who used the simplex design method to find the optimal ratio for jute, coir and polypropylene matrix when all the parameters were maximized.

The present findings are also in agreement with those found by Yusof et al [40], who used the Taguchi approach to determine the optimal parameters of kenaf reinforced epoxy resin composites.

**4.5. Value of research**

This research has a significant impact in the development of sustainable nanocellulosic nanocomposites and minimizing environmental impact. Furthermore, the finite element method of composite modelling and the optimization technique formulated has contributed to the increase in database of knowledge on the production of nanocellulosic nanocomposites. The Finite element method and the minitab optimization technique are also relevant from an economic perspective, since they can be used to accurately predict the properties of the nanocomposites at minimal costs.

**5. Conclusions**

Finite element analysis was used to formulate novel models of both mono and hybrid nanocomposites, and also to predict their flexural properties. This study also formulated the optimal combination mix ratio of fibres and matrix of the flexural properties for use in different applications. The predicted properties were verified using the experimental method with the hypothesis being whether the flexural properties were significant. This study showed that the flexural properties were significant through the use of ANOVA. This study also found that the flexural properties of all the composites depicted an increase with fibre additions up to a certain optimum point, beyond which they began to fall gradually with further fibre additions. There was also evidence of transverse matrix fracture in rice husk nanoparticle-reinforced epoxy resin composites' flexural stiffness graphs. Moreover, this study found that the increases in flexural properties of mono composites were about 2 times lower in magnitude than hybrid composites. With regards to the formulation of optimal combination mix ratio of fibres and matrices for different applications, this study found that the optimal combination mix ratio of fibres to the matrix for applications where all the flexural properties were maximized was 4:1. Moreover, the optimal combination mix ratio of fibres to the matrix for applications where the flexural properties were minimized was 1:2. Furthermore, the optimal combination mix ratio of fibres to the matrix for applications with a target of 10 MPa flexural strength properties and 10 GPa for flexural stiffness properties was 2:3.

This research has a significant impact in the development of sustainable nanocellulosic nanocomposites and minimizing environmental impact. Furthermore, the finite element method of composite modelling and the optimization technique formulated has contributed to the increase in database of knowledge on the production and properties of nanocellulosic nanocomposites. The Finite element method and the minitab optimization technique are also relevant from an economic perspective, since they can be used to accurately predict the properties of the nanocomposites at minimal costs.

The effect of span to depth ratio on the modulus of rupture of sisal nanofibre reinforced epoxy resin composites, rice husk nanoparticle reinforced epoxy resin composites and sisal/rice husk hybrid reinforced epoxy resin nanocomposites was not studied in the present work. It should, therefore, be pursued by future researchers with the objective of coming up with optimum dimensions for standard test pieces.

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**Data availability statement**

All data that support the findings of this study are included within the article.
Declarations

Availability of data and material
All data generated or analysed during this study are included in this published article.

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There is none to be declared.

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