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Original article

# Enhancing the properties of date palm fibre reinforced bio-epoxy composites with chitosan – Synthesis, mechanical properties, and dimensional stability



Siti Noorbaini Sarmin<sup>a,b</sup>, Mohammad Jawaid<sup>a,c,\*</sup>, Sheikh Ahmad Zaki<sup>c</sup>, Ali Mohd Radzi<sup>a</sup>, Hassan Fouad<sup>d</sup>, Ramzi Khiari<sup>e</sup>, Sri Rahayu<sup>f</sup>, Mohd Hazim Mohamad Amini<sup>g</sup>

<sup>a</sup> Department of Bio-composite Laboratory, Institute of Tropical Forestry and Forest Products (INTROP), Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia

<sup>b</sup> Department of Wood Technology, Faculty of Applied Science, Universiti Teknologi MARA, 26400 Jengka, Pahang, Malaysia

<sup>c</sup> Malaysia-Japan International Institute of Technology, Universiti Teknologi Malaysia, Jalan Sultan Yahya Petra, 54100 Kuala Lumpur, Kuala Lumpur, Malaysia

<sup>d</sup> Applied Medical Science Department, Community College, King Saud University, P.O. Box 10219, Riyadh 11433, Saudi Arabia

<sup>e</sup> University Grenoble Alpes, CNRS INP, LGP2, F-38000 Grenoble, France

<sup>f</sup> Faculty of Forestry, Universitas Gadjah Mada, Yogyakarta, Indonesia

<sup>g</sup> Faculty of Bioengineering and Technology, Universiti Malaysia Kelantan, Jeli Campus, 17600 Jeli, Kelantan, Malaysia

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## ABSTRACT

The present work deals with the mechanical behavior of dual bonding filler; chitosan (CTS) and date palm (DP) fibre in bio-epoxy composites. The primary objective of this research was to find out if the addition of CTS particles to a DP/bio-epoxy composite could enhance its mechanical properties. Bio-composites are being developed to better understand and manipulate the unique features of these versatile polysaccharides. CTS was included into the bio-epoxy matrix through solution mixing. Bio-epoxy composites with 40% DP fibre loading were developed by dispersing different CTS filler ratios - 5%, 10%, 15%, and 20%. For the purpose of comparison, a control sample was prepared without the inclusion of the CTS filler. The mixture was then manually disseminated for 7–10 min before being uniformly drenched into a steel cast mould with dimensions of 150 mm (width) 150 mm (length) 3 mm (thickness) and taken to the hot press, after which it underwent heating at a temperature of 110 °C for a duration of 10 minutes, while being subjected to a significant pressure of 250 bar. Tensile properties, impact properties, flexural properties, morphological characterization, water absorption, and thickness swelling were all measured on the prepared bio-composites. Among the CTS fractions, DP/CTS20 has the highest tensile strength (24.04 MPa) and tensile modulus (4.93 GPa), flexural strength (45.11 MPa), and impact strength (2.70 J/m<sup>2</sup>). The scanning electron microscope exposes the properties of fibres and polymers, as well as the reason for tensile test deficiency. Remarkably, bio-composites with higher CTS content have elevated moisture content and dimensional instability. Generally, we diagnosis that DP/CTS20 had the best tensile, flexural, and impact properties, as well as the highest water absorption and thickness swelling of any CTS compositions. The discovery improved our knowledge of bio-composite implementation trends, allowing us to assess their potential for more dependable implementations of much more sustainable green products. Furthermore, material enhancements such as bio-epoxy toughening to change the ductility behavior to improve energy absorption or natural fibre pretreatment can improve interbonding between the matrix and filler.

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\* Corresponding author at: Department of Bio-composite Laboratory, Institute of Tropical Forestry and Forest Products (INTROP), Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia.

E-mail address: [jawaid@upm.edu.my](mailto:jawaid@upm.edu.my) (M. Jawaid).

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## 1. Introduction

Epoxy resins are generated after petroleum resources and are one of the most versatile thermosetting resins, serving an important role in a diverse range of industry sectors. With the increasing scarcity of petroleum resources and the resulting environmental impact, much greater emphasis has been placed on epoxy resins

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made from renewable resources in latest years. The synthesis of bio-epoxy using natural resources such as tannins (Basak et al., 2021; Zhang et al., 2022), cardanol (Huang et al., 2022; Liu et al., 2021), lignin (Ortiz et al., 2019; Pappa et al., 2022), and vegetable oils (Fombuena et al., 2019; Li et al., 2020) has begun. Rapid advances in the synthesis of bio-epoxy resins, conjunction over their excellent properties, have positioned them as promising candidates to replace the traditional diglycidyl ether of bisphenol A (DGEBA) inferred from petroleum.

Bio-epoxy resins are of great interest when correlated to synthetic epoxy resins because they offer numerous advantages including low cost, biodegradability, biocompatibility, nontoxicity, superior heat stability, wide availability, flame retardancy, and comprising of naturally derived building blocks that are chemically reactive for the modification of polymers when used in bio-engineering applications, biomedical, and pharmaceutical (Decsov et al., 2019; Kumar et al., 2018; Parameswaranpillai, Rangappa, Siengchin, & Jose, 2020). The vast majority of bio-based monomers are composed of long, flexible side chain or alicyclic structures (Chen et al., 2015; Jia et al., 2019; Zhang et al., 2017), which can significantly improve the thermal stability (Wang et al., 2019) and mechanical properties (Xu et al., 2019) of the resultant bio-based epoxy resins. There are also numerous rigid bio-based monomers with aromatic structures that improve the glass transition temperature significantly (Khalilullah et al., 2022; Liu et al., 2021; Thiagamani et al., 2019) and mechanical properties (Chen et al., 2015) of the subsequent bio-based epoxy resins.

The date palm trees are indigenous to various regions, including the Middle East, Northern Africa, the Canary Islands, Pakistan, India, and the United States (specifically California). (AL-Oqila et al., 2022; Rajeshkumar et al., 2021). Date palm (DP) fibres are among the large amounts of biowaste accumulate without adequate management. These amounts have the prospective to aid in industrial sustainability by creating cheap environmentally friendly materials. When compared to other fibre types, DP fibre performs best in terms of a variety of evaluation criteria, including specific strength to cost ratio (Awad et al., 2021). Studies in the past have looked into the effects of using DP fibres in natural fibre composites. Most of the findings suggested that DP fibre reinforcement have a promising future and can be utilized in a diverse selection of applications (Asim et al., 2021; Jawaid et al., 2021; Supian et al., 2021). Benmansour et al. (2014) investigated the use of date palm fibre as a filler in mortar for use as an insulating medium in buildings and discovered that date palm fibre incorporation appears to be a very promising option for use as thermal insulation materials in buildings. According to AL-Oqila et al. (2022) research's on date palm fibre reinforced polypropylene composites, date palm fibre accumulation improved the thermal stability of polypropylene composites. Amroune et al. (2021) investigated the date palm fibre for composites reinforcement and discovered that chemical treatment of the fibres can improve their tensile strength properties up to threefold.

Chitosan (CTS) is an aminogluco-pyran composed of glucosamine (GlcN) and N-acetylglucosamine (GlcNAc) (Wang et al., 2020). Because these polysaccharides are renewable resources that are being extensively researched, chitosan-based composites have broad relevancy and probable in the fields of biomedical, packaging, and water purification (Arumugam et al., 2020; Gupta et al., 2019; Oladzadabbasabadi et al., 2022). Blending chitosan with cellulose and/or incorporating nanofibre isolated from cellulose, can be beneficial because chitosan is highly compatible with other biopolymers (Liu et al., 2020). Because of the existence of functional groups like hydroxyl, acetamido, and amine, these distinctive polymers have developed as a new class of physiological materials with a good solubility profile, fewer crystallinity, and are responsive to chemical modifications (Oladzadabbasabadi

et al., 2022; Qin et al., 2018). Despite the fact that chitosan has several benefits, such as biodegradability, nontoxicity, and profusion in nature, chitosan-based materials have poor water blocking capabilities due to their hydrophilic character, which primarily affects their mechanical, gas penetrability, and thermal properties (Arumugam et al., 2020; Wang et al., 2020). Hence, blending biopolymers to make composites is one procedure for reducing chitosan's hydrophilic nature. Blending chitosan with other polysaccharides resulted in the increase of obstruction, mechanical, and aesthetic composite characterization (Cazón et al., 2018; Tang et al., 2018).

To summarize, while date palm fibre is a prevalent material for preparatory fibre reinforced composites, few studies have preferred chitosan particles as an additive filler to mend the composite's properties. The primary goal of this study is to improve the mechanical properties of a DP/bio-epoxy composite by using CTS particles as an additive filler. The resultant of CTS particle weight percentages of 0, 5%, 10%, 15%, and 20% on the mechanical properties of bio-composites is investigated. The novel aspect of this work is the investigation of the synergistic interaction between DP fibre and CTS particle in the bio-epoxy based composite. The results of this study will strengthen the appealing interpretation of using natural plant fibres to fabricate composites with CTS filler for various lightweight purposes in a wide range of industrial applications such as biomaterials and engineering.

## 2. Materials and method

### 2.1. Raw material preparation

The present study employed a green bio-epoxy polymer, specifically the SR GreenPoxy 56 resin and SD 280x hardener, manufactured by Sicomin Epoxy Systems in France. Notably, this polymer possesses a molecular structure that is derived from plants, constituting approximately 56% of its composition. Table 1 lists the selected properties of the GreenPoxy 56 resin. The resin to hardener weight ratio was 100:37. Chitosan 38,906 (molecule weight 345,500 g mol<sup>-1</sup> with degree of deacetylation 84.5 %) was purchased from Chemiz (M) Sdn Bhd, Selangor Malaysia.

The date palm (DP) was obtained through the harvesting of date palm trees located in Jeddah, Saudi Arabia. These fibres were being shattered into short fibres and contained 6 to 8 % moisture (during the fabrication stage). The DP fibres were sieved to a size between 0.8 and 1 mm. To remove moisture from the samples, they were placed in an oven at 60 °C for 24 h. The DP fiber's chemical composition (Table 2) was determined at the Malaysian Agricultural Research and Development Institute (MARDI) in Selangor, Malaysia. Table 2 lists the chemical components of DP fibre.

**Table 1**  
Bio-epoxy resin type SR GreenPoxy 56 and SD 280x hardener.

Specification	Bio-epoxy: SR GreenPoxy 56	Hardener: SD 280x
Density @ 20 °C (g/ml)	1.198	0.958
% Bio-based Carbon content	50–58	0
Color	Clear liquid	1 max (Gardener)
Tension-Modulus of elasticity @7 days 23 °C (N/mm <sup>2</sup> )	3200	–
Flexion-Modulus of elasticity @7 days 23 °C (N/mm <sup>2</sup> )	3300	–
Charpy impact strength @7 days 23 °C (kJ/m <sup>2</sup> )	20	–

**Table 2**  
Chemical constituent of date palm leaf sheath (DP) fibre.

Chemical Composition	Date palm (DP)
Cellulose (%)	43.5
Hemicellulose (%)	24.0
Lignin (%)	18.0
Ash, extractives, others (%)	14.5

## 2.2. Fabrication of bio-composites

Table 3 displays the different loading of CTS particles employed in DP/bio-epoxy composites. The process used to produce the composite material is illustrated in Fig. 1. At this stage, CTS was incorporated into the bio-epoxy matrix via solution mixing. In the formulations, four independent weight fractions of CTS particles were utilized as matrix substitutions in DP/ bio-epoxy composite, with loadings of 5, 10, 15, and 20% wt. The required volume of bio-epoxy resin had first been placed in a beaker, accompanied by the appropriate amount of CTS. For 7 to 10 minutes, the mixture was mixed manually. The required quantity of hardener was then added and mixed thoroughly for 3 to 5 minutes prior to degassing for 15 minutes. In addition, during this phase, the mixture was introduced into DP fibres with a fibre loading of 40% and manually agitated at ambient temperature for a duration of 7 to 10 minutes. At the third stage, the mixture was then uniformly flooded into the open steel mould with dimensions 150 mm (width) × 150 mm (length) × 3 mm (thickness) and transferred to the curing treatment process, where it was heated at 110 °C under intense pressure of 250 bar for 10 min. The composites were post-cured at 105 °C for 3 h before being cut into specified replication sizes as per ASTM Standard.

## 2.3. Characterizations of the bio-composites

To determine tensile strength, the specimens composite was cut to the ASTM D3039 standard of 120 mm × 20 mm × 3 mm and the test was then examined using five replicates. The experiments were conducted using a 5 kN Blue Hill INSTRON Universal Testing Machine, and each specimen of the composite material was assessed and documented for subsequent analysis. The tensile test specimen was collected, and a cross-sectional surface of the fractured region was prepared for microscopic analysis of its structure. An EM-30AX scanning electron microscope was used to analyze the shattered morphological characteristics of the composite samples (Model: COXEM, Daejeon, Korea). The specimen's test was gold-coated and placed in a machine for testing.

In order to ascertain the flexural characteristics of a composite material, a three-point bending test was conducted following the guidelines outlined in ASTM D790. Five replication samples with measurements of 120 mm × 20 mm × 3 mm were evaluated employing a three-point jig at a crosshead speed of 2 mm/min by a Universal Testing Machine of 20 kN.

The sample underwent an ASTM D256 impact test utilising Izod impact testing equipment (Geotech GT-7045-MD, Gotech, Tai-

**Table 3**  
Formulations of filler (CTS and DP fibre) in the bio-epoxy composites.

Composites code	Bio-epoxy (g)	Hardener (g)	CTS (g)	DP fibre (g)
DP	58.39	21.61	0	32
DP CTS 5	55.47	20.53	4	32
DP CTS 10	52.55	19.45	8	32
DP CTS 15	49.4	18.36	12	32
DP CTS 20	46.72	17.28	16	32

chung City, Taiwan). Samples were cut to 80 mm × 10 mm × 3 mm impact test dimensions and notched in the center. The samples were horizontally placed on the machine, with a 60 mm gap between the support lines. The gauge was initially read before a dangling handle, which rotates and breaks the sample, was released. The reading was recorded after the sample had been broken. Each sample went through five test pieces. Each sample was subjected to five test pieces, and the average was then calculated.

The ASTM-D 570-98 standard for water absorption of plastic specimens was accustomed to regulating the frequency of water absorption and swelling test thickness. The sample was trimmed evenly to 20 mm × 20 mm × 3 mm for all duplicates. All specimens were dipped in distilled water for 24 h at room temperature. For 12 days, the weight and thickness of the specimens were determined.

## 3. Results and discussions

### 3.1. Tensile properties

The tensile properties of the composite are investigated in order to determine the effect of different weight percentages of CTS in the DP/CTS bio-epoxy composites. Fig. 2 depicts the tensile strength and modulus of the DP/CTS composites while Fig. 3 shows the tensile stress–strain curve.

Fig. 2 clearly shows that DP/CTS 20 composites have greater tensile strength (24.04 MPa) and modulus (4.93 GPa) than other composites due to the existence of extra CTS percentage in the composite maintaining great loads. This enhancement could be attributed to further effective stress transfer (Fig. 3) among adjoining chitosan chains as a result of intense electrostatic bonding between the –NH<sub>2</sub> and –NH<sub>3</sub> groups (Abdurrahim, 2019). By efficiently transferring stresses from the bio-epoxy matrix, the CTS particle performs well as a reinforcement, improving the mechanical properties of the composites significantly over those of the DP bio-epoxy composite. The improved mechanical properties of the DP/CTS bio-epoxy composite can be attributed primarily to the homogeneous dispersion (Fig. 4) of great rigid CTS particles with an elevated aspect ratio in the bio-epoxy matrix, ensuing in ultra-high interfacial adhesion and ionic bonds between CTS, DP fibres, and bio-epoxy resin.

DP/CTS5, DP/CTS10, and DP/CTS15 composites, conversely, have lesser tensile strength than DP/CTS20 composites due to irregular dispersion and agglomeration of chitosan particles in DP bio-epoxy affiliation. This interface transforms the DP/CTS5, DP/CTS10, and DP/CTS15 composites into amorphous structures, resulting in inopportune default even at little strain rates. In the absence of CTS, DP fibres exhibit significantly higher tensile strength (22.53 MPa) than DP/CTS5 (20.46 MPa) and DP/CTS10 (20.58 MPa), indicating that the bio-composite containing DP fibres has higher rigidity than DP/CTS5 and DP/CTS10, and the latter has higher flexibility than the former.

It is clear that the elastic modulus of bio-composite containing CTS increases as the percentage of CTS increases. Tensile strength exhibits similar behavior. The utmost elastic modulus is observed at 20 wt% CTS content, where it is enhanced by 25%. The expansion in elastic modulus could be attributed to CTS's reinforcing effect, its improved aspect ratio and surface area, excellent dispersion of CTS particles all over the DP bio-epoxy composite, and strong interaction between bio-epoxy resin and CTS particles via hydrogen bond formation (Abdurrahim, 2019; Arumugam et al., 2020). The same trend was observed for the mechanical properties as a capacity of chitosan filling, as previously informed in numerous polymer-based composites such as sisal fibre (Arumugam et al., 2020), clay (Abdurrahim, 2019), and cellulose (Cazón et al., 2018).

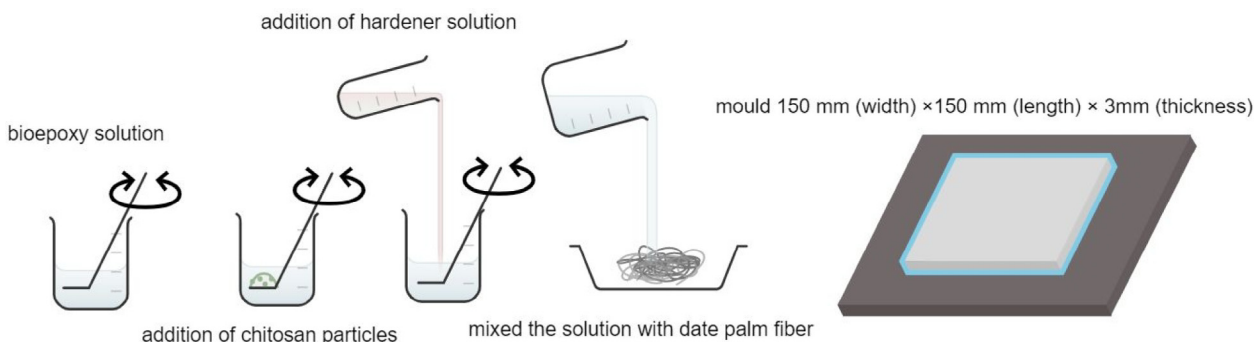


Fig. 1. Schematic diagram of fabrication of DP/CTS bio-epoxy composites.

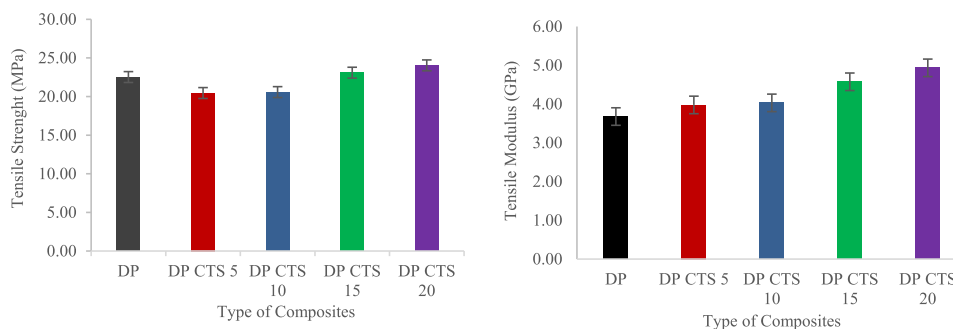


Fig. 2. Tensile strength and tensile modulus of DP/CTS bio-epoxy composites.

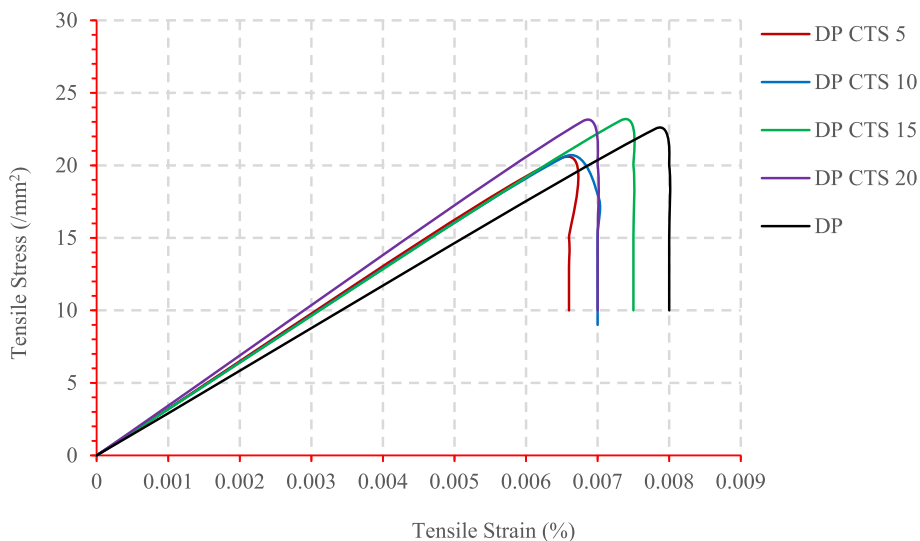


Fig. 3. Tensile stress–strain curve of of DP/CTS bio-epoxy composites.

Fig. 4 (a-e) depicts the composite structure’s uniform dissemination of DP fibre and CTS particles. The failure occurs in the bend direction due to the longitudinal tensile load, indicating the existence of a decent interfacial connection among the matrix and the fibre. The fractured surfaces of the bio-composites become rougher as the CTS content increases. Fig. 3 (d) and (e) show that no cracks are observed in bio-composites containing 15 and 20 wt% CTS, respectively, implying that DP/CTS15 and DP/CTS20 composites have superior mechanical properties when equated to other combinations. Figs. 3 (b) and 2 (c) display matrix damage and voids due to their low longitudinal load carrying capacity. The stress is spread unevenly in the matrix due to the random disse-

mination of CTS and fibres, resulting in the advancement of internal glitch in the bio-composite structures.

Because the overall surface is rough, no obvious CTS aggregation is observed, even for DP/CTS15 and DP/CTS20. The SEM analysis, however, does not allow us to estimate the extent of CTS dispersiveness in the bio-composites. However, we assume that the dispersed phase of CTS coalesces, causing the particles to become greater and non-uniform, resulting in an unsteady morphology. Voids manage to form at the boundary in this case, resulting in poor mechanical properties for bio-composites. The ruptured surfaces of the bio-composites are more heterogeneous, and micro-phase separation is possible.



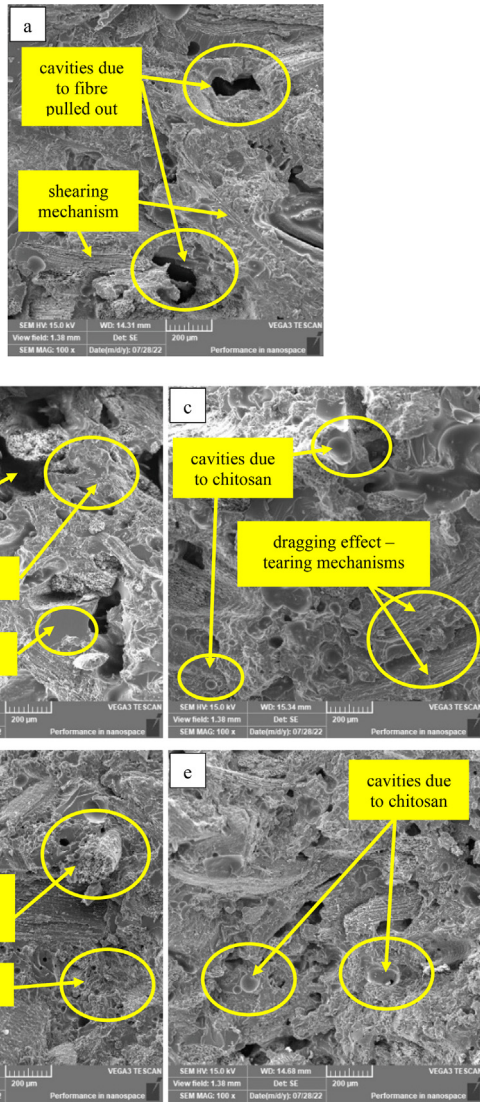


Fig. 4. SEM micrograph of tensile fractured surface of DP/CTS bio-epoxy composite: (a) DP, (b) DP/CTS5, (c) DP/CTS10, (d) DP/CTS15, (e) DP/CTS20.

The DP fibres, CTS particles, fibre breakage, fibre matrix debonding, and fiber/matrix interface are clearly visible in the micrographs of the bio-composites (Figure a - e). The DP fibres are evenly distributed throughout the matrix. SEM micrographs show a strong fibre matrix interface with a few fibres pulling

out, which explains the strong fibre matrix interaction. Large domains of CTS and DP fibres are distributed throughout the matrix of DP/CTS bio-composites (Figure b-e). The SEM images also show multiple DP fibre pull-outs. The fibre pull-out suggests the debonding of the DP fibre from the bio-epoxy matrix at high loads.

### 3.2. Flexural properties

Fig. 5 depicts the effect of varying CTS reinforcement on the flexural and modulus strength of bio-composites. The DP/CTS composite average ultimate flexural strength and modulus are 45.11 MPa and 4.20 GPa at 20 wt% CTS filler loading.

The accumulation of CTS particles improves the bio-flexural composite's stability. Fig. 4 demonstrates that due to the random dispersion of CTS, the DP/CTS15 and DP/CTS20 bio-composites have higher flexural strengths of 44.11 and 45.11 MPa, respectively, than the DP/CTS5 and DP/CTS10 bio-composites (38.33 and 38.82 MPa). Flexural properties improve dramatically as a result of the eventual distribution of chitosan in DP/CTS15 composites. According to study, increasing the amount of fibre in composites could boost their flexural strength up to certain critical loading points (Gheith et al., 2019). Flexural modulus is greatest at 20 wt% CTS of 4.20 GPa. The flexural modulus increases linearly from 5 to 20 wt% reinforcement with CTS particles, which could be attributed to the filler, which provides good reinforcement with the bio-epoxy with well dispersed particulate. This demonstrates that chitosan particles are effective reinforcement materials under flexural loading.

The decrease in flexural properties (from DP - 43.57 MPa to DP/CTS5 - 38.33 MPa and DP/CTS10 - 38.82 MPa) can be attributed to inability of the current polymer matrix to moisten or completely encase every reinforced DP fibre and CTS particle, resulting in lacking interfacial attachment between DP, CTS, and bio-epoxy matrix to impart the applied stress under observaance. This assertion is consistent with other observers' findings that fibres agglomerate at higher loadings in polyurethane matrix due to scarce matrix enclosed of the particles, along with the fact that the reinforcement's hydrophilicity and the polyurethane matrix's hydrophobicity make them incompatible (Oushabi et al., 2017).

### 3.3. Impact strength

Impact strength is the ability of composite materials to endure higher energy impacts without breaking, and it is greatly influenced by the characteristics of each fibre and the interfacial bonding between any of the fibres and the matrix. (AL-Oqla et al., 2022; Awad et al., 2021). Fig. 6 makes it evident that the lack of stiffness in the composite causes the higher impact characteristics of the DP bio-composite (2.39 J/m<sup>2</sup>) to noticeably decrease with the addition of CTS filler. The impact strength of 40 wt% DPF bio-composite is

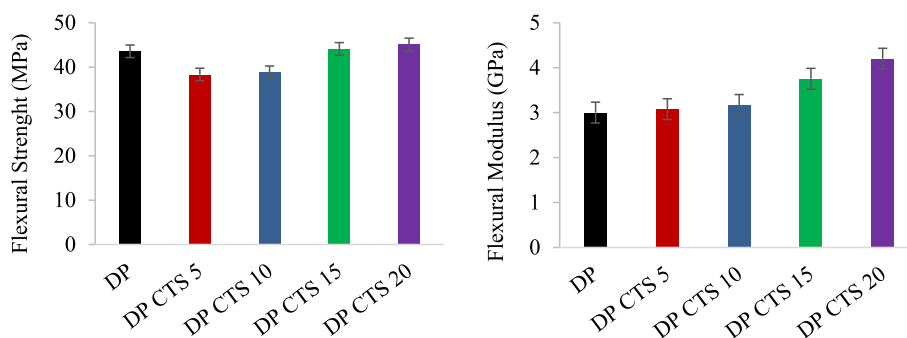


Fig. 5. Flexural properties of DP/CTS bio-epoxy composite.

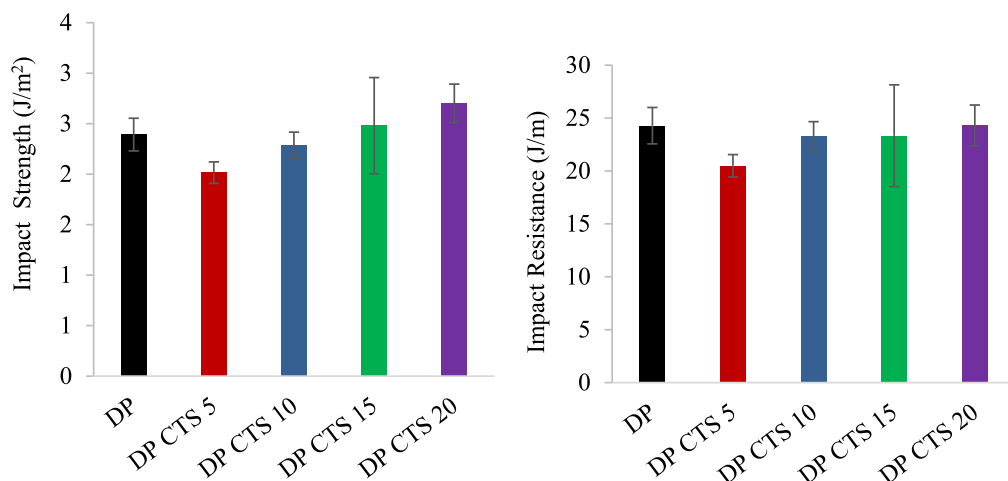


Fig. 6. Impact strength of DP/CTS bio-epoxy composites.

significantly improved compared to 15 and 20 wt% CTS filler loadings, which may be attributed mostly to DPF's improved adherence to the bio-epoxy matrix to withstand the elevated impact pressure applied. Additionally, the improvement in stress capability that results in the improvement in impact strength will reduce the contribution of fibre-related mechanisms such fibre pulls out.

Fig. 6 shows that CTS particles escalation the impact strength of the bio-composites (DP/CTS5 and DP/CTS10) from 2.02 to 2.29 J/m<sup>2</sup>. This can be due to the multiple filler arrangement around the matrix being able to absorb more strain energy. Expected to the uniform dispersion of CTS in the composites, the maximum impact strength of 2.70 J/m<sup>2</sup> is obtained at 20 wt% CTS (DP/CTS20). This demonstrates that the CTS particulate reinforcement is effective in dissipating strain energy throughout the composites. This claim and its supporting evidence are entirely consistent with published literature, which shows that strong interfacial adhesions cause impact strength and toughness to enhance natural fibre composites as fibre loading increases (Asim et al., 2021; Supian et al., 2021; Thiagamani et al., 2019). Aside from an expand in impact strength due to the increment of CTS filler to the DP bio-epoxy composite, a significant diminish in impact strength is observed during the izod impact test (for DP - 2.39 J/m<sup>2</sup> to DP/CTS5 - 2.02 J/m<sup>2</sup> and DP/CTS10 - 2.29 J/m<sup>2</sup>). Lower impact strength is more likely to be affiliated through the total of inner abrasion caused by the impact load.

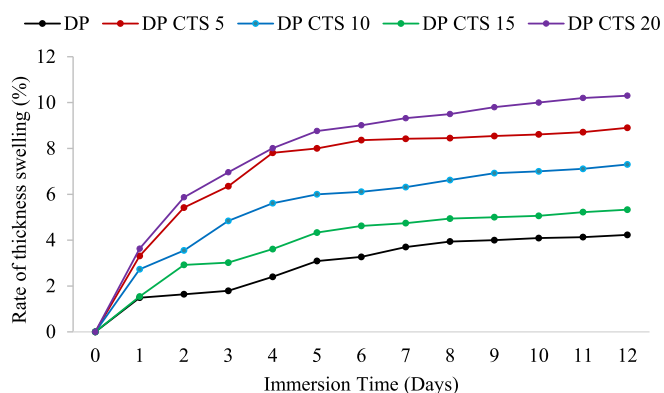
### 3.4. Dimensional stability

Fig. 7 depicts the rate of water accumulates as a function of dipping time for DP/CTS bio-epoxy composites with varying CTS loading. In each case, a foremost linear correlation between water accumulates and immersion time was monitored, pursued by saturation. This demonstrates that the water absorption attitude of bio-composites adhere to Fick's law (according to Fick's law, the rate of diffusion of an element across a unit area (such as a surface or membrane) is correlated to the concentration gradient) (Devan and Bachchan, 2021).

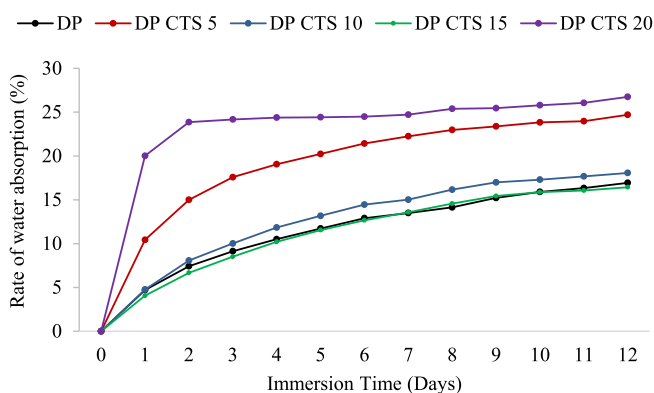
Fig. 7 shows that the water uptake increases as the CTS content increases. The water absorption of DP bio-epoxy composite is 1.64%, whereas 5, 10, 15, and 20 wt% CTS incorporation increases water uptake by 5.42%, 3.55%, 2.92%, and 5.87% after two days immersion, respectively. The increased percentage of water uptake for DP/CTS bio-epoxy composite is due to the chitosan's inherent hydrophilic nature (Cervera et al., 2004). This is also due to the

substantial interaction of water molecules with chitosan's hydroxyl and amino groups.

As demonstrated in Fig. 7 (a) and (b), the DP/CTS10 and DP/CTS15 possess lower water absorption and thickness swelling rate compared to DP/CTS5 and DP/CTS20 bio-composites. According to the



(a)



(b)

Fig. 7. Water sorption curves of DP/CTS bio-epoxy composite at various chitosan weight percentages: (a) rate of thickness swelling, (b) rate of water absorption.

findings, chitosan has a significant impact on water diffusion. The inner chitosan particles entangled in the bio-epoxy matrix are not attainable to form hydrogen bonds with water molecules due to poor filler dissemination in the composites. Chitosan particles, on the other hand, are tightly packed in bio-epoxy composites, resulting in farther steady dispersion in the matrix (Fig. 4). The high surface area of these uniformly distributed chitosan particles favors water absorption. Water grasp in the composite are primarily caused by chitosan particles that are exhibit at or beside the surface and intrigue moisture faster than those that are acquaint in the inner.

#### 4. Conclusions

As a result of this work, we were able to draw the following conclusions about the effectiveness of using CTS particles as an additive filler to enhance the mechanical properties of a DP/bio-epoxy composite:

- Following a thorough analysis of the bio-composites' characteristics, the mechanical properties of bio-epoxy composite get intensified by the increment of CTS, however 20 wt% CTS loading showed enhancement equated to the rest of the composites, attributed to improved dispersion, wettability, and interfacial bonding between DPF, CTS and bio-epoxy matrix.
- In tensile testing, the ultimate tensile strength increases from 22.53 to 24.04 MPa as the volume fraction of CTS increases from 0% to 20% with 5% increments.
- In the flexural test, as the volume fraction of CTS increases from 0% to 20% with a 5% increment, the ultimate flexural strength increases from 43.57 to 45.11 MPa. The same thing happened with the impact strength test, which went from 2.39 to 2.37 J/m<sup>2</sup>.
- Nonetheless, in line with CTS weight percentage, water absorption rose. These results demonstrate the hydrophilic nature of CTS.
- The comparable and satisfactory mechanical properties of DPF/CTS20 bio-epoxy composites in comparison to rattan/polypropylene, jute/hemp/poly(lactic acid), oil palm/pineapple leaf/phenolic, and hemp/epoxy composites govern its potential usages, with the primary benefits of renewability, sustainability, and economic issues, in addition to being a lightweight structural material.

Despite the fact that the composite's properties improved as the CTS loading increased, chitosan is prone to agglomeration and will reduce the composite's strength. Mechanical interlocking and adhesion of composite fragments will also be disrupted (depending on the disintegrated reinforcements and their morphology). The success of this work will most likely reduce massive date palm waste deposition and accommodate an effort to reduce the current utilization of synthetic fibres in polymer composite manufacturing for advanced engineering operations such as automotive, modern manufacturing, and outdoor applications. It is suggested that future research investigate multiscale modelling techniques to understand the micromechanical behavior of the composite and predict its micromechanical properties with the optimal CTS content.

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#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### References

- Abdurrahim, I., 2019. Water sorption, antimicrobial activity, and thermal and mechanical properties of chitosan/clay/glycerol nanocomposite films. *Heliyon* 5 (8).
- AL-Oqla, F.M., Hayajneh, M.T., Al-Shrida, M.A.M., 2022. Mechanical performance, thermal stability and morphological analysis of date palm fiber reinforced polypropylene composites toward functional bio-products. *Cellul.* 29 (6), 16.
- Amroune, S., Bezazi, A., Dufresne, A., Scarpa, F., Imad, A., 2021. Investigation of the date palm fiber for green composites reinforcement: thermo-physical and mechanical properties of the fiber. *J. Nat. Fibers* 18 (5), 17.
- Arumugam, S., Kandasamy, J., Md Shah, A.U., Hameed Sultan, M.T., Safri, S.N.A., Abdul Majid, M.S., Mustapha, F., 2020. Investigations on the mechanical properties of glass fiber/sisal fiber/chitosan reinforced hybrid polymer sandwich composite scaffolds for bone fracture fixation applications. *Polymers* 12 (7).
- Asim, M., Jawaid, M., Fouad, H., Althman, O.Y., 2021. Effect of surface modified date palm fibre loading on mechanical, thermal properties of date palm reinforced phenolic composites. *Compos. Struct.* 267.
- Awad, S., Hamouda, T., Midani, M., Zhou, Y., Katsou, E., Fan, M., 2021. Date palm fibre geometry and its effect on the physical and mechanical properties of recycled polyvinyl chloride composite. *Ind. Crop. Prod.* 174.
- Basak, S., Raja, A.S.M., Saxena, S., Patil, P.G., 2021. Tannin based polyphenolic biomacromolecules: creating a new era towards sustainable flame retardancy of polymers. *Polym. Degrad. Stab.* 189.
- Benmansour, N., Agoudjil, B., Gherabli, A., Kareche, A., Boudenne, A., 2014. Thermal and mechanical performance of natural mortar reinforced with date palm fibers for use as insulating materials in building. *Energ. Build.* 81, 6.
- Cazón, P., Vázquez, M., Velázquez, G., 2018. Novel composite films based on cellulose reinforced with chitosan and polyvinyl alcohol: Effect on mechanical properties and water vapour permeability. *Polym. Test.* 69, 8.
- Cervera, M.F., Karjalainen, M., Airaksinen, S., Rantanen, J., Krogars, K., Heinamaki, J., Colarte, A.I., Yliruusi, J., 2004. Physical stability and moisture sorption of aqueous chitosan-amylose starch films plasticized with polyols. *Eur. J. Pharm. Biopharm.* 58, 7.
- Chen, J., Nie, X., Liu, Z., Mi, Z., Zhou, Y., 2015. Synthesis and application of polyepoxide cardanol glycidyl ether as biobased polyepoxide reactive diluent for epoxy resin. *ACS Sustain. Chem. Eng.* 3, 7.
- Decsov, K., Bocz, K., Szolnoki, B., Bourbigot, S., Fontaine, G., Vadas, D., Marosi, G., 2019. Development of bioepoxy resin microencapsulated ammonium-polyphosphate for flame retardancy of polylactic acid. *Molecules* 24 (22). <https://doi.org/10.3390/molecules24224123>.
- Devan, K., Bachchan, A., 2021. Homogeneous reactor and neutron diffusion equation. In: *Physics of Nuclear Reactors*. Academic Press, p. 69.
- Fombuena, V., Petrucci, R., Dominici, F., Jordá-Vilaplana, A., Montanes, N., Torre, L., 2019. Maleinized linseed oil as epoxy resin hardener for composites with high bio content obtained from linen byproducts. *Polymers* 11 (2).
- Gheith, M.H., Aziz, M.A., Ghorri, W., Saba, N., Asim, M., Jawaid, M., Althman, O.Y., 2019. Flexural, thermal and dynamic mechanical properties of date palm fibres reinforced epoxy composites. *J. Mater. Res. Technol.* 8 (1), 7.
- Gupta, P., Agrawal, A., Murali, K., Varshney, R., Beniwal, S., Manhas, S., Lahiri, D., 2019. Differential neural cell adhesion and neurite outgrowth on carbon nanotube and graphene reinforced polymeric scaffolds. *Mater. Sci. Eng. C* 97, 12.
- Huang, J.L., Ding, H.L., Wang, X., Song, L., Hu, Y., 2022. Cardanol-derived anhydride cross-linked epoxy thermosets with intrinsic anti-flammability, toughness and shape memory effect. *Chem. Eng. J.* 450.
- Jawaid, M., Awad, S., Fouad, H., Asim, M., Saba, N., Dhakal, H.N., 2021. Improvements in the thermal behaviour of date palm/bamboo fibres reinforced epoxy hybrid composites. *Compos. Struct.* 277.
- Jia, P., Zheng, M., Ma, Y., Feng, G., Xia, H., Hu, L., Zhou, Y., 2019. Clean synthesis of epoxy plasticizer with quaternary ammonium phosphotungstate as catalyst from a byproduct of cashew nut processing. *J. Clean. Prod.* 206, 11.
- Khalilullah, A., Sayeed, K., Uddin, I., 2022. Bioinspired GO/Au nanocomposite synthesis: Characteristics and use as a high-performance dielectric material in nanoelectronics. *S. Afr. J. Bot.* 151, 8.
- Kumar, S., Krishnan, S., Mohanty, S., Nayak, S.K., 2018. Synthesis and characterization of petroleum and biobased epoxy resins: a review. *Polym. Int.* 67 (7), 24.
- Li, R., Bahadori, A., Xin, J., Zhang, K., Muhunthan, B., Zhang, J., 2020. Characteristics of bioepoxy based on waste cooking oil and lignin and its effects on asphalt binder. *Constr. Build. Mater.* 251.
- Liu, G., Tsen, W.C., Jang, S.C., Hu, F., Zhong, F., Zhang, B., Gong, C., 2020. Composite membranes from quaternized chitosan reinforced with surface-functionalized

- PVDF electrospun nanofibers for alkaline direct methanol fuel cells. *J. Membr. Sci.* 611 (118242).
- Liu, G., Jin, C., Huo, S., Kong, Z., Chu, F., 2021. Preparation and properties of novel bio-based epoxy resin thermosets from lignin oligomers and cardanol. *Int. J. Biol. Macromol.* 193, 8.
- Oladzadabababadi, N., Nafchi, A.M., Ariffin, F., Wijekoon, M.J.O., Al-Hassan, A.A., Dheyab, M.A., Ghasemlou, M., 2022. Recent advances in extraction, modification, and application of chitosan in packaging industry. *Carbohydr. Polym.*, 277.
- Ortiz, P., Wiekamp, M., Vendamme, R., Eevers, W., 2019. Bio-based epoxy resins from biorefinery by-products. *BioResources* 14, 9.
- Oushabi, A., Sair, S., Abboud, Y., Tanane, O., El Bouari, A., 2017. An experimental investigation on morphological, mechanical and thermal properties of date palm particles reinforced polyurethane composites as new ecological insulating materials in building. *Case Stud. Constr. Mater.* 7, 9.
- Pappa, C., Feghali, E., Vanbroekhoven, K., Triantafyllidis, K.S., 2022. Recent advances in epoxy resins and composites derived from lignin and related bio-oils. *Current Opinion in Green and Sustainable Chemistry*.
- Parameswaranpillai, J., Rangappa, S.M., Siengchin, S., Jose, S., 2020. *Bio-based Epoxy Polymers, Blends and Composites : Synthesis, Properties, Characterization and Applications*. Wiley-VCH, Weinheim.
- Qin, H., Wang, J., Wang, T., Gao, X., Wan, Q., Pei, X., 2018. Preparation and characterization of chitosan/ $\beta$ -glycerophosphate thermal-sensitive hydrogel reinforced by graphene oxide. *Front. Chem.* 6 (565).
- Rajeshkumar, G., Hariharan, V., Devnani, G.L., Prakash Maran, J., Sanjay, M.R., Siengchin, S., Al-Dhabi, N.A., Ponnuragan, K., 2021. Cellulose fiber from date palm petioles as potential reinforcement for polymer composites: Physicochemical and structural properties. *Polym. Compos.* 42 (8), 10.
- Supian, A.B.M., Jawaid, M., Rashid, B., Fouad, H., Saba, N., Dhakal, H.N., Khiari, R., 2021. Mechanical and physical performance of date palm/bamboo fibre reinforced epoxy hybrid composites. *J. Mater. Res. Technol.* 15, 11.
- Tang, Y., Zhang, X., Zhao, R., Guo, D., Zhang, J., 2018. Preparation and properties of chitosan/guar gum/nanocrystalline cellulose nanocomposite films. *Carbohydr. Polym.* 197, 8.
- Thiagamani, S.M.K., Krishnasamy, S., Muthukumar, C., Tengsuthiwat, J., Nagarajan, R., Siengchin, S., Ismail, S.O., 2019. Investigation into mechanical, absorption and swelling behaviour of hemp/sisal fibre reinforced bioepoxy hybrid composites: Effects of stacking sequences. *Int. J. Biol. Macromol.* 140, 637–646. <https://doi.org/10.1016/j.ijbiomac.2019.08.166>.
- Wang, S., Ma, S., Li, Q., Xu, X., Wang, B., Yuan, W., Zhu, J., 2019. Facile in situ preparation of high-performance epoxy vitrimer from renewable resources and its application in nondestructive recyclable carbon fiber composite. *Green Chem.* 21 (6), 13.
- Wang, W., Meng, Q., Li, Q., Liu, J., Zhou, M., Jin, Z., Zhao, K., 2020. Chitosan derivatives and their application in biomedicine. *Int. J. Mol. Sci.* 21 (2).
- Xu, X., Ma, S., Wu, J., Yang, J., Wang, B., Wang, S., et al., 2019. High-performance, command-degradable, antibacterial Schiff base epoxy thermosets: synthesis and properties. *J. Mater. Chem. A* 7 (25), 11. 7(25), 15420–15431.
- Zhang, C., Garrison, T.F., Madbouly, S.A., Kessler, M.R., 2017. Recent advances in vegetable oil-based polymers and their composites. *Prog. Polym. Sci.* 71, 52.
- Zhang, T., Yu, C., Yu, M., Huang, Y., Tan, J., Zhang, M., Zhu, X., 2022. Multifunctional tannin extract-based epoxy derived from waste bark as a highly toughening and strengthening agent for epoxy resin. *Ind. Crop. Prod.* 176.