



Characterization of functional bio-plasticizer from *Millettia pinnata* leaf biomass as a green alternative to petroleum-based plasticizers

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Abstract

With the increasing demand for sustainable and non-toxic alternatives, bio-based plasticizers derived from renewable sources are being developed as environmentally friendly replacements for conventional synthetic plasticizers such as phthalate esters, adipates, trimellitates, benzoates, sebacates, etc. This study investigated the extraction of solid plasticizers from the leaves of the abundantly available *Millettia pinnata* plant (MPL). It was chemically treated through processes including phytoremediation, slow pyrolysis, alkylation, and filtration to extract the plasticizers. Scanning electron microscopy revealed a porous, smooth surface, while atomic force microscopy further supported the morphological suitability of these materials for biofilm and composite preparation. Fourier transform infrared spectroscopy identified functional groups such as alcohol, amine, amide, hydrocarbon, alkene, and aromatic compounds, while UV analysis confirmed the presence of alcoholic, amino, and carboxyl constituents. The primary phytoconstituents detected in the MPL were molecularly docked to determine binding affinity. Thermal analysis demonstrated that the extracted plasticizer can withstand temperatures up to 267 °C. Furthermore, X-ray Diffraction analysis yielded a high crystallinity index (47.5%) and a low crystalline size (11.3 nm), desirable characteristics in plasticizers. These findings suggest that plasticizers extracted from MPL leaves could serve as a viable, eco-friendly alternative to conventional synthetic plasticizers, offering a sustainable replacement with considerable functional benefits.

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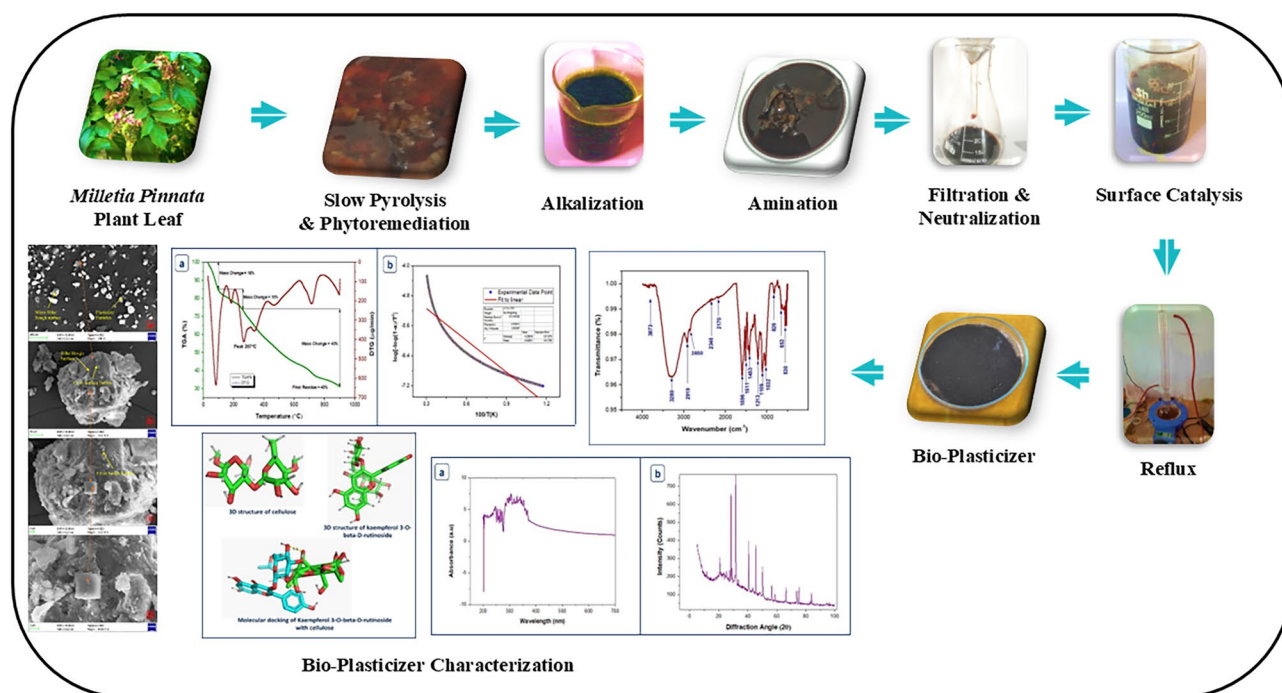
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Graphical abstract



Keywords *Millettia pinnata* leaf · Biofiller · Bioplasticizer

1 Introduction

Plasticizers are essential additives widely used in polymer industries to enhance the flexibility, workability, and durability of rigid materials. They work by embedding within the polymer matrix, reducing intermolecular forces between polymer chains and allowing them to move more freely, resulting in improved elasticity and toughness [1]. The demand for plasticizers is vast, spanning applications in packaging, building and construction, automotive, and pharmaceutical sectors [2]. Traditionally, plasticizers have been derived from synthetic, petroleum-based sources like phthalates, adipates, trimellitates, and sebacates. However, synthetic plasticizers, particularly phthalates, have raised significant environmental and health concerns due to their toxicity, persistence in ecosystems, and potential as endocrine disruptors [3]. Traditional plasticizers have few polar capabilities, resulting in increased chain interactions between polymers. This reduces polymer chain free volume and enhances the crystallinity [4]. These drawbacks have accelerated the search for sustainable and non-toxic alternative.

Researchers were currently involved in developing bio-based plasticizers from renewable resources such as plant, curcumin, citric acid, sucrose, cardanol, succinate

esters, etc. Plants are rich in bioactive compounds like glycols, glycerol, and polyphenols, which can be chemically modified to produce effective plasticizers [5]. By utilizing natural and renewable sources, bio-based plasticizers reduce dependency on fossil fuels and mitigate ecological risks. They also provide an environmentally friendly disposal path, often exhibiting enhanced biodegradability compared to their synthetic counterparts. This shift to bio-based plasticizers addresses both sustainability goals and the increased regulatory restrictions on synthetic plasticizers. Bio-based plasticizers have more polar groups such as aromatic ring, diglycidyl ester groups and long alkyl chains interact with the polar parts of polymer molecules, reducing polymer-polymer interactions and increasing polymer molecule free space. Additionally, bio-based plasticizer alkyl chains migrate, creating polymer space.

Bio-based plasticizers increase the amount of the amorphous portion of the plasticized polymer matrix and the indiscriminate mobility of polymer chains in non-connected polymer regions [4]. Bio-based plasticizers can replace synthetic ones in products, such as cables, flooring, and medical tubing, polyurethane for flexible foams and elastomers, and cellulose derivatives in coatings and films. Chen et al. extracted a new natural plasticizer (epoxidized cardanol phenyl phosphate ester plasticizer), from cardanol and added

in the PVC films [6]. Modified PVC films improved the flexibility, hardness and toughness of the films compared to the PVC film added with the commercial plasticizer dioctyl terephthalate. The TGA results indicate that adding epoxidized cardanol phenyl phosphate ester plasticizer to PVC films increases their thermal stability. Volatility, extraction, and exudation resistance testing indicate that epoxidized cardanol phenyl phosphate ester plasticizer has a better stability. In another study, Chen et al carried out an investigation to determine the plasticising properties of PVC films made with a new soybean oil-based plasticizer (epoxidized glycidyl ester of soybean oil fatty acids) [7]. TGA indicated soybean oil-based plasticizer had superior thermal stability than commercial dioctyl phthalate, with T10 and T50 weight loss increasing from 242.6 and 291.2 °C to 266.5 and 311.4 °C. The tensile and hardness test showed that soybean oil-based plasticizer enhances the PVC film flexibility. Surface characterisation of films also indicates that adding soybean oil-based plasticizer had improved PVC matrix interaction. The castor oil fatty acid (COFA) is plasticized with ethyl cellulose films and assessed in making high-quality films [8]. Results show improved mechanical characteristics and thermal stability at concentrations of 15–25% in ethyl cellulose films. This outstanding performance was attributed to the chemical structure and intermolecular interactions between plasticizers and ethyl cellulose chains. A bioplasticizer (camphorquinone diketal glycerol) extracted from the camphor was investigated to replace phthalate-based plasticizers [9]. Due to their solubility qualities, these plasticizers were found to be very compatible with PVC. Due to its strong and bulky structure, PVC with 20% camphorquinone diketal glycerol was thermally stable up to 251.9 °C and had high Glass transition temperatures, strength, and flexibility. A biofilm was fabricated using polylactic acid reinforced with the solid plasticizers extracted from the leaves of *Pandanus tectorius* [10]. The five-percentage filler loading enhanced the elongation break percent by 70.46%. Similar investigation was done on the plasticizers extracted from the *Petalium murex* plant in PLA film. The results show increments in elongation at break by 25.6% compared to pure PLA at 1% plasticizers reinforcement. In another study plasticizers extracted from *Agave sisalana* were used in PLA biofilm [11]. Increasing the plasticizers content resulted in decreased tensile and Young's modulus. But elongation at break was found to rise up rapidly due to the plasticizing effects.

There are large liquid plasticizers available commercially compared to solid plasticizers. Still there is a gap in finding suitable solid bio-plasticizers. *Leguminous Millettia pinnata* trees are indigenous to Southeast Asia and India. The plant's usage in traditional medicine dates back a long time, and its fibers have also been used to manufacture textiles and other items [12, 13]. The utilization of *Millettia pinnata* leaf (MPL)

fibers as a reinforcing material for bio-composites has drawn more attention in recent years [14]. This plant has spread around the world in humid and subtropical conditions, from sea level to 1200 meters. It is referred to locally by several names, including “Pongamia,” “Karanja,” “Honge,” and “Pungai.” [15]. The *Millettia pinnata* plant, which produces seeds whose oil can be used as a biodiesel fuel, has been the subject of research. The ability to withstand high temperatures, adaption to challenging terrain that reduces competition for fertile land with agriculture, and lack of known pests or diseases are only a few of the advantageous traits of *pogamia*. It is frequently used in landscaping as a windbreak or for shade because of its broad canopy and beautiful, fragrant blooms. The various plant parts have been suggested for the treatment of a variety of ailments in Indian Ayurvedic literature. Several inflammatory diseases such as leucoderma, leprosy, lumbago, muscular and articular rheumatism, and other infectious disorders have all been successfully treated using this plant's seed and seed oil [16]. *Millettia* sp are widely used to treat rheumatic arthritis and gynecologically diseases. The Pongam tree's leaves have both anti-inflammatory and anti-diarrheal properties, and the leaf extracts are antioxidant.

Alternative uses can be investigated to add value in addition to its use in conventional medicine, fibers and biofuel. *Millettia pinnata* leaves (MPL) were chosen for bioplasticizer extraction due to their abundant availability, non-edible nature, and rich phytochemical composition including polyphenols and fatty acid derivatives, which are known to enhance plasticizing performance. Additionally, MPL is an underutilized biomass that does not compete with food crops and thrives in marginal lands, making it a sustainable and environmentally friendly raw material. The novelty of using MPL for solid bioplasticizer development further adds value to this multipurpose plant and promotes the diversification of bio-based plasticizer sources.

To ensure the efficacy of extracted bio-plasticizers from the MPL, several analytical techniques such as FTIR, XRD, SEM, EDX, AFM, and TGA are used to characterize their physical, chemical, morphology, and thermal properties [4]. Hence, the bio-based plasticizers from the MPL sources represent a promising advancement toward sustainable, eco-friendly plasticizer production. The use of natural resources combined with advanced chemical processing and characterization techniques is driving the development of effective, safe, and biodegradable plasticizers that can replace their synthetic counterparts in diverse industrial applications.

2 Materials and methods

2.1 Materials

The fresh leaves of *Millettia pinnata* were collected from Dharmapuri district, Tamil Nadu, India. The leaf was washed three times in double-distilled water and then dried in the shade for 5 days. All chemical used in this research work was purchased from Sigma-Aldrich, India.

2.2 Extraction of bio-plasticizers

Initially, air-dried *Millettia pinnata* leaves were meticulously cut into small pieces and underwent gradual pyrolysis under moderate chemical activation conditions. The biomass underwent treatment with a 1:10 (w/v) solution of hydrochloric acid (HCl) and potassium chloride (KCl), and the mixture was held at room temperature ($25 \pm 2^\circ\text{C}$) for 12 hours to promote regulated thermal and acid-assisted breakdown. This procedure facilitated partial carbonisation, demineralisation, and preliminary surface activation of the lignocellulosic matrix. Subsequent to pyrolysis, the residue was subjected to alkali delignification with 5% sodium hydroxide (NaOH) at ambient temperature for 8 hours, therefore augmenting the extraction of hemicellulose and lignin fractions, and revealing the reactive hydroxyl groups on the cellulose backbone. The alkali-treated biomass underwent amination with 5% dimethylformamide (DMF) and 5% amination

aqueous ammonia (NH_3), facilitating nucleophilic substitution and the grafting of amine functional groups onto the polymeric chains, thereby improving plasticising efficiency through enhanced polarity and intermolecular interactions. The aminated matrix was treated with benzalkonium chloride (BKC), a cationic surfactant that serves as a surface modifier to stabilise and promote further surface reactions. The concluding stage entailed reflux purification with a 1:2 v/v ratio of acetic anhydride and ethanol, which enabled the acetylation of remaining hydroxyl groups and eliminated unbound reactants, yielding a chemically modified, high-performance bioplasticizer appropriate for biocomposite and polymer applications. The whole process carried out for the extraction is represented graphically in Fig. 1.

2.3 Characterization of the MPL plasticizers

For a plant source to be effective in bio-plasticizer production, it should be rich in bioactive constituents like glycols, glycerols, and polyphenols [17]. These compounds exhibit molecular structures that are compatible with polymers, allowing them to reduce intermolecular forces and increase flexibility. The ideal constituents should also have thermal stability, lower density, higher yield, good morphological properties and sufficient functional groups (e.g., hydroxyl, carboxyl, or amino groups) to ensure strong interactions with polymer matrices. To ensure the efficacy of extracted bio-plasticizers, several analytical techniques were used to



Fig. 1 Extraction of bio-plasticizers from *Millettia pinnata* plant leaf

characterize their chemical, physical, morphology, and thermal properties.

2.3.1 Physical properties

Density is a crucial parameter as it influences material properties such as strength, flexibility, and compatibility with polymers, which are essential for assessing suitability in industrial applications [18]. The Pyc II 1340 pycnometer gas analyser uses gas displacement to measure density accurately by filling a sealed chamber with helium and recording the pressure difference between the sample-filled and empty chambers [19]. The volume and density of the sample were calculated by dividing the sample mass by this volume. Yield is a critical factor in determining the effectiveness of the extraction method and the bio-material's potential as a viable plasticizer source. The yield percentage was calculated by dividing the extracted plasticizer mass by the raw bio-source mass and multiplying it by 100 [20].

2.3.2 Fourier transform infrared spectroscopy (FT-IR) analysis

FTIR analysis is a precise and non-destructive method for identifying the chemical structure and functional groups within the sample [21]. The FTIR instrument (FTIR-8400S; Bruker-Alpha, Germany) was set to scan from 4000 to 400 cm^{-1} with a resolution of 2–4 cm^{-1} , performing 16–32 scans to improve signal-to-noise ratio. Environmental conditions were controlled at $25 \pm 2^\circ\text{C}$ and <50% humidity to minimize spectral interference. Background scans were conducted prior to sample analysis, and measurements utilized an MCT detector for enhanced sensitivity, ensuring accurate detection of functional groups.

2.3.3 X-ray diffraction (XRD) analysis

For XRD analysis, bio-plasticizer samples were placed on a flat sample holder to ensure even distribution. Measurements were conducted using a D8-Bruker XRD machine with a $\text{Cu-K}\alpha$ radiation source ($\lambda = 1.5406 \text{ \AA}$). Scanning was performed from 10° to 80° 2θ at a step size of 0.02° and a scanning speed of $1^\circ/\text{min}$. The equipment environment was stabilized at ambient temperature ($25 \pm 2^\circ\text{C}$) to ensure consistency and reduce instrumental drift, optimizing data quality for phase identification and crystallinity analysis. The crystallinity index (CI) was calculated using the Segal method (Eq. 1) and Crystallite size (D) was calculated using the Scherrer equation (Eq. 2) [22, 23]

$$\text{CI} = \frac{I_{\text{cr}} - I_{\text{am}}}{I_{\text{cr}}} \times 100\% \quad (1)$$

$$\text{CS}(\text{nm}) = \frac{k\lambda}{\beta \cos \theta} \quad (2)$$

where I_{cr} is the crystalline curve region, I_{am} is the amorphous curve region, β is the peak full width at half maximum, λ is the radiation wavelength, k (Scherrer's constant) is 0.94, and θ is the diffraction angle.

2.3.4 UV spectroscopy analysis

For UV-Vis analysis, bio-plasticizer samples were dissolved in an appropriate solvent to a concentration of 0.01–0.05 mg/mL to ensure clear spectral readings without saturation [24]. Using a Shimadzu UV-2101 PC spectrometer, measurements were conducted from 200 to 700 nm at a 1 nm interval, with baseline correction performed using a solvent blank. Quartz cuvettes with a 1 cm path length were used to maintain optical clarity. The testing environment was maintained at $25 \pm 2^\circ\text{C}$ to ensure stable readings, allowing for precise determination of absorbance peaks and electronic transitions of the bio-plasticizer.

2.3.5 Scanning electron microscopy analysis with energy dispersive X-ray spectroscopy (SEM-EDX)

For SEM-EDX analysis, bio-plasticizer samples were coated with a thin layer of gold using a sputter coater to enhance conductivity and reduce charging artifacts. Using a ZEISS Gemini 360 SEM machine, imaging was performed at an accelerating voltage of 5–15 kV with a working distance of 5–10 mm, optimizing surface morphology visualization. For EDX analysis, the accelerating voltage was set to 15–20 kV to ensure sufficient energy for element identification [25]. Samples were analyzed under vacuum at room temperature ($25 \pm 2^\circ\text{C}$) to maintain stability, enabling accurate assessment of surface features and elemental composition.

2.3.6 Particle size analysis

For particle size analysis, SEM images of the bio-plasticizer samples were analysed using ImageJ software [26]. The images were calibrated for scale, and the threshold was adjusted to enhance particle visibility. Particles were identified using the “Analyze Particles” feature, allowing for the measurement of individual particle sizes and the calculation of size distribution. Additionally, a histogram analysis was performed to represent the size distribution of the particles visually. This histogram provided a clear graphical representation of particle size frequencies, enabling easy interpretation of the particle size range and the predominant sizes

present in the bio-plasticizer samples, which is essential for evaluating their suitability for various applications.

2.3.7 Atomic force microscopy (AFM) analysis

For the AFM analysis, bio-plasticizer samples were prepared by depositing a thin film on a clean silicon wafer substrate to ensure optimal surface characterization. The measurements were conducted using a Park Systems XE-70 AFM, operating in tapping mode to minimize sample damage and enhance topographical imaging. The scan size was set to $1\ \mu\text{m} \times 1\ \mu\text{m}$ or $5\ \mu\text{m} \times 5\ \mu\text{m}$, with a scan rate of 0.5–1 Hz for high-resolution imaging. The AFM was calibrated before use, and images were acquired at ambient conditions ($25 \pm 2\ ^\circ\text{C}$) to ensure stability. Data analysis was performed using Park Systems software to extract roughness parameters and grain size, providing detailed insights into the surface morphology and characteristics of the bio-plasticizer samples [27].

2.3.8 Molecular docking analysis

Molecular docking studies are computational methods used to predict how two molecules, such as a bio-plasticizer and a protein or enzyme, interact. They help determine binding strength and biological activity, which is useful for applications in drug discovery and materials science [28]. For plant-based bio-plasticizers, docking can reveal interactions with enzymes or structural proteins, offering insights into bioactivity, toxicity, environmental impact, and suitability for applications involving other biological or synthetic materials. For docking bio-plasticizers, PyRx offers an efficient, low-cost solution to understand binding behaviors. PyRx is a user-friendly molecular docking software that integrates a variety of computational tools to perform molecular docking and virtual screening.

2.3.9 Thermal analysis

For thermal analysis, bio-plasticizer samples were prepared by weighing approximately 5–10 mg and placing them in alumina crucibles to ensure minimal interference during testing. Thermogravimetric analysis (TGA), differential thermal analysis (DTA), and differential thermogravimetric (DTG) analysis were performed using a TG/DTA model SIINT-6300 (Japan). The analyses were conducted under a nitrogen atmosphere to prevent oxidation, with a heating rate set at $10^\circ\text{C}/\text{min}$, ranging from room temperature to $600\ ^\circ\text{C}$. Kinetic activation energy was determined using the Coats–Redfern Eq. 3, which facilitated the analysis of the thermal decomposition kinetics [29]. The testing environment was maintained

at a stable temperature to ensure consistent thermal readings, providing insights into the thermal stability and degradation behaviour of the bio-plasticizers.

$$\ln \left[\frac{-\ln(1 - \alpha)}{T^2} \right] = \ln \left(\frac{AR}{\beta E_a} \right) - \frac{E_a}{RT} \quad (3)$$

where T is the absolute temperature in Kelvin, R is the universal gas constant ($8.314\ \text{J mol}^{-1}$), A is the pre-exponential factor, β is the linear heating rate in $^\circ\text{C}/\text{min}$, α is the fraction of the material reacted.

3 Result and discussions

3.1 Physical and chemical properties

The plasticizers extracted from plant leaves were pale yellow in colour, odourless, and had a density of $0.916\ \text{g}/\text{cm}^3$ with an extraction yield of 44%. They showed excellent solubility in both water and organic solvents, which enhanced their compatibility across various chemical environments. This was important because water insolubility can cause phase separation resulting in loss of flexibility or formation of discontinuity zones during film drying. The density was significantly lower than that of many synthetic plasticizers, such as diethyl phthalate ($1.12\ \text{g}/\text{cm}^3$), citrate ($1.7\ \text{g}/\text{cm}^3$), ethylene glycols ($1.11\ \text{g}/\text{cm}^3$), and glycerol ($1.26\ \text{g}/\text{cm}^3$), making it ideal for applications requiring lightweight materials [17, 30–32]. Furthermore, the low molecular weight of this plant-based plasticizer can enhance the flexibility of low-grade polymers, allowing them to exhibit improved flexibility. This characteristic broadens the application range of these polymers and enhances their mechanical properties, making them suitable for more demanding uses. Being odourless is a notable advantage, as it allows the plasticizer to be used in consumer-facing products where scent neutrality is preferred, such as food packaging or medical devices. The extraction yield reflects the effectiveness of the process, though some reduction in yield is due to the elimination of minerals and other organic compounds, which enhances the purity and overall performance of the plasticizer. Optimization of the extraction parameters, such as solvent concentration and duration, could potentially improve yield.

3.2 FT-IR analysis

The FTIR analysis of the plant leaf-derived plasticizer extract revealed a complex profile of functional groups (Fig. 2), indicative of its diverse molecular composition. High wavenumber peaks observed at 3937, 3873, 3829,

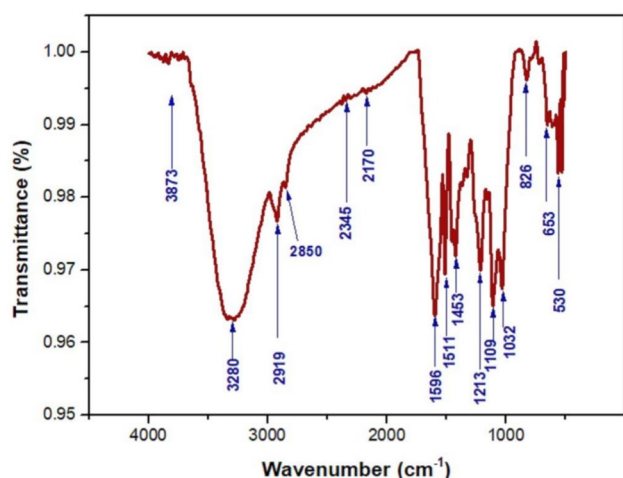


Fig. 2 FTIR spectra of MPL plasticizers

and 3748 cm^{-1} correspond to O–H stretching vibrations, characteristic of hydroxyl groups prevalent in polysaccharides, confirming a strong presence of cellulose and similar polysaccharide structures (Fig. 2). The distinct peak at 3280 cm^{-1} may also be attributed to O–H stretching in hydrogen-bonded hydroxyl groups or N–H stretching, suggesting the potential inclusion of proteinaceous compounds alongside polysaccharides. The peaks at 2919 and 2850 cm^{-1} correspond to C–H stretching in aliphatic $-\text{CH}_2$ and $-\text{CH}_3$ groups, indicating the presence of fatty acid chains or wax-like aliphatic hydrocarbons commonly found in plant materials, which contribute to the plasticizing properties of the extract. Peaks at 2345 and 2170 cm^{-1} , which are likely due to $\text{C}\equiv\text{N}$ or $\text{C}\equiv\text{C}$ groups, suggest minor components or possible contamination, as these groups are less common in natural polysaccharide structures. The prominent peaks at 1596 and 1511 cm^{-1} are associated with $\text{C}=\text{C}$ stretching in aromatic rings,

indicating the presence of phenolic compounds, potentially lignin or other aromatic plant constituents that add rigidity and hydrogen-bonding capability to the extract. Further, peaks observed at 1453 and 1422 cm^{-1} correspond to C–H bending vibrations in methyl and methylene groups, reinforcing the presence of aliphatic hydrocarbons likely from fatty acids or waxes. The peak at 1327 cm^{-1} , possibly due to O–H bending, and the one at 1213 cm^{-1} , likely due to C–O stretching, point to ester linkages or hydroxyl groups that are consistent with plant-derived waxes or polysaccharides. Significant peaks at 1152 , 1109 , and 1032 cm^{-1} correspond to C–O–C stretching in glycosidic bonds, a characteristic feature of polysaccharides like cellulose or hemicellulose, thereby confirming the presence of glycosidic linkages in the extract. Additionally, peaks at 826 and 722 cm^{-1} , associated with out-of-plane bending in aromatic rings, support the presence of aromatic compounds such as lignin or other phenolic structures. The lower-frequency peaks at 653 , 610 , 563 , 530 , and 515 cm^{-1} could represent complex bending modes in aromatic structures or halogenated compounds, though halogen presence in plant-derived materials is less common and might suggest contamination. In conclusion, the FTIR data reveals that the plasticizer extract comprises a mixture of polysaccharides, particularly cellulose, along with phenolic compounds and aliphatic hydrocarbons.

The functional groups and associated chemical fractions for the observed peak are listed in Table 1. This composition, characterized by a combination of O–H groups, aromatic structures, glycosidic linkages, and aliphatic chains, renders the extract a suitable natural plasticizer, with potential applications in biodegradable polymers or eco-friendly packaging due to its flexibility, hydrogen-bonding capability, and compatibility with diverse polymer matrices.

Table 1 FTIR peak observed and their related chemical fraction [33–35]

Peak (cm^{-1})	Functional group	Related chemical fraction
3937–3748	O–H stretching	Hydroxyl groups in polysaccharides
3280	O–H or N–H stretching	Hydrogen-bonded hydroxyls, proteinaceous compounds
2919, 2850	C–H stretching	Aliphatic hydrocarbons (fatty acids, waxes)
2345, 2170	$\text{C}\equiv\text{N}$ or $\text{C}\equiv\text{C}$ bonds	Nitriles or alkynes (minor components/contaminants)
1596, 1511	$\text{C}=\text{C}$ stretching	Aromatic rings in phenolic compounds (lignin)
1453, 1422	C–H bending	Methyl and methylene groups in aliphatic chains
1327	O–H bending	Hydroxyl groups in polysaccharides or waxes
1213	C–O stretching	Esters or alcohols in polysaccharides and waxes
1152, 1109, 1032	C–O–C stretching	Glycosidic bonds in polysaccharides (cellulose, hemicellulose)
826, 722	Aromatic ring deformation	Aromatic compounds (e.g., lignin, phenolics)
653, 610, 563, 530, 515	Bending modes (potential C–Cl or C–Br)	Aromatic structures or minor halogenated compounds

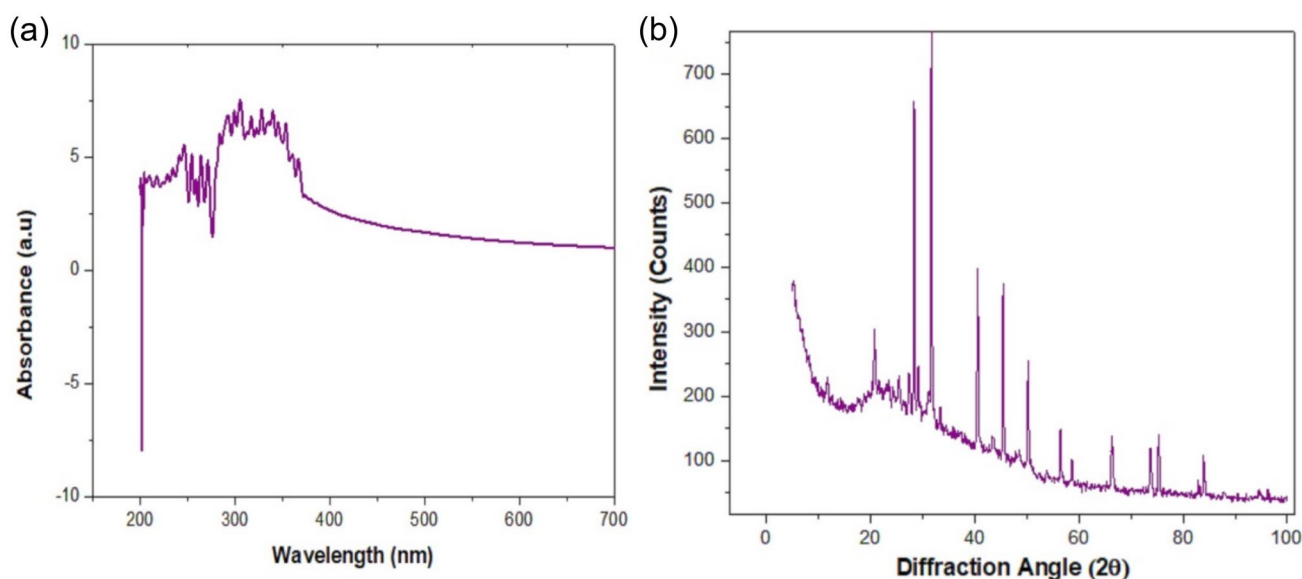


Fig. 3 MPL plasticizers **a** UV-Vis spectra, **b** XRD spectra

3.3 UV-Vis analysis

The UV analysis of the extracted plasticizer revealed absorbance peaks (Fig. 3a) at various wavelengths, indicating the presence of specific phytochemical compounds. Notably, peaks around 240–270 nm, particularly at 251 nm and 286 nm, suggest the presence of flavonoids, known for their antioxidant properties and potential functionality as plasticizers [36]. Additionally, the peaks at 202, 213, and 221 nm may correspond to alkaloids, which are nitrogen-containing compounds that enhance the plasticizer's performance. The peaks observed in the 280–320 nm range, specifically at 296, 302, and 310 nm, indicate the presence of polyphenols, which contribute to the plasticizer's thermal and oxidative stability [37]. Overall, the range of absorbance peaks signifies a complex mixture of bioactive compounds that can enhance the plasticizer's functional properties, making it a viable alternative to synthetic plasticizers. The combination of flavonoids, alkaloids, and polyphenols may provide synergistic effects, improving flexibility, durability, and resistance to environmental degradation, thus highlighting the sustainability and safety of using plant-derived materials in plasticizer production.

3.4 XRD analysis

The XRD analysis of the bio plasticizer extracted from plant leaves revealed both crystalline and amorphous peaks, indicating a mixed structural composition as shown in Fig. 3b. The crystalline peaks, particularly at 28.47°, 31.78°, 40.78°, 45.6°, and 50.4°, reflected a defined crystalline structure, while the broader amorphous peaks 22.17°, 33.73°, 43.44°,

reflecting the presence of polysaccharides and other amorphous components in the bio plasticizers [38]. The presence of these amorphous regions may contribute to the flexibility and processability of the materials, balancing the rigidity provided by the crystalline structure. Similar peaks were also seen in the investigation of bioplastic made with cassava starch, water, acetic acid (22.27°), and another bioplastic made with gum ghatti and glycerol (33.16°) [39, 40]. An optimal balance between crystalline and amorphous content is often desirable, allowing the bio plasticizer to maintain essential mechanical properties while still offering the benefits of lower crystallinity. The crystallinity index was calculated to be 47.5% implies a moderate degree of crystallinity, which contributes to enhanced mechanical stability without compromising flexibility, an advantage in polymer applications [41]. This crystallinity is similar to the value of ethylene glycol and glycerol plasticizers (48% and 55%). Furthermore, the calculated crystalline size of 11.3 nm supports the presence of nanoscale crystalline domains within the bioplasticizer matrix, which could improve compatibility with other polymeric materials. When compared to conventional plasticizers, which often exhibit broader and less distinct peaks due to their predominantly amorphous nature, the sharp peaks combined with amorphous characteristics in the bioplasticizers highlight their potential as effective alternatives in polymer applications. This unique structural combination suggests that the bio-derived plasticizers could improve the performance and sustainability of plastic formulations, leveraging the advantages of both crystalline and amorphous phases. Overall, the findings indicate that the structural attributes of these bioplasticizers, particularly their polysaccharide content, can be strategically

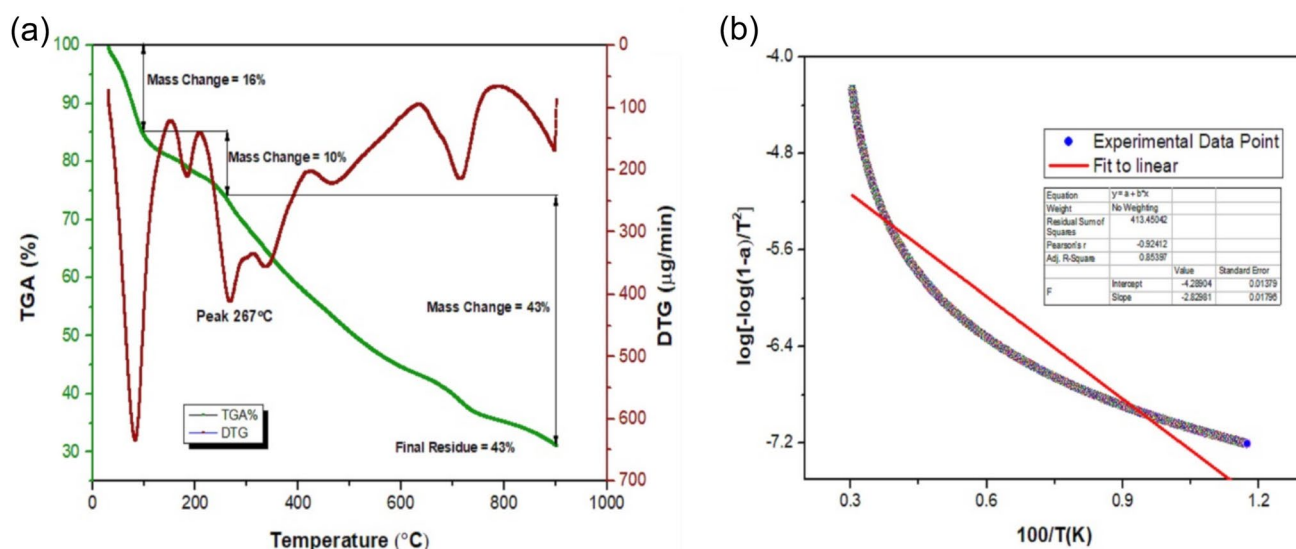


Fig. 4 MPL plasticizers **a** TGA/DTG analysis; **b** Coats-Redfern activation energy

harnessed to optimize their functional properties in various applications.

3.5 TGA

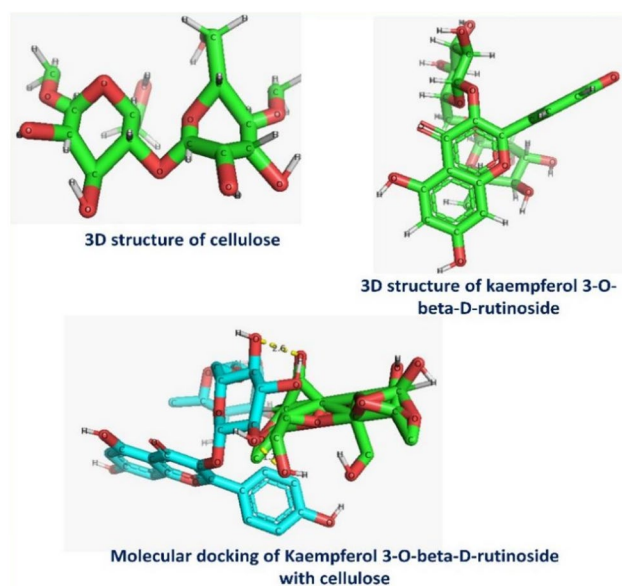
The thermal degradation analysis followed multi-stage decomposition of distinct molecular components as seen in Fig. 4a. The initial stage, which occurred at 50 to 98 °C with a mass loss of 16%, mass loss was due to residual moisture, as well as other volatile compounds present in the MPL bio plasticizer [42]. This stage had a smaller mass loss and it was associated with the drying of residual water and low-boiling-point organics that might be naturally present or retained from the extraction process. The second stage occurred between 98 and 260 °C and involves the breakdown of major plasticizing components like glycols, smaller fatty acids, esters, polyphenols, simple sugars, or other low-molecular-weight components [43]. This stage showed moderate mass loss (10%) and represents the initial degradation of the plasticizer's molecular structure. The third degradation phase happened at higher temperatures, typically from 260 to 898 °C. This stage involves the breakdown of tannins, saponins, non-ferrous components, or more complex plant compounds. During this stage, large molecules like polysaccharides and lignin-derived structures decompose, leading to the release of gases (CO₂, CO, and H₂O). The significant mass loss (43%), reflecting the core thermal breakdown of the bio plasticizer in this stage. Further degradation beyond 898 °C resulted in the formation of carbonaceous char [44]. This residue is produced due to the plant material's carbon content and structural complexity.

Further insight is provided by the derivative thermogravimetry (DTG) analysis, where distinct peaks were observed at 84 °C, 267 °C, and 716 °C, indicating key phases of thermal degradation. Typical bioplasticizers, such as those derived from triglycerides or polysaccharides, also exhibit multiple DTG peaks but at lower temperatures (commonly between 100 and 500 °C). The higher decomposition peaks for MMPL plasticizers suggest a complex or cross-linked structure, enhancing the overall thermal stability and possibly broadening the potential for applications requiring elevated heat resistance.

The Coats-Redfern method (Fig. 4b) enables the precise estimation of the energy required to initiate the degradation process. Using equation (2), the activation energy of the decomposition process was calculated to be 54.16 kJ/mol, suggesting moderate thermal stability, which is slightly higher than bio-based plasticizers (range between 40 and 60 kJ/mol) [43]. This level of activation energy aligns with the observed multi-stage degradation profile, wherein different compounds within the plasticizer such as glycols, polyphenols, and saponins decompose at distinct temperature ranges. This moderate activation energy is advantageous for applications involving moderate thermal exposure, as it suggests the bio-plasticizer can withstand standard processing conditions without excessive breakdown. This thermal resilience allows it to maintain its plasticizing function effectively in composite materials or packaging applications, thereby enhancing material flexibility and durability. Additionally, an activation energy of 54.16 kJ/mol implies that while the bio-plasticizer remains stable under moderate thermal conditions, it is also capable of decomposing under high temperatures commonly found in composting or waste treatment environments [11].

Table 2 Degradation behaviour of the MPL compared with synthetic and bio-plasticizers [10, 32, 45, 48–51].

S. no	Plasticizers	Degradation temperature (°C)	Kinetic activation energy (kJ mol ⁻¹)
1.	Di-n-butyl phthalate	305	78.5
2.	Phthalic isobutanol	260–300	62
3.	Calotropis gigantica	244	53.98
4.	Diethyl phthalate	295	66.45
5.	Dimethyl phthalate	230–260	44.3
6.	Ethylene glycol	230–250	-
7.	Glycerine	130–170	67.57
8.	Pandanus tectorius	341	-
9.	Agave sisalana	345	25.34
10.	Propylene glycol	200 below	55.80
11.	Di isobutyl phthalate	255	79.2
12.	Jatropha curcas L	290	28.08
13.	Phthalic butanol	240–285	62
14.	Nelumbo nucifera	277	62.20
15.	Pedaliium murex	262	55.77

**Fig. 5** MPL plasticizers molecular docking

Thus, the MPL bio-plasticizer's activation energy highlights a balanced profile. In Table 2, the degradation behaviour, and kinetic activation energy of MPL were compared with synthetic and bio-plasticizers. The findings confirm the potential of this bio-plasticizer for sustainable applications where moderate thermal resilience and environmentally friendly decomposition are both prioritized.

Table 3 Major phytoconstituents in MPL

S. no	Phytoconstituents	Binding affinity
1	Quercetin	– 2.4
2	Kaempferol	– 2.8
3	Kaempferol 3-O-beta-D glucopyranoside	– 2.8
4	Isoquercitrin	– 2.1
5	Vitexin	– 2.8
6	Rutin	– 3.2
7	Kaempferol 3-O-beta-D-rutinoside	– 3.8

3.6 Molecular docking analysis

The molecular docking study (Fig. 5) of phytoconstituents extracted from *Milletia pinnata* leaves, conducted using PyRx software, revealed valuable insights into their binding affinities and potential contributions to bio-plasticizer applications. The findings indicate that various phytoconstituents exhibit different degrees of interaction with target sites, which is reflected in their binding affinity values. The higher negative docking score represented a higher binding affinity [49]. The binding affinity for the compounds were listed in Table 3. All seven compounds have binding affinities in the range of – 2.1 to – 3.8 kcal/mole. Among these, kaempferol 3-O-β-D-rutinoside showed the strongest binding affinity (– 3.8), suggesting it can form stable interactions that enhance the bioactivity and environmental compatibility of the bio-plasticizer. Rutin followed with a binding affinity of – 3.2, indicating robust interactions that may contribute to the material's durability and effectiveness in bio-based applications. Additionally, kaempferol, kaempferol 3-O-β-D-glucopyranoside, and vitexin each showed a binding affinity of – 2.8, suggesting moderate interactions that could support the bio-plasticizer's structural stability and biodegradability. The results for quercetin and isoquercitrin, with binding affinities of – 2.4 and – 2.1 respectively, suggest weaker interactions, yet these compounds may still contribute antioxidant properties that enhance the bio-plasticizer's longevity and resistance to environmental degradation. Overall, these findings provide a clearer understanding of how each phytoconstituent may enhance the performance and sustainability of the bio-plasticizer derived from *Milletia pinnata* leaves. The high binding affinities of specific compounds indicated that the bio-plasticizer was promising for applications requiring environmental compatibility and bioactivity.

3.7 SEM-EDX analysis

The SEM analysis of MPL plasticizers revealed distinct surface morphology and structural characteristics across different magnifications (200X, 3000X, and 10,000X) as shown in Fig. 6. The particles appear to be randomly

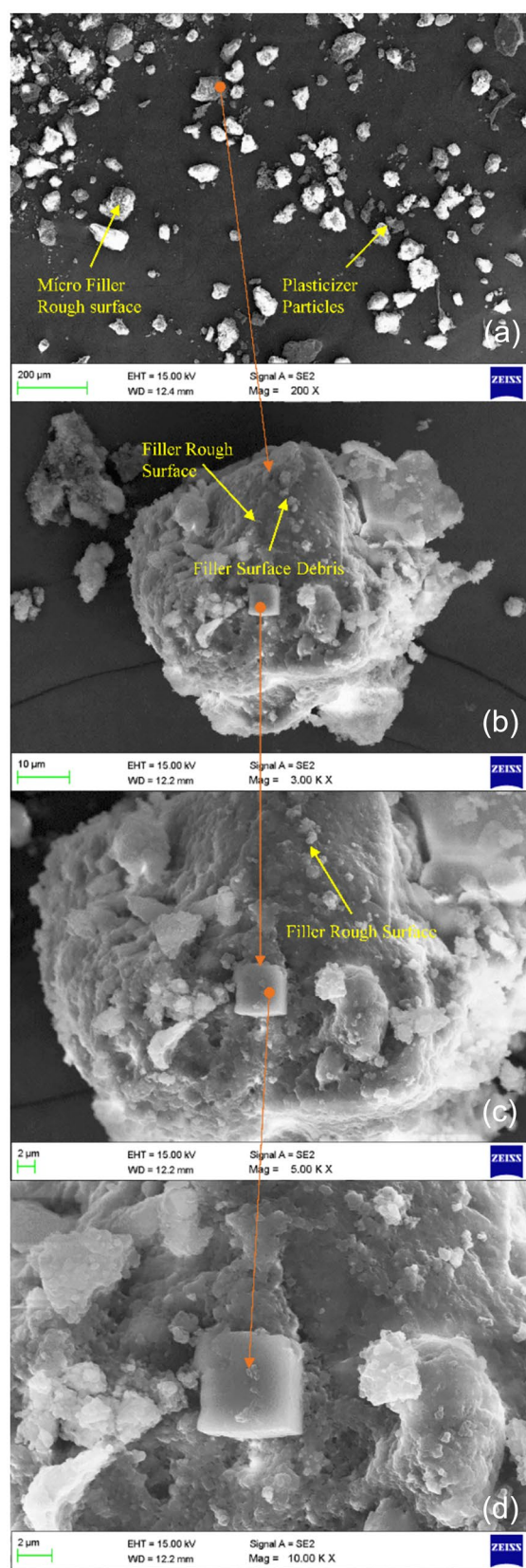


Fig. 6 MPL plasticizers SEM image at various magnification

scattered, resulting in a partially uneven surface distribution. This was due to the milling process, which tends to fragment the material into varied particle sizes. The absence of agglomeration enhanced the effectiveness of the bioplasticizers based on free volume theory by ensuring uniform distribution throughout the polymer matrix. This uniformity maximizes free volume, allowing polymer chains to move more freely, which improves flexibility, lowers the glass transition temperature, and enhances mechanical strength [50]. At higher magnifications, the SEM images revealed the regions resembling honeycomb structures with visible voids and holes. These honeycomb-like voids add a unique dimension to the surface morphology, suggesting possible applications where energy absorption or lightweight characteristics are desired. The honeycomb voids may facilitate enhanced polymer entrapment and physical interlocking, contributing to improved mechanical integrity. Moreover, the voids could enhance the material's impact resistance and compatibility with low-density matrices, making these plasticizers potentially useful in applications requiring lightweight fillers with cushioning or energy dissipation properties [51].

The surface characteristics were predominantly granular, with the granules appearing relatively smooth. A smooth surface can significantly enhance the properties of a plasticizer by promoting uniform distribution, improving compatibility, and reducing surface imperfections that may disrupt the plasticizer's interaction with the polymer matrix. In addition, in applications where optical clarity is required, a smooth surface can help maintain transparency, which is beneficial for flexible films and coatings where aesthetic and functional performance are critical. These findings highlight the potential for improved dispersion, adhesion and impact resistance, which are critical to optimizing the performance of the material in future applications.

3.8 Particle size analysis

The particle size of the MPL bio-plasticizers was measured using ImageJ software using SEM image as shown in Fig. 7. The clear contrast and visibility of particle boundaries were enhanced by adjusting the threshold. The scale was set by using the “Set Scale” function, referencing the image's scale bar to calibrate measurements in micrometers. Using the freehand selections tool, each particle was outlined individually and recorded the dimension. This process was repeated for 50 particles to obtain the individual size measurements and average particle size was measured. To better understand the distribution of particle sizes, the recorded data were transferred to a spreadsheet, where a histogram was created to visualize the particle size distribution and determine the mean dimension and standard deviation. The mean particle size of MPL is 39.37 μm with a standard deviation of 14.43.

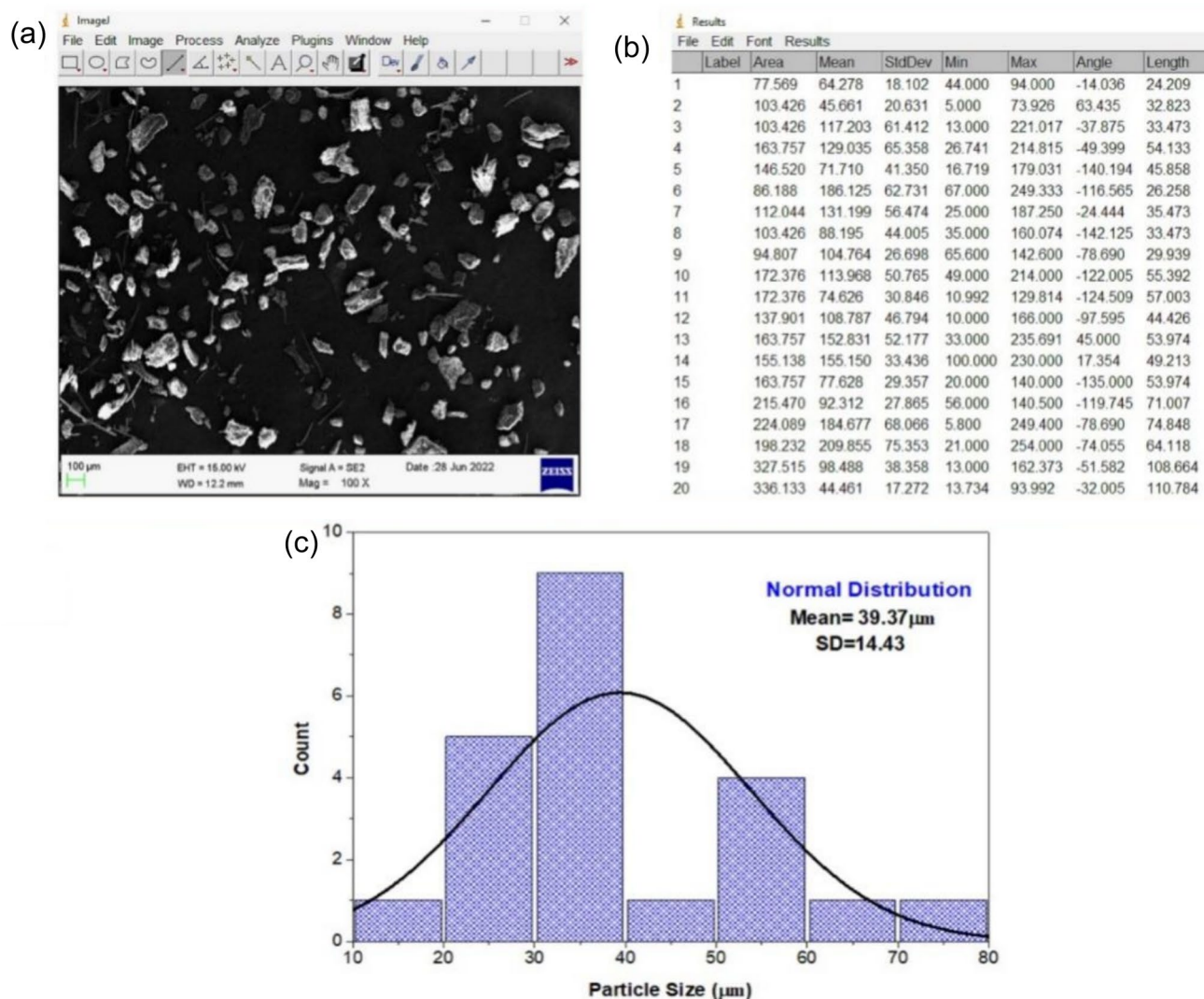


Fig. 7 MPL plasticizers a SEM image, b Image J measurements, c Histogram analysis

3.9 EDX analysis

The major element detected (Fig. 8b) was carbon (C) with a weight percentage of 60.58%, indicating the organic nature of the plasticizers and their high carbon content contributing to improved flexibility and reduced brittleness in films and composites. Chlorine (Cl) and sodium (Na) were present at 11.36% and 11.82% respectively, probably due to specific chemical treatments applied during extraction which may have introduced or retained these elements. Chlorine may provide improved resistance to degradation, while sodium may improve thermal stability or specific conductivity, depending on the matrix used. Oxygen (O), at 15.70%, reflects the presence of functional groups such as hydroxyls or carbonyls, which are commonly found in bio-derived materials which can improve compatibility with polymer matrices, enhancing dispersion and bonding [52, 53]. This

property can be particularly useful in bio-based films or eco-friendly composites where good adhesion between the matrix and filler is essential. Magnesium (Mg) is detected in a smaller amount, at 0.54%, and may originate from the plant material itself or be introduced during processing. Although its minimal, can act as a reinforcement, potentially enhancing the composite's mechanical properties slightly. Overall, this EDX characterization highlights a carbon-rich composition with essential elemental constituents that contribute to the functionality and performance of the bio-plasticizers.

3.10 Atomic force microscopic analysis

The average roughness (R_a) of 3.83 nm indicated a low roughness level, which could contribute to a more transparent and uniform film when the bio-plasticizer is used in thin layers [54]. Additionally, the negative Skewness (R_{sk})

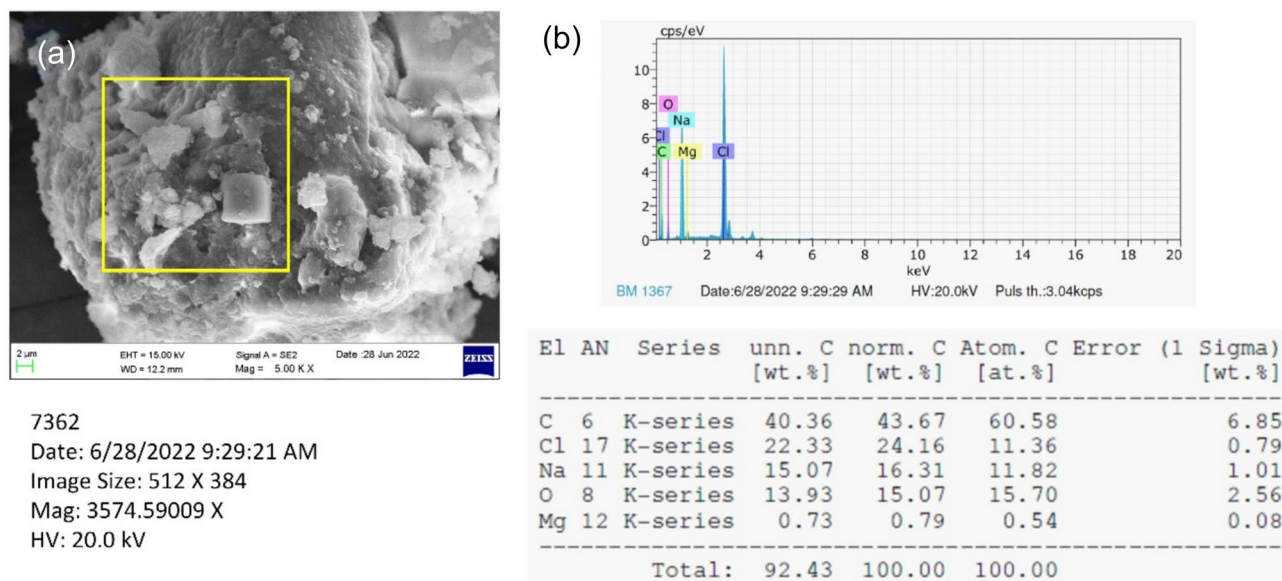


Fig. 8 EDX analysis of MPL plasticizers

values of -2.060 and -2.442 in both scans indicated that the surface had a porous structure, which could be beneficial in applications where retaining bonding agents or adhesives was important [55]. The Kurtosis (Rku) values of 12.429 (horizontal) and 19.832 (vertical) were relatively high (greater than 3), signifying a relatively smooth texture [56, 57]. This smoother surface texture is beneficial in applications requiring uniformity, such as thin films where even distribution is essential for performance. The other parameter peak-to-valley (Rpv) (43.272 nm, 53.186 nm), root mean square roughness (Rq) (5.571 nm, 5.288 nm), average maximum height (Rz) (23.456 nm, 26.503 nm) indicating a moderate level of surface roughness. The AFM three dimensional, two dimensional and roughness parameters were illustrated in Fig. 9. This could be advantageous in bio-composites, as a moderately rough surface may enhance adhesion between the bio-plasticizer and other composite materials. Overall, these surface characteristics suggest that the bio-plasticizer, with its balanced texture, moderate roughness, and complex peak-valley structure, could be highly suitable for use in bio-composite materials where enhanced adhesion and interlocking are required, as well

as in films that benefit from a smooth yet slightly textured surface for mechanical stability.

4 Conclusions

The bioplasticizer extracted from the leaf of the MPP plant presented a versatile alternative to synthetic plasticizers due to its unique physico-chemical, structural, thermal and morphological properties. The material achieved a high extraction yield of 44% and exhibited a low density of 0.916 g/cm³, making it suitable for lightweight polymer formulations. It demonstrated excellent solubility in both aqueous and organic solvents, enhancing its versatility across different processing environments. FTIR and UV analyses confirmed a complex composition of polysaccharides, aliphatic hydrocarbons, phenolic compounds, and bioactive groups such as flavonoids and polyphenols, contributing to its thermal stability, flexibility, and environmental resistance. The XRD analysis revealed a crystallinity index of 47.5% and a crystallite size of 11.3 nm, indicating a balanced crystalline-amorphous structure that supports both mechanical strength and elasticity ideal for biodegradable and eco-friendly applications. Thermal analysis revealed a multi-stage degradation with an activation energy of 54.16 kJ/mol, indicating suitability for moderate thermal applications and potential for compostable end-of-life. AFM analysis showed a surface roughness average (Ra) of 3.83 nm, with negative skewness and high kurtosis values, indicating a smooth yet porous texture. The SEM and particle size analyses highlighted a smooth, honeycomb-like morphology conducive

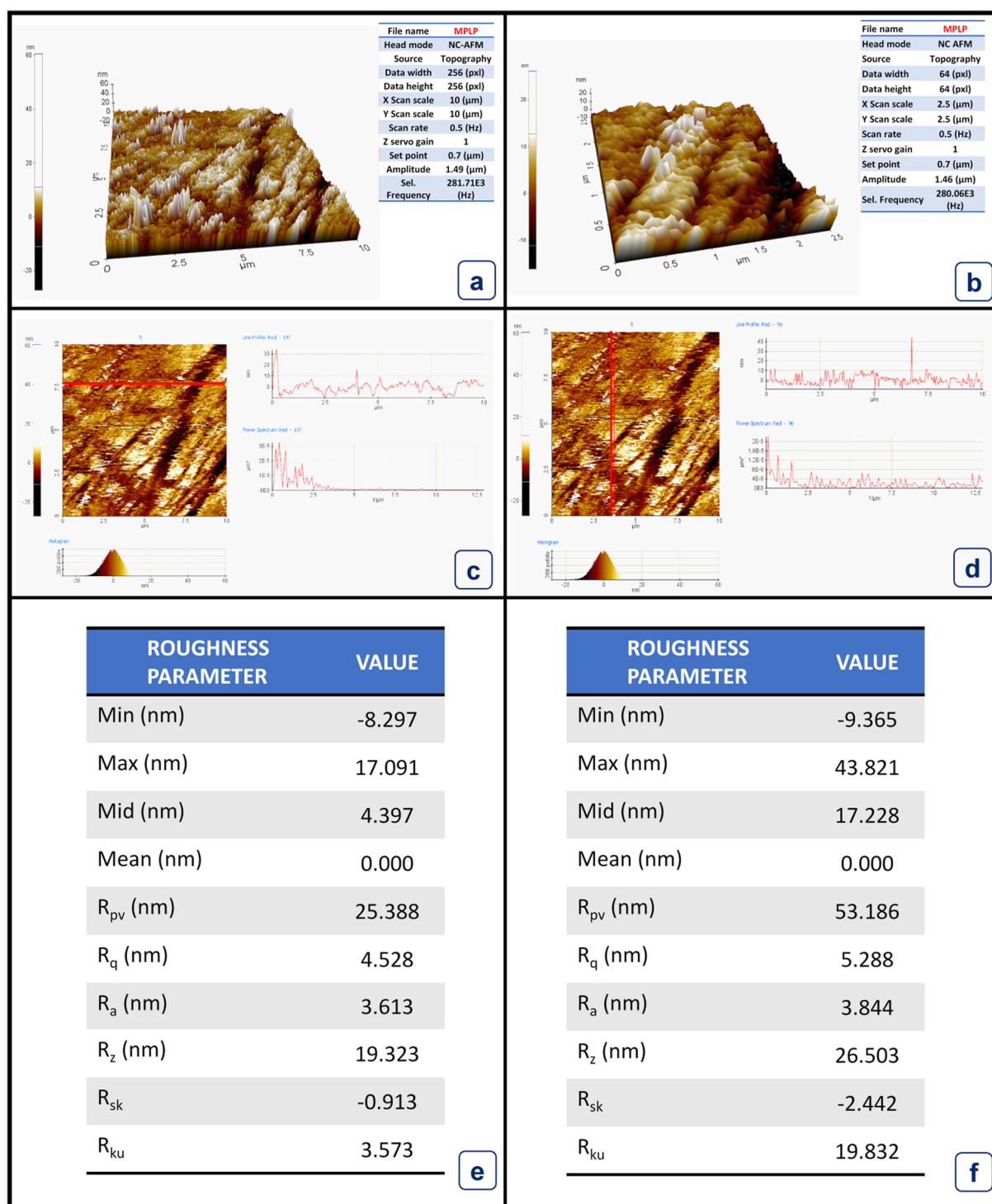


Fig. 9. MPL plasticizers AFM analysis **a** and **b** horizontal and vertical 3D plot; **c** and **d** horizontal and vertical 2D plot; **e** and **f** roughness parameters

to improved dispersion and energy absorption, enhancing compatibility with various matrices. Collectively, these attributes establish the MPP bio plasticizer as a promising, sustainable substitute for conventional plasticizers, offering

compatibility with environmentally friendly processing and performance requirements polymer applications, such as packaging, coatings, and flexible films.

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Availability of data and materials The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

Declarations

Conflict of interest The author(s) declared no competing interest with respect to the research, authorship, and/or publication of this article.

Ethical approval Not Applicable

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