

Cold Plasma-Induced Surface Modification of Microfibrillated Cellulose Using Lauric Acid to Enhance Compatibility in Polymer Composites

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Abstract: Green materials, such as microfibrillated cellulose (MFC), are increasingly used as fillers in polymer composites for academic and industrial applications. However, their inherent hydrophilic property limits compatibility with polymer matrix. This study employs an environmentally friendly cold plasma technique to modify the surface of MFC, improving its compatibility with the polymer. Plasma treatment was performed at a voltage of 60 V for 30 min by making three molar ratios (3:1, 4:1, and 5:1) between lauric acid as a hydrophobic precursor and anhydroglucose (AGU). The results indicate several changes in the modified MFC properties, as evidenced by the appearance of a new peak at a wavenumber of 1742 cm^{-1} (ester's C=O) in FTIR spectra, indicating successful plasma-induced grafting. XPS results also confirm the formation of O-C=O bond at a binding energy of 289.3 eV. The optimum conditions were obtained at a molar ratio of 4:1 (lauric acid:AGU). There was a decrease in the hydrophilic property of MFC, indicated by an increase in the water contact angle from 50.16° to 71.26° . Moreover, the surface tension difference between MFC and polypropylene was significantly reduced from 136.99 to 47.51%, suggesting improved compatibility.

Keywords: microfibrillated cellulose; surface modification; cold plasma; compatibility

■ INTRODUCTION

Composite polymers have gained popularity in various industries, such as automotive, maritime, and construction, owing to their lightweight and rust-resistant advantages while exhibiting high strength and stiffness [1]. Polymer composites combine the primary materials, matrices with fillers, or reinforcements. The combination of these two materials can generate products with better strength based on the design [1-2]. Cellulose fibers as filler materials for composite polymers are gaining increasing attention in academic and industrial circles [3-5]. This trend aligns with the increasing concern for the environment, where more environmentally friendly and

sustainable materials are needed. Generally, fillers/reinforcements often used in composite polymer applications are synthetic fibers, such as glass fibers derived from non-renewable resources. They may pose environmental or health risks due to their abrasive and non-biodegradable nature [6]. In contrast, cellulose is the biopolymer material that can be obtained from biomass waste in the highest amount. Consequently, the use of cellulose as an alternative material for replacing synthetic fibers has excellent potential due to its availability, non-toxic, low cost, and biodegradable natures, also owing to its relatively lightweight and non-abrasive properties [7]. Besides, compared to synthetic fibers, such as E-glass,

cellulose offers several advantages, such as comparable specific strength and higher specific modulus [8].

Cellulose, one type of which is microfibrillated cellulose (MFC), is a polysaccharide composed of a repeating unit known as cellobiose, namely an anhydroglucose (AGU) dimer connected by β -1,4 glycosidic linkages [9]. Even though MFC offers the benefits mentioned before, its inherent hydrophilic property—due to the high number of hydroxyl groups in its chemical structure—poses a major challenge when used as fillers/reinforcements in polymer composites, especially those composed of hydrophobic matrix, such as polypropylene (PP). This hydrophilic nature causes significant surface tension mismatch, reducing compatibility and poor interfacial adhesion [7,10]. For this reason, improving the compatibility between cellulose and polymer matrix is crucial for enhancing their interfacial adhesion, which ultimately affects the properties of composite materials. Many studies have explored various techniques to reduce the hydrophilic property of MFC, one of which is surface modification.

Surface modification techniques are widely carried out chemically by grafting more hydrophobic chains on cellulose surfaces using chemical solvents and catalysts [9,11-15]. For example, Pasquini et al. [12] carried out surface esterification of cellulose fibers by chemically reacting dodecanyl chloride, toluene, and pyridine reagents for reinforcement in low-density polyethylene polymer matrices. Similarly, Kim et al. [11] grafted lauryl alcohol by chemically reacting cellulose and lauryl alcohol using DMF solvent and pyridine catalyst. However, although they produced more hydrophobic cellulose and increased interfacial adhesion with the polymer matrix, these methods still used harmful chemicals, such as toxic toluene, DMF, and pyridine, which can generate chemical waste, endangering the environment. Therefore, developing more environmentally friendly methods for modifying cellulose surfaces is necessary.

Cold plasma is one of the more environmentally friendly techniques to modify the surface of various materials, as it requires fewer chemicals and a relatively short duration compared to conventional wet chemical techniques [16-18]. Cold plasma can be used to modify

the material surface in two ways: first, by condensing the precursor to form a physically deposited layer, and second, by reacting the precursor with the activated substrate to form a stronger, wash-resistant covalent bond [16]. Another form of modification is plasma-induced grafting. Several studies have succeeded in grafting monomers to various material substrates—such as starch, natural fiber, natural rubber, and cotton—using the cold plasma method [16,19-22]. However, generating an efficient reaction between substrate and precursor has still proven challenging, as it depends on the precursor's reactivity, substrate type, and plasma parameters.

Therefore, this study—through the plasma technique—provides an initial assessment of the feasibility of MFC surface modification using lauric acid as a hydrophobic precursor, with the aim to enhance MFC compatibility with polymer matrices. The plasma treatment is expected to functionalize the MFC surface through the grafting of lauric acid, thereby reducing the surface tension of MFC. As a result, the surface energy gap between MFC and PP is anticipated to decrease, improving interfacial compatibility. As a preliminary investigation, this study focused on the feasibility of lauric acid as a plasma precursor to modify the MFC surface. The molar ratio of cellulose:lauric acid was varied, while the plasma treatment condition was kept constant. Attenuated total reflectance-Fourier transform infrared (ATR-FTIR) and X-ray photoelectron spectroscopy (XPS) were performed to observe the changes in surface chemistry of MFC. Moreover, water contact angle measurement and surface tension analysis were also conducted to evaluate MFC changes in hydrophilicity and surface wettability. Additionally, thermogravimetric analysis (TGA) was carried out to determine the thermal stability of the pristine and modified MFC surfaces.

■ EXPERIMENTAL SECTION

Materials

In this study, the materials used for cellulose surface modification were MFC, lauric acid, and acetone. Additionally, aquadest and ethylene glycol were used for contact angle testing. All materials were

analytical grade and purchased from Sigma Aldrich, Germany.

Instrumentation

The instruments used in this study were a plasma generator, oven, desiccator, ATR-FTIR spectroscopy Bruker Alpha 11, XPS Kratos Supra+, Ossila Contact Angle Goniometer, and TGA-TG 209 F3 Tarsus Netzsch.

Procedure

Surface modification of MFC

Low-pressure air plasma was used to modify the surface of MFC. The plasma circuit consisted of a commercial AC to AC voltage regulator (Variac, OKI model TDGC 2, China)–with an output of up to 220 V, 3 kVA, and a frequency of 50/60 Hz–that was connected to a full bridge converter with a DC output, which was then amplified using a plasma driver with a maximum output of approximately 21 kV connected to the electrodes. The pressure was set at approximately –60 cmHg. Initially, the MFC was exposed to plasma for 1 min with the AC input setting at 60 V to activate its surface. Following this, a solution of lauric acid in acetone was added, and then plasma treatment was conducted again for 30 min, also at the setting input voltage of 60 V. The plasma treatment condition was determined based on preliminary exploratory trials and the operational limits of the available equipment. The variations and conditions of plasma treatment are displayed in Table 1. After the treatment, the samples were washed using acetone to remove any remaining unreacted lauric acid.

MFC characterization

Characterization was performed for the surface and bulk MFC before and after plasma treatment. Surface characterization was performed using ATR-FTIR, XPS, and water contact angle (WCA) measurement. Bulk

characterization was performed using TGA to analyze the thermal stability of MFC. The detailed methods were described as follows.

ATR-FTIR analysis. The spectra were recorded at a wavenumber range of 4000–400 cm^{-1} with a resolution of 4 cm^{-1} and 32 scans.

XPS analysis. XPS measurements were performed with a monochromatic Al K-alpha X-ray source. Wide scan measurements were performed at the range of 0–600 eV, and high-resolution spectra were taken for C1s (280–298 eV) with a pass energy of 20 eV and a resolution of 0.05 eV/step to measure the binding energy and atomic concentration quantitatively. The test was performed on a sample with a depth of 10 nm. The quantitative chemical analysis was based on method number 21. The ESCApe XPS software was used for background subtraction (Shirley-type), peak integration, fitting, and quantitative chemical analysis. The C1s (C–C) peak at 285 eV was used to calibrate the binding energy scale [21].

WCA and surface tension. WCA was measured to assess changes in the hydrophilic properties of the pristine and modified MFC surfaces. A microsyringe was used to drop 10 μL of aquadest onto the sample surface. A camera incorporated within the goniometer was used to capture the droplet images, which were then processed to determine the contact angle. The contact angle was used to analyze the surface wettability. A contact angle above 90° indicates a hydrophilic surface, while a lower than 90° signifies a hydrophobic surface [23].

Surface tension was determined using the Owens-Wendt method. Following the WCA testing, the contact angle was also measured using ethylene glycol to calculate the energy. The determination of surface tension was determined using the Owens-Wendt method [24–26]. Following the WCA testing, the contact

Table 1. Precursor composition, plasma parameter condition, and sample code

Sample code	Molar ratio of lauric acid:AGU	Plasma voltage (V)	Duration of plasma treatment (min)
MFC	-	-	-
p-MFC 1	3:1	60	30
p-MFC 2	4:1	60	30
p-MFC 3	5:1	60	30

angle was also measured using ethylene glycol to calculate the energy. The determination of surface tension using this method was based on the polar component (γ_s^p) and the dispersive component (γ_s^d) of the surface tension of a solid (γ_s), where $\gamma_s = \gamma_s^p + \gamma_s^d$. The formula used to determine the surface tension is presented in Eq. (1) [24,26].

$$\frac{\gamma_l(1+\cos\theta)}{2\sqrt{\gamma_l^d}} = \sqrt{\gamma_s^p} \sqrt{\frac{\gamma_l^p}{\gamma_l^d}} + \sqrt{\gamma_s^d} \quad (1)$$

In Eq. (1), γ_l is the surface tension of the liquid, γ_l^d is the dispersive component of the liquid, and γ_l^p is the polar component of the liquid. By using this equation, the surface tension of the solid, which consists of γ_s^p and γ_s^d , can be determined.

TGA characterization. TGA was performed to evaluate

the thermal stability and thermal degradation of the pristine and modified MFCs. Approximately 20 mg of the sample was inserted into an alumina crucible, then analyzed at a temperature range of 25–750 °C at a heating rate of 10 °C/min. Next, a 20 mL/min nitrogen gas flow was used as a purge gas.

RESULTS AND DISCUSSION

Surface Chemistry Analysis

Fig. 1 presents the FTIR and XPS analysis results on the pristine and modified MFC samples, while Table 2 presents their atomic composition. As seen in the FTIR spectra in Fig. 1(a), characteristic peaks for cellulose were detected at six wavenumbers, namely 3331, 2899, 1624, 1315, 1023, and 893 cm^{-1} , attributed to strong O–H stretching, C–H stretching, O–H bending, C–H bending,

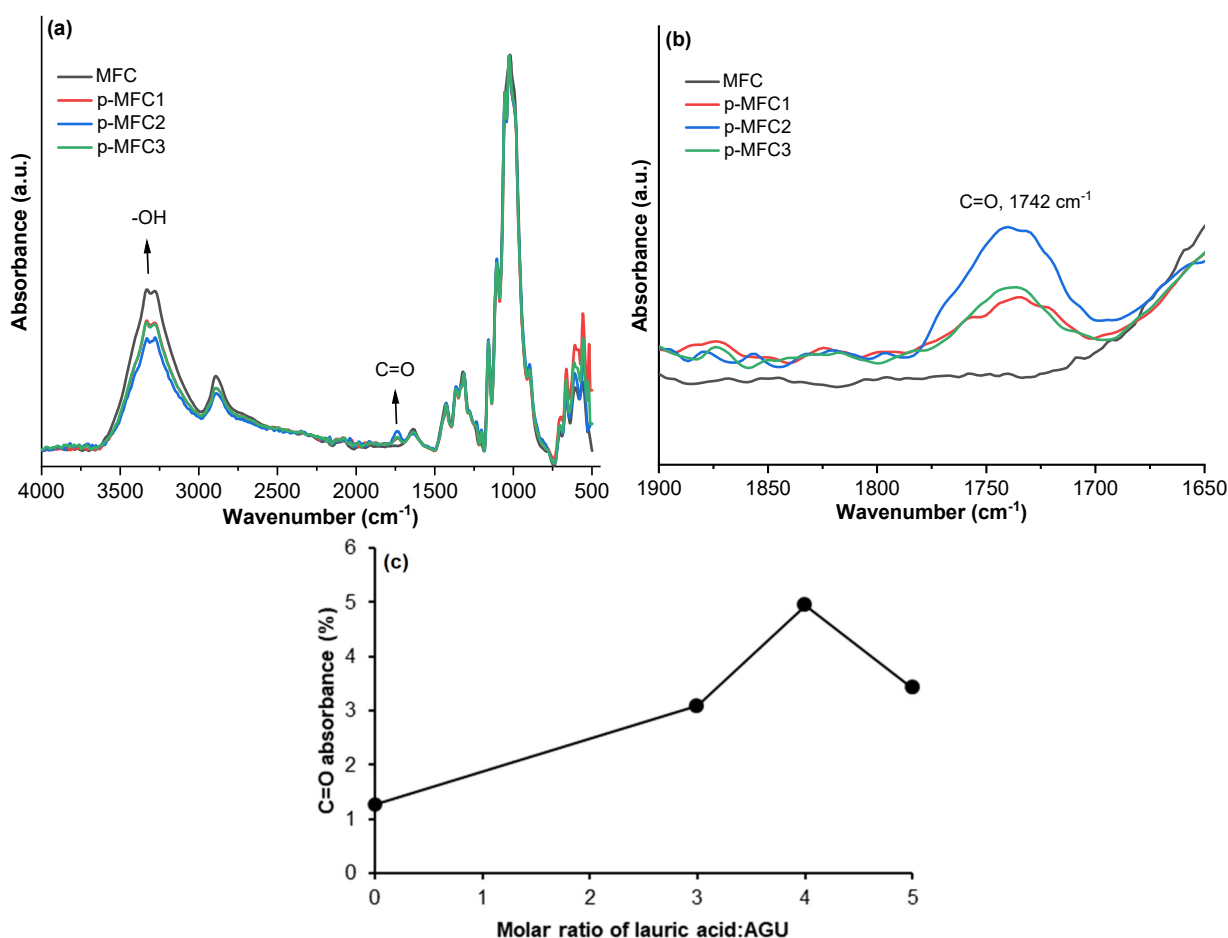


Fig 1. FTIR spectra at a wavenumber range of (a) 4000–400 cm^{-1} , (b) at 1742 cm^{-1} (C=O) normalized to the C–O absorbance of the pyranose units at 1023 cm^{-1} , and (c) trend of absorbance intensity at 1742 cm^{-1} (C=O) after normalized

Table 2. Atomic composition and C/O ratio of the pristine and modified MFCs

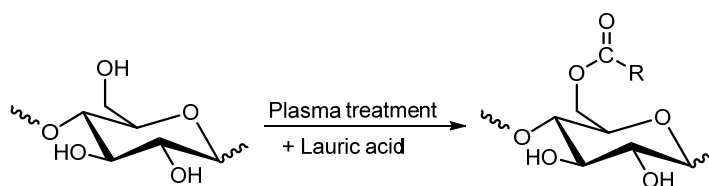
Sample	C (%)	O (%)	N (%)	Si (%)	Na (%)	C/O
MFC	64.6	35.0	0.4	-	-	1.84
p-MFC 1	65.8	32.7	0.2	1.2	0.1	2.01
p-MFC 2	65.9	32.9	0.2	1.0	-	2.01
p-MFC 3	66.6	32.9	-	0.4	-	2.01

C–O–C, and β -glycosidic linkage vibrations, respectively [27–28]. After plasma treatment, based on the normalized spectra with a peak at 1023 cm^{-1} , a new peak was observed at 1742 cm^{-1} in all variations, as seen in Fig 1(b). This new peak indicates the presence of ester's carbonyl (C=O) group, suggesting that esterification occurred between the OH groups in cellulose and COOH in lauric acid. This was further supported by a decrease in OH intensity of modified MFC at $\sim 3400\text{ cm}^{-1}$ compared to that of pristine MFC. The decrease in the same case also occurred in other studies by Yu et al. [14] and Lease et al. [29]. The possible occurrence of esterification is also illustrated in Fig. 2.

It is important to note that side reactions, such as cellulose oxidation, can occur during plasma treatment, forming carboxylate functional groups [29–32]. However, in this study, such oxidation cannot be conclusively confirmed based on FTIR spectra. This is because the C=O stretching vibrations of carboxylic acids typically appear around 1700 cm^{-1} [33], which overlaps with the peak of ester's C=O, making it difficult to distinguish these two using FTIR alone. To ascertain this possibility, further surface analysis using XPS was conducted to detect the presence of oxygen-containing functional groups. Based on ATR-FTIR results, a low-pressure plasma technique using lauric acid can initiate the grafting of cellulose. In this study, lauric acid, the hydrophobic precursor, was introduced into the plasma chamber after the MFC had been activated, followed by another round of plasma treatment for post-grafting. A possible mechanism involves free radicals, where the

plasma reactive species (such as free radicals and ions) bombard the cellulose surface, activating its cellulose's OH groups. This activation increases the reactivity of these groups, allowing them to react chemically with lauric acid [17,34–35]. However, the absorbance intensity of the formed C=O is still very low. This is probably due to the abundance of OH groups in cellulose molecules. Additionally, the formation of radicals during the plasma process occurs randomly on both the polymer substrate and monomer, thus affecting the effectiveness of the grafting process [18]. The free radicals formed are highly reactive. Those formed on the substrate can recombine before reacting with the monomer. Similarly, radicals on the monomer can also recombine without interacting with the substrate, which reduces the grafting efficiency. However, this is not always viewed as a drawback, as the plasma technique focuses on altering the surface, conserving chemicals, and enabling environmentally friendly grafting (without an initiator). Therefore, further studies are needed to improve grafting effectiveness by optimizing plasma parameters, such as voltage, treatment duration, or type of carrier gas.

The degree of grafting was qualitatively estimated by normalizing the absorbance intensity of the peak of ester's C=O at 1742 cm^{-1} to the C–O–C absorption of the pyranose unit in the cellulose backbone at around $1023\text{--}1025\text{ cm}^{-1}$, which was unaffected during modification [36]. As the concentration of lauric acid increased to a molar ratio of 4:1 (lauric acid:AGU), the intensity of the ester's peak also increased, indicating a

**Fig 2.** Possible cellulose structures after plasma treatment esterified cellulose where R is lauric acid chain

higher degree of grafting. However, beyond this ratio, the intensity began to decrease, as shown in Fig. 1(b) and 1(c). This finding is supported by a related study by Panda et al. [16], which reported similar behavior when textile surfaces were modified using helium glow discharge plasma with dodecanoic acid. In their study, the efficiency of fabric functionalization initially increased with higher precursor concentrations but then decreased after reaching an optimal point. This likely occurred due to the saturation of the substrate surface or the formation of an uneven layer of the precursor.

XPS testing was performed to confirm the possible reactions that occurred. The wide-scan XPS spectra, as seen in Fig. 3 along with Table 2, provide information on the atomic composition of MFC surfaces before and after plasma treatment. Notably, there is an increase in the carbon-to-oxygen (C/O) atomic ratio, which supports the substitution of OH groups in the MFC surface. Other output about plasma-induced grafting results was revealed through high-resolution XPS C1s spectra, as

illustrated in Fig. 4 and Table 3. The high-resolution C1s spectrum, as shown in Fig. 4(a), indicates that the AGU of cellulose contains three carbon components, namely C1, C2, and C3, which possess binding energies of 285.0, 286.7, and 288.2 eV, respectively. C1 indicates a C–C bond,

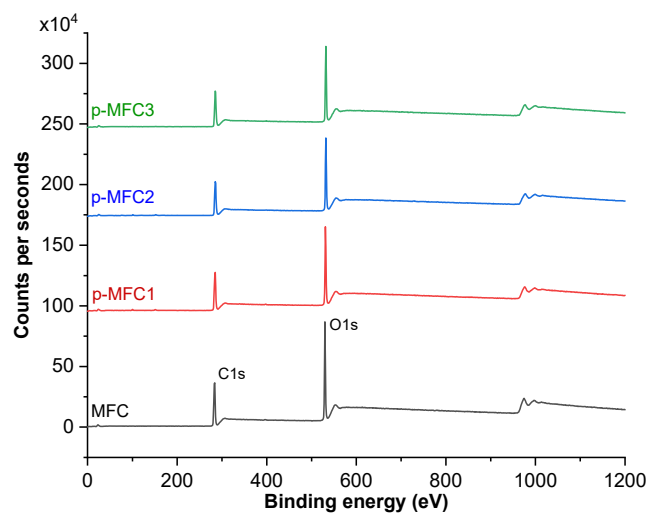


Fig 3. XPS spectra of the pristine and modified MFCs

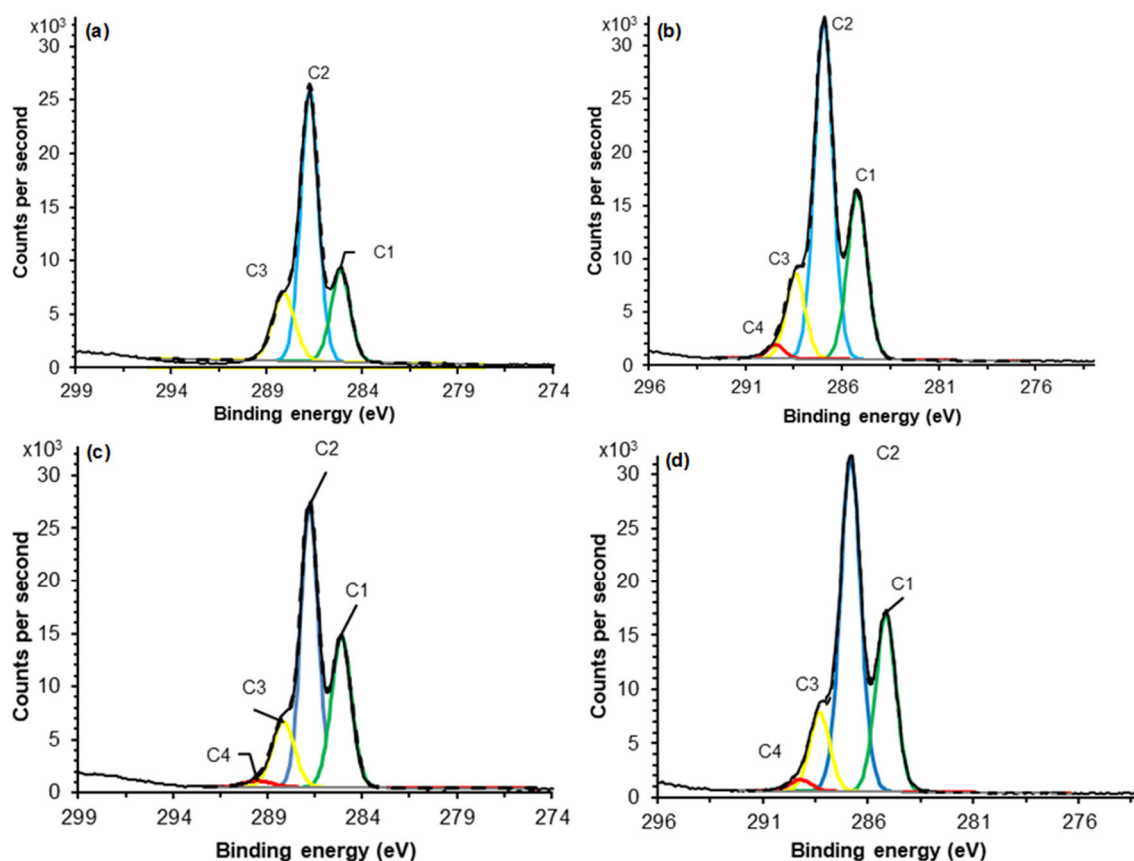


Fig 4. C1s spectra of (a) MFC, (b) p-MFC 1, (c) p-MFC 2, and (d) p-MFC 3

Table 3. Binding energy and C1s peak area of the pristine and modified MFCs

Carbon type	Binding energy (eV)	Sample			
		MFC	p-MFC 1	p-MFC 2	p-MFC 3
C1 (C–C/C–H)	285.0	22.11	27.66	31.31	29.40
C2 (C–O)	286.7	59.13	56.06	51.16	55.02
C3 (C=O/C–O–C)	288.2	18.75	13.96	15.46	13.36
C4 (O–C=O)	289.3	-	2.32	2.08	2.22

C2 indicates a C–O bond, and C3 indicates a C–O–C bond [15,37].

On the other hand, after plasma treatment (Fig. 4(b), 4(c), and 4(d)), MFC formed a new bond at a binding energy of 289.3 eV (C4 peak), indicating the presence of an O–C=O bond for esters or carboxylates. These results align with a previous study by Men et al. [15], who conducted esterification of cellulose with long-chain fatty acids using 1-butyl-3-methylimidazolium acetate. They reported that a new peak was formed at a binding energy of 289.2 eV, indicating the formation of an ester bond. Along with the emergence of the C4 peak, a decrease in the C2 and C3 peak areas and an increase in the C1 peak area were also observed, consistent with previous studies on the chemical esterification of cellulose fibers [21]. As a result, the hydrophilic nature of MFC will be reduced due to the chemical surface modification of cellulose, where the OH groups that contribute to its high hydrophilicity are replaced by esters, which will be able to increase the compatibility and adhesion of MFC with the synthetic or petroleum-based polymer matrix, which is usually hydrophobic.

The percentages of C4 peak area for p-MFC 1, p-MFC 2, and p-MFC 3 samples were 2.32, 2.08, and 2.22%, respectively. It is notable that these slight differences are still within the experimental margin of error, indicating that the O–C=O content among all modified MFC samples was relatively similar. These results differ from the trend intensity of ester's C=O observed in the FTIR spectra, as shown in Fig. 3. This discrepancy may be attributed to the overlapping binding energy of ester and carboxylate groups in the XPS analysis, which appears near 289 eV, as discussed previously. This supports the possibility that, in addition to esterification, other reactions, such as cellulose oxidation, also occurred during plasma treatment, forming carboxylic acid groups.

Surface Wettability

In fabricating polymer composites, the compatibility between filler and matrix is important for obtaining strong interfacial adhesion. PP has a low surface tension due to its hydrophobic nature, while cellulose has a higher surface tension due to its hydrophilic nature. The significant difference in surface tension between PP and cellulose can cause incompatibility and poor interfacial adhesion. Therefore, WCA measurement is important to determine the compatibility of MFC with polymer matrix. As previously stated, WCA higher than 90° indicates that the material surface tested is hydrophobic, and conversely, WCA lower than 90° signifies that the material surface tested is hydrophilic [23,38].

As shown in Fig. 5(a), after plasma treatment using lauric acid, the highest increase in WCA was generated in p-MFC 2 at 42.01%, followed by p-MFC 3 at 26.44% and p-MFC 1 at 19.28%. This increase in WCA indicates the success of cellulose surface modification. Besides, this increase is also in line with the percentage area of C1 in the high-resolution XPS C1s spectra, indicating an enhancement in C–C bonds due to lauric acid grafting. However, this result is inconsistent compared to the C4 area (O–C=O) in the high-resolution XPS C1s spectra, where p-MFC 1 and p-MFC 3 exhibit larger C4 areas than p-MFC 2. This is most likely due to side reactions that possibly occurred alongside esterification, particularly in the form of cellulose oxidation by plasma reactive species, which produced a carboxylic acids group (HO–C=O) that is hydrophilic compared to esters. This reasoning aligns with previous studies, which stated that cellulose would be further oxidized after being treated using air plasma from carboxylic acid functional groups [30,33]. These carboxylic acids have the same binding energy as esters (RO–C=O), making them difficult to

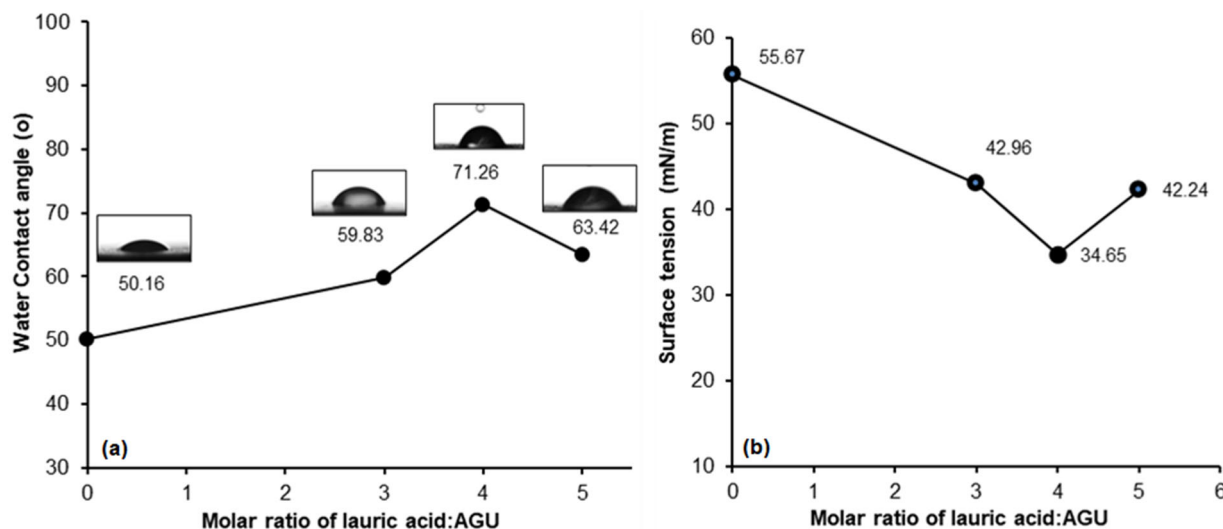


Fig 5. (a) Water contact angle and (b) surface tension of the pristine and modified MFCs

distinguish [39]. However, as previously discussed, the carboxylate peak in the FTIR spectrum at a wavenumber of $\sim 1700\text{ cm}^{-1}$ was not detected, possibly because it overlaps with the peak of ester's C=O. Consequently, the formation of carboxylates can affect the contact angle. p-MFC 1 and p-MFC 3 exhibited lower contact angles than p-MFC 2, even though their C4 areas were larger than that of p-MFC 2. This can occur because, apart from ester formation, p-MFC 1 and p-MFC 3 produced more carboxylic acids than that of p-MFC 2.

Fig. 5(b) illustrates the impact of lauric acid addition on the surface tension of MFC. A significant decrease in surface tension was observed following plasma treatment, with the lowest surface tension resulting in a molar ratio of 4:1 (lauric acid:AGU) in p-MFC 2, which showed a reduction of 37.75%. This was followed by p-MFC 3, with a reduction of 24.12%, and p-MFC 1, with a decrease of 22.83%. This trend corresponds with the increase in WCA, indicating a decrease in hydrophilic property, as previously explained. Regarding the use of MFC as a filler/reinforcement in polymer composites, especially with a hydrophobic matrix like PP, the difference in surface tension between PP and MFC becomes a critical factor. PP has a relatively low surface tension, 23.49 mN/m, while pristine MFC exhibits a significantly higher surface tension, 55.67 mN/m. This significant surface tension gap can lead to poor interfacial adhesion, which may result in unfavorable stress concentration

points and ultimately reduce the mechanical performance of the composite. Notably, the pristine MFC-PP mismatch was 136.99%, but this difference was significantly reduced to 47.51% in the p-MFC 2 sample. This reduction will improve compatibility between MFC and PP so as to enhance their interfacial adhesion and facilitate better stress transfer at the matrix-filler interface in the resulting composite.

Thermal Stability of the Pristine and Modified MFCs

This study used TGA to figure out the thermal stability of the pristine and modified MFCs, the results of which are shown in Fig. 6 and Table 4. Overall, plasma treatment of MFC using lauric acid as a hydrophobic precursor slightly increased the initial degradation temperature compared to that of pristine MFC, with the highest increase (3.8%) resulting in p-MFC 2. This increase in the initial degradation temperature is related to the increase in thermal stability. This finding is similar to a previous study by Wen et al. [13] who reported a rise in cellulose laurate ester's initial thermal degradation temperature because chains grafted onto the cellulose surface can form new structures that may be responsible for the increased thermal stability.

Thermal degradation of pristine MFC shows a single-step decomposition with a maximum temperature of $346.64\text{ }^{\circ}\text{C}$, as depicted in Fig. 6. Both p-MFC 1 and

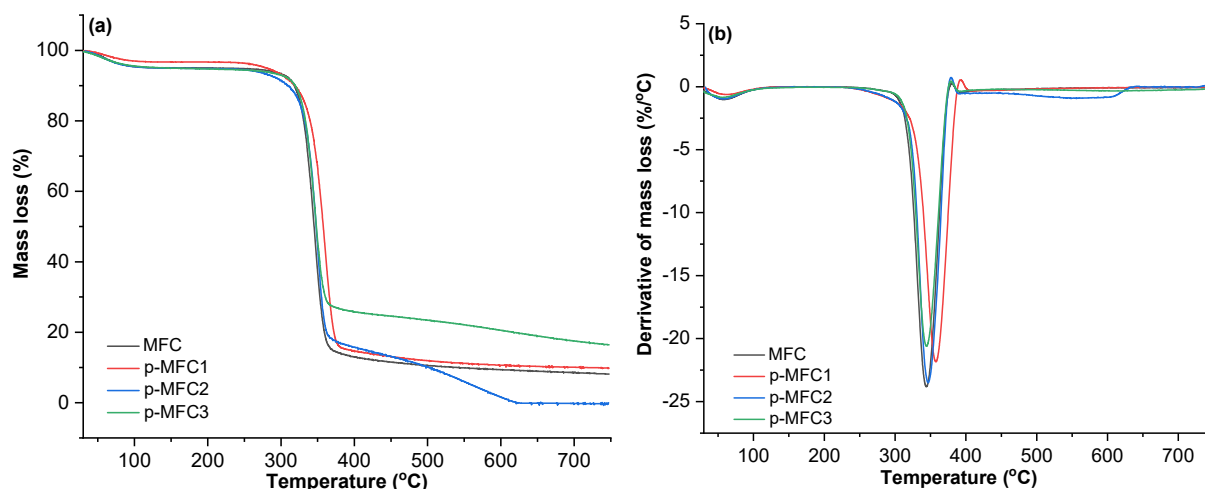


Fig 6. (a) TGA and (b) DTG thermograms of the pristine and modified MFCs

Table 4. Temperature degradation and mass loss of the pristine and modified MFCs

Sample	Initial degradation temperature (°C)	Degradation step				Char at 750 °C (%)
		1 st step		2 nd step		
		T _{max} (°C)	Mass loss (%)	T _{max} (°C)	Mass loss (%)	
MFC	326.5	346.64	86.37	-	-	8.28
p-MFC 1	333.0	360.39	86.86	-	-	9.97
p-MFC 2	339.0	351.16	80.37	571.96	14.06	1.02
p-MFC 3	329.2	347.60	78.16	-	-	16.71

p-MFC 3 exhibit the exact single-step decomposition. Still, there is an increase in the two's maximum decomposition temperature compared to that of pristine MFC. In p-MFC 2, there are two decomposition steps: the first occurred in a temperature range of 200–450 °C, and the second happened in a range of 450–700 °C. This finding aligns with a study by Jandura et al. [40] who noticed changes in thermal degradation characteristics after cellulose esterification, resulting in two distinct steps. The second decomposition peak may be attributed to the thermal degradation of a new cross-linked material formed through thermal cross-linking. Furthermore, differences were also noticed in the char formed through pyrolysis, which likely occurred due to differences in the resistant by-product of decomposition [13]. However, as previously mentioned, this phenomenon was only observed in the p-MFC 2 sample. This is presumably due to the grafting degree in p-MFC 2 being at an optimal level, as discussed in the FTIR analysis before.

CONCLUSION

The MFC surface has been successfully modified with lauric acid as the hydrophobic precursor to improve its compatibility with the polymer matrix for polymer composite applications. Surface modification was performed using a more environmentally friendly technique, cold plasma, as the alternative for lessening conventional wet chemical techniques. The FTIR and XPS results revealed chemical changes in the hydrophilic MFC surface. In addition to plasma-induced grafting to form esters, oxidation also occurred in MFCs, which affected their polarity. Optimum conditions were achieved at a lauric acid:AGU molar ratio 4:1 (p-MFC2). This treatment increased 42.01% in WCA compared to pristine MFC, indicating a decrease in its hydrophilic properties. Likewise, this treatment succeeded in reducing the surface tension of MFC; the resulting surface tension of p-MFC 2 is 34.65 mN/m, while that of pristine MFC is 55.67 mN/m. Therefore, compared to

the surface tension of PP, which is 23.49 mN/m, it is conclusive that there is a decrease in the difference in the surface tension between PP and modified MFC. This signifies the increasing compatibility between the matrix and the filler, thus increasing the interfacial adhesion. Moreover, this study's findings have provided information for future studies to optimize further plasma technique parameters, such as by employing various exposure durations, gas types, and power levels.

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■ CONFLICT OF INTEREST

The authors declare no conflicts of interest to report regarding the present study.

■ AUTHOR CONTRIBUTIONS

Mochamad Chalid contributed to the study's concept, data analysis, and supervision. Annisa Rifathin conducted the experimental design, experiment, characterization, data analysis, wrote the original manuscript, and revised the manuscript. Ade Mundari Wijaya conducted the experiment. Achmad Nandang Roziafanto, Joddy Arya Laksmo, and Adam Febriyanto Nugraha contributed to data analysis and supervision. All authors agreed to the final version of this manuscript.

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