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Research article

Effects of cold plasma treatment on the surface and properties of poly(lactic acid) fibers

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Abstract. This study examines the interface of biodegradable polylactic acid (PLA) fiber-reinforced thermoplastic starch composites to address market demand for their mechanical properties. We first used a combination of solution treatment (acid/alkali) and plasma modification to modify fibers. This method improves surface roughness and increases oxygen-containing groups of the fibers, which enhances the mechanical interlocking at the interface and promotes stronger hydrogen bonds between fibers and starch, respectively. Their synergy effect significantly strengthens interfacial adhesion and interfacial shear strength (IFSS). In contrast, cold plasma modification alone results in a smaller increase in IFSS because of its lower roughness and fewer oxygen-containing groups. Alkaline treatment and increased cold plasma power level are more conducive to higher IFSS through stronger synergistic effects. Although IFSS improved from 2.41 to 4.40 MPa after alkali and plasma treatments of 200 W, the tensile strength decreased from 811.46 to 351.55 MPa. To optimize the mechanical properties of composite materials, it is crucial to choose a kind of modification method that balances IFSS enhancement and tensile strength reduction.

Keywords: *PLA fiber, cold plasma treatment, tensile strength, surface morphology*

1. Introduction

Polylactic acid (PLA) fiber is a synthetic fiber known for its favorable mechanical properties and exceptional biocompatibility [1, 2]. It has the potential to substitute petroleum-based materials across diverse domains, making a significant contribution to enhancing the ecological environment [3]. The degradation of PLA fibers results in the emission of CO_2 and H_2O, generally considered non-toxic and harmless under normal environmental conditions [4–6]. PLA fiber, derived from renewable resources, is considered a sustainable material. The material has remarkable mechanical properties, processability, biocompatibility, and moisture absorption. This makes it a promising alternative to petroleum-based materials in various industries, thereby significantly contributing to environmental improvement [7, 8]. Particularly noteworthy is the frequent use of PLA fibers in the medical industry for applications such as heart stents [9], surgical sutures, gauze [10], tissue scaffolds, and artificial tissue implants [11, 12], thanks to their exceptional biocompatibility and non-toxic nature. PLA fibers possess inherent moisture conductivity, enabling rapid drying. Additionally, they exhibit outstanding resilience and wrinkle resistance, making them highly suitable for the development of sports fabrics [13].

Biodegradable thermoplastic starch is widely reported as a matrix for composites, but research on the interface of PLA fiber-reinforced thermoplastic starch

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composites is limited. Natural fibers are often favored over costly synthetic biodegradable fibers. The low-density functional groups on PLA fibers, including $–CH_3$, –C=O, –OH, and –COOH, impede hydrogen bond formation with the matrix, leading to poor interfacial bonding and inadequate mechanical properties. Thus, improving interfacial compatibility between PLA fibers and starch is necessary for enhancing composite performance.

One method is starch modification. Li *et al*. [14] introduced functional groups with chemical similarities to the ester groups in PLA molecules into starch molecules through a blending modification to enhance adhesion. Another method is fiber modification, which can be accomplished by ultraviolet (UV) grafting [15, 16], surface hydrolysis followed by reaction [17, 18], or plasma treatment [19, 20]. The UV grafting method utilizes a photoinitiator decomposed into $R^{\bullet}$ radicals to capture a hydrogen atom from the PLA backbone, forming reactive free radicals that can subsequently be grafted onto different substrates. The combination of surface hydrolysis and reaction involves the cleavage of ester bonds, which endows PLA chains with –COOH and –OH groups; these newly formed chemical groups can facilitate the coupling of additional functional groups. Plasma treatment entails the exposure of fiber surfaces to high-energy electrons and various active species, leading to etching effects and the introduction of polar groups. Among the three methods, surface modification of fibers by low-temperature plasma has emerged as a rapidly advancing area of research in recent years [21–24]. This technique offers several advantages, including high particle energy, near-ambient gas temperatures, mild reaction conditions, and less processing times. Furthermore, plasma treatment can significantly reduce damage to the material body and mitigate side reactions typically associated with high temperatures [25]. Donate *et al*. [26] employed plasma treatment on PLA fibers to enhance the hydrophilicity of the PLA surface while maintaining its performance. Mehboob *et al*. [27] examined the effect of exposing biological glass fibers to air plasma for 30 s on the tensile strength, flexural strength, and interfacial shear strength of the composite. The results demonstrated a significant increase of 31% in tensile strength, 13.5% in flexural strength, and 33% in interfacial shear strength. Hosseini *et al*. [28] used plasma treatment to modify the surface of PLA/ethyl cellulose blend films, increasing their surface roughness. Jordá-Vilaplana *et al*. [29] demonstrated an enhancement in the roughness of fibers and the amount of polar oxygen-containing groups on their surfaces through plasma technology. In contrast, UV grafting necessitates the use of initiators and grafting agents, and the reaction steps after surface hydrolysis also require the introduction of reactants, thereby increasing the complexity of the process. Furthermore, the use of surface hydrolysis methods can lead to damage to the fiber body and a reduction in tensile properties because the immersion time is long. To further improve the interfacial bonding between the fibers and the matrix, we intend to combine plasma modification with solution treatment. Short-term solution treatment endows the fibers with more oxygen-containing functional groups while only slightly damaging the fiber body. In addition, the slight hydrolysis of the fiber surface makes plasma modification easier to exert its effectiveness because the surface becomes more brittle.

In this paper, PLA fibers were modified through acid/alkali solution treatment followed by cold plasma modification at varying power levels. The surface morphology and chemical composition of the PLA fibers under different treatment methods were analyzed through weight changes, ATR-FTIR, SEM, and AFM. Their tensile strength and elongation at the break of the fibers were evaluated. Corn starch was used as a representative matrix, and the IFSS was tested through microsphere debonding experiments. These characterizations reveal the effects of surface morphology and chemical composition, which are influenced by various treatment conditions, on tensile strength and IFSS.

2. Experimental

2.1. Materials

PLA fiber (75D/72F PLA FDY) was purchased from Rixin Ecological Textile Garment Co., Ltd. (Xinxiang, China). Ethanol (C_2H_6O, ≥99.7%), sodium hydroxide (NaOH, ≥96%), hydrogen peroxide (H_2O_2, ≥30%), corn starch (≥98%), glycerol (≥99%), urea (≥99%), and sorbitol (≥98%) were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Deionized water was self-made in the laboratory.

2.2. Methods

2.2.1. PLA fibers pretreatment

PLA fibers were trimmed to approximately 10 cm in length and agitated using a self-made slingshot to ensure complete dispersion and contact with the water. Subsequently, the loose bundle of PLA fibers was immersed and stirred in absolute ethanol for 60 min, followed by a 30 min wash in an ultrasonic cleaner. Following the cleaning process, the PLA fibers were rinsed with deionized water and then dried in a heating air-blower at 35 °C. Once dry, the PLA fibers were placed in a sealed bag for preservation purposes.

2.2.2. Acidic or alkaline solutions treatment

The PLA fibers, from which the surface sizing agent had been removed, were submerged in a 5 wt% NaOH solution or a 5 wt% H_2O_2 solution with a fiber-to-solution mass ratio of 1:100. The sample was then subjected to a 30 min reaction at 40 °C in a constant temperature oscillation box. Subsequently, the PLA fibers were rinsed with deionized water until the fiber surface reached a neutral pH. Following the cleaning process, the samples were placed in a heating air-blowing drier and dried at 35 °C. The fibers on the solution-treated surface were modified using cold plasma.

2.2.3. Cold plasma modification

After solution treatment or not, cold plasma modification was carried out, and the schematic diagram of the device is shown in Figure 1. The PLA fibers were uniformly dispersed on the glass stage within the vacuum chamber. The vacuum pump pressure was maintained at a fixed value of approximately 10 Pa by using two electromagnetic vacuum pumps.

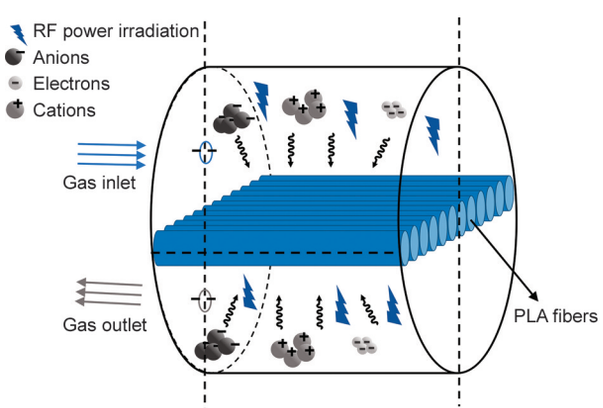

Figure 1. Cold plasma modification treatment device (HD-1A).

Throughout the plasma treatment process, the output power level was adjusted by controlling the RSG500RF generator to 100, 150, and 200 W, respectively. The glow discharge time was set to 3 min in the presence of air (20.9% O_2, 79.1% N_2). Following the completion of the treatment, the PLA fibers were retrieved, sealed, and dried for storage.

2.2.4. Preparation of corn starch/PLA fiber micro debonding samples

First, gelatinized starch was prepared by mixing plasticizer and corn starch in a 1:1 mass ratio. The plasticizer consists of glycerol, urea, and sorbitol in a mass ratio of 4:4:2. Using a rubber-tipped dropper to apply a small amount of the mixture onto PLA fiber filament. Continue this process until microspheres form around the filament. Place the sample in a constant temperature air-drying oven at 150 °C for 1 h to obtain a thermoplastic starch micro-debonding sample. Ensure that the PLA fibers used have undergone solution pretreatment and cold plasma modification at various power levels.

2.3. Characterization

2.3.1. Weight loss test

PLA samples were dried in a heating air-blowing drier at 35 °C for 120 min before the weight loss test. Equation (1) was used to calculate the weight loss of samples after cold plasma modification:

$$\text{Weight loss}\,[\%] = \frac{W_1 - W_2}{W_1} \cdot 100 \qquad (1)$$

where W_1 and W_2 are the weights of the dried samples before and after treatment.

2.3.2. ATR-FTIR spectroscopy

The ATR-FTIR spectra of PLA fibers was recorded to measure the changes in surface chemical properties of PLA fibers treated with different plasma power levels and different solutions. The spectra was performed using a Thermo Scientific Nicolet 6700 infrared spectrometer (Thermo Fisher Scientific, Waltham, MA, USA).

2.3.3. SEM analysis

The surface morphology of the PLA fibers before and after treatment was investigated by S-3400N (Hitachi, Chiyoda City, Japan) scanning electron microscope.

2.3.4. AFM analysis

The surface micromorphology and fiber surface roughness of PLA fibers before and after treatment were investigated by SPM-9700 (Shimadzu, Kyoto, Japan) atomic force microscope. The area at which the fiber surface is scanned by the probe is 2×2 μm.

2.3.5. Tensile measurements

The tensile properties of monofilament PLA fibers were tested according to GB/T 9997-1988, using YG004A single fiber strength tester (Changzhou, China), as shown in Figure 2. The clamping length of the tested fiber monofilament was 10 mm, the stretching speed was 2 mm/min, and the preset tension was 3 cN. Thirty valid tests were performed on each of the samples, and the force at break (marked as F_n) and the breaking elongation of the monofilament fiber (Marked as η_n) were acquired through a desktop computer interfaced with the testing machine. The average force at break (marked as F_c) of the monofilament fiber and the average breaking elongation (marked as η_c) of the monofilament fiber were obtained by calculating 30 groups of data. The tensile strength of the monofilament fiber was calculated by the Equation (2):

$$\sigma_b = \frac{F_c}{\frac{D^2\pi}{4}} \tag{2}$$

where F_c is the average breaking strength of the monofilament fiber, D is the average diameter of the PLA fibers, and σ_b is the tensile strength of the monofilament fiber.

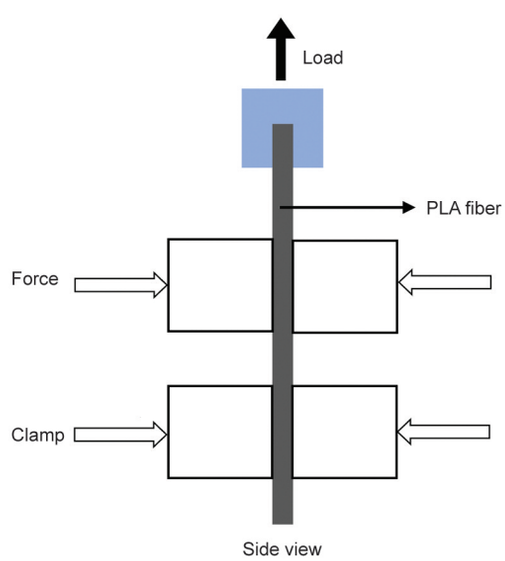

Figure 2. YG004A single fiber strength tester.

2.3.6. Interface shear performance

Using micro-debonding experiments, the effective data from 20 samples modified with cold plasma at different powers were measured. The average of these 20 data points was calculated to determine the IFSS of the samples, along with the standard deviation.

3. Results and discussion

3.1. Weight loss analysis of PLA fibers

As illustrated in Table 1, the presence or absence of solution treatment before cold plasma modification significantly influences the weight loss of PLA fibers. Following cold plasma modification, PLA fibers without solution treatment exhibit only minimal weight loss, whereas the weight loss of the solution-treated PLA fibers was markedly greater. This phenomenon can be attributed to the susceptibility of PLA molecules to hydrolysis in acidic or alkaline solutions, with the mechanism of the hydrolysis reaction depicted in Figure 3. Compared with an acidic solution (5 wt% H_2O_2), the weight loss of PLA fibers subjected to an alkaline solution treatment (5 wt% NaOH) is significantly greater. When the power level of the cold plasma is set at 200 W, the weight loss rate of PLA fibers treated with the alkaline solution is measured 2.010%. This is because PLA molecules exhibit higher sensitivity to hydrolysis in alkaline environments [30]. Hydroxyl attacks are easier to initiate than attacks from hydrated hydrogen

Table 1. Weight loss rate of PLA fibers treated with different power levels and different solutions.

Plasma power [W]	Solution	Weight loss [%]
100	–	0.095±0.011
150		0.136±0.015
200		0.168±0.013
100	5 wt% H_2O_2	0.511±0.011
150		0.762±0.012
200		0.971±0.021
100	5 wt% NaOH	1.343±0.014
150		1.771±0.019
200		2.010±0.017

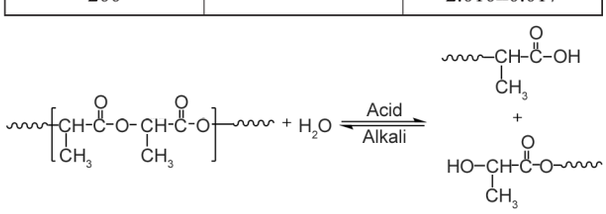

Figure 3. Hydrolysis process of PLA polymer.

ions under acidic conditions. The ether connected to the tetrahedral intermediate (CH_3CO–) causes hydrolysis of the PLA chain by generating oligomers with –OH and –COOH, resulting in chain scission and hence polymer degradation. When these segments lack entanglement or bonding with the segments of the fiber body, they are prone to detachment from the fiber surface, which consequently results in a reduction in fiber weight.

3.2. Analysis of the surface chemical composition of PLA fibers

The ATR-FTIR spectra of PLA fibers before and after the cold plasma treatment are shown in Figure 4. It was standardized by selecting the $–CH_3$ peak at 1450 cm^{-1} in untreated PLA fibers as the reference peak, based on the assumption that the methyl content remains unchanged before and after treatment. The intensity of various characteristic peaks after standardization is detailed in Table 2. The main characteristic peaks [31] of the PLA fibers include the C–O asymmetric vibration peaks at 1082 and 1180 cm^{-1}, as well as the C=O stretching vibration peak at 1750 cm^{-1}. The peak corresponding to the hydroxyl group at approximately 3500 cm^{-1} is relatively weak. As hydroxyl groups are primarily located at the terminus of PLA molecular chains, their content is relatively low. The spectral curve indicates that regardless of the cold plasma power level, the hydroxyl peak of the treated PLA fiber remains almost unchanged. As cold plasma treatment solely affects the surface of the PLA fiber, its effects do not penetrate deeper. Increasing the power of cold plasma processing without altering the processing time did not result in a significant change in the hydroxyl content on the fiber surface. As the cold plasma processing power increases, the peak transmittance of the C=O stretching vibration peak and the C–O asymmetric vibration peak slightly decreases.

Table 2. The intensity of various standardized characteristic peaks of fibers with cold plasma modification.

Power level [W]	Wavenumber [cm^{-1}]			
	1082	1180	1750	3500
0	48.086	54.088	53.896	97.923
100	42.668	49.862	48.091	97.936
150	42.361	48.517	47.169	98.115
200	41.335	47.902	46.427	97.998

The FTIR spectrum of PLA fibers treated with H_2O_2 or NaOH solution followed by cold plasma modification is presented in Figure 5. The standardization process employed is consistent with the previous methodology, and the intensity of various characteristic peaks after standardization is detailed in Table 3. Since the PLA molecular chain can undergo a hydrolysis reaction with acid or alkali in the aqueous solution, the content of C=O, C–O and –OH on the fiber surface is higher compared to PLA fibers treated with cold plasma alone. Figure 5 illustrates that the most prominent alteration in the FTIR spectrum occurs in the sample treated with NaOH solution treatment followed by cold plasma modification of 200 W. Due to

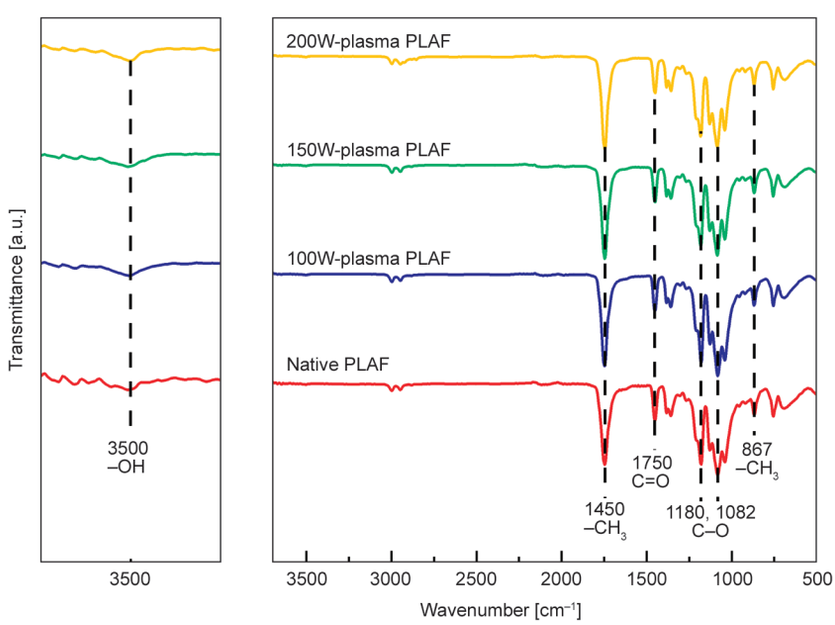

Figure 4. FTIR spectra of PLA fibers treated with cold plasma of different power levels.

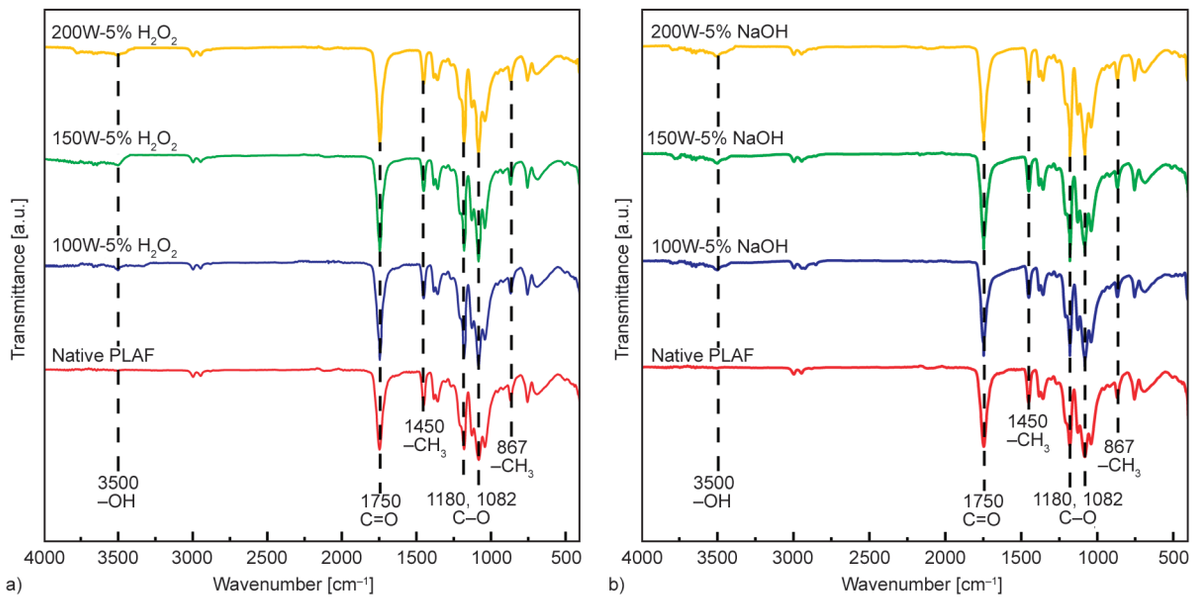

Figure 5. The FTIR spectra of PLA fibers treated with a) H_2O_2 solution and b) NaOH solution followed by cold plasma modification of different power levels.

Table 3. The intensity of various standardized characteristic peaks of fibers after solution treatment and cold plasma modification.

Power level [W]	Wavenumber [cm^{-1}]							
	5 wt% NaOH				5 wt% H_2O_2			
	1082	1180	1750	3500	1082	1180	1750	3500
100	38.869	45.862	45.352	92.239	39.624	45.983	46.187	94.368
150	38.165	45.517	45.224	92.311	39.151	45.379	46.245	94.112
200	37.511	45.902	44.865	92.465	39.186	45.678	45.141	93.587

its susceptibility to hydrolysis reactions, PLA fiber exhibits greater reactivity in alkaline solutions [30]. In acidic solutions, the presence of abundant hydrogen ions leads to the gradual attainment of reaction equilibrium as the reaction progresses.

In conclusion, the combination of solution treatment and plasma modification offers greater benefits in enhancing the concentration of oxygen-containing functional groups on the fiber surface, as opposed to utilizing plasma modification in isolation.

3.3. Surface morphology of PLA fibers

Figure 6 illustrates the surface morphology of the PLA fibers before and after treatment. Upon subjecting the PLA fiber to a plasma modification with a 100 W power level (Figure 6b), the surface displayed shallow, thin, and irregular pores. Increasing the plasma power to 150 W (Figure 6c) resulted in more numerous and larger pores on the PLA fiber surface. Elevating the plasma power level to 200 W (Figure 6d) resulted in the uniform distribution of small, dense holes across the surface of the PLA fiber. Additionally, these holes enlarged, deepened, and even coalesced to form a single rectangular crack. This phenomenon originates from the interaction between fibers and electrons. In plasma, fibers are continuously subjected to electrons with energies ranging from 5 to 10 eV, which induces the breaking of C–C and C–H bonds within polymer chains [32]. As the power level increases, a greater number of electrons collide with the fibers, leading to an increased detachment of segments within the same time frame, thereby resulting in more significant surface damage to the fibers. When the accumulation of detached segments reaches a critical threshold, the formation of cracks occurs. Figure 6e and Figure 6f demonstrate that, in comparison to cold plasma modification alone, the application of 200 W plasma modification following treatment with acid/alkali solutions increases crack sizes. This is because the interaction between fibers and electrons is influenced not only by the power level but also by the inherent properties of the fibers themselves. Following treatment with acid/alkali solutions, hydrolysis leads to

the degradation of polymer chains on the fiber surfaces, resulting in a loosening of the entanglement between these chains and a reduction in covalent bonding. Consequently, the weakened points of interaction are broken under subsequent electronic action, leading to the detachment of chain segments in fragmented pieces. Furthermore, the PLA fibers treated with an alkaline solution (Figure 6f) have a more pronounced degree of surface etching, an increased crack size, and a more irregular and complex surface morphology, which results from the fact that PLA chains are more easily degraded in alkaline media than in acidic media [30].

To analyze changes in PLA fiber morphology induced by various treatments, AFM was employed to examine alterations in the micro morphology and

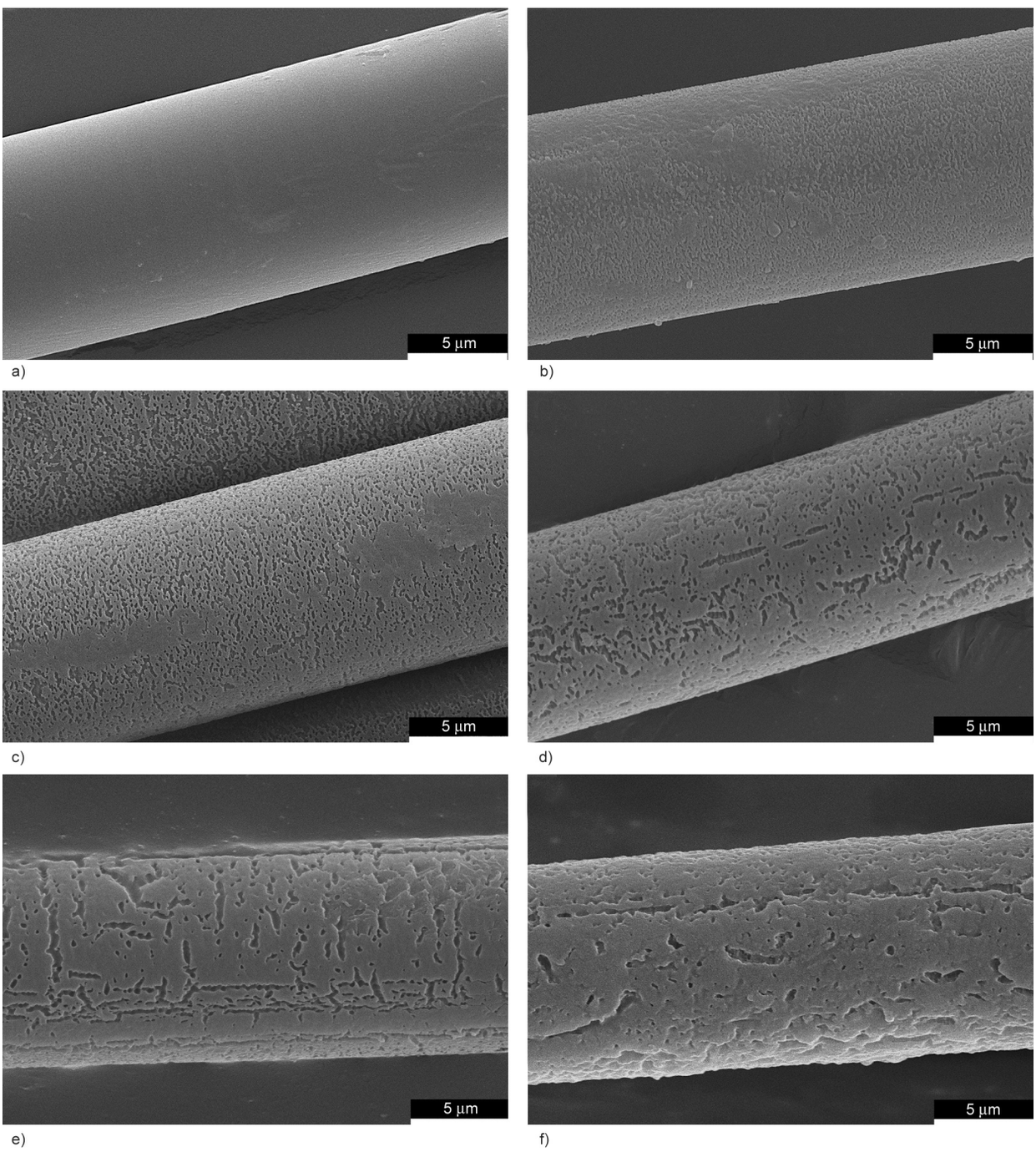

Figure 6. SEM images of surface morphology of PLA fibers with different treatments; a) untreated PLA fibers, b) PLA fibers modified with 100 W plasma, c) PLA fibers modified with 150 W plasma, d) PLA fibers modified with 200 W plasma, e) PLA fibers modified with 200 W plasma after pretreatment with H_2O_2, f) PLA fibers modified with 200 W plasma after pretreatment with NaOH.

roughness of the PLA fibers (Figure 7). The roughness is quantified using root mean square R_q. A comparison of Figure 7a and Figure 7b reveals that a 100 W plasma modification rendered the surface of the PLA fiber slightly rougher. As illustrated in Figure 7c, a 150 W plasma treatment reduced the number of projections and depressions present on the surface of the PLA fiber. Figure 7d depicts that elevating the processing power to 200 W resulted in prominent ripples on the surface of the PLA fiber, displaying an even wavy texture. Compared with plasma treatment alone, the significantly increased roughness (Table 4) demonstrates that PLA fibers treated with acid or alkali solutions before cold

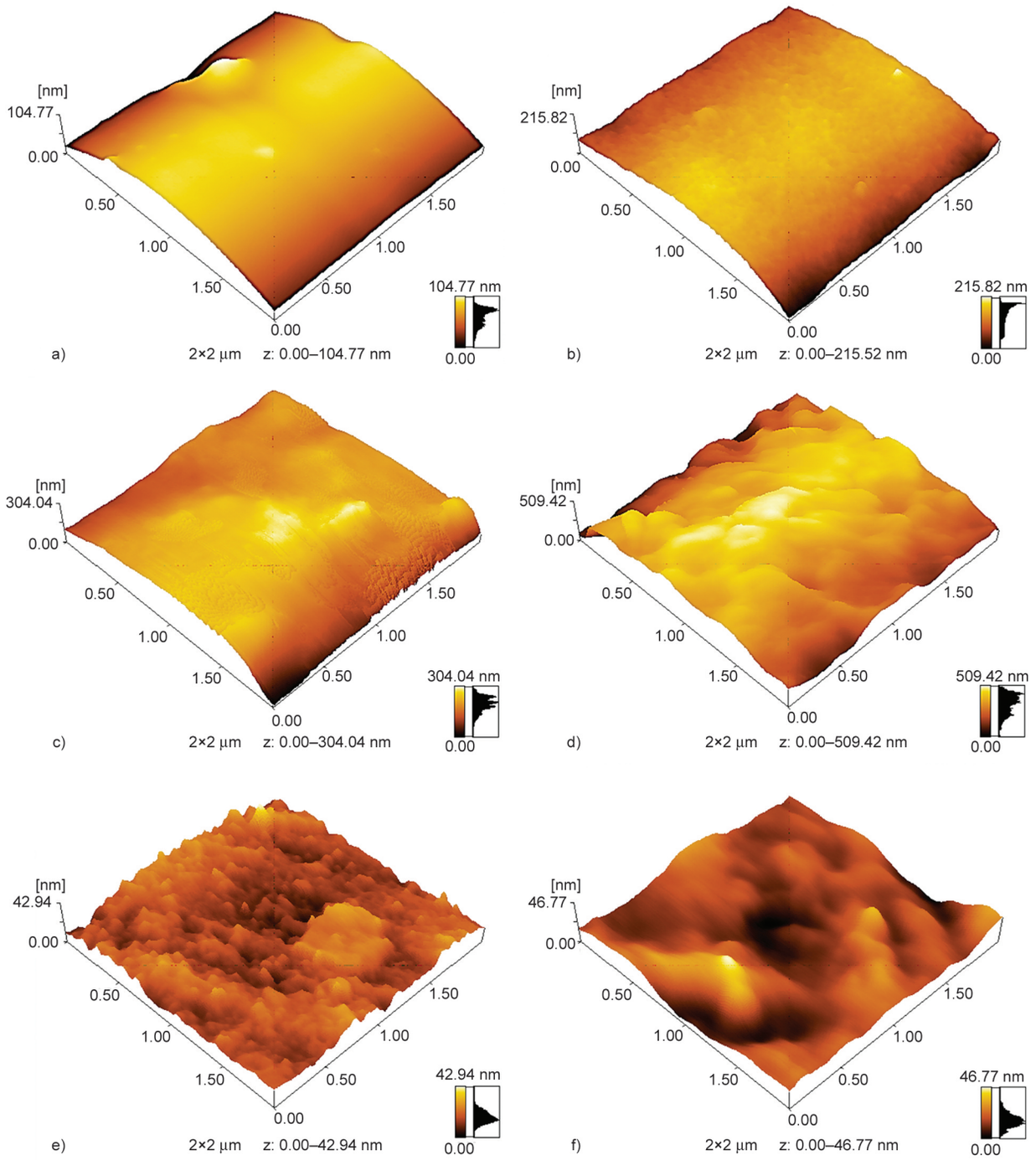

Figure 7. AFM images of surface micromorphology of PLA fibers with different treatments; a) untreated PLA fibers, b) PLA fibers modified with 100 W plasma, c) PLA fibers modified with 150 W plasma, d) PLA fibers modified with 200 W plasma, e) PLA fibers modified with 200 W plasma after treatment with H_2O_2, f) PLA fibers modified with 200 W plasma after treatment with NaOH.

Table 4. Root mean square R_q of PLA fibers with different treatments.

	Native	100 W	150 W	200 W	200W-5% H_2O_2	200W-5% NaOH
R_q [nm]	0.211	0.236	0.354	0.763	4.870	6.713

plasma modification possess a rougher morphology. Furthermore, fibers subjected to alkaline treatment exhibit greater roughness (6.413 > 4.870).

3.4. PLA fibers tensile strength

In Figure 8a, the initial tensile strength of the PLA fiber is 811.46 MPa. With increasing cold plasma power level, the tensile strength of the treated PLA fiber monofilament decreases after cold plasma treatment. A one-way analysis of variance indicates that cold plasma modification can cause significant damage to fibers. Cold plasma treatment induces the impact of high-energy particles on the fiber surface, leading to the formation of varying degrees of pits and faults. The application of external tension to the fibers causes stress concentration, increasing their susceptibility to fracture and ultimately leading to a decrease in the average tensile strength of the fibers. Nevertheless, in comparison to the combined effects of solution treatment and plasma modification, the observed damage is minimal, resulting in a relatively minor reduction in tensile strength.

Figure 8b and Figure 8c illustrate the tensile strength and elongation at break of the fiber monofilament after treatment with different solutions, followed by plasma modification at varying power levels. Solution treatment reduces tensile strength, especially when treated with a 5 wt% NaOH solution. The greater decrease in tensile strength can be attributed to the ester hydrolysis reaction occurring on the surface of PLA fibers in an acid/alkaline solution. Mineral cavities and defects form on the surface, accompanied by relatively deep surface etching. Consequently, the mechanical properties of the fibers decline, and this effect becomes more prominent with increasing plasma power. A 5 wt% H_2O_2 solution treatment before cold plasma modification causes fiber surface hydrolysis in an acidic solution, albeit

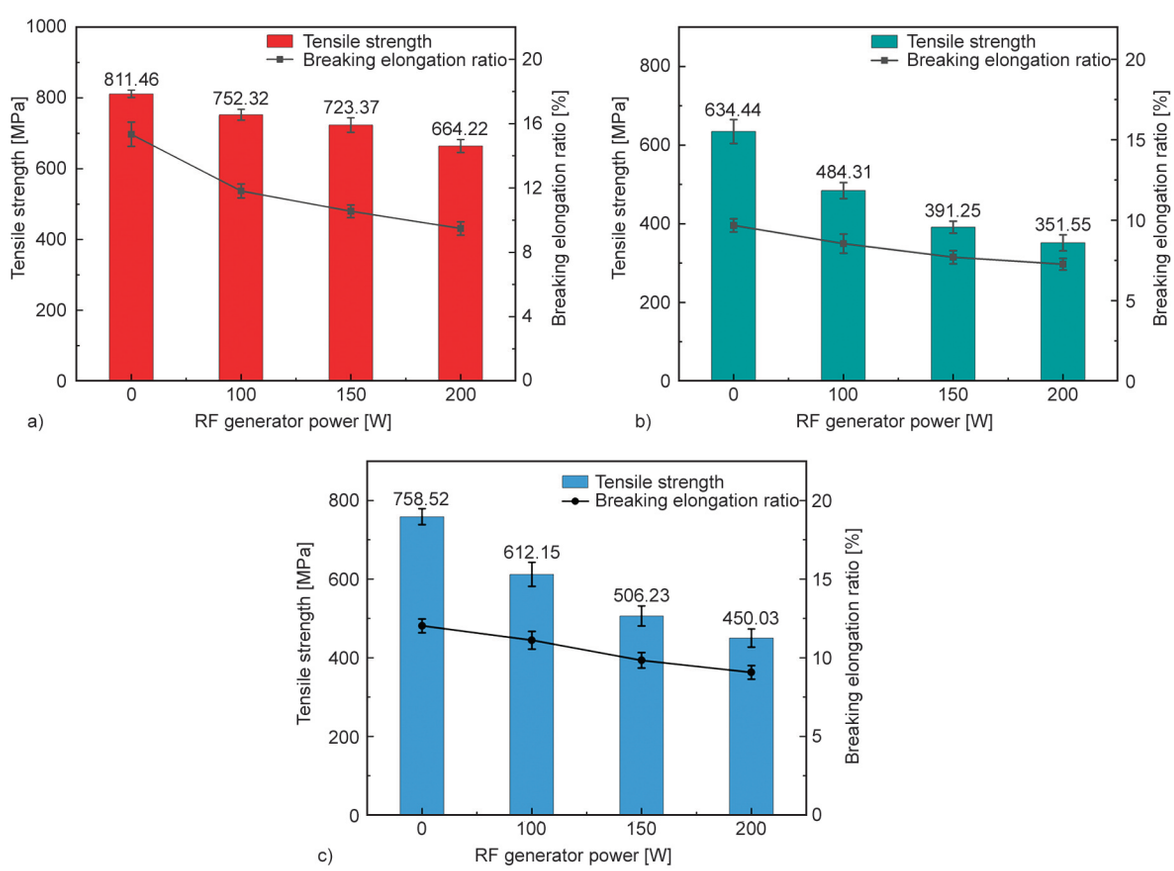

Figure 8. The tensile strength and elongation at break of PLA fibers treated with a) untreated, b) H_2O_2 solution, and c) NaOH solution.

to a lesser extent compared to an alkaline solution. Consequently, the mechanical properties of the fibers experience a less pronounced decline.

3.5. Interface properties of PLA fiber/corn starch

The IFSS between cold plasma-modified PLA fiber and corn starch is shown in Figure 9a. After micro-debonding experiments, the typical morphology of the starch resin micro-debonding area on the fiber surface is illustrated in Figure 9b. Only a small amount of starch resin particles remains on PLA fibers, indicating complete adhesive failure between the PLA fibers and the thermoplastic starch resin matrix. The IFSS between non-modified PLA fibers and corn starch is the lowest at only 2.41 MPa. Due to the smooth surface and low number of active groups on PLA fibers, they exhibit strong inertness, preventing mechanical interlocking or chemical bonding with the resin. When the PLA fiber surface is modified by cold plasma, the IFSS improves significantly. After cold plasma modification at power levels of 100, 150, and 200 W, the IFSS of the fibers reach 3.71, 4.01, and 4.40 MPa, respectively. This improvement is attributed to the introduction of numerous oxygen-containing active groups (C=O, –OH, –COOH) on the fiber surface post-modification. These functional groups form hydrogen bonds with hydroxyl groups in the corn starch resin, enhancing the interfacial bonding. Additionally, the high-energy particles during plasma modification create irregular protrusions and depressions on the fiber surface, significantly increasing surface roughness and mechanical interlocking with corn starch, thereby further improving the IFSS.

PLA fibers with only solution treatment do not significantly alter the IFSS between the fibers and corn starch. This is because the fiber surface only undergoes a minor hydrolysis reaction, leading to molecular chain breakage with minimal impact on surface morphology. PLA fibers treated with NaOH solution and subjected to plasma modification exhibit higher IFSS compared to those treated with deionized water and H_2O_2 solution. This is due to the more intense hydrolysis reaction induced by the base treatment, causing more PLA molecular chain breakage and destroying the surface structure of the fibers to a greater extent. Under the impact of high-energy particles in plasma, more fragile surface structures are more prone to peeling off, resulting in more irregular and complex surface morphology. A higher plasma power level intensifies this damage. Consequently, the friction coefficient and mechanical interlocking force at the PLA fiber-corn starch interface increase. Additionally, the specific surface area of PLA fibers and the content of oxygen-containing groups (–C=O, –OH, –COOH) significantly increase, enhancing the sites for hydrogen bond formation with corn starch and strengthening intermolecular interaction. These combined effects substantially improve the interfacial bonding and compatibility between PLA fibers and corn starch.

4. Conclusions

To address the market demand for the mechanical properties of biodegradable PLA fiber-reinforced

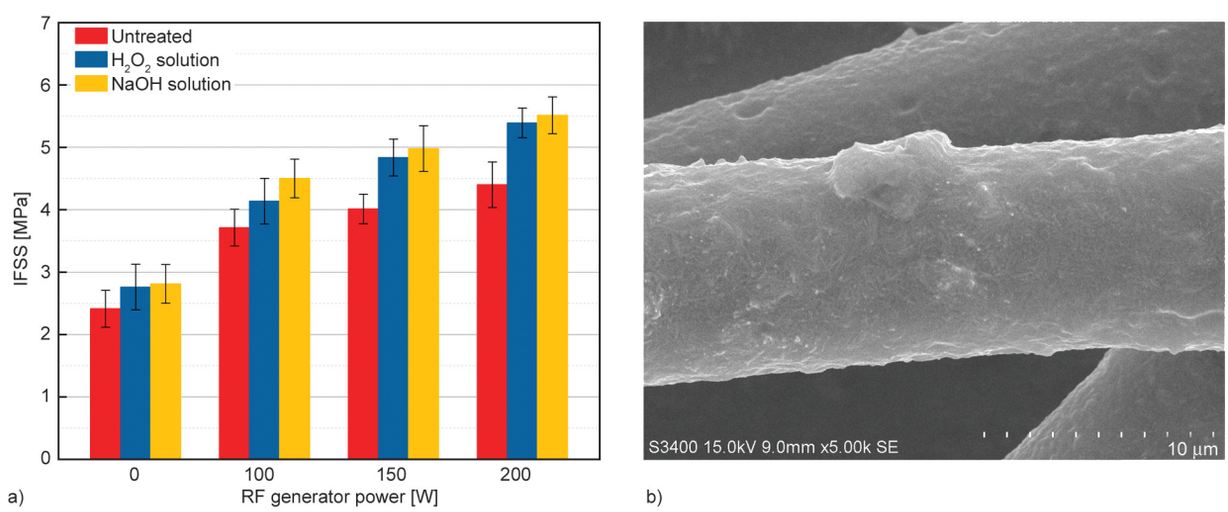

Figure 9. a) IFSS of PLA fiber/corn starch modified by cold plasma with different power levels after treatment of different solutions; b) the typical morphology of the starch resin microsphere debonding area after micro-debonding experiments.

thermoplastic starch composites, this study investigates the material's interface. The surface modification of PLA fibers was carried out by combining acid (H_2O_2 solution) or alkali (NaOH solution) treatment with cold plasma modification at varying power levels. The application of this method, while damaging the mechanical properties of PLA fibers, effectively improves surface roughness and increases oxygen-containing groups of the fibers, which enhances the mechanical interlocking at the interface and promotes stronger hydrogen bonds between fibers and starch, respectively. Their synergy effect significantly strengthens interfacial adhesion and IFSS. In contrast, although the use of cold plasma modification alone has a smaller impact on the tensile strength of PLA fibers, its lower roughness and fewer oxygen-containing groups result in a smaller increase in IFSS. In addition, alkaline treatment and higher cold plasma power levels have a more positive impact on IFSS due to their stronger synergistic effect. Without any modification of PLA fibers, the IFSS is only 2.41 MPa, and the tensile strength is 811.46 MPa. After alkali treatment and plasma modification of 200 W, the IFSS increased to its maximum value of 4.40 MPa; however, the tensile strength decreased to 351.55 MPa. To optimize the mechanical properties of composites, it is crucial to compromise between the increase of IFSS and the decrease in tensile properties of the fibers. Therefore, further testing of composites is necessary. In summary, this study advances the practical production and application of low-cost, fully biodegradable materials, and at the same time, also offers valuable insights for interface research involving other fibers and matrices.

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