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Polyimide (PI) Composite Spunlace Nonwovens for Hygiene Material with Excellent Comfort and Antimicrobial Properties

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Abstract: Nonwoven fabrics with appropriate hydrophilicity and potent antimicrobial properties hold important promise for hygiene applications. However, existing materials with certain limitations and complex manufacturing steps, along with the unavoidable use of chemicals in the process, are limited to a certain extent in terms of the balance between comfort and antimicrobial properties. In this paper, a polyimide (PI) fiber was reported to be used for the preparation of PI composite nonwoven fabrics (5-P), which can effectively enhance the surface hydrodynamic and antimicrobial properties of the nonwoven by a one-step plasma treatment on one side. After treatment, the one-sided water contact angle (WCA) changed from 121.5° to 68.5°, and the permeation volume from 0.7 to 2.1 g, with a relative increase of 181.9%. Meanwhile, the reverse osmosis amount was only 0.5 g, achieving rapid permeation while keeping a low reverse osmosis amount. The antimicrobial experiment showed that plasma-treated 5-P exhibited 64.3% and 91.6% inhibitory properties against Escherichia coli and Staphylococcus aureus, respectively. Notably, the production process of antimicrobial 5-P was fast and efficient without the addition of any chemicals. This method has great potential for the industrial preparation of antimicrobial comfort materials on a large scale, which is competitive in the medical, sanitary materials, and personal care fields.

Keywords: polyimide; spunlace nonwovens; air plasma treatment; facing materials; antibacterial

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1. Introduction

Hygienic surface materials are critical to modern medical [1], health protection, and lifestyle products. In order to provide users with a more comfortable and safer experience [2], these materials need to possess a series of critical properties, including water absorbency [3,4], the ability to stay dry and clean [5], and antimicrobial properties [6], which are essential for preventing moisture build-up and the growth and spread of bacterial pathogens. To this end, researchers often conduct a wide range of studies and innovations in the addition of antimicrobial substances to materials, including studies on antimicrobial nanoparticles [7], antimicrobial polymers [8], natural antimicrobial plant extracts [9], and antimicrobial drug encapsulation. Recently, many studies have been conducted on the addition of antimicrobial substances. Wu et al. [10] loaded Ag+ and tannic acid (TA) onto cotton fabrics by esterification, which effectively improved the antimicrobial properties and reduced the cytotoxicity of TA. Wang et al. [11] used chitosan-grafted bacterial cellulose (BC-CS) as a hydrophilic agent sprayed on PP nonwoven fabrics to achieve a uniform distribution across the nonwoven thickness gradient, achieving the effect of improving hydrophilicity while having excellent antimicrobial properties against *S. aureus* and *E. coli*. Iqbal et al. [12] added peppermint extract to viscose fibers, resulting in 100% viscose fabrics with 97% and 94% inhibitory properties against *E. coli* and *S. aureus*, respectively. Agrawal et al. [13] reported a facile synthesis technique for the fabrication of superhydrophobic antibacterial fabrics by employing fluorine-free silane coupling agents as cross-linkers for

enhanced durability and antimicrobial effect. Lian et al. [14] extended the practical application of EOs to antibacterial surfaces and fabrics, which is promising for use in personal care products and medical settings.

However, the addition of antimicrobial substances carries some potential drawbacks [15]. Some commonly used antimicrobial compounds may have negative impacts on the environment, e.g., residues of antimicrobial agents may lead to resistance problems in microorganisms of the environment [16]. Long-term uses of antimicrobial substances may lead to the development of resistance in bacteria, posing a potential threat to public health. In addition, the antimicrobial properties of additive methods degrade over time, and the development of new fibers with stable antimicrobial properties is urgently necessary. Polyimide (PI) is a high-performance fiber with high strength, large modulus, heat resistance, chemical resistance, and super hydrophobicity, which has long been applied in leading-edge fields such as aerospace, fireproof materials [17,18], filter material [19,20], and so on [21]. Recently, it has been gradually discovered that PI fibers exhibit strong antimicrobial activity against both Gram-positive and Gram-negative bacteria. As a result, they have been recognized as an entirely new type of high-performance fiber containing inherent antibacterial capabilities. According to Yang et al. [22], PI fibers can be used as effective antibacterial wound dressings because of the specific groups on their surface that can directly damage bacteria's cell walls and have significant antibacterial activity against *E. coli* and *S. aureus*. It is considered to be the perfect antimicrobial material for surgical procedures as it inhibits bacterial growth and speeds up wound healing.

Polyimide fiber is an organic fiber with an imide ring in the main chain of the polymer, which has a high orientation and hydrophobic surface. Typically, hydrophilic elements must be added to the surface material [23], or the fabric is surface-treated to improve its hydrophilicity. A type of green, moisture-absorbing fiber known for its superior performance, lyocell combines the softness of silk with the durability of cotton [24]. It exhibits reduced shrinkage and higher toughness and modulus than viscose fiber, particularly when wet. The liquid conductivity of the composite fabric can be increased by combining lyocell and polyimide fibers [25]. For satisfying the characteristics of rapid penetration and low backflow of sanitary surface, it is necessary to further enhance the hydrophilicity of one side of nonwoven materials to achieve better use. For this feature, the advantages of plasma treatment as a fast and efficient method of fabric surface modification become apparent.

As a commonly used method for the surface modification of inert polymers [26], the plasma attack of high-energy particles can open the chemical structure of the fiber surface, making the free radicals combine with the broken chemical bonds to generate new polar groups, improving the surface activity of the fiber [27]. On the other hand, the impact of energetic particles can produce uneven defects on the polymer surface, thus changing the surface morphology of the fibers. Therefore, plasma treatment has important applications in the study of hydrophilic and hydrophobic modification of polymer surfaces and their properties [28]. Apart from other surface modification techniques, the plasma surface modification technique is one of the most effective and economical surface treatments, which will provide a pathway for designing novel materials, with enhanced physiochemical properties for emerging technologies. Liu et al. [29] focused on a comparative study of surface modification of polyethylene by parallel-field and cross-field atmospheric pressure plasma jets. The surface modification of a polyethylene (PE) sheet by the two types of atmospheric pressure plasma jet (APPJ) was comparatively studied. Results show that the two types of APPJ can improve the surface wettability of the PE sheet obviously through the comprehensive effect including plasma etching and inducing of hydrophilic groups. Kim et al. [30] investigated the pullout qualities of carbon fiber bundles from a cementitious matrix after the fibers had been changed using one of three plasma treatments: argon, nitrogen, or oxygen. The maximum interfacial shear strength for the argon plasma-treated sample can be explained by an increase in surface roughness without a reduction in mechanical strength, as well as a change in surface chemistry from hydrophobic to hydrophilic. Studies using both atmospheric and low-pressure plasmas are included.

In order to minimize the PA surface defects caused by nanomaterial incorporation and improve the membrane surface hydrophilicity for reverse osmosis (RO) application [31], hydrophilic acrylic acid (AA) was deposited onto the PA surface of TFN membrane using an environmentally friendly surface modification technique based on plasma-enhanced chemical vapor deposition (PECVD). It was then subjected to 15 s plasma deposition of AA monomer to establish an extremely thin hydrophilic layer atop a PA nanocomposite layer. Dhanumalayan et al. [32] demonstrated the effective hydrophilicity of polyvinylidene difluoride (PVDF), polyacrylonitrile (PAN), and NH2-functionalized multi-walled carbon nanotube (MWCNT) mixtures in a 5 min air plasma treatment for the modification. Said et al. [33] demonstrated in their work that the PECVD technique is a promising surface modification method that could be employed to rapidly improve membrane surface hydrophilicity (15 s) for the enhanced protein purification process without using any organic solvent during the plasma modification process.

In this paper, the plasma treatment of hydroentangled nonwoven fabrics with a 5/5 blend of PI and lyocell (5-P) had improved the liquid-conducting properties and increased the total comfort of the nonwoven fabrics. The performance of fast permeation and low reverse osmosis was achieved, with a certain exploration of the effect of plasma treatment on antimicrobial properties. Hydroentangled composite nonwovens were prepared by combining the natural antimicrobial fiber PI and the cellulosic fiber lyocell, and their surface morphology, surface elemental analysis, comfort properties and antimicrobial properties were characterized. This study provides a new idea and method for the development of high-level comfortable and antibacterial hygienic surface-layer materials, meanwhile providing a new reference for the enhancement of antimicrobial properties of hygienic materials.

2. Materials and Methods

2.1. Materials

PI fibers(1.67dtex·51 mm) and lyocell fibers (1.33dtex·38 mm) were purchased from Changchun Gao qi Polyimide Materials Co., Ltd. (Changchun, China) and Quanzhou Smartian Import&Export Trading Co., Ltd. (Quanzhou, China). Special standardized synthetic test solution was purchased from Yunfei Precision Equipment Co., Ltd. (Dongguan, China). Peptone from yeast and agar powder were purchased from Shanghai Adamas-beta Reagent Co., Ltd. (Shanghai, China). *Escherichia coli* (*E. coli*) ATCC 25922 and *Staphylococcus aureus* (*S. aureus*) ATCC 25923 were obtained from Shanghai LuWei Biotechnology Co., Ltd. (Shanghai, China). All chemicals were analytically pure and used without further purification.

2.2. Preparation

2.2.1. Preparation of PI Composite Spunlace Nonwoven Fabric

The PI fibers and lyocell fibers were opened and mixed uniformly in the proportions of 1/9, 2/8, 3/7, 4/6, and 5/5 by weight, respectively, and then fed into a single-string double-doffer flat-top carding machine, with the areal densities controlled at 60 ± 5 g/m^2 carding. After overlapping, the web composite was fed into a pilot hydroentanglement line (Fei long, Changshu, China) for two stages of hydroentanglement, as shown in Figure 1a. During hydroentanglement, the web was first prewetted by water jets with a low pressure (20 bar), and then hydroentangled under a high pressure (40 bar). The resultant composite spunlace nonwoven was dried under an infrared heater at 90 °C.

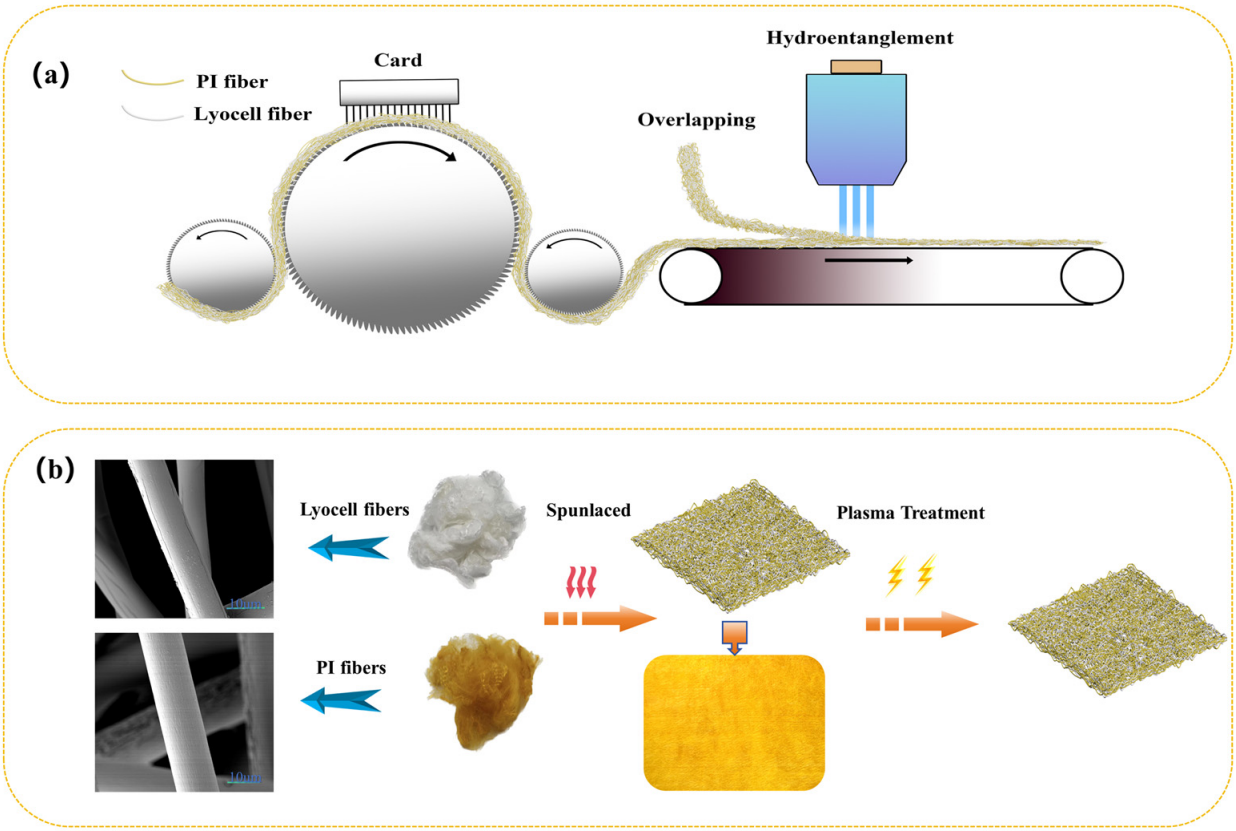

Figure 1. Schematic diagram of the preparation of PI/lyocell composite spunlace nonwoven fabric: (**a**) hydroentanglement process schematic and (**b**) plasma processing 5-P process diagram.

2.2.2. Plasma Treatment of 5-P

The plasma treatment on 5-P was carried out with a PSPT-2000C Atmospheric Pressure Glow Discharge Plasma Machine (Nanjing Presepe Electronic Technology Co., Ltd., Nanjing, China), air employed as both the carrier and reactive gases, operating under standard temperature (25 °C) and pressure conditions (101 kPa). Samples were placed on a quartz glass plate, with the treatment height set to 2 mm, the electrode moved at a speed of 40 mm/s, and exposed for 120 s at 60 W, 90 W, 120 W, and 150 W, respectively. The treated fabrics were stored for 24 h under standard circumstances (21 ± 1 °C, 65 ± 2% humidity) before being evaluated further.

2.3. Characterization

The surface morphology of the nonwoven was investigated using a scanning electron microscope (SEM, ZEISS, Oberkochen, Japan) after being coated with gold. Elemental analyses of the nonwoven samples were carried out using an Escola 250Xi X-ray photoelectron spectrometer (Thermos Scientific, Waltham, MA, USA). The maximum breaking strength of nonwoven fabrics was measured using an electronic tensile strength tester YG026HB (Quanzhou, China). The WCA of the sample was measured on a contact angle meter CA-100A (Shanghai, China). The air permeability of the nonwoven was evaluated on an air permeability tester YG461E (Quanzhou, China) based on ASTM D737 [34]. According to GB/T 30133 [35], the permeability properties and liquid penetration time of the nonwoven fabrics were tested on a sanitary napkin permeability tester CVOK-1040 (Dongguan, China) and a sanitary napkin absorption speed tester SGJ351A (Fuzhou, China).

2.4. Antibacterial Properties

Antibacterial tests were performed according to the standard GB/T15979-2002 method [36], and the tested strains were Escherichia coli (*E. coli*, ATCC 25922, Gram-negative) and Staphylo-

coccus aureus (*S. aureus*, ATCC 25923, Gram-positive). Prior to each experiment, the bacteria were placed in broth (LB) liquid nutrient medium (pH = 7.4) and incubated for 24 h at 37 °C. The composition of the liquid medium was 5 g/L yeast peptone and 10 g/L NaCl. Finally, bacteria were inoculated at standard density (108 CFU/mL). On this basis, bacterial tests were carried out for both strains. Block fabric samples (0.5 g) were introduced into a buffer solution (50 mL) containing 2×10^4–3.0×10^4 colony-forming units (CFU)/mL of bacteria, and then incubated for 18 h at 24 °C temperature with oscillation. Then, after making gradient dilutions, 100 µL of each solution was inoculated onto nutrient agar by the plate-coating method.

The colonies were counted after performing the antimicrobial test and the antimicrobial rate was calculated according to the following equation:

$$R = \frac{(A0 - A1)}{A0} \times 100\% \quad (1)$$

where $A0$ and $A1$ were the numbers of the bacterial colonies of the blank group and the samples. Each sample was measured three times.

3. Results

3.1. Preparation of PI Composite Spunlace Nonwoven Fabric

Since pure PI fibers generate static electricity during carding, the fibers are hard to card into a web, and the resulting nonwoven fabrics do not meet the specifications due to their inability to conduct liquids. Therefore, the lyocell fiber, which contains a large number of hydroxyl groups in its chemical structure and possesses fast moisture absorption and effective water retaining property, was selected to blend with PI fiber (Figure 1a).

Table 1 shows that PI composite hydroentangled nonwoven textiles with varied ratios have outstanding air permeability (>1500 mm·s^{-1}), but the maximum breaking strength does not change significantly. Figure 2 shows that when the proportion of PI fibers in the composite nonwoven fabric was lower than 40%, it was difficult to mix the two fibers evenly, making the final fabric surface uneven.

Table 1. Air permeability, tensile strength, and sample photo of PI/lyocell spunlace nonwoven with different blending ratio.

PI/lyocell Ratio	Air Permeability (mm·s^{-1})	Tensile Strength (N)
1/9	1806	30.24
2/8	1827	32.68
3/7	1839	35.36
4/6	1856	34.57
5/5	1906	35.58

Figure 2. Photos of hydroentangled nonwoven fabrics with PI/Lyocell ratios of 1/9, 2/8, 3/7, 4/6, and 5/5.

The penetration performance test of spunlace nonwoven is shown in Figure 3. It can be seen that with 50% PI content, the reverse osmosis of the nonwoven fabric is only 0.1 g. Considering the requirement of low reverse osmosis during the use of sanitary materials, the composite spunlace nonwoven with a PI/lyocell ratio of 5/5 was selected for the next step.

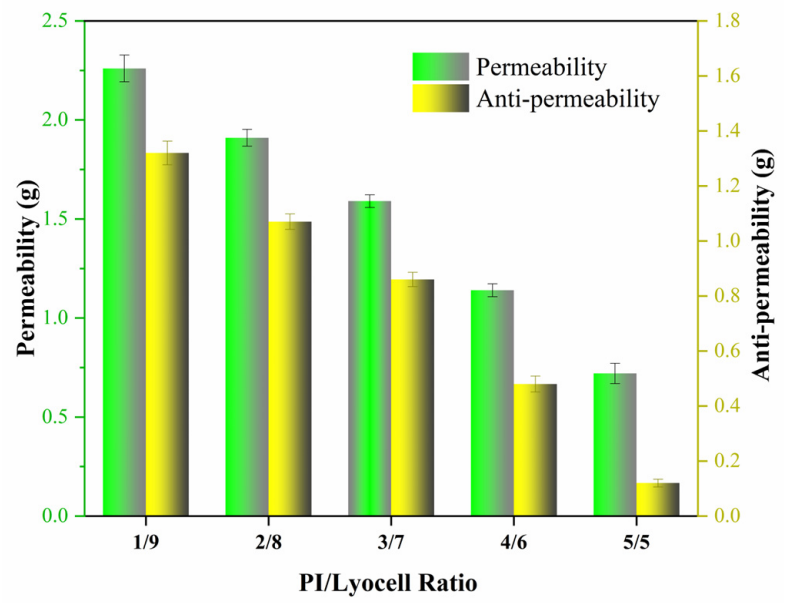

Figure 3. Permeability of PI/lyocell composite spunlace nonwoven fabric with different blending ratios.

3.2. Plasma Treatment of 5-P

To further improve the comprehensive performance of the nonwoven fabrics, one side of the 5-P was exposed to plasma treatments. The morphological changes in the fiber surface are shown in Figure 4.

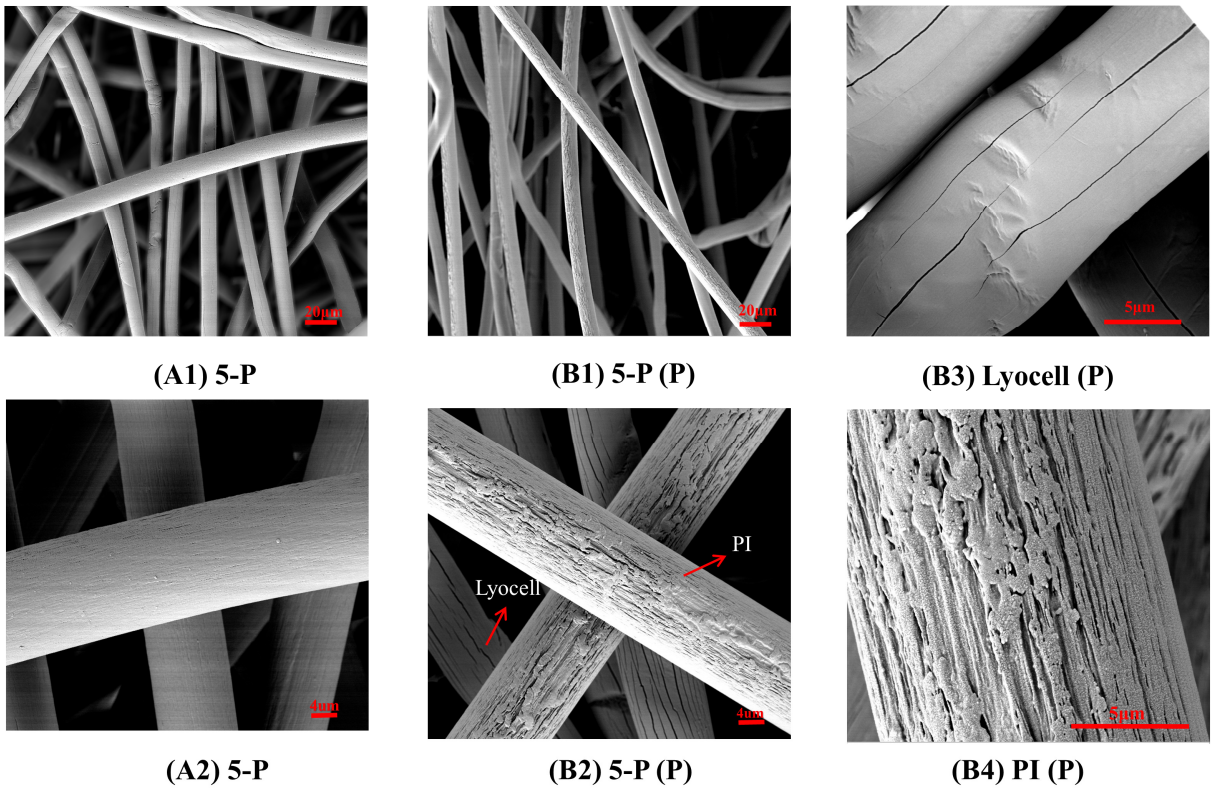

Figure 4. SEM photographs of spunlace nonwoven, (**A1**,**A2**) raw 5-P and (**B1**–**B4**) 5-P plasma after treatment.

As shown in Figure 4, the PI fibers have a diameter of 12 μm, while that of the lyocell fiber is 9 μm. The fiber surface is smooth and flat regardless of PI fibers and lyocell fibers. After the plasma treatment, it can be seen that the surface of the PI fiber was etched

with irregular defects (Figure 4(B4)), which increased the roughness of the surface of the nonwoven fiber. As for the lyocell fiber, there was no significant change in the surface morphology after the treatment, and only insignificant slight cracks existed on the surface (Figure 4(B3)). This is due to the fact that plasma is able to react on the surface of conductive polymers, thus affecting the surface of PI fibers, whereas lyocell fibers are cellulosic fibers that are not conductive, and therefore plasma is not able to affect lyocell fibers in any significant way.

3.3. XPS Analysis

XPS analysis was used to analyze the surface elements of 5-P before and after plasma treatment. Full spectral analysis of the fiber surface was performed to obtain the atomic fraction content of the C, N, and O elements.

The content of the three elements on the fiber surface was altered following treatment with atmospheric pressure air plasma 120 W for two minutes, as indicated in Table 2. The intensity of the N peak at 400 eV on the nonwoven fabric's surface is noticeably reduced, as seen in Figure 4A,B. The main reason for the drop in N content is that certain N atoms and N-containing groups were peeled off onto the fiber surface and into the air, causing damage to the N-containing crystal structure of the fiber surface caused by the attack of overwhelming plasma particles. The small decrease in oxygen content, from 21.1% to 19.04%, is the consequence of some oxygen atoms being sprayed from the fiber surface.

Table 2. C, N, and O atomic content of 5-P before and after plasma treatment.

	Element Content/%, Atomic Fraction		
	C	**N**	**O**
Untreated Sample	77.53	1.37	21.1
Treated Sample	80.15	0.81	19.04

The XPS spectra of nonwoven surfaces with and without plasma treatment exhibited changes in the composition of distinct functional groups by elemental peak mapping. As shown in Figure 5a, the C1s spectrum shows three peaks of -C-C-/-C=C-, -C-O-, and -O-C=O groups at 284.80 eV, 286.19 eV, and 288.65 eV, respectively. The content of the three groups on the untreated sample was 60.65%, 29.71%, and 9.64%, while the ratio of polar to non-polar groups was 0.65. After plasma treatment, the content of non-polar groups C-C-/-C=C- was reduced to 21.94%, while the content of polar groups -C-O- and -O-C=O was elevated to 65.42% and 12.64%, and the ratio of polar groups to non-polar groups was increased to 3.56. The reason for this is that the excited-state atoms, charged ions, and other energetic particles in the plasma interact with the free groups on the surface of the PI fibers to generate more polar groups. Polar groups can affect the charge balance inside and outside the bacterial cell, thereby disrupting the cell barrier and leading to cell inactivation. It is believed that a grafting reaction on the fiber surface caused by the plasma induces the significant decrease in the number of -C-C-/-C=C- groups [37]. This reaction is oxidized to produce oxygen-containing groups including -C-O- and -O-C=O, which is compatible with an immense rise in the content of -C-O- and -O-C=O groups after treatment. From Figure 5d N1s, it is shown that after plasma treatment, the main chain group, C(O)NH, in the PI fibers decreased from 61.92% to 30.06% and the content of NH_4^+ increased from 18.72% to 43.40%. This is due to the reaction of the C(O)NH groups under plasma treatment, splitting into -C=O, NH_4^+, and other oxides of nitrogen, which creates more positive charges on the fiber surface. The above results indicate that plasma treatment can introduce many reactive oxygen-containing groups and positively charged amino groups on the surface of PI fibers, thus improving the surface wettability and polarity of the fibers.

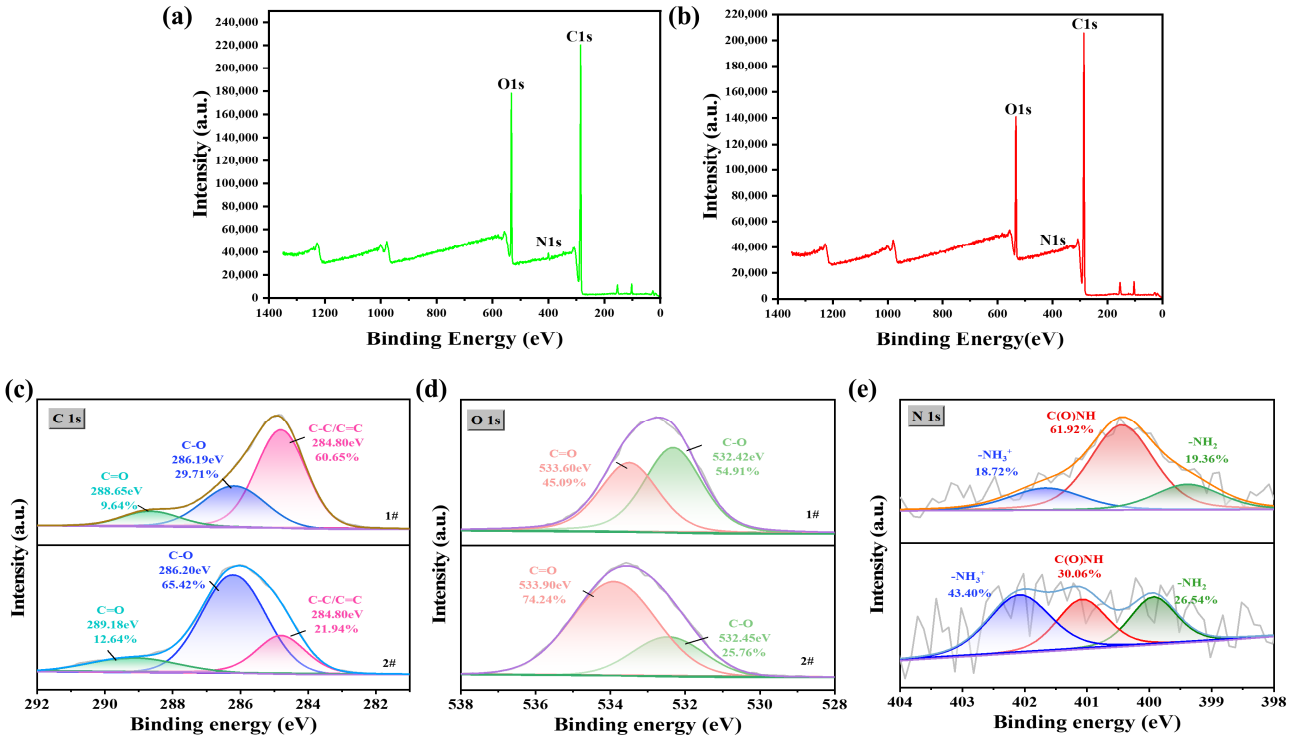

Figure 5. (**a**) XPS spectra of raw 5-P, (**b**) XPS spectra of plasma treatment 5-P, (**c**–**e**) C1s, N1s, and O1s spectra of 5-P before and after plasma treatment with deconvoluted peaks.

3.4. Comfort Properties

3.4.1. Air Permeability

Air permeability is an important indicator of hygienic materials, which affects the comfort of hygienic skin-fitting materials. The air permeability of 5-P was characterized at different plasma treatment powers and the results are shown in Figure 6.

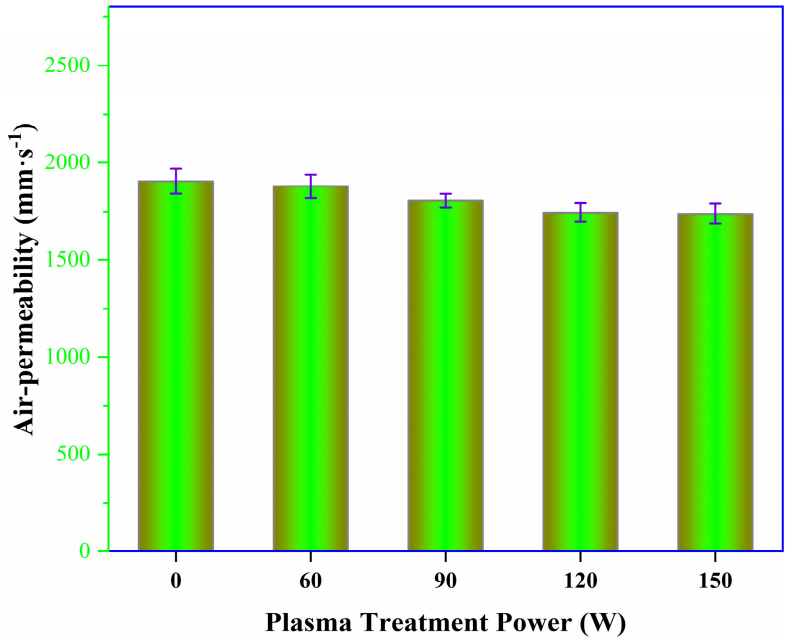

Figure 6. Air permeability of 5-P at different plasma treatment powers.

The results show that increasing the plasma treatment power had no significant effect on the air permeability of the nonwoven textiles. The air permeability of fabrics is mainly

related to the pore size and porosity of fibers, which do not change before and after plasma treatment. This is due to the plasma only working on the fibers' surfaces, eroding them and altering their surface morphology. The change in microscopic morphology of the fibers has no effect on the macroscopic properties of the total nonwoven fabric; hence, the overall air permeability of 5-P remains unchanged appreciably when exposed to plasma.

3.4.2. Liquid Conductivity

The increase in roughness improves the specific surface area of the fabric, which is the main reason for the increase in hydrophilicity. Following plasma treatment, surface channels were opened to allow for water transfer, as seen in Figure 7d. The amount of energy in the system grows with increasing treatment power, which leads to a growth in free radicals (oxygen-containing functional groups, such as aldehyde, carboxyl, carbonyl, ester groups, etc.). The fabric's wettability improves when more active groups are grafted onto the fiber surface. As can be seen from Figure 7a, after plasma treatment, the water contact angle (WCA) of 5-P changed from 121.5° to 68.5°, and the surface of nonwoven fabric changed from hydrophobic to hydrophilic, with significant improvement in hydrophilicity.

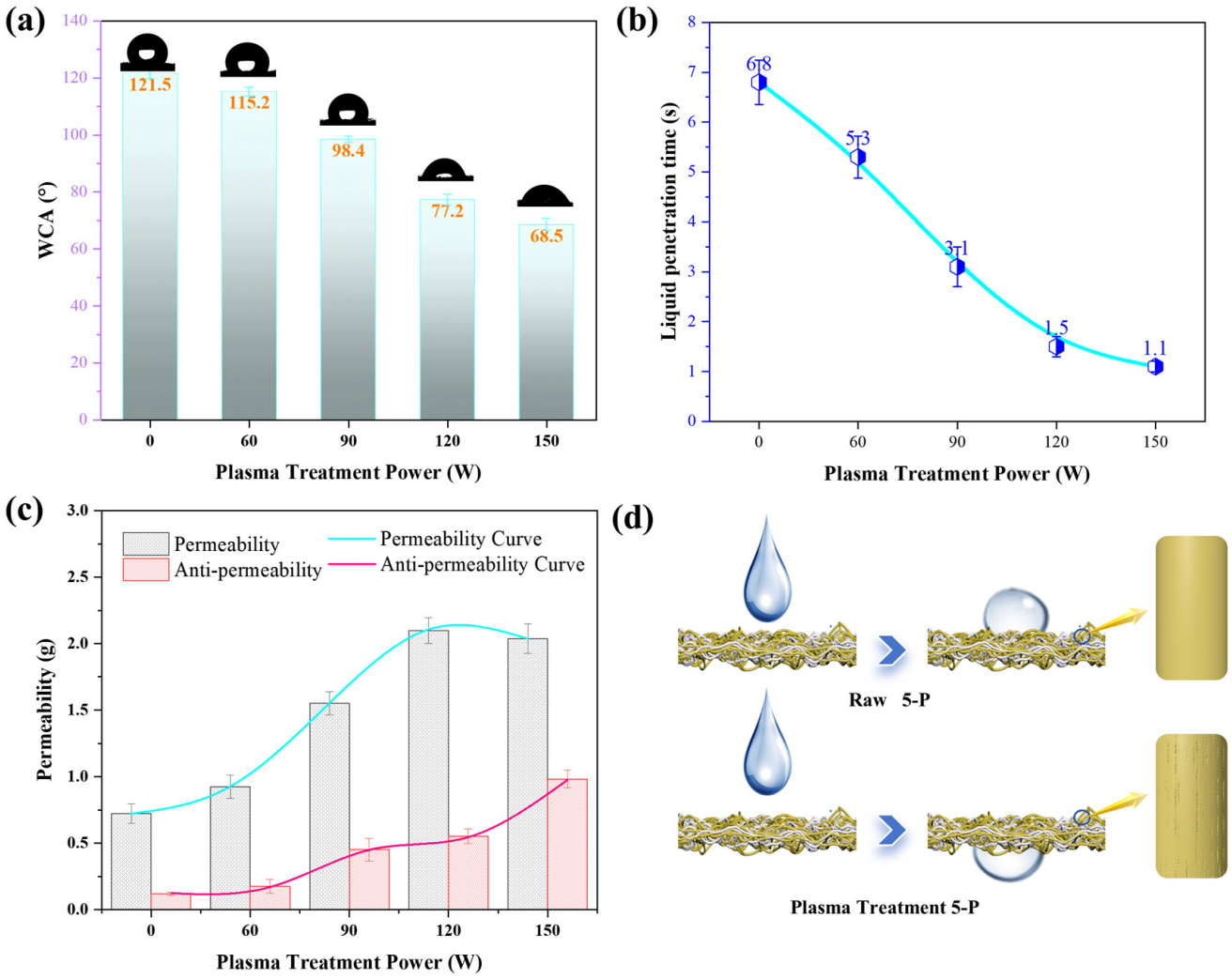

Figure 7. Effect of different plasma treatment power on (**a**) WCA, (**b**) liquid penetration time, and (**c**) permeability performance; (**d**) schematic of water conductivity of 5-P before and after plasma treatment.

Liquid penetration time is an essential factor for the liquid conductivity of sanitary materials, which directly affects the user's skin comfort. The results in Figure 7b show that after plasma treatment, the penetration time changed from 6.8 s to 1.1 s, achieving the effect of rapid liquid penetration and realizing the function of rapid liquid penetration. This result

is consistent with the change in WCA above, and this also benefited from the increase in the surface roughness of the nonwoven and the change in the number of oxygen-containing functional groups after plasma treatment.

The permeability of 5-P was analyzed as shown in Figure 7c. After plasma treatment, the permeability of the nonwoven fabrics was significantly improved, with a general trend of increasing and then decreasing, reaching a maximum value of 2.1 g at 120 W. This is due to the saturation of the fiber surface with the amount of grafting and etching, which is no longer evident as the treatment power continues to increase. Instead, the excessive build-up of etch points on the fiber surface causes the roughness to begin to decrease slightly, thus affecting the amount of liquid penetration. The amount of 5-P reverse osmosis did not change significantly before or after treatment; it was only 0.5 g at 120 W, as shown in Figure 7c. This is due to the plasma treatment only working on the surface of the fiber layer and being unable to penetrate deep into another untreated surface. The super-hydrophobicity of the PI fiber will prevent the liquid from flowing backwards when reverse osmosis occurs.

3.5. Mechanical Properties

The mechanical properties of 5-P were tested, and the results are shown in Figure 8. The tensile strength increased slightly from initial 36.5 N to 50.9 N. This was attributed to the increased roughness of the fiber surface after plasma treatment. The original two fibers hydroentangled formed a stable 3D structure. The increased fiber roughness reduced the slip effect and enhanced the inter-fiber adhesion [38]. As a result, the tensile strength of the nonwoven fabric was enhanced. As the processing power was increased to 120 W, the ion etching on the fiber surface reached its peak. Under continued increase in power, the tensile properties no longer changed significantly.

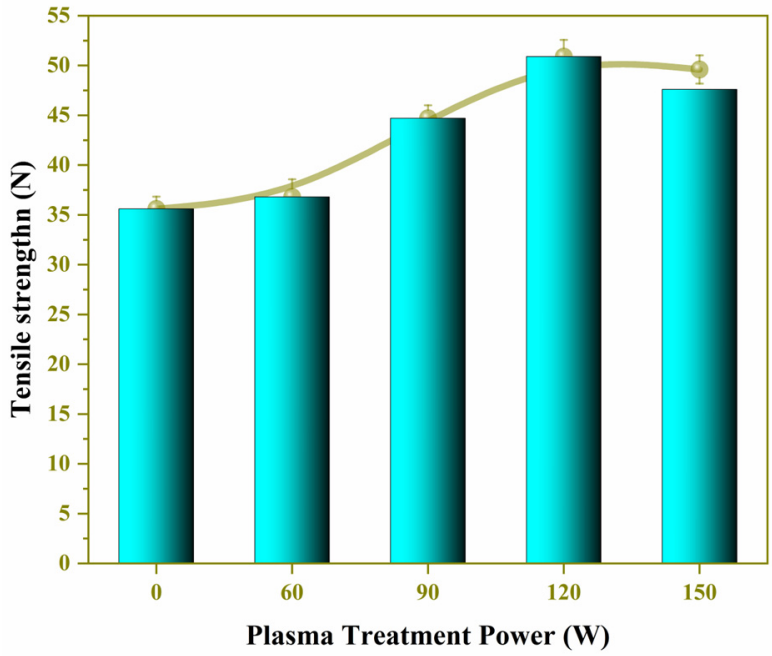

Figure 8. Mechanical properties of 5-P at different plasma treatment powers.

3.6. Antimicrobial Properties

The antimicrobial activity of 5-P is shown in Figure 9a,b. It can be seen that raw 5-P exhibited 73.3% and 42.7% bacterial inhibition against *S. aureus* and *E. coli*, indicating that PI fiber has an effective antimicrobial effect. After being plasma-treated, 5-P exhibited 91.57% and 64.3% against *S. aureus* and *E. coli*, respectively. As compared to untreated samples, the antimicrobial rate increased by 24.9% and 50.6% against the two species of bacteria.

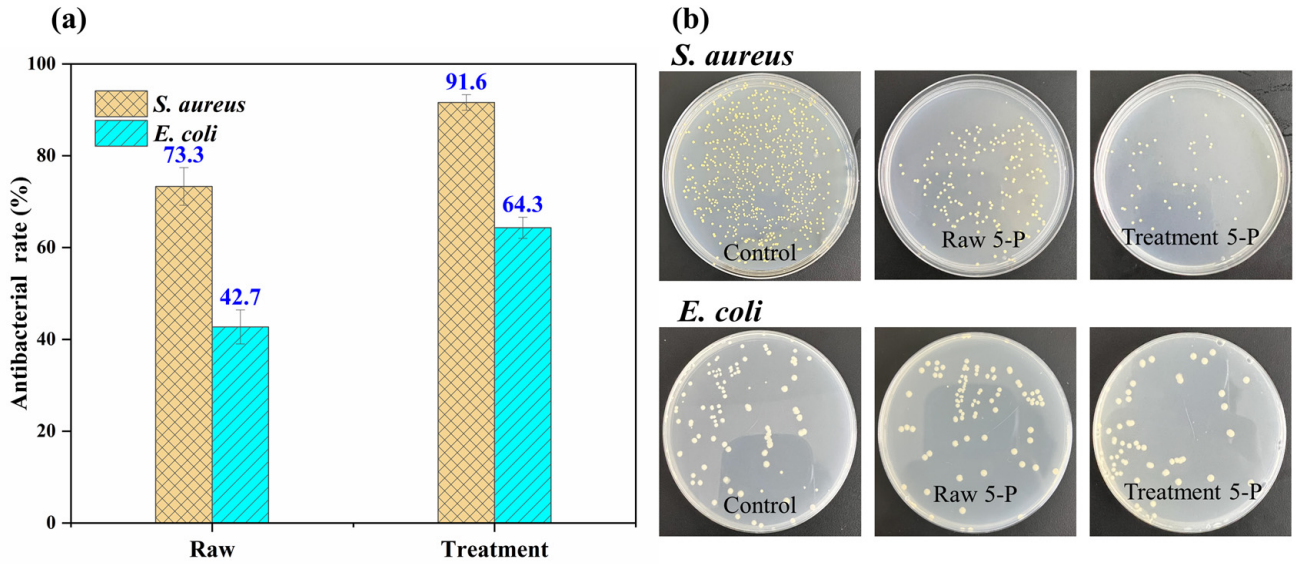

Figure 9. Antibacterial rate (**a**) and colony number photos (**b**) of 5-P against *S.aureus* and *E.coli* before and after plasma treatment.

This is possibly due to the production of positively charged substances on the surface of the nonwoven fabrics under the action of plasma, as shown in Figure 10. More N-containing groups with antimicrobial properties were generated and exposed on the fiber surface, as can be seen in Figure 5d. After plasma treatment, NH_3^+ and NH_4^+ on the fiber surface increased by 37.0% and 131.8%, respectively. Meanwhile, under the action of high-energy particles, free locally reactive oxygen and locally reactive nitrogen can be generated on the fiber surface [39], including the atoms O, O_3, H_2O_2 and the N atoms NO, NO_2, and their derivatives.

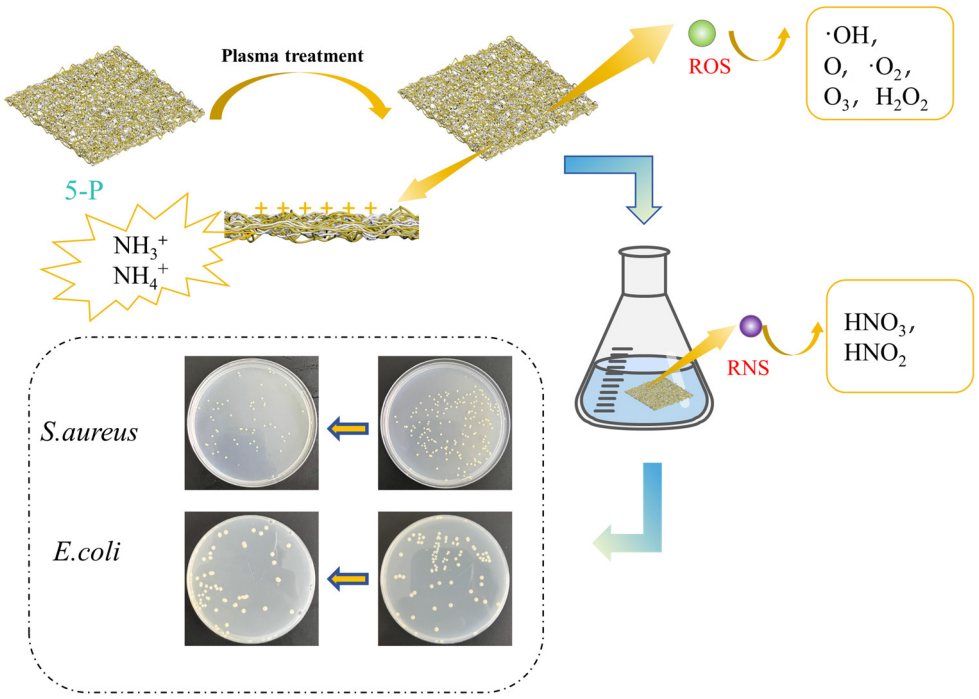

Figure 10. Antimicrobial mechanism of plasma-treated 5-P.

O_3 and H_2O_2 can directly inactivate bacteria, whereas NO and NO_2 can be dissolved in liquid media and reacted with water to form acids such as nitrite and nitric acid [40]. The acidity of the liquid leads to oxidative stimuli in the bacteria and their intracellular

components, as well as the coagulation and inactivation of proteins. From Figure 11a, it can be seen that the pH value of the plasma-treated 5-P solution gradually decreases, and the solution as a whole is acidic, while the untreated 5-P solution is alkaline. This demonstrates that acidic substance is generated on the surface of 5-P, and the concentration of acid gradually increases with time, which indicates the success of localized reactive nitrogen conversion. The interaction of positively charged fiber surfaces with negatively charged bacterial membranes can lead to ionic imbalances by destroying cell membrane functions (e.g., respiration, solute transport, and cell wall biosynthesis), which can initiate cell death. As shown in Figure 11b, the charge of the untreated 5-P solution diminishes to 0 in air over time, while the plasma-treated 5-P solution has a positive charge of 106 eV, and even after 5 days, it still has a positive charge of 102. This indicates that there are stable positively charged atoms and groups on the surface of the nonwoven fabric and that the positive charge is able to inactivate the bacterial cells on a microscopic scale.

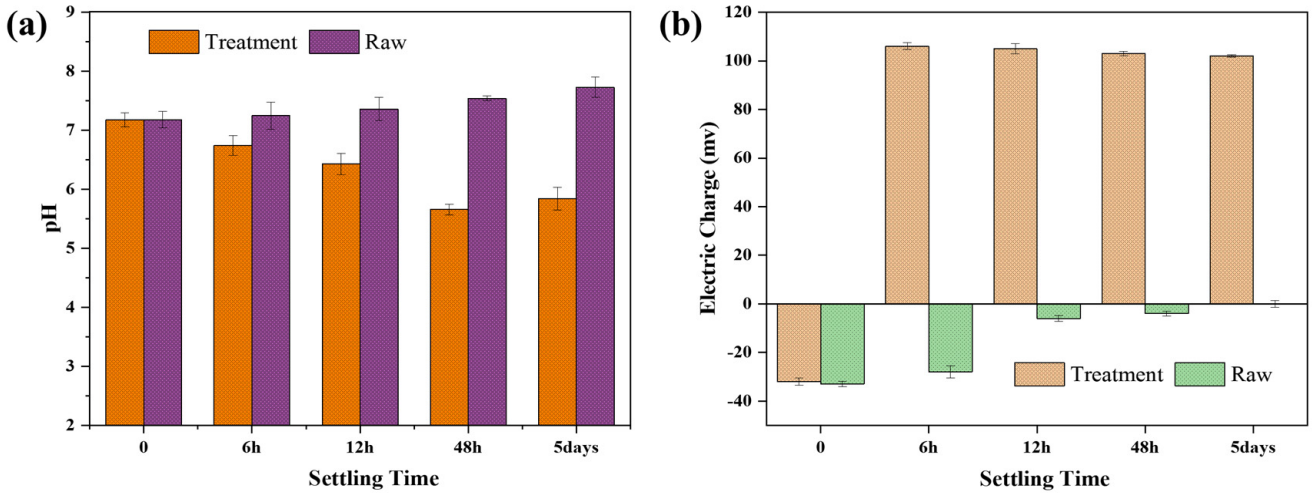

Figure 11. Charge (**a**) and Ph (**b**) of raw and plasma-treated 5-P in deionized water at different times.

4. Conclusions

In conclusion, this study developed a comfortable spunlace nonwoven fabric with rapid liquid penetration capability and good antimicrobial properties through a simple and effective one-step plasma treatment of 5-P. The application of natural antimicrobial fiber PI follows the principle of green chemistry and has a broad development prospect in the field of hygienic materials. Fast and efficient plasma treatment enhances the overall performance of nonwovens by modifying the fiber surface morphology and air graft reactions. After the treatment, one side of the nonwoven was successfully converted from a hydrophobic to a hydrophilic surface (WCA from 121.5° to 68.5°), resulting in high liquid permeability (2.1 g) and low impermeability (0.5 g). Meanwhile, the antibacterial rate against *S. aureus* reaches 91.6%, and against *E. coli* up to 64.3%. Moreover, the nonwoven fabrics were manufactured quickly and without the addition of any chemicals, which is therefore potentially more significant in the practical application of disposable sanitary materials.

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References

1. Barman, A.; Katkar, P.M.; Asagekar, S.D. Natural and Sustainable Raw Materials for Sanitary Napkin. *J. Text. Sci. Eng.* **2017**, *7*, 3.
2. Das, D. Composite Nonwovens in Absorbent Hygiene Products. In *Composite Non-Woven Materials*; Woodhead Publishing: Cambridge, UK, 2014; pp. 74–88.
3. Atasağun, H.G.; Kara, S. Investigation of Moisture Management and Frictional Characteristics of Top Layers Used in Disposable Absorbent Hygiene Products. *Fibers Polym.* **2022**, *23*, 2577–2585. [CrossRef]
4. Miao, D.; Cheng, N.; Wang, X.; Yu, J.; Ding, B. Sandwich-Structured Textiles with Hierarchically Nanofibrous Network and Janus Wettability for Outdoor Personal Thermal and Moisture Management. *Chem. Eng. J.* **2022**, *450*, 138012. [CrossRef]
5. Xiao, Y.-Q.; Kan, C.-W. Review on the Development and Application of Directional Water Transport Textile Materials. *Coatings* **2022**, *12*, 301. [CrossRef]
6. Ajmeri, J.R.; Ajmeri, C.J. Nonwoven Personal Hygiene Materials and Products. In *Applications of Nonwovens in Technical Textiles*; Woodhead Publishing: Cambridge, UK, 2010; pp. 85–102.
7. Cheng, H.; Han, N.; Bai, Z.; Li, Z.; Xu, W.; Xiao, S. Ag-Decorated PP Nonwoven for Excellent Catalytic Reduction of 4-Nitrophenol and Antibacterial Study. *Surf. Interfaces* **2023**, *39*, 102929. [CrossRef]
8. Yadav, S.; Illa, M.P.; Rastogi, T.; Sharma, C.S. High Absorbency Cellulose Acetate Electrospun Nanofibers for Feminine Hygiene Application. *Appl. Mater. Today* **2016**, *4*, 62–70. [CrossRef]
9. Sun, L.; Feng, Y.; Qian, X. Preparation and Properties of Wormwood Extract/Viscose Spunlaced Nonwovens. *J. Text. Inst.* **2021**, *112*, 709–717. [CrossRef]
10. Wu, Y.; Yang, S.; Fu, F.; Zhang, J.; Li, J.; Ma, T.; Liu, X.; Yao, J. Amino Acid-Mediated Loading of Ag NPs and Tannic Acid onto Cotton Fabrics: Increased Antibacterial Activity and Decreased Cytotoxicity. *Appl. Surf. Sci.* **2022**, *576*, 151821. [CrossRef]
11. Wang, Y.; Liu, X.; Yang, R.; Ma, Q. Asymmetric Wetting and Antibacterial Composite Membrane Obtained by Spraying Bacterial Cellulose Grafted with Chitosan for Sanitary Products Surface Layers. *Carbohydr. Polym.* **2021**, *256*, 117602. [CrossRef]
12. Iqbal, W.; Yexiong, Q.; Yaming, J.; Owais Raza Siddiqui, M.; Khalid, R.; Iqbal, A.; Usman Munir, M.; Hussain, A.; Xu, S. Antimicrobial Property of Functional Viscose Fibre by Using Mint Extract. *Ind. Textila* **2023**, *74*, 464–469. [CrossRef]
13. Agrawal, N.; Low, P.S.; Tan, J.S.J.; Fong, E.W.M.; Lai, Y.; Chen, Z. Durable Easy-Cleaning and Antibacterial Cotton Fabrics Using Fluorine-Free Silane Coupling Agents and CuO Nanoparticles. *Nano Mater. Sci.* **2020**, *2*, 281–291. [CrossRef]
14. Lian, L.; Pang, C.; Wei, H.; Hong, L. Citronellol-Based Long-Lasting Antibacterial Cotton Fabrics without Bacterial Resistance. *Macromol. Biosci.* **2023**, *23*, 2300169. [CrossRef]
15. Cheng, W.; Yu, Y.; Liu, W.; Wang, X.; Zhou, M.; Xu, B.; Wang, P.; Wang, Q. Self-Assembly Amino-Quinone Network Coatings onto Polyester Fabric via Single-Sided Spraying of Natural Polyphenols and Polyethyleneimine for Highly Efficient Moisture Conducting and Bacteriostatic Properties. *Appl. Surf. Sci.* **2022**, *606*, 154913. [CrossRef]
16. Liu, J.; Lin, X.; Liang, H. Dyed Fabrics Modified via Assembly with Phytic Acid/Berberine for Antibacterial, UV Resistance, and Self-Cleaning Applications. *J. Eng. Fibers Fabr.* **2019**, *14*, 155892501988897. [CrossRef]
17. He, H.; Qin, Y.; Zhu, Z.; Jiang, Q.; Ouyang, S.; Wan, Y.; Qu, X.; Xu, J.; Yu, Z. Temperature-arousing self-powered fire warning e-textile based on p-n segment coaxial aerogel fibers for active fire protection in firefighting clothing. *Nano-Micro Lett.* **2023**, *15*, 226. [CrossRef]
18. Yu, Z.; Wan, Y.; Qin, Y.; Jiang, Q.; Guan, J.P.; Cheng, X.W.; Wang, X.; Ouyang, X.; Qu, X.; Zhu, Z.; et al. High fire safety thermal protective composite aerogel with efficient thermal insulation and reversible fire warning performance for firefighting clothing. *Chem. Eng. J.* **2023**, *477*, 147187. [CrossRef]
19. Wang, Q.; Bai, Y.; Xie, J.; Jiang, Q.; Qiu, Y. Synthesis and Filtration Properties of Polyimide Nanofiber Membrane/Carbon Woven Fabric Sandwiched Hot Gas Filters for Removal of PM 2.5 Particles. *Powder Technol.* **2016**, *292*, 54–63. [CrossRef]
20. Wang, L.; Cui, L.; Liu, Y.; Riedel, J.; Qian, X.; Liu, Y. Electrospun Polyimide Nanofiber-Coated Polyimide Nonwoven Fabric for Hot Gas Filtration. *Adsorpt. Sci. Technol.* **2018**, *36*, 1734–1743. [CrossRef]
21. Qiao, S.; Kang, S.; Zhu, J.; Wang, Y.; Yu, J.; Hu, Z. Facile Strategy to Prepare Polyimide Nanofiber Assembled Aerogel for Effective Airborne Particles Filtration. *J. Hazard. Mater.* **2021**, *415*, 125739. [CrossRef]
22. Yang, X.; Ma, W.; Lin, H.; Ao, S.; Liu, H.; Zhang, H.; Tang, W.; Xiao, H.; Wang, F.; Zhu, J.; et al. Molecular Mechanisms of the Antibacterial Activity of Polyimide Fibers in a Skin-Wound Model with Gram-Positive and Gram-Negative Bacterial Infection in Vivo. *Nanoscale Adv.* **2022**, *4*, 3043–3053. [CrossRef]
23. Song, Y.; Chen, X.; Xu, K.; Li, H.; Xu, Y.; Wu, H.; Wu, J.; Huang, C. Green and Scalable Fabrication of Nonwoven Composites Featured with Anisotropic Water Penetration. *ACS Sustain. Chem. Eng.* **2019**, *7*, 19679–19685. [CrossRef]
24. Zhang, H.; Shen, Y.; Edgar, K.J.; Yang, G.; Shao, H. Influence of Cross-Section Shape on Structure and Properties of Lyocell Fibers. *Cellulose* **2021**, *28*, 1191–1201. [CrossRef]
25. Adnan, M.; Moses, J. A Study on the Thermophysiological and Tactile Comfort Properties of Silk/Lyocell Blended Fabrics. *Matéria* **2020**, *25*, e12796. [CrossRef]
26. Hanh Le, T.M.; Singto, S.; Sajomsang, W.; Mongkolnavin, R.; Nuisin, R.; Painmanakul, P.; Sairiam, S. Hydrophobic PVDF Hollow Fiber Membrane Modified with Pulse Inductively Coupling Plasma Activation and Chloroalkylsilanes for Efficient Dye Wastewater Treatment by Ozonation Membrane Contactor. *J. Membr. Sci.* **2021**, *635*, 119443. [CrossRef]

27. Kan, C.; Lam, Y. The Effect of Plasma Treatment on Water Absorption Properties of Silk Fabrics. *Fibers Polym.* **2015**, *16*, 1705–1714. [CrossRef]
28. Urbaniak-Domagala, W.; Krucinska, I.; Wrzosek, H.; Komisarczyk, A.; Chrzanowska, O. Plasma Modification of Polylactide Nonwovens for Dressing and Sanitary Applications. *Text. Res. J.* **2016**, *86*, 72–85. [CrossRef]
29. Liu, X.; Wang, C.; Liu, J.; Wang, G.; Yang, Z.; Chen, F.; Song, J. Comparative Study of Surface Modification of Polyethylene by Parallel-Field and Cross-Field Atmospheric Pressure Plasma Jets. *J. Appl. Phys.* **2019**, *125*, 123301. [CrossRef]
30. Kim, J.H.; Han, J.H.; Hong, S.; Kim, D.-W.; Park, S.H.; Wee, J.-H.; Yang, K.S.; Kim, Y.A. Effect of Plasma Surface Modification on Pullout Characteristics of Carbon Fiber-Reinforced Cement Composites. *Carbon Trends* **2021**, *3*, 100030. [CrossRef]
31. Khoo, Y.S.; Lau, W.J.; Liang, Y.Y.; Karaman, M.; Gürsoy, M.; Ismail, A.F. Eco-Friendly Surface Modification Approach to Develop Thin Film Nanocomposite Membrane with Improved Desalination and Antifouling Properties. *J. Adv. Res.* **2022**, *36*, 39–49. [CrossRef]
32. Dhanumalayan, E. Studies on the Surface and Wetting Properties of Poly (Vinylidene Fluoride)/Poly (Acrylonitrile)/Multiwalled Carbon Nanotube-NH2 Blends as a Function of Air Plasma Treatment. *J. Mater. Eng. Perform.* **2021**, *30*, 7343–7353. [CrossRef]
33. Said, N.; Khoo, Y.S.; Lau, W.J.; Gürsoy, M.; Karaman, M.; Ting, T.M.; Abouzari-Lotf, E.; Ismail, A.F. Rapid Surface Modification of Ultrafiltration Membranes for Enhanced Antifouling Properties. *Membranes* **2020**, *10*, 401. [CrossRef]
34. *ASTM D737-18*; Standard Test Method for Air Permeability of Textile Fabrics. ASTM: West Conshohocken, PA, USA, 2023.
35. *GB/T 30133-2014*; General Technical Specification for Surfaces for Sanitary Napkins. China Standard Press: Beijing, China, 2014.
36. *GB/T 15979-2002*; Hygienic Standard for Disposable Sanitary Articles. China Standard Press: Beijing, China, 2002.
37. Ding, C.; Chen, C.; Yuan, Z.; Chen, L. Molecular Simulation of the Effect of Plasma Modification on the Microscopic Properties of Polyimide. *Front. Mater.* **2022**, *9*, 1018882. [CrossRef]
38. Yang, D.; Lin, W.; Xin, B.; Miao, Y.; Chen, X. Research on the Surface Roughness of Aramid Fibers via the Surface Modification. *J. Text. Inst.* **2021**, *112*, 1743–1752. [CrossRef]
39. Xu, Z.; Cheng, C.; Shen, J.; Lan, Y.; Hu, S.; Han, W.; Chu, P.K. In Vitro Antimicrobial Effects and Mechanisms of Direct Current Air-Liquid Discharge Plasma on Planktonic Staphylococcus Aureus and Escherichia Coli in Liquids. *Bioelectrochemistry* **2018**, *121*, 125–134. [CrossRef]
40. Patange, A.D. Inactivation Efficacy of Atmospheric Air Plasma and Airborne Acoustic Ultrasound against Bacterial Biofilms. *Sci. Rep.* **2021**, *11*, 2346. [CrossRef]