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# Fabrication and Changes in Physical and Functional Properties after Repeated Laundering of PU Nanocomposite Laminates Loaded with Electrospun *Juniperus Chinensis* Extracts

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**Abstract:** To investigate the potential of nanoweb laminates that contain juniperus chinensis extracts as functional garments or specialty textiles, a layered fabric system with PU nanoweb was inserted between the PET fabric and the mesh. Of particular interest in this study was the development of nanoweb laminates by electrospraying polyurethane resin onto the substrate fabrics. Therefore, to minimize damage to the nanoweb, the PU adhesive resin was electrosprayed on the polyester plain weave fabric and mesh, respectively, and the PU nanoweb laminate was prepared by inserting PU nanowebs containing juniperus chinensis extracts (JCE) between two fabrics and heat-pressing them together at 110 °C for 30 s. To examine the compatibility of the fabricated PU nanoweb laminates (PNLs), 1, 5, and 10 launderings were performed, and changes in the physical and functional properties due to the laundering were investigated. The nanofiber structure of the PNL was maintained after repeated laundering. The tensile strength of the PNL was maintained partially after repeated laundering. There was no significant change in the moisture permeability after repeated laundering, whereas the air permeability decreased somewhat after the launderings, but an acceptable level of air permeability still was maintained. Laundering made the PNLs less waterproof, and their adhesive strength was reduced, which implied that different laminating techniques or substrate materials should be investigated. However, the anti-bacterial properties of the PNLs were very good for both Staphylococcus aureus and Pneumoniae.

**Keywords:** PU nanoweb laminate, *Juniperus Chinensis* extract, Laundering, Antibacterial, Physical property

## Introduction

Recently, there have been reports on the merits of nanofiber webs made by electrospinning to improve their performance by combining the properties of natural extracts or to make new functional materials [1-5]. To maximize the utility and usefulness of natural extracts, their function should be ensured through atomization, dispersion, and combination with useful material [6].

*Juniperus chinensis* (Cupressaceae), commonly known as *Chinese juniper* is native to countries in the East Asian region, and it is used extensively as an ornamental plant. It has been found to possess strong anti-microbial, anti-fungal, and anti-tumor activities [7-10]. The main components of *Juniperus chinensis* extract are terpene components composed of monoterpenes and sesquiterpenes [11]. The chain and cyclic hydrocarbons that occupy most of the essential oil component of terpene are known to have sterilization, disinfection, and anti-inflammatory properties [12,13].

Electrospinning is a technique for obtaining nanofibers that have three-dimensional structures, and the electrospinning is conducted in an electric field that is formed by applying a high voltage to a polymer solution or melt [14-16]. The nanofibers that are produced have beneficial properties, such as excellent breathability, permeability to moisture, light weight, soft to the touch [17-20]. In addition, since the polymer solution can be maintained stably without changing the anti-microbial substance by electrospinning at room temperature, the nanofiber webs that are manufactured are useful in making many materials, such as filter materials, reinforcing fibers, functional clothing materials, and wound-healing materials [21-25]. While ultrathin nanofiber webs offer some exciting properties, it has been reported that they have limited mechanical properties [26]. In order to provide strength and durability, nanofiber webs must be laminated by adding an adhesive to the substrate fabric system [27,28]. However, the lamination method is a secondary process, such as bonding, it is applied with an adhesive, and it does not exhibit appropriate breathability and permeability to moisture. These disadvantages result from the pores of the nanofiber being blocked by the adhesive, and there also are secondary problems, such as emissions to the environment [29].

Kang *et al.* [30] proposed the application of the nanofiber web as a breathable and waterproof material with improved permeability to air and water vapor, heat retention, and comfort. In addition, compared with conventional polyurethane coating or polytetrafluoroethylene (PTFE) laminated fabric, the nanofiber web treatment material has the potential for use as a new, breathable, waterproof material that has satisfactory performance, e.g., excellent water resistance, breathability, moisture permeability, and soft to the touch [31,32]. Without disclosing the details of the lamination process, Lee *et al*. [33] reported that the physical properties of the nanofiber web laminate fabricated by two methods, i.e., hot-melt laminating and solvent laminating, were

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different and that the hot-press method was more suitable for laminating the nanofiber web.

In order to validate the functionality of the nanofiber web laminate material for commercial use, its basic properties and its moisture permeability, durability, and functional changes must be evaluated after washing.

Therefore, in this study, to minimize damage to the nanoweb, the PU adhesive resin was electrosprayed on the polyester plain weave fabric and the mesh, respectively. Then, the PU nano-web laminate composites were prepared by inserting PU nano-webs containing *Juniperus chinensis* extracts (JCE) between the two fabrics and heat-pressing them together. To examine the functional and physical properties of the fabricated PU nano-web laminates, 1, 5, and 10 washes were performed, and the physical properties, air and moisture transfer characteristics, and anti-bacterial properties before and after washing were compared and evaluated.

## Experimental

### Materials

Polyurethane (PU, MW=80,000) was obtained from Lubrizol (pellethane 2103-80AE, USA). As the adhesive resin, a polyester-based polyurethane resin was used from Ilsam (Korea). N-dimethylformamide (DMF, >98 %) and methyl ethyl ketone (MEK, >98 %) (Daejung Chemicals (Korea) were used as the solvent. *Juniperus chinensis* extracts were obtained from the Kyungdong Traditional Market (Korea). The substrate fabrics on which the resin was electrosprayed were polyester plain fabric and polyester mesh. Table 1 shows the characteristics of the substrate fabrics supplied by Seong-An Synthetics Co. (Korea).

### Preparation of the Electrospun PU/JCE Nanofibers

The solution of JCE/PU was prepared with a JCE concentration of 1.0 wt% in the PU solution. The PU concentration was kept constant at 16 wt% with respect to the DMF/MEK (1:1) solution. The polymer solutions were prepared with constant stirring at room temperature.

Since it is difficult to fabricate wide-width nano-webs with lab-level electrospinning, we ordered a 60 cm-wide nano-web from FTENE Co., which has a system that is capable of manufacturing wide-width nano-webs using multi-channel multi-nozzles. The electrospinning system is composed of a high-voltage DC power supply (NanoNC, Korea) capable of supplying voltage in the range of 0 to 40 kV, and it has a rotating drum collector (NanoNC, Korea). The drum collector was wrapped with a teflon sheet and then electrospun using the multi-nozzle. A 5 m*l* plastic syringe tip that had an inner diameter of 0.6 mm was used to inject solutions into the electrospinning apparatus. The parameters associated with the electrospinning process were a voltage at 8 kV, the tip-to-collector distance of 15 cm, and a flow rate of 0.5 m*l*/h. All of the electrospinning process were conducted at room temperature at ~40 % humidity. Figure 1 shows the image and Table 2 shows the characteristics of the PU/JCE nanofiber web.

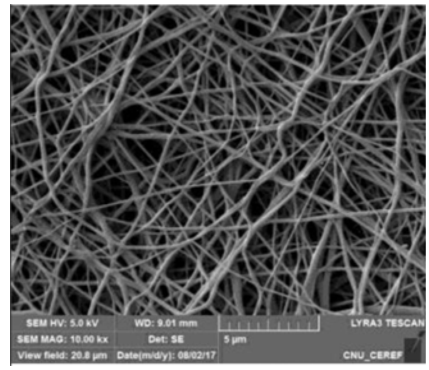


**Figure 1.** SEM image of PU/JCE nanofibers.

**Table 2.** Characteristics of the PU/JCE nanofiber

| | |
|---|---|
| Nano web thickness (micron) | 20 |
| Average diameter of fibers (nm) | 213±44 |
| Weight (g/m$^2$) | 10 |

### Preparation of Polyurethane Resin Electrosprayed Fabrics

A polyester-based polyurethane resin (MW=~50,000 g/mol) was prepared with DMF solution to make a 33 wt% resin solution for electrospraying. The electrospray condition was 18 kV of applied voltage, tip-to-collector distance of 15 cm, and a flow rate of 0.5 m*l*/h. The prepared resin solution was electrosprayed on a PET plain fabric and a PET mesh with a thickness of 2 g/m$^2$, respectively.

### Heat Pressing for Lamination

The two prepared substrate fabrics, i.e., the PET plain fabric and the PET mesh in which the adhesive resin solution was electrosprayed and the PU/JCE nanofiber web, were heat-pressed together at 110 $^o$C for 30 s sec using a plate heat-press (MAXXTM 38x38, STAHLS, USA) to laminate.

**Table 1.** Specifications of the substrate fabrics

| Fabric | Contents (%) | Yarn count (d/f) | Fabric count (w×f/in$^2$) | Weave | Weight (g/m$^2$) | Thickness (mm) |
|---|---|---|---|---|---|---|
| Taffeta | Polyester 100 | 75/36×75/36 | 97×78 | Plain | 49 | 0.083 |
| Mesh | Polyester 100 | 36/22×40/24 | 83×61 | Plain | 25 | 0.118 |

**Laundry Machine Washing of PU Nanoweb Laminate Composites**

The PU nanoweb laminated composites were washed according to the KS K ISO 6330 (Textiles: Domestic washing and drying procedures for textile testing). The wash was performed using a commercial electric laundry machine based on KS K ISO 6330, because it is the most common washing method used by consumers. A neutral, commercial, liquid laundry detergent for outdoor sportswear (Woolshampoo, AK Co., Korea) also was used. Then, all of the properties of the PU nanocomposite laminates (PNLs) were measured again after one, five, and 10 washing cycles.

**Characterization**

***Scanning Electron Microscopy***

A Scanning Electron Microscope (SEM, JSM-6010LA by JEOL, Tokyo, Japan) was used to observe the PU nanocomposite laminates. The acceleration voltage was 10 kV and the magnification was ×10,000 for the surface of the nanoweb and ×5,000 for the cross-section of the nanocomposites.

***Tensile Strength***

To measure tensile strength, testing was performed according to the ASTM D 5035 (Standard test method for breaking force and elongation of textile fabrics (strip method)). The test specimen with a width of 2.54 cm, length 15 cm were respectively prepared 5 each in a warp direction, the weft direction.

***Air Permeability***

The air permeability of the PU nanocomposite laminates (PNL) was evaluated according to KS K ISO 9237: 1995 (air permeability measurement of textile fabric). The amount of air passing through the vertical direction of the sample was measured for a test area of 20 $cm^2$ and an air pressure of 100 Pa, and the base fabrics (PET) were subjected to air pressure.

***Water-vapor Permeability***

The moisture permeability of PU/JCE-LC was measured by KS K 0594 (2008, calcium chloride method). The moisture permeability was calculated using a moisture-permeable cup containing calcium chloride ($CaCl_2$) based on the increase in weight of the moisture permeable cup at 40±2 °C per unit time at a relative humidity of 90±5 % RH. The area of the moisture-permeable cup was 0.00283 $m^2$, and the height of the moisture-permeable cup was 25 mm.

***Water Resistance***

The water resistance was measured in accordance with KS K ISO 811: 2015 (Textile Cloth - Water Resistance Measurement - Water Pressure Method). When the water pressure was applied at a constant rate per unit area using a hydrostatic pressure tester, the water pressure (pressure expressed by the height of the water column) was measured when three water droplets were seen on the surface of the fabric. Then, the water pressure was measured at three points on the back side of the specimen at a rate of 600 $mmH_2O$/min. The temperature of the water was adjusted to 20±2 °C.

***Adhesive Strength***

A peel test was performed to measure the adhesive strength between the nanofiber web and the woven fabrics. The peel test was performed according to ASTM D 4851 (Standard test method for coated and laminated fabrics for architectural use).

***Anti-bacterial Properties***

*Staphylococcus aureus* ATCC 6538 and *Klebsiella pneumoniae* ATCC 4352 were used as test strains in order to investigate the anti-bacterial properties of PU/JCE-LC. The control bacteria were cultured for 18 hours, the number of viable cells was measured, and the rate of decrease of the bacteria was calculated. The bacterial reduction rate (%) in the experiment was calculated as the bacteriostatic rate using the following equation;

$$\text{Anti-bacterial rate (\%)} = (M_b - M_c)/M_b \times 100\ \%$$

where $M_b$ is the number of bacteria recovered from the control specimen incubated for 18 hours, $M_c$ is the number of bacteria recovered from the treated test specimen incubated for 18 hours.

## Results and Discussion

**Morphology of PU Nanocomposite Laminates**

The surface and cross-sectional structures of the 3-layer nanocomposite laminated with PET fabric and mesh were analyzed by SEM in order to observe the changes in the morphology of the nanofibers after laundering. Figure 2 shows the appearance of the molten adhesive resin between the nanofibers on the surface of the PNL from which the PET fabric was removed. This is because the adhesive resin solution was electrospun to the PET plain weave fabric and the mesh fabric, and, then, the nano web was sandwiched and calendered in the middle. It seems that the adhesive resin applied to the PET and mesh fabrics was melted by the heat treatment, and the parts adhered to the nanoweb were mixed with the parts that did not adhere to the nanoweb.

There was no structural change in the surface or the cross section of the PNL due to the number of washes. But it is considered that the bonding was loosened due to the number of times the fabric was washed in the portion where the three-layered nanocomposite structure had not been adhered by the adhesive.

**Tensile Strength and Elongation**

Tensile strength and elongation are important factors that determine the durability of garment material. The tensile strength and elongation of PNL were measured before washing and after washing 1 time, 5 times, and 10 times in

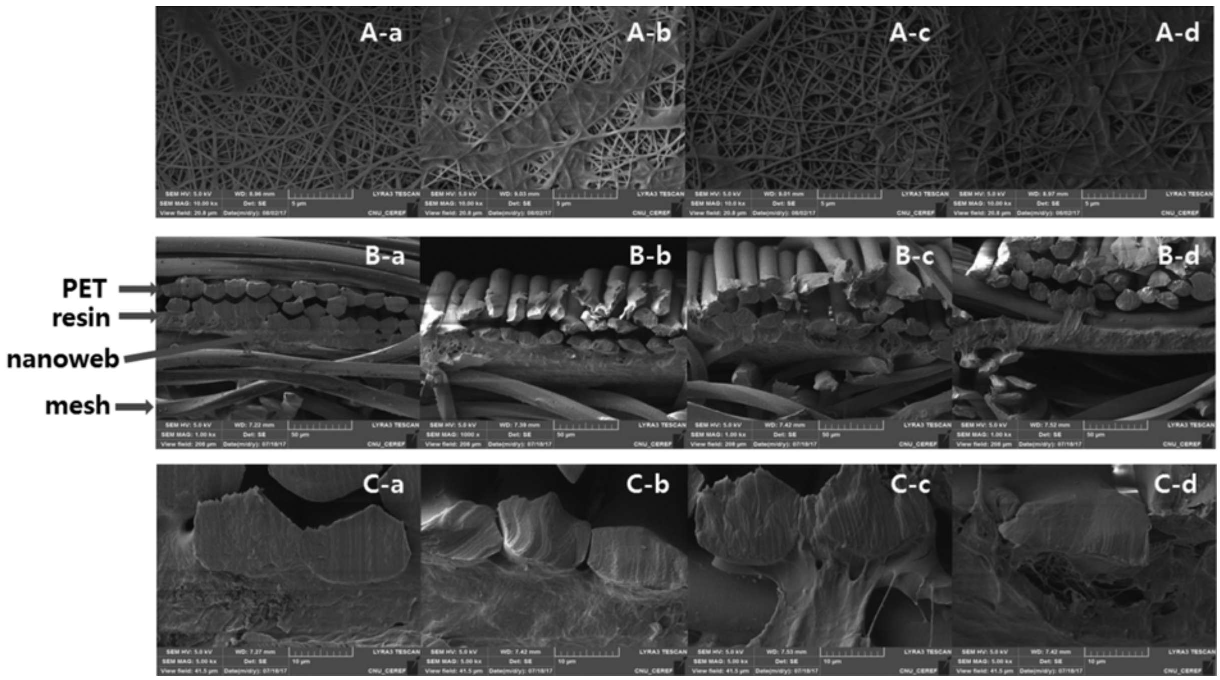


**Figure 2.** SEM images of PU nanocomposite laminates (PNL) according to the number of launderings; (A) surface of PNL ×10,000, (B) cross-section of PNL ×1,000, (C) cross-section of PNL ×5,000; (a) no wash, (b) washed 1 time, (c) washed 5 times, and (d) washed 10 times.

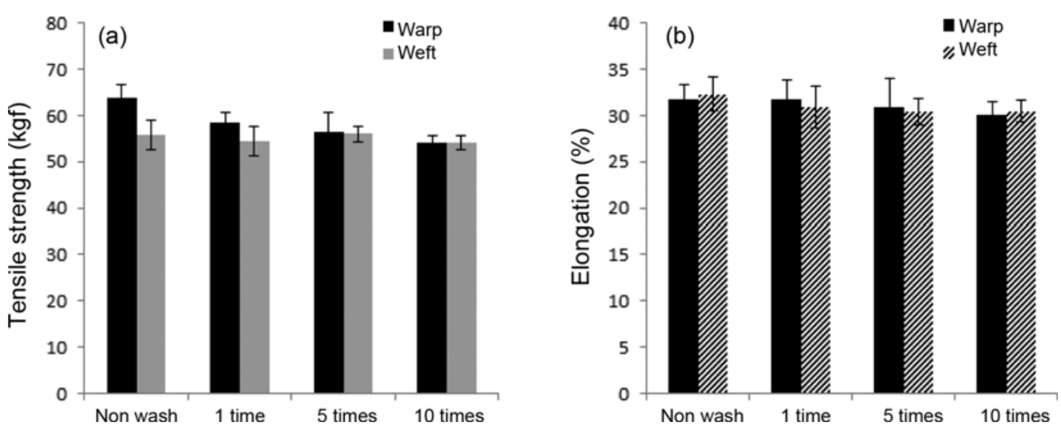


**Figure 3.** Tensile strength (a) and elongation (b) of PNL based on the number of launderings.

the direction of warp and weft. Figure 3 shows the results of the tensile strength and elongation tests. PNL originally had high tensile strength in the direction of warp, and it decreased slightly after repeated washing, but it was not affected by washing in the direction of weft. Also, the elongation of PNL was not affected by repeated washing.

A study of the change of moisture permeability and durability after repeated washings of the nanofiber web laminate materials indicated that the tensile strengths of the base fabrics and the laminated structures were affected, so, when designing the nano-fiber web laminate, it is necessary to consider the durability of the base fabric and the laminated structure in addition to the moisture-permeable and waterproof performance [34]. In this study, the PNL seemed to have the properties of both the base fabric and the adhesive since the polyester plain weave fabric, the nanoweb, and the mesh were heat treated together with an adhesive.

**Air Permeability**

Figure 4 shows the change of air permeability of PNL after repeated washing. The air permeability of PNL after repeated washing showed a tendency to be slightly lower than that before washing. This is because it is believed that

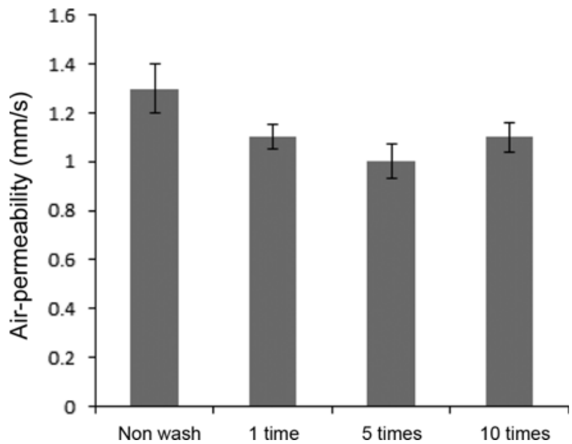


**Figure 4.** Air-permeability of PNL according to the number of launderings.

repeated physical and chemical forces are applied by repeated washing, resulting in rearrangement of the pores due to changes in the fiber and fabric layered system, and thus the air permeability of the PNL is reduced. Previous studies have shown that air permeability decreases and the moisture permeability increases as the specific gravity of small pores increases in the same pore area [35].

**Water-vapor Permeability**

Moisture permeability means the amount of water vapor that passes through per unit time per unit area. Good moisture permeability prevents the humidity in the garment from rising when sweating, and it minimizes condensation in the garment when the outside temperature is low. Therefore, the permeability of clothing to moisture is very important from the perspective of the wearer's comfort. Since the average rate of sweat production during hard work is about 2,880 $g/m^2/24$ hr at 20 °C, it is sufficient if the permeability of a garment to moisture is more than 3,000 $g/m^2/24$ hr. It has been reported that when the ambient temperature is high or in the case of heavy exercise, a large amount of perspiration occurs temporarily, and, thus, it is preferred for the clothing to have high permeability [36]. Figure 5 shows that the permeability of PNL to moisture before washing was 5327 $g/m^2/24$ hr, which showed that the material's characteristics effectively allowed the amount of perspiration produced during hard work to pass through the clothes effectively. The change in the moisture permeability of the PNL after one washing to 5357 $g/m^2/24$ hr indicated that the water vapor transmission rate was a slight increase compared to before washing to. Also, it was slightly reduced to 5173 $g/m^2/24$ hr after 5 washings, and increased to 5449 $g/m^2/24$ hr after 10 washings. This result is consistent with a previous study in which the water-vapor permeability was increased due to the rearrangement of the pores in fabric layered system when nanofiber web laminate was washed repeatedly [34].

**Water Resistance**

Water resistance means resistance to wetting or penetration of water. In general, when measured by the low-pressure method, it is classified into a low water pressure type of 300 to 800 $mmH_2O$, a heavy water pressure type of 1,000 to 2,500 $mmH_2O$ (universal type), and a high water pressure type moisture permeable material of 5,000 to 10,000 $mmH_2O$. Figure 6 shows the variation of water resistance with one, five, and ten repetitive washes of PNL. The water resistance of the PNL before washing was 832 $mmH_2O$, a low resistance. As the number of washings increased, the PNL decreased to 633 $mmH_2O$, 537 $mmH_2O$, and 487 $mmH_2O$. This was considered to be the result of the damage

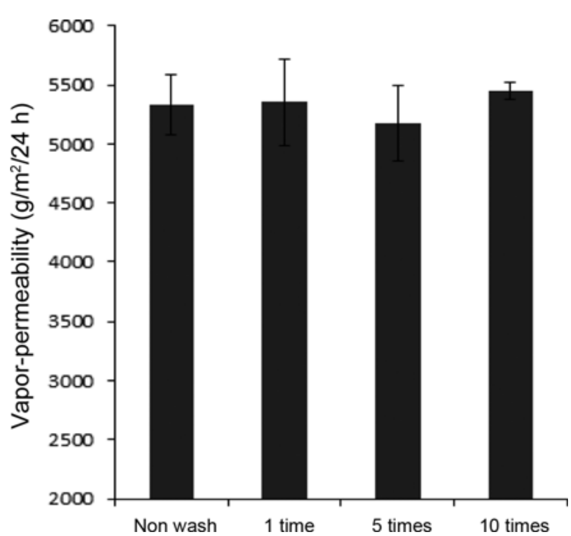


**Figure 5.** Vapor-permeability of PNL according to the number of launderings.

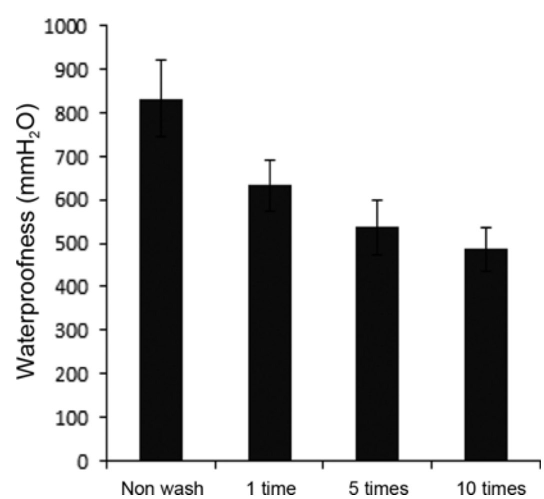


**Figure 6.** Waterproofness of PNL according to the to the number of launderings.

of the bonding state between the fabric and the nano-web that resulted from the number of times the fabric was washed. And it may not be the electrospraying method of the adhesive selected as a method to minimize the damage of the nanoweb. Therefore, improving the laminating method and adding a cross-linking agent and an acid catalyst with an appropriate ratio to electrospinning using polyurethane resin are expected to improve the resistance to increases in the water pressure as the strength of the nano web increases. In previous study showed that the waterproofness of the nanofiber web laminate material manufactured by the two laminating methods after washing was observed, and it was found that even after washing 10 times, the water resistance property required for the high water pressure type breathable waterproof material was satisfied [28]. In this study, a 100 % nylon woven fabric was used as the base fabric, and the nanofiber web laminate material was prepared by two methods, hot melt laminating and solvent laminating. Therefore, it would be possible to manufacture a nanofiber web laminate material that can exhibit excellent water resistance even after repeated washing, by improving the laminating method and selecting a base fabric suitable for maintaining the waterproof performance.

**Adhesive Strength**

Laminated composite nanomaterials must be durable to withstand the external forces of physical deformation, such as bending, stretching, and abrasion, when they are used as clothing, because the film is bonded or coated to the base fabric. In order to evaluate durability, the peel strength was measured, and the results are shown in Figure 7. The peel strength of the PNL before washing was 15.1 N/25 mm in the warp direction and 14.7 N/25 mm in the weft direction. After one wash, the peel strength decreased to 11.6 N/25 mm

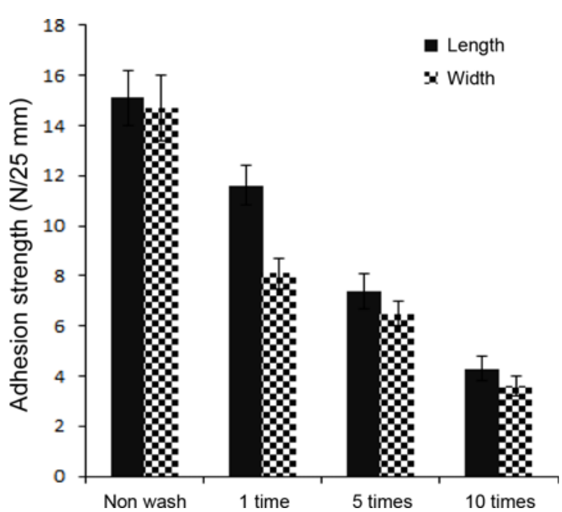


**Figure 7.** Adhesion strength of PNL according number of launderings.

in the warp direction and to 8.1 N/25 mm in the weft direction; after being washed five times the peel strength in the warp direction was 7.4 N/25 mm, and it was 6.5 N/25 mm in the weft direction. After washing 10 times, the peel strength of PNL decreased sharply to 4.3 N/25 mm in the oblique direction and to 3.6 N/25 mm in the weft direction. This result suggests that the nano-web laminate fabric has been subjected to continuous chemical and physical forces due to repeated washing. Lee *et al*. [33], also reported that the hot-melt laminating nano-web decreased the peel strength after washing on the study of the washing effect on the mechanical properties of the mass-produced nanofiber web. In order to maintain the durability of the moisture permeable and waterproof material for outdoor use, the peel strength should be 1.4 lb/in (6.212 N/in) or more [34]. It showed a durability exceeding the minimum requirement until the fifth wash, and the minimum was not reached after being washed 10 times. Therefore, the durability of the nano-web composite may vary depending on the type and amount of adhesive to various attempts to components and bonding method of the adhesive used for bonding of the base fabric and the nano web that it is necessary to implement the durability suitable for end-use.

**Antibacterial Property**

The anti-microbial activity of PNL was evaluated after repeated launderings. Table 3 shows the results of the antibacterial tests using the PET fabric in the control group and the PNL containing the JCE 1.0 % in the test group. The polyester fabric showed a bacteriostatic reduction rate of zero as shown in Table 3. Figure 8 shows that colonies formed on the agar plate with PNL and the PET fabric after incubation at 37 $^{o}$C for 24 hr. The photographs showed that a large number of colonies were formed in the PET of the control group in both the anti-bacterial test using *Staphylococcus aureus* and in the anti-bacterial test using pneumococcus. However, the pre-wash PNL showed very good anti-bacterial activity against *Staphylococcus aureus* with 99.9 % before washing, 99.9 % after one wash, 99.6 % after five washings and 99.9 % after 10 washings. The antimicrobial activity against *Klebsiella pneumoniae* was 96.5 % before washing, and 99.9 % after one, five, and 10 washings, which showed very good anti-microbial activity. These results were consistent with previous studies that reported that *juniperus chinensis* extracts containing terpenes had anti-microbial properties [37]. The anti-microbial activity of the extracts of *Juniperus chinensis* could, in part, be associated with their major constituents, i.e., a-pinene, β-phellandrene, a-terpinyl acetate, and cedrol [38,39]. Therefore, investigation into the effect of JCE in PNL suggests that JCE-loaded PNL has very effective anti-bacterial activity against both gram-positive and gram-negative bacteria even after repeated washing.

**Table 3.** Anti-microbial abilities of PNL according to the number of launderings

| Sample \ Bacteria | *Staphylococcus aureus* (gram-positive) | | | *Klebsiella pneumoniae* (gram-negative) | | |
|---|---|---|---|---|---|---|
| | 0 (hr) | 18 (hr) | Reduction of bacteria (%) | 0 (hr) | 18 (hr) | Reduction of Bacteria (%) |
| Blank | $2.2\times10^4$ | $4.1\times10^6$ | - | $1.8\times10^4$ | $4.0\times10^7$ | - |
| PET | $2.2\times10^4$ | $3.5\times10^6$ | 0 | $1.8\times10^4$ | $2.6\times10^7$ | 0 |
| Non wash | $2.2\times10^4$ | $4.5\times10^3$ | 99.9 | $1.8\times10^4$ | $1.4\times10^6$ | 96.5 |
| 1 time | $2.2\times10^4$ | $1.2\times10^3$ | 99.9 | $1.8\times10^4$ | $6.0\times10^4$ | 99.9 |
| 5 times | $2.2\times10^4$ | $1.8\times10^4$ | 99.6 | $1.8\times10^4$ | $6.0\times10^4$ | 99.9 |
| 10 times | $2.2\times10^4$ | $4.1\times10^3$ | 99.9 | $1.8\times10^4$ | $4.9\times10^4$ | 99.9 |

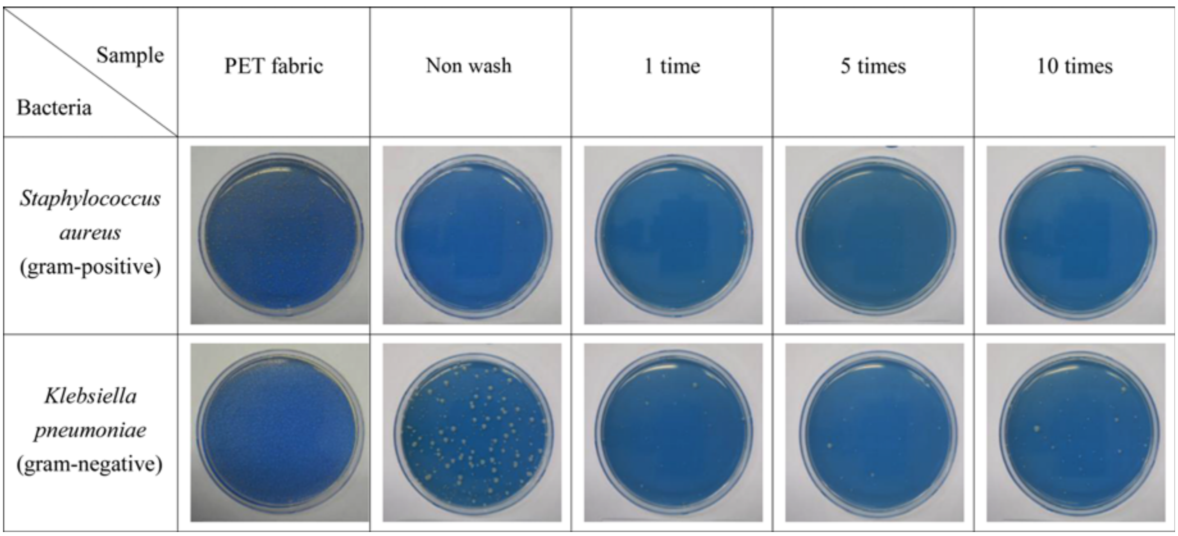


**Figure 8.** Images of anti-bacterial activities to *S. aureus* and *K. pneumoniae* of PNL according to the number of launderings.

## Conclusion

In this study, PU adhesive resin was electrosprayed on polyester, plain-weave fabric and mesh, respectively, and the PU nanoweb laminate composites (PNLs) were prepared by inserting PU nanowebs that contained *Juniperus chinensis* extracts (JCE) between two fabrics and heat-pressing them together. The physical and functional properties of the PNL were investigated according to the number of launderings. Surface images of the nanofiber webs were taken using SEM after one, five, and 10 wash cycles in order to investigate changes in morphology. The nanofiber structure of the PNL was maintained even after 10 washings. The tensile strength of PNL was found to be maintained to some extent after repeated laundering. The air permeability of PNL after repeated washings showed a tendency to be lower than before washing. Also, there was no significant change in the moisture permeability after repeated laundering. It was observed that PNL maintained its high water vapor transmission rate even after repeated washings. However, it was found that the water proofness and the adhesive strength decreased after washing. Among moisture permeable and waterproof fabrics, PNL was found to be a low water pressure type.

In order to maintain the durability of the moisture permeable and waterproof material for outdoor use, the peel strength should be 1.4 lb/in (6.212 N/in) or more. Durability exceeded the minimum requirement until the fifth wash, but after 10 washings, the minimum value was not reached. Investigation into the effect of JCE in PNL suggested that JCE loaded PNL has very effective anti-bacterial activity against both gram-positive and gram-negative bacteria, even after repeated washing.

## Acknowledgments

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## References

1. H. Lee, M. Nishino, D. Sohn, J. S. Lee, and I. S. Kim,

*Cellulose,* **25**, 2829 (2018).
2. H. Lee, G. Xu, D. Kharaghani, M. Nishino, K. H. Song, J. S. Lee, and I. S. Kim, *Int. J. Pharm.,* **531**, 101 (2017).
3. W. Chae, Z. Xing, Y. Kim, H. Sang, M. Huh, and I. Kang, *Polym.(Korea),* **35**, 210 (2011).
4. L. Jeong and W. H. Park, *Int. J. Mol. Sci.,* **15**, 6857 (2014).
5. E. H. Kim and J. H. Kim, *Text. Sci. Eng*., **53**, 75 (2016).
6. J. R. Venugopal and S. Ramakrishna, *Plant Sci. Today,* **1**, 151 (2014).
7. A. Ali, M. Macjen, M. Hamid, N. Lajis, El-Sharkawy, and M. S. Murakoshi, *J. Ethnopharmacol.,* **53**, 165 (1996).
8. C. I. Chang, W. C. Chen, Y. Y. Shao, G. R. Yeh, N. S. Yang, W. Chiang, and Y. H. Kuo, *Nat. Prod. Res.,* **22**, 1158 (2008).
9. J. H. Kim, H. Lee, A. W. Jatoi, S. S. Im, J. S. Lee, and I. S. Kim, *Mater. Lett.,* **181**, 367 (2016).
10. L. L. Shiu, W. C. Chen, and Y. H. Kuo, *Chem. Pharm. Bull.,* **47**, 557 (1999).
11. Y. H. Kuo and L. L. Shiu, *Chem. Pharm. Bull.,* **44**, 1758 (1996).
12. K. Cimanga, K. Kambu, L. Toga, S. Apers, T. Bruyne, N. Hermans, J. Totte, L. Pieters, and A. J. Vlietinck, *J. Ethnopharmacol.,* **79**, 213 (2002).
13. J. H. Kim, S. M. Park, G. S. Sim, B. H. Lee, and H. B. Pyo, *J. Soc. Cosmet. Sci. Korea*, **30**, 63 (2004).
14. A. Frenot and I. Chronakis, *Curr. Opin. Colloid Interface Sci.,* **8**, 64 (2003).
15. Z.-M. Huang, Y.-Z. Zhang, M. Kotaki, and S. Ramakrishna, *Compos. Sci. Technol.,* **63**, 2223 (2003).
16. K. W. Kim, K. H. Lee, M. S. Khil, Y. S. Ho, and H. Y. Kim, *Fiber. Polym.,* **5**, 122 (2004).
17. J. M. Deitzel, J. Kleinmeyer, D. Harris, and N. C. Beck Tan, *Polymer,* **42**, 261 (2001).
18. Z. D. Jia, Q. Li, J. Liu, Y. Yang, L. Wang, and Z. Guan, *J. Polym. Eng.,* **28**, 87 (2008).
19. S. Megelski, J. S. Stephens, D. B. Chase, and J. F. Rabolt, *Macromolecules,* **35**, 8456 (2002).
20. J. Gunn and M. Q. Zhang, *Trends Biotecnol.,* **28**, 189 (2010).
21. E. R. Kneawy and Y. R. Abdel-Fattah, *Macromo. Biosci.,* **2**, 261 (2002).
22. W. J. Li, C. T. Laurencin, E. J. Carterson, R. S. Tuan, and F. K. Ko, *J. Biomed Mater Res.,* **60**, 613 (2002).
23. H. L. Schreuder-Gibson, P. Gibson, K. Senecal, M. Sennett, J. Walker, and W. Yeomans, *J. Adv. Mater.,* **34**, 44 (2002).
24. S. Y. Ji, Y. G. Jeong, and W. H. Park, *Text. Sci. Eng*., **53**, 285 ( 2016).
25. Y. K. Jeong, S. Choi, and H. S. Kim, *Text. Sci. Eng*., **54**, 1 (2017).
26. A. Pedicini and R. J. Farris, *Polymer,* **44**, 6857 (2003).
27. A. K. Haghi and M. Akbari, *Phys. Status Solidi A,* **204**, 1830 (2007).
28. S. M. Lee, D. Kimura, A. Yokoyama, K. H. Lee, J. C. Park, and I. S. Kim, *Text. Res. J.,* **79**, 1085 (2009).
29. J. H. Yang, N. S. Yoon, I. K. Kim, and J. H. Yeum, *Text. Coloration Finish.,* **23**, 107 (2011).
30. Y. K. Kang, C. H. Park, J. Kim, and T. J. Kang, *Fiber. Polym.,* **8**, 564 (2007).
31. B. Yoon and S. Lee, *Sci. Emot. Sensib.,* **13**, 391 (2010).
32. B. Yoon and S. Lee, *Fiber. Polym.,* **12**, 57 (2011).
33. S. Lee, D. Kimura, K. H. Lee, J. C. Park, and I. S. Kim, *Text. Res. J.,* **80**, 99 (2010).
34. K. Lee, B. Yoon, and S. Lee, *J. Kor. Soc. Cloth. Ind.,* **14**, 122 (2012).
35. S. Lee and S. K. Obendorf, *J. Text. Inst.,* **98**, 87 (2007).
36. E. A. Kim, S. Yoo, and J. Kim, *Fiber. Polym.,* **4**, 215 (2003).
37. H. J. D. Dorman and S. G. Deans, *J. Appl. Microbiol.,* **88**, 308 (2000).
38. S. Cosentino, C. I. G. Tuberoso, B. Pisano, V. Mascia, E. Arzedi, and F. Palmas, *Lett. Appl. Microbiol.,* **29**, 130 (1999).
39. A. L. Oliveira, R. B. Lopes, F. A. Cabral, and M. N. Eberlin, *Food Chem.,* **99**, 1 (2006).