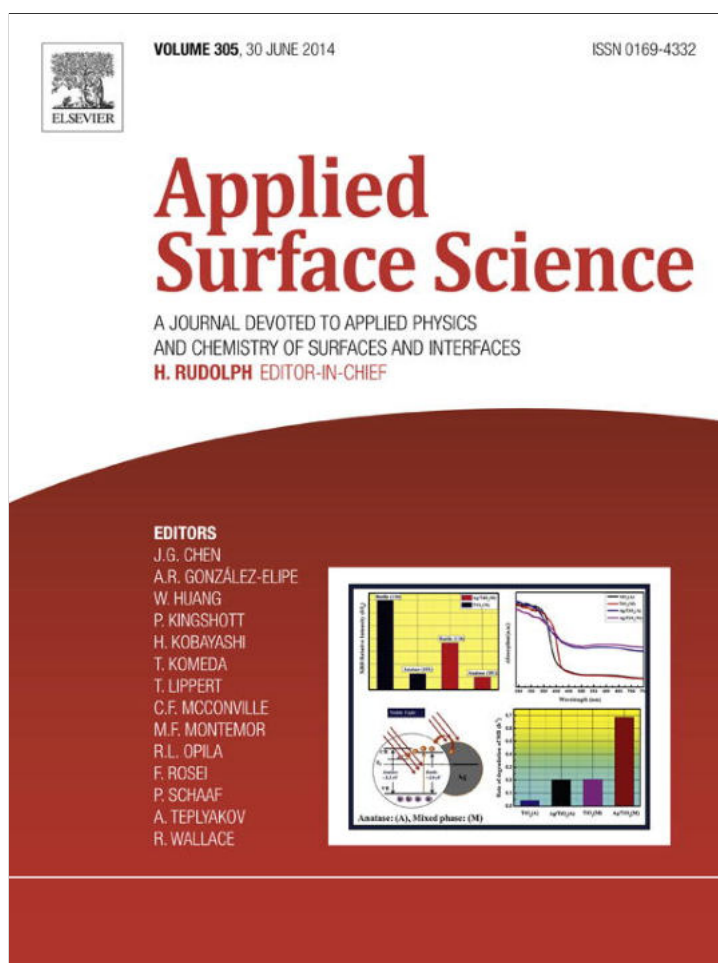


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Laser-drilled micro-hole arrays on polyurethane synthetic leather for improvement of water vapor permeability



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ABSTRACT

Three kinds of lasers at 1064, 532 and 355 nm wavelengths respectively were adopted to construct micro-hole arrays on polyurethane (PU) synthetic leather with an aim to improve water vapor permeability (WVP) of PU synthetic leather. The morphology of the laser-drilled micro-holes was observed to optimize laser parameters. The WVP and slit tear resistance of the laser-drilled leather were measured. Results show that the optimized pulse energy for the 1064, 532 and 355 nm lasers are 0.8, 1.1 and 0.26 mJ, respectively. The diameters of the micro-holes drilled with the optimized laser pulse energy were about 20, 15 and 10 μm , respectively. The depths of the micro-holes drilled with the optimized pulse energy were about 21, 60 and 69 μm , respectively. Compared with the untreated samples, the highest WVP growth ratio was 38.4%, 46.8% and 53.5% achieved by the 1064, 532 and 355 nm lasers, respectively. And the highest decreasing ratio of slit tear resistance was 11.1%, 14.8%, and 22.5% treated by the 1064, 532 and 355 nm lasers, respectively. Analysis of the interaction mechanism between laser beams at three kinds of laser wavelengths and the PU synthetic leather revealed that laser micro-drilling at 355 nm wavelength displayed both photochemical ablation and photothermal ablation, while laser micro-drilling at 1064 and 532 nm wavelengths led to photothermal ablation only.

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1. Introduction

Water vapor permeability (WVP) of polyurethane (PU) synthetic leather is of great importance when PU synthetic leather is used for the manufacture of shoes and garments [1]. In order to keep human bodies warm and comfortable, shoes and garments should have high WVP value which allows perspiration to evaporate promptly, especially when human bodies are in hot environments. Relative humidity inside the shoes and garments will decrease when water vapors transfer through the PU synthetic leather into the environment. This subsequently leads to a decrease in thermal conductivity of the insulating air, resulting in guarding against a damp feeling [2–6]. In this way, WVP has significant influence on thermal comfort properties of shoes and garments. However, PU synthetic leather exhibits low WVP, despite the excellent physical properties it has. The low WVP of the PU synthetic leather is mainly caused by the PU film on the top layer. The PU film has a remarkably negative impact on the WVP, though its thickness is small [3,7,8]. According

to the results reported by Lewin [9], WVP of PU synthetic leather decreased by 30–50% when compared with unfinished leather. Many researches have been done to improve the WVP of PU film. The present investigations mainly focused on the ways to promote surface hydrophilic property and porous structures of the PU film by chemical process. Zuo et al. [10] prepared blend membranes of PU and superfine chitosan powder by immersion precipitation phase inversion method. They found that the WVP was improved remarkably with increasing superfine chitosan powder content. Chen et al. [11] enhanced WVP of PU synthetic leather by oxygen plasma treating. Liu et al. [12] used solution casting method to blend down powder with PU to improve the WVP of PU membrane. They found that large pore size of composite membranes and hydrophilic groups on the surface of down powder lead to improvement of the WVP. Han et al. [13] modified PU by hydrophilic DMPA segments to improve the WVP of PU. Chwang et al. [14] used ethylene glycol and other strong hydrophilic compounds to prepare modified PU. In this way, the WVP was raised.

Slit tear resistance is of great value to leather when they are used to manufacture shoes and garments, since better slit tear resistance enables leather to withstand tearing stresses. As a result, decrease in slit tear resistance can affect durability of leather.

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Compared with other manufacturing methods, laser micro-drilling has many advantages. For example, it has good flexibility, high efficiency, as well as the ability to control the duration of the energy deposition process [15,16]. Many researches on laser micro-drilling have been reported up to now. Pan et al. [15] fabricated hole arrays with a 50 μm thick PI film by using a 248 nm excimer laser. They found that laser fluence has a great influence on the diameter of the hole. Chen et al. [16] used UV excimer laser drilling a submicron via hole inside a bigger via hole. They thought that the refocusing of the reflected laser light from the side-wall of the bigger hole and the wave-guide effect of the light trapped inside the smaller hole led to the formation of the smaller via hole. Tokarev et al. [17] developed a model of multipulse excimer laser drilling in polymers. They tried to investigate the particular mechanism of radiation propagation and absorption inside the keyhole, to identify the factors that control the formation of the keyhole and to optimize the drilling. Tan [18] achieved 20 μm holes in a 250 μm thick silicon substrate by using a nanosecond UV laser. He successfully eliminated the deformation caused by plasma shielding with a multi-burst pulse train. Seet et al. [19] got hole arrays on PVC templates by using a 355 nm diode-pumped solid state Nd:YAG laser. He pointed out that the laser fluence, laser irradiation time, number of pulses and focal point all could affect the quality of the drilled holes. Liu et al. [20] formed circular and rectangular via holes in 300 μm thick bulk 4H-SiC substrates by a 193 nm UV laser. Yalukova et al. [21] compared the drilled holes in fiber reinforced polymer and non-reinforced thermoplastic sheets using three wavelengths, 1064, 532, and 266 nm. They pointed out that by using UV light, bond breaking rather than thermal material removal occurred.

Rather than promoting neither the surface hydrophilic property nor the porous structures of PU synthetic leather by chemical process, we fabricated micro-hole arrays on the PU synthetic leather by a laser micro-drilling process to improve the WVP of the PU synthetic leather in this study. In order to obtain small drilled micro-holes, lasers with different wavelengths were used. The influence of pulse energy on the morphology of the micro-holes was investigated. The interaction mechanism between laser beams at three kinds of wavelengths and the PU synthetic leather was studied. After micro-hole arrays being drilled on the PU synthetic leather, the WVP and slit tear resistance were then tested.

2. Materials and methods

In this study, the thickness of the PU synthetic leather is about 1.3 mm, and the thickness of the PU layer is about 60–70 μm . Laser micro-drilling of PU synthetic leather was carried out by adopting three kinds of lasers with different wavelengths, i.e., a SPI 20W/HS laser at 1064 nm wavelength, a Huaray Cedar-532/20B diode-pumped Nd:YAG laser at 532 nm wavelength and a Huaray Cedar-355/10B diode-pumped Nd:YAG laser at 355 nm wavelength. The specifications of the lasers are given in Table 1. Focal length of objective lens adopted by the 1064, 532 and 355 nm lasers is 100, 120 and 100 mm, respectively. Numerical aperture of objective lens equipped the 1064, 532 and 355 nm lasers is 0.22, 0.15 and 0.18, respectively. For the 1064 nm laser, magnification of beam expanding lens is 75. For the 532 and 355 nm laser, magnification of beam expander is 10. The morphology of the laser-drilled micro-holes was observed by a JEOL-7600F scanning electron microscope (SEM) and a Dino-Lite digital microscope. The micro-hole density was varied from 10,000 to 40,000 micro-holes per square centimeter to investigate the effect of the micro-hole density on WVP and slit tear resistance.

WVP was measured in a round mouth plastic cup filled with distilled water. Laser-drilled PU synthetic leather was placed over

the top of the cups. The cups were placed in a chamber where the temperature was constant at 20 °C and the relative humidity was at 70%. The weight loss after 24 h was measured. For each WVP measurement, on an average of three different readings was used. The result of WVP was calculated using the following equation:

$$\text{WVP} = \frac{G}{tA} \quad (1)$$

where G is weight change in milligram, t the duration of measurement in hour and A the measurement area in square centimeter.

The slit tear resistance of laser-drilled PU synthetic leather was evaluated with a universal material testing machine (Zwick/Roell, Z020) according to ASTM D2212. The speed of the clip was 100 mm/min.

3. Results

3.1. Laser micro-drilling parameters of the PU synthetic leather

The morphology of micro-hole arrays, front view of micro-holes and cross section of micro-holes drilled by different lasers under variable pulse energy are shown in Figs. 1–4, respectively. When the 1064 nm laser was adopted, it could be seen from Fig. 1 that no micro-hole but only some thermal degradation and shrinkage were observed at the laser pulse energy lower than 0.2 mJ, and obvious micro-hole was observed at the laser pulse energy higher than 0.3 mJ. Fig. 2 shows that micro-holes could be drilled using the 532 nm laser at the pulse energy higher than 0.5 mJ, and there was no burning but a heat affected zone (HAZ) around the micro-holes. Micro-holes could be drilled using the 355 nm laser at the pulse energy higher than 0.18 mJ, as shown in Fig. 3. It was clear that the variation of pulse energy had a remarkable influence on diameter and depth of the micro-holes. With an increase in pulse energy, diameter of the laser-drilled micro-hole at 1064 nm wavelength increased from 15 μm to 20 μm ; diameter of the micro-hole drilled with the focused 532 nm laser beam increased from 12 μm to 15 μm ; the diameter of the micro-hole drilled by the 355 nm laser increased from 8 μm to 10 μm . In addition, diameter of the HAZ in Fig. 2 increased from 27 μm to 32 μm , with an increase in pulse energy. Moreover, depth of the micro-hole drilled with the focused 1064 nm laser beam increased from 31 μm to 69 μm with an increase in pulse energy from 0.23 mJ to 0.26 mJ; depth of the micro-hole drilled by the 532 nm laser increased from 34 μm to 60 μm with an increase in pulse energy from 0.7 mJ to 1.1 mJ; depth of the laser-drilled micro-hole at 355 nm wavelength increased from 31 μm to 69 μm with an increase in pulse energy from 0.23 mJ to 0.26 mJ. Furthermore, in comparison with 1064 nm-laser and 532 nm-laser drilled specimens, thermal influence was significantly reduced when the 355 nm laser was adopted.

It is of great significance that the depth of the laser-drilled micro-holes should be deep enough in order to penetrate the PU films that cover the synthetic leather. Taking the results of the earlier parameters studies into consideration, the optimized pulse energy was used for different kinds of lasers to drill different micro-hole densities. The optimized laser parameters for three kinds of laser wavelengths are summarized in Table 2.

3.2. The effect of laser wavelength and micro-hole density on the WVP

Fig. 4 shows the WVP value of the laser-drilled PU synthetic leather. The samples for WVP test were prepared according to the optimized laser parameters listed in Table 2. It could be observed that the WVP showed an upward trend with a decrease in laser wavelength. The WVP values of all laser-drilled samples were significantly influenced by laser beam wavelength at the fixed

Table 1
Specification of lasers used in the experiments.

Wavelength (nm)	Laser mode	M^2	Maximum power (W)	Pulse duration (ns)	Frequency (kHz)
IR: 1064	TEM ₀₀	≤ 2	20	200	25
Green: 532	TEM ₀₀	≤ 2	11	25	10
UV: 355	TEM ₀₀	≤ 2	7	11	30

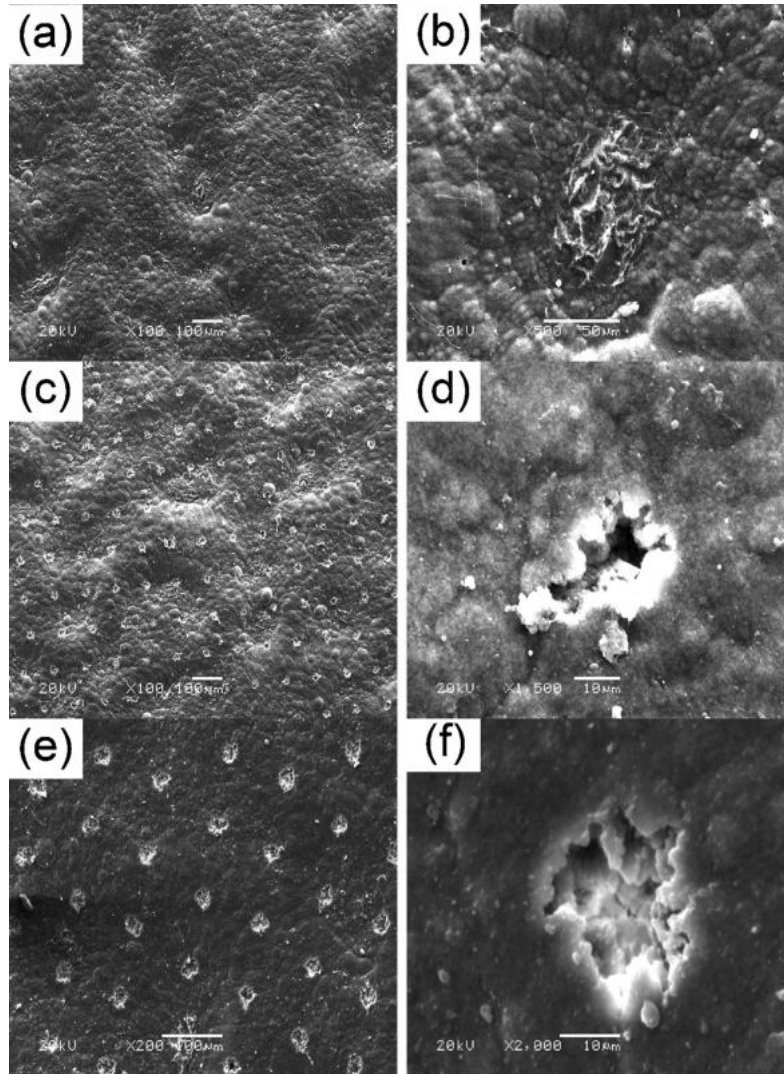


Fig. 1. Micro-hole arrays and micro-holes drilled with a focused beam at 1064 nm. (a) and (b) The pulse energy is 0.2 mJ. (c) and (d) The pulse energy is 0.3 mJ. (e) and (f) The pulse energy is 0.8 mJ.

Table 2
The optimized laser parameters for three kinds of lasers.

Laser	Expt. No.	Pulse energy (mJ)	Micro-hole density (number of micro-holes per sq cm)	Micro-hole spacing (mm)
IR: 1064 nm	I1	0.8	10,000	0.1
	I2		20,000	0.07
	I3		30,000	0.06
	I4		40,000	0.05
Green: 532 nm	G1	1.1	10,000	0.1
	G2		20,000	0.07
	G3		30,000	0.06
	G4		40,000	0.05
UV: 355 nm	U1	0.26	10,000	0.1
	U2		20,000	0.07
	U3		30,000	0.06
	U4		40,000	0.05

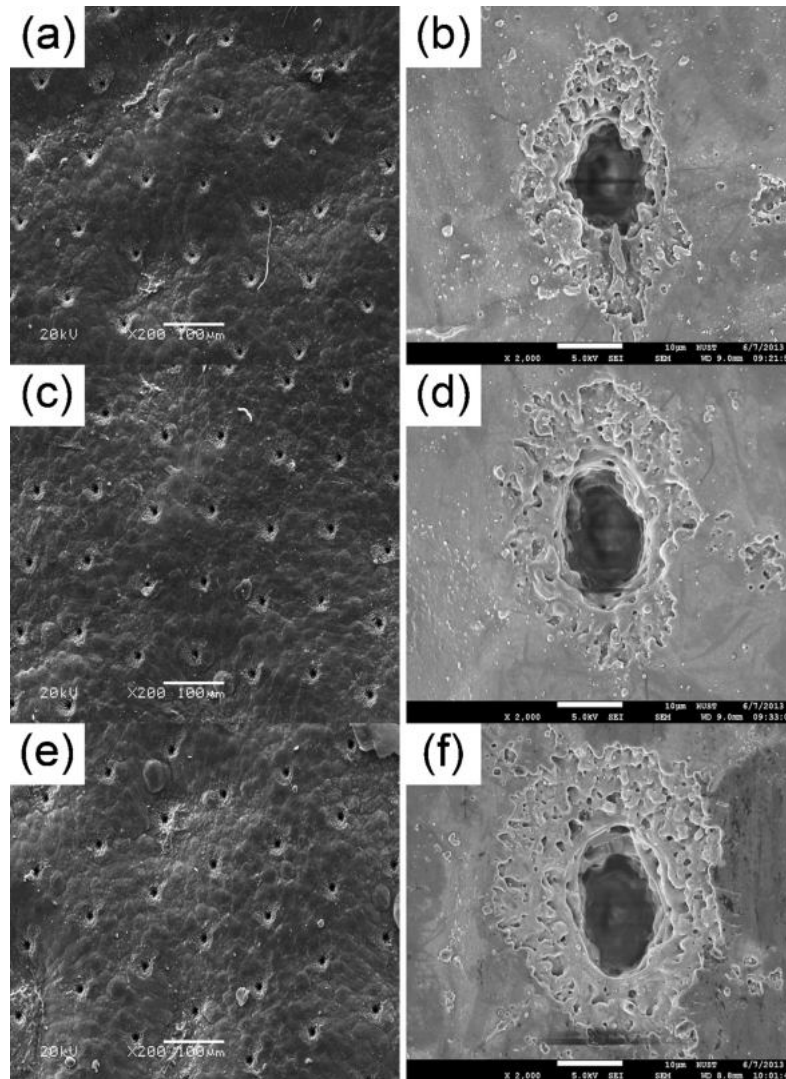


Fig. 2. Micro-hole arrays and micro-holes drilled with a focused beam at 532 nm. (a) and (b) The pulse energy is 0.5 mJ. (c) and (d) The pulse energy is 0.7 mJ. (e) and (f) The pulse energy is 1.1 mJ.

micro-hole density. The shorter the laser wavelength, the higher the WVP value was. Meanwhile, the greater the micro-hole density, the higher the WVP value was. The highest WVP growth ratio was 38.4%, 46.8% and 53.5% achieved by the 1064 nm laser, the 532 nm laser and the 355 nm laser, respectively. In comparison with oxygen plasma treating method (with the highest WVP growth ratio of 40.2%) reported by Chen et al. [11], both 532 nm-laser and 355 nm-laser drilled PU synthetic leathers present higher WVP growth ratio. In addition, it takes 120 s for the oxygen plasma treatment to get the highest WVP growth ratio. Meanwhile, laser micro-drilling process spends only about 10 s to get the maximum WVP value. It is obvious that laser micro-drilling process is characterized by a much higher efficiency.

3.3. The effect of laser wavelength and micro-hole density on the slit tear resistance

Fig. 5 reveals the slit tear resistance of the laser-drilled samples. The samples for slit tear resistance test were prepared according to the optimized laser parameters listed in Table 2. The slit tear resistance showed an upward trend with an increase in laser wavelength. The 1064 nm laser-drilled samples presented the highest slit tear resistance while the 355 nm laser-drilled samples showed

the lowest slit tear resistance. Additionally, the slit tear resistance decreased with an increase in micro-hole density. Amongst all laser-drilled samples, the slit tear resistance minimized when the micro-hole density was 40,000 micro-holes per square centimeter. The highest decreasing ratio of slit tear resistance was 11.1%, 14.8%, and 22.5% treated by the 1064 nm laser, the 532 nm laser and the 355 nm laser, respectively. The slit tear resistance of the laser-drilled samples is still higher than that of natural leather, although laser micro-drilling can lead to decrease of slit tear resistance [22–24]. Consequently, laser micro-drilling will not affect performance of the laser-drilled PU synthetic leather.

4. Discussion

During the micro-drilling process, laser energy was supplied to the surface of the target material. By absorbing the laser energy, the molecular vibrations of the PU synthetic leather increased. Then the material was heated up, since the photo energy was transferred to internal potential energy of the PU synthetic leather. The rising temperature would result in the burning, melting or vaporization of the drilled region.

It could be seen from Figs. 1–4 that the diameters of the micro-holes, the depths of the micro-holes and the sizes of the HAZ

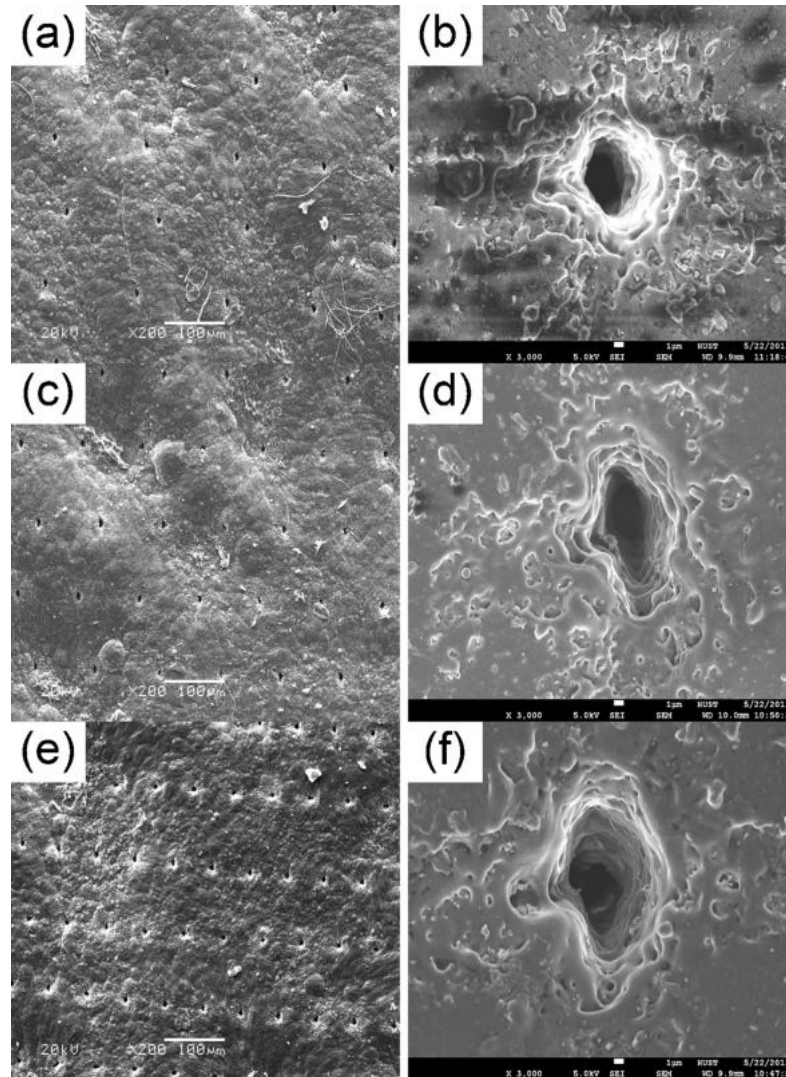


Fig. 3. Micro-hole arrays and micro-holes drilled with a focused beam at 355 nm. (a) and (b) The pulse energy is 0.18 mJ. (c) and (d) The pulse energy is 0.23 mJ. (e) and (f) The pulse energy is 0.26 mJ.

increased with an increase of the pulse energy. At higher pulse energy, more energy would be supplied to the sample. Laser irradiation would cause more materials to be ablated. As a result, a larger and deeper micro-hole would be produced.

It could be observed from Fig. 4 that the depths of the micro-holes drilled by the 532 nm laser and the 355 nm laser are much deeper than those of the micro-holes drilled by the 1064 nm laser at the fixed pulse energy. As mentioned early, laser energy could be absorbed and converted to the internal potential energy of the PU synthetic leather, resulting to the ablation of materials. The photon energy of 1064 nm laser, 532 nm laser and 355 nm laser are 1.17 eV, 2.33 eV and 3.56 eV, respectively. This meant that less energy could be transferred to the internal potential energy when the 1064 nm laser was adopted. A shallow micro-hole would be produced, as the depth of the micro-holes was dependent on the rate of heat transfer.

It could be found from Figs. 1–4 that the thermal effects were significantly reduced when the 355 nm laser was utilized. Micro-drilling with the 1064 nm laser and the 532 nm laser led to photothermal ablation only. The laser micro-drilling by the 355 nm laser had both photochemical ablation and photothermal ablation. The absorption of laser energy is mainly determined by the applied wavelength. The main chemical bonds that PU synthetic

leather consists of are C=O, N–H, C–O, and C–N bonds. The strength of a C–O bond is approximately 3.39 eV. The strength of a C–N bond is approximately 3.17 eV. The strength of an N–H bond is approximately 4.05 eV. The strength of a C=O bond is approximately 7.57 eV. Neither the 1064 nm laser nor the 532 nm laser can break the chemical bonds, because of the fact that the photon energy of the 1064 nm laser and the 532 nm laser are 1.17 eV and 2.33 eV, respectively. For both of the two kinds of lasers, the thermal influence is dominant. They can only decompose the PU synthetic leather by photothermal ablation. On the other hand, the photon energy of the 355 nm laser is 3.56 eV, which enables it to break the C–O and C–N covalent bonds by photochemical dissociation. Therefore, the thermal effects decrease remarkably, resulting to the significantly improved quality of the micro-holes.

Fig. 5 shows that the WVP value increases with an increase of the micro-density. This can be explained by the fact that with more micro-holes, a higher amount of water vapor molecules can be able to transmit through the PU films easily. The micro-hole arrays provide the water vapor molecules with a passage to get into the environment with less resistance, resulting to a significantly improved WVP.

Fig. 5 also illustrates that with a decrease of the wavelength, the WVP shows an upward trend at the fixed micro-hole density. The

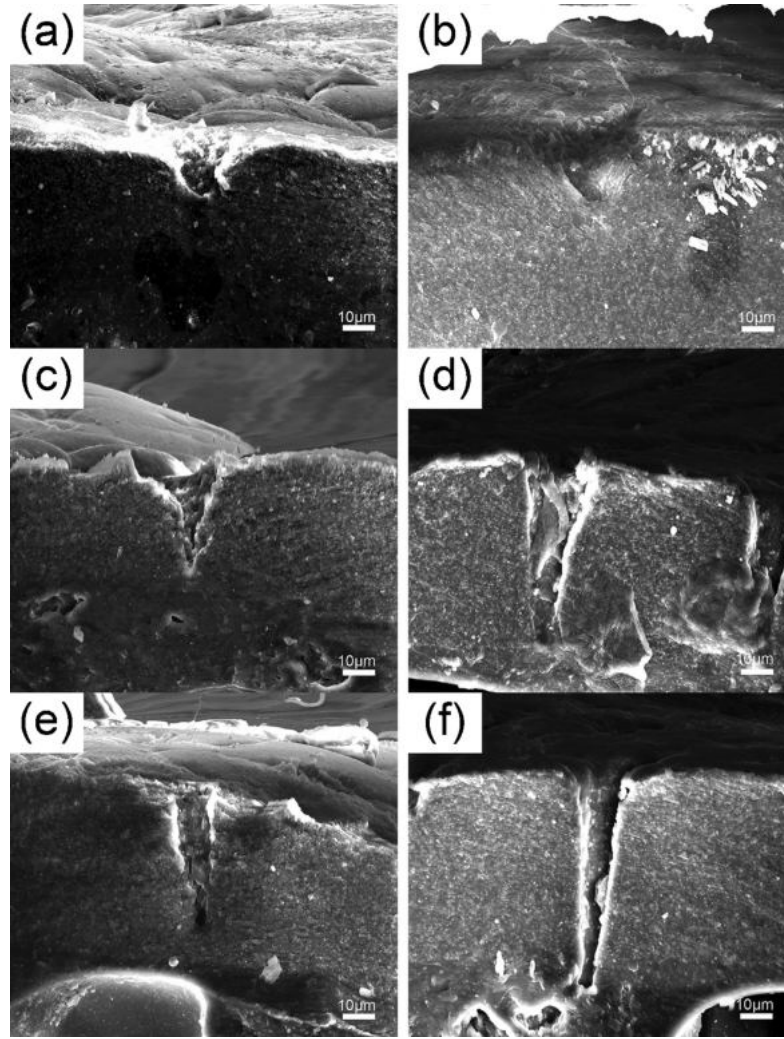


Fig. 4. Cross section of the micro-holes drilled by different lasers. (a) Drilled by a 1064 nm laser at 0.3 mJ. (b) Drilled by a 1064 nm laser at 0.8 mJ. (c) Drilled by a 532 nm laser at 0.7 mJ. (d) Drilled by a 532 nm laser at 1.1 mJ. (e) Drilled by a 355 nm laser at 0.23 mJ. (f) Drilled by a 355 nm laser at 0.26 mJ.

WVP is determined by the depth and the diameter of the laser-drilled micro-hole when the micro-hole density is fixed. Larger depth means that the micro-hole is more likely to penetrate the PU film. In this way, water vapor molecules are able to transmit

through the PU films with fewer blockages. Additionally, larger diameter can make more water vapor molecules to pass through the PU film, leading to a higher WVP value. The samples drilled by the 1064 nm laser gained the least amount of WVP value, because

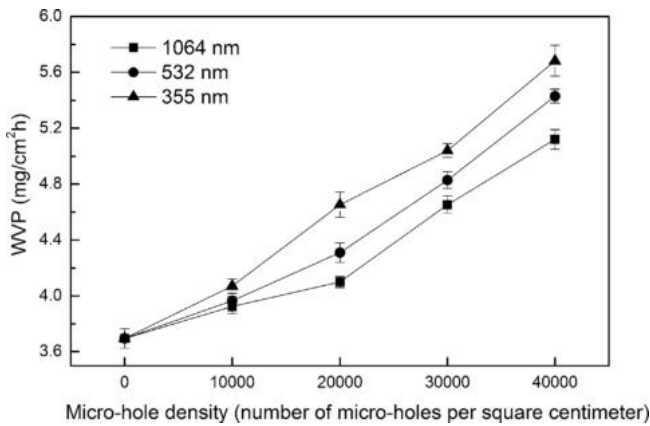


Fig. 5. WVP value of the laser-drilled PU synthetic leather. The WVP value increases with an increase of the micro-hole density. The samples drilled by the 355 nm laser exhibit the maximum WVP.

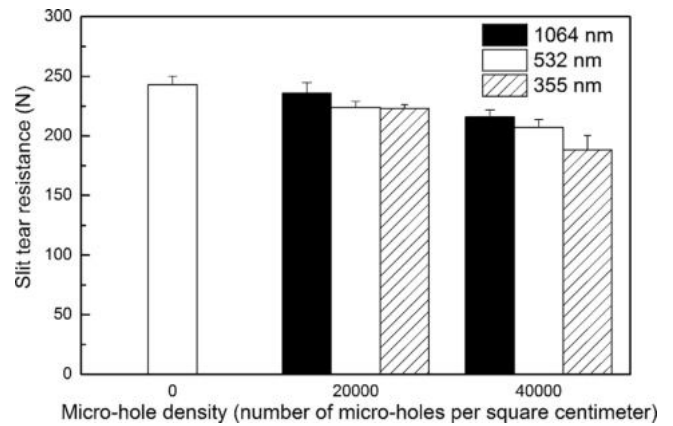


Fig. 6. Slit tear resistance of the laser-drilled PU synthetic leather. Slit tear resistance decreases with a decrease of the micro-hole density. The samples drilled by the 1064 nm laser exhibit the maximum slit tear resistance.

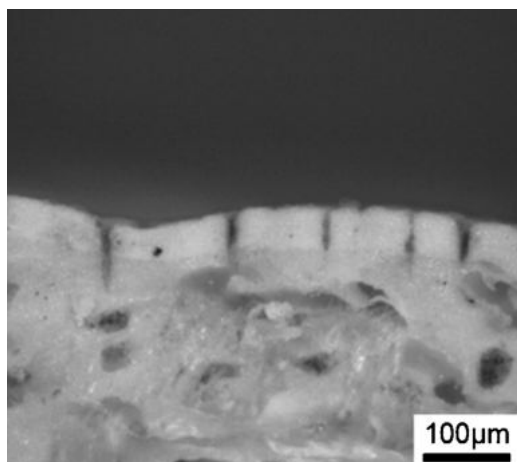


Fig. 7. Cross section of micro-hole arrays drilled by the 355 nm laser at 0.26 mJ.

the depth of the micro-hole is the minimum as shown in Fig. 4. In this case, though the diameter of the laser-drilled micro-hole is 20 μm , the depth of the micro-hole is so small that the PU film can still prevent the evaporation of water significantly. Therefore, the samples produced by the 1064 nm laser exhibited the lowest WVP value. Moreover, the samples drilled by the 355 nm laser displayed the maximum WVP value, although the diameter of the micro-hole drilled by the 355 nm laser is the minimum. The micro-hole drilled by the 355 nm laser is more likely to penetrate the PU film, since the depth of the micro-hole drilled by the 355 nm laser is the maximum. Fig. 7 shows cross section of the micro-hole arrays drilled by the 355 nm laser at 0.26 mJ. It could be seen from Fig. 7 that some micro-holes penetrated the PU film. Consequently, a higher amount of water vapor molecules could transmit through the PU films promptly. Based on the discussion mentioned above, it is believed that depth of the micro-hole is the most important factor for improvement of WVP.

It can be found from Fig. 6 that slit tear resistance decreases with an increase of the micro-hole density. The micro-hole arrays on the PU film lead to the decline of the mechanical property of the PU synthetic leather, due to the stress concentration around the micro-holes. Under the effect of outside force, micro-cracks are more likely to occur near the micro-holes initially. Then these micro-cracks will grow and eventually connect with other micro-cracks which initiates from nearby micro-holes, thus forming a crack which is large enough to lead to the break of the PU synthetic leather. The increase of micro-hole density will cause a decline of the mechanical property, since more micro-holes mean a more serious stress concentration. Besides, as more micro-holes in the PU film, there will be more crack sources. What is more, when there are more micro-holes, the distance between the adjacent micro-holes will get smaller, resulting to an easy connection of the micro-holes.

Fig. 6 reveals that with an increase of the wavelength, the slit tear resistance shows an upward trend at the fixed micro-hole density. The diameter and depth of the micro-holes have a remarkable influence on the slit tear resistance. Larger diameter and depth will lead to a lower slit tear resistance due to a much more serious stress concentration. The samples drilled by the 1064 nm laser exhibit the greatest slit tear resistance, because the depth of the micro-holes is the minimum as shown in Fig. 4. In this case, though the diameter of the laser-drilled micro-hole is 20 μm , the depth is so low that the samples drilled by the 1064 nm laser have the greatest slit tear resistance. It is clear in Fig. 4 that the micro-holes drilled by the 355 nm laser have a larger depth than the micro-holes drilled by the 532 nm laser do. In this way, the samples drilled by the 355 nm laser display the minimum slit tear resistance.

5. Conclusion

Three different wavelengths, 1064, 532 and 355 nm were applied to construct micro-hole arrays on the surface of the PU synthetic leather for improvement of WVP, and the optimized pulse energy for the 1064, 532, and 355 nm laser are 0.8, 1.1, and 0.26 mJ, respectively. The diameter of the laser-drilled micro-holes increases with an increase in pulse energy. The influences of laser beam wavelength and pulse energy on the diameter of the micro-holes are concluded as follows: for the 1064 nm laser-drilled samples, the diameter changes from 15 μm to 20 μm at the pulse energy ranging from 0.3 mJ to 0.8 mJ; for the 532 nm laser-drilled samples, the diameter changes from 12 μm to 15 μm at the pulse energy ranging from 0.5 mJ to 1.1 mJ; for the 355 nm laser-drilled samples, the diameter changes from 8 μm to 10 μm at the pulse energy ranging from 0.18 mJ to 0.26 mJ. In addition, increase in pulse energy makes the depths of the micro-holes increase as well. The maximum depths of the micro-holes are about 21, 60 and 69 μm achieved by the 1064 nm laser, the 532 nm laser and the 355 nm laser, respectively. Analysis of the interaction mechanism between laser beams at three kinds of laser wavelengths and the PU synthetic leather shows that laser micro-drilling at 355 nm wavelength displays both photochemical ablation and photothermal ablation, while laser micro-drilling at 1064 and 532 nm wavelengths leads to photothermal ablation only. The WVP increases significantly after laser micro-drilling. Compared with the untreated samples, the highest WVP growth ratio is 38.4%, 46.8% and 53.5% achieved by the 1064 nm laser, the 532 nm laser and the 355 nm laser, respectively. Laser micro-hole drilling also leads to decrease of slit tear resistance. Compared with the untreated samples, the highest decreasing ratio of slit tear resistance is 11.1%, 14.8%, and 22.5% treated by the 1064 nm laser, the 532 nm laser and the 355 nm laser, respectively. The decrease in slit tear resistance does not influence performance of PU synthetic leather, since the slit tear resistance of the laser-drilled samples is still higher than that of natural leather. Samples drilled by the 355 nm laser display the maximum WVP value as well as satisfactory slit tear resistance. Consequently, the best laser system for patterning PU synthetic leather is the 355 nm laser.

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