

Optimization of Recovery and Relaxation of Acrylic Pressure-Sensitive Adhesives by Using UV Patterning for Flexible Displays

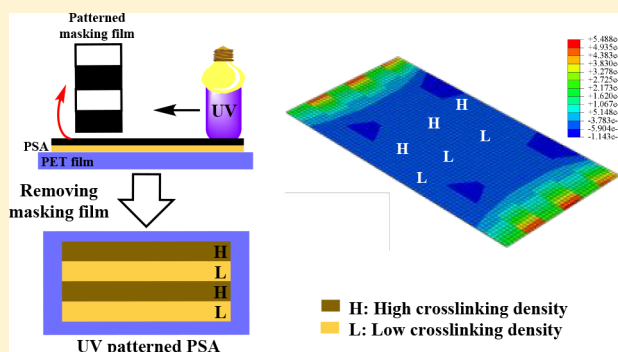
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ABSTRACT: For flexible displays, recovery and relaxation of acrylic pressure-sensitive adhesives (PSA) must be enhanced; however, only a few studies have focused on their optimization. High cross-linking density of the PSA leads to improved recovery but deteriorates the stress relaxation; thus, it is difficult to perform optimization by simply controlling the cross-linking density. Herein, it was determined that a UV-patterned PSA with both high and low cross-linking densities in a single layer enables the optimization of both recovery and relaxation. By introducing the UV-patterned PSA, the elasticity and recovery largely deteriorated but the stress relaxation was significantly improved, compared to that of the nonpatterned PSA, and this effect was enhanced with increases in the applied strain. Thereby, the recovery and relaxation were well-optimized with both values above 71% only at 300% strain. The recovery and relaxation of PSA had, respectively, a positive and negative correlation with the storage modulus.



1. INTRODUCTION

Pressure-sensitive adhesives (PSAs) are polymeric materials that are permanently tacky at room temperature in dry film form. PSAs firmly adhere to various substrates and have the ability to bind two different substrates.^{1,2} PSAs are classified by their composition, such as acrylate, silicone, rubber, and so on. In particular, acrylic PSA has many advantages, including high transparency, adhesion performance, UV-curable property, and resistance to aging, and thus, it has been widely used for various applications, such as labeling, protective films, medical application, automotive parts, and displays.^{3–10}

In display applications, PSA is widely used for the assembly of the display components.^{11–15} Since the display has developed from a rigid plate shape to having a curved shape or flexible nature, the flexibility of display components has been investigated.^{16–18} PSA, which is one of the important components of a flexible display (Figure 1), has also been investigated in order to characterize and enhance flexibility,¹⁹ because poor flexibility of PSA may lead to serious defects such as delamination during the period of use of the display. Our research group previously attempted to determine the factors influencing the flexibility of a PSA, such as molecular weight and cross-linking density. The effect of cross-linking density on the flexibility of PSA was evaluated by comparing the recovery and relaxation of PSA; however, the optimization of the two properties has not been investigated extensively yet.^{20–22}

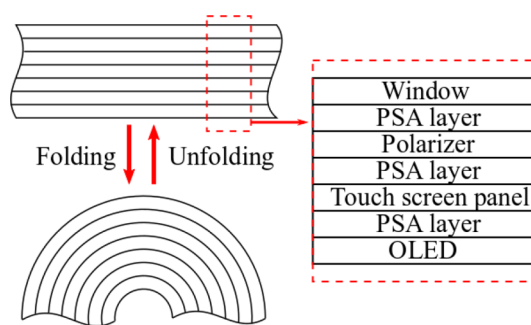


Figure 1. Structure of flexible display and its composition.

The recovery of PSA is important for flexible displays, because a high recovery of PSA prevents it from sagging under repeated folding–unfolding conditions. Furthermore, relaxation is also required because low stress relaxation causes high levels of residual stress, which can lead to delamination, stress corrosion, cracking, and reduction in fatigue life of the assembled display.²³

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Despite the significance of recovery and relaxation for the PSA for flexible displays, only a few studies have attempted to optimize the two properties. Lee et al. determined that increasing the cross-linking density of PSA resulted in enhancement of recovery but degradation of relaxation.²² Zhang et al., also suggested that cross-linking lead to improvement in the elasticity and recovery of the PSA but caused a reduction in the relaxation.²⁴ However, all these studies only focused on characterizing the recovery and relaxation according to the cross-linking density, rather than optimizing the two properties.

In this research, we prepared a UV-patterned PSA containing both high and low cross-linking densities into a single PSA layer to optimize both the recovery and relaxation. The cross-linking density of PSA was controlled by a UV dose and the curing

behavior and mechanical strength of the PSA were estimated. Furthermore, the recovery and relaxation for nonpatterned and patterned PSAs were evaluated.

2. MATERIALS AND METHODS

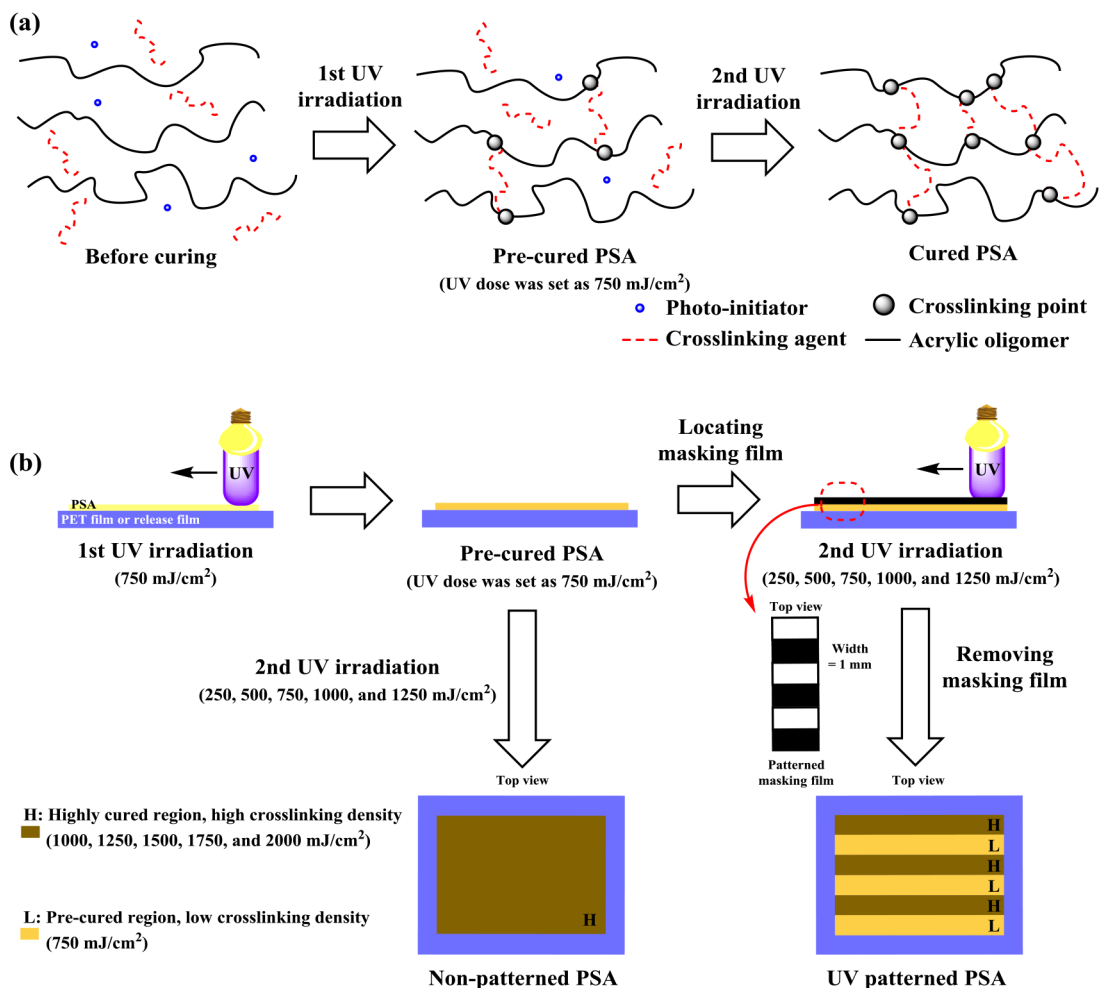
2.1. Preparation of UV-Patterned Acrylic PSA. Acrylic oligomer was synthesized by bulk polymerization of the acrylic monomer.^{11,25,26} The specific composition of the oligomer is presented in Table 1. The cross-linking agent (polyethylene glycol 200 dimethacrylate, Sartomer SR210, Sartomer, USA) and photoinitiator (2-hydroxy-2-methyl-1-phenyl-propan-1-one, Irgacure 1173, BASF, Germany) were added as 1 wt % of the synthesized oligomer.

Acrylic PSA was prepared by UV curing of the mixture, the curing mechanism of which is shown in Scheme 1a. The PSA was coated on a silicone release film by film applicator with 50 μm . Through UV irradiation (intensity = 100 mW/cm^2), a radical was formed from the photoinitiator, followed by coupling reaction with a C=C double bond in both cross-linking agent and acrylic oligomer. After the first UV irradiation, the precured PSA, which had low cross-linking density, was prepared into a film form. The minimum UV dose for the formation of the PSA film from viscous liquid state was 750 mJ/cm^2 , and thus, we set the UV dose for the precured PSA as 750 mJ/cm^2 in the process. As shown in Scheme 1b, the precured PSA with the masking film was exposed to a

Table 1. Composition of Acrylic Oligomer

classification	composition	content (wt %)
acrylic monomer	2-ethylhexyl acrylate	64
	isobornyl acrylate	19
	acrylic acid	3
	methyl methacrylate	4
	2-hydroxyethyl acrylate	10
photo-initiator	2-hydroxy-2-methyl-1-phenyl-propan-1-one	0.1 phr

Scheme 1. Preparation of UV-Patterned PSA: (a) Curing Mechanism of PSA and (b) Process of UV Patterning of PSA



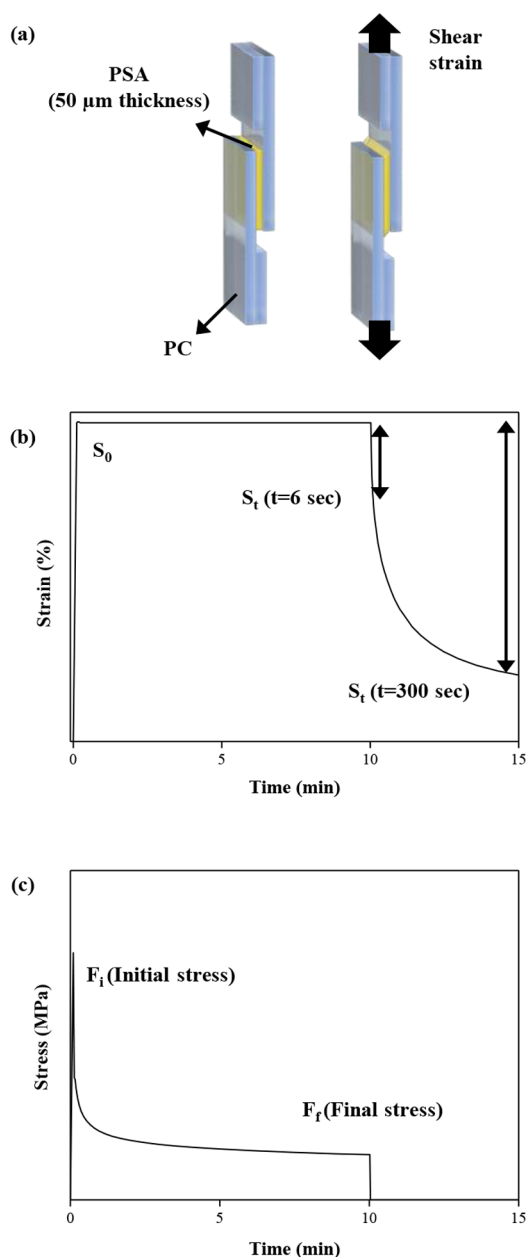


Figure 2. Methodology for evaluating recovery and relaxation of PSA: (a) test specimen, (b) strain recovery, and (c) stress relaxation.

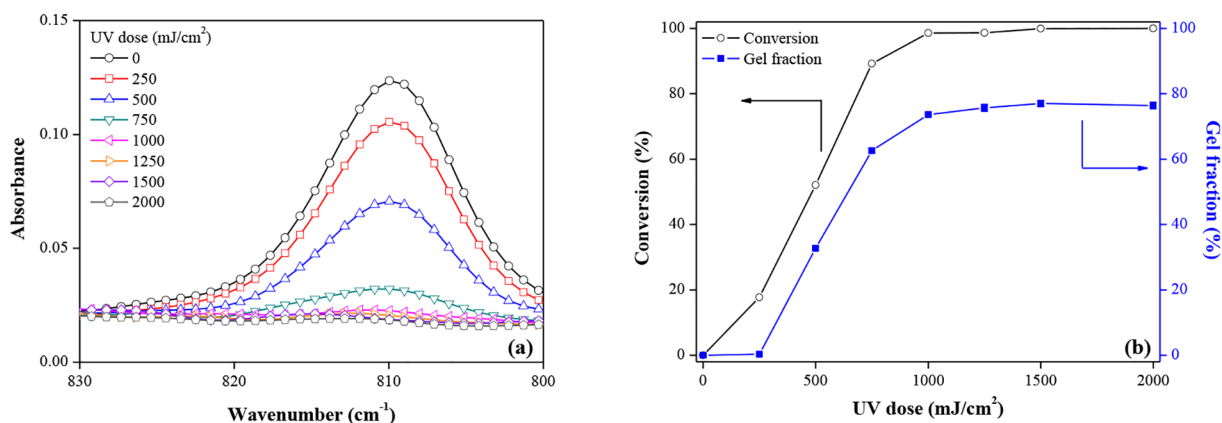


Figure 3. Curing behavior of PSA: (a) FT-IR and (b) conversion and gel fraction.

second UV irradiation. The masking film had a line pattern—the UV radiation is blocked by the lines, although it can penetrate the regions in which a line is not printed. The masking film was prepared by printing the line pattern on the back side of the silicone release film. Therefore, after exposure to the second UV irradiation with the masking film, the line patterned PSA was prepared, which included both high and low cross-linking densities in a single PSA layer. By controlling the second UV irradiation, we prepared the patterned PSA that had different cross-linking densities for a highly cured region. All samples were cured by a UV conveyor with constant speed. The UV dose of the UV conveyor was fixed as 250 mJ/cm², and the total UV dose was controlled by the number of passing onto the UV conveyor.

2.2. Fourier Transform Infrared Spectroscopy and Gel Fraction. As a radical is generated by the photoinitiator via UV irradiation, it reacts with the C=C bond so that the acrylic PSA is cured. By comparing the conversion of the C=C bond, we could quantitatively estimate the reaction by using FT-IR (JASCO, Model FTIR-6100). As the C=C bond of acrylate shows an FT-IR peak at 810 cm⁻¹, the conversion of the C=C bond was evaluated using eq 1:

$$\text{conversion (\%)} = \frac{(A_{810})_0 - (A_{810})}{(A_{810})_0} \times 100 \quad (1)$$

where $(A_{810})_0$ and (A_{810}) denote the intensity integration of the 810 cm⁻¹ peak at 0 mJ/cm² and after curing, respectively.

The gel fraction is the weight ratio of the total polymer weight to the cross-linked polymer weight, and it was estimated using the following steps: cured PSA was soaked in toluene for 24 h and then filtered using a 200-mesh filter. After drying the filtered mesh, the weight filtered by the mesh was measured to determine the cross-linked polymer fraction. The remaining mass was the linear polymer, which penetrated the mesh. Therefore, the gel fraction was calculated by dividing the cross-linked polymer weight by total weight.

2.3. Lap Shear Test and Adhesion Test. We used a polyethylene terephthalate (PET) film (75 μm thick) as the substrate for the lap shear test specimen, and the adhesion area was 25 mm × 25 mm. A universal testing machine (AllroundLine Z010, Zwick, Germany) pulled the specimen in the tensile direction at 1 mm/s. The maximum stress for the fracture was determined as the lap shear strength and the strain was evaluated by dividing the displacement by the thickness of the PSA (50 μm).

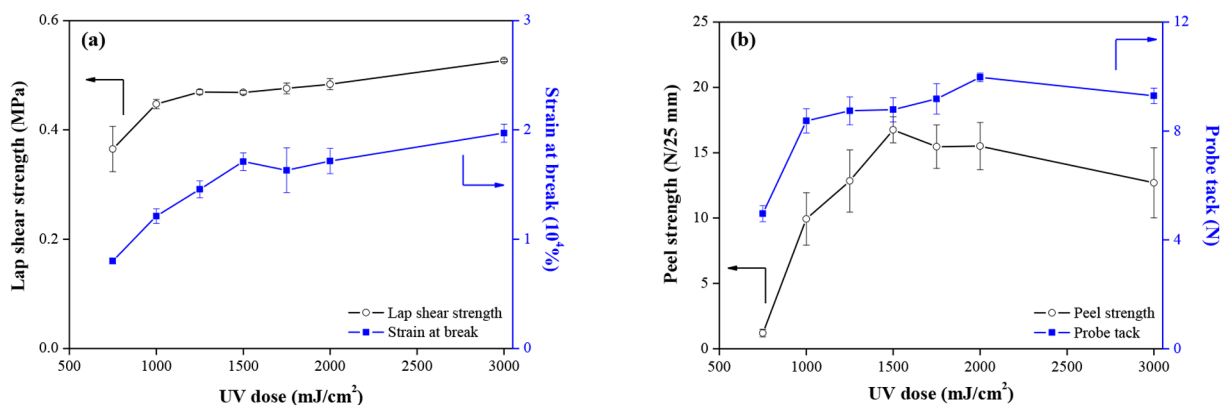


Figure 4. (a) Lap shear strength and (b) adhesion performance of PSA.

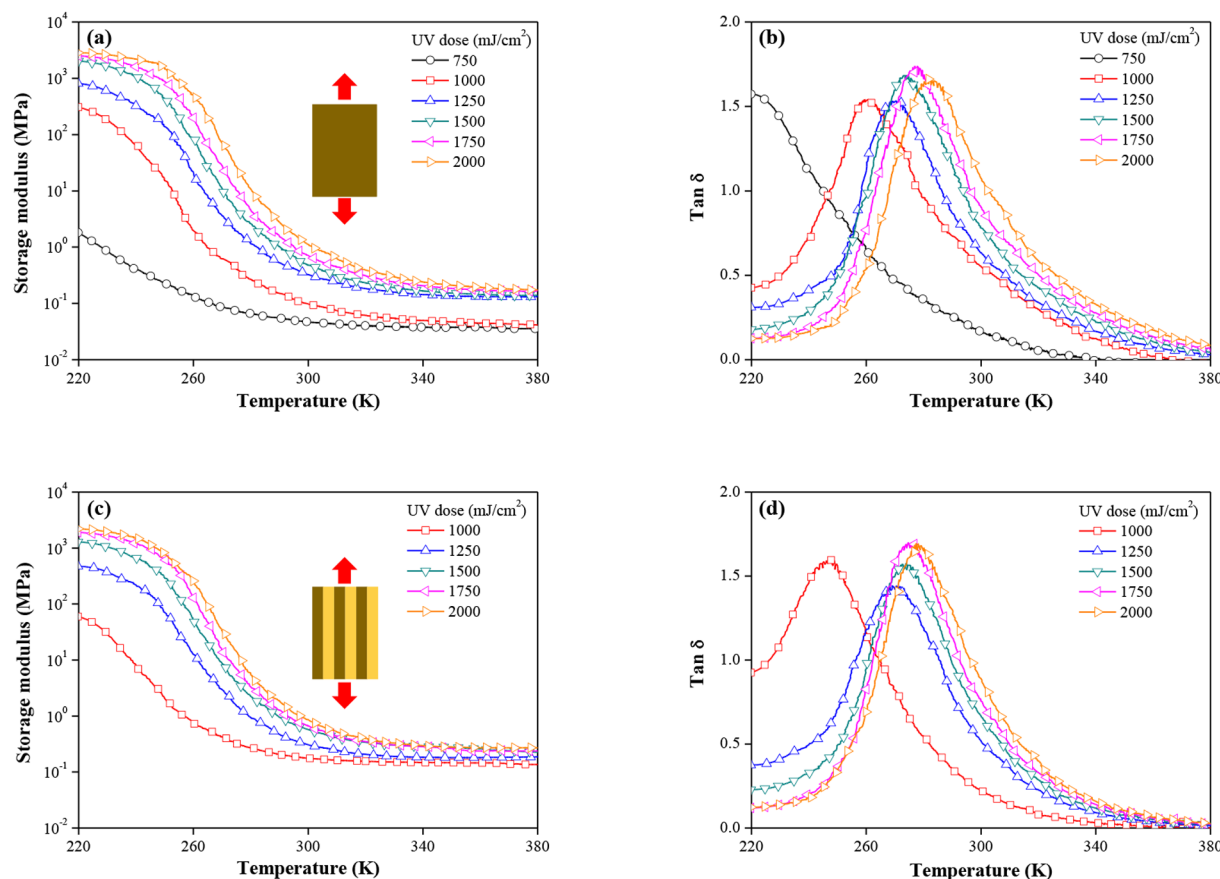


Figure 5. Viscoelastic property of nonpatterned and patterned PSA: (a) storage modulus and (b) damping factor of the nonpatterned PSA, (c) storage modulus and (d) damping factor of the patterned PSA. The preparation procedure of the UV-patterned PSA is illustrated in Scheme 1.

The peel strength and probe tack were investigated by using a texture analyzer (TA-TX plus, Micro Stable Systems, U.K.). The peel strength was measured at an angle of 180° with a crosshead rate of 5 mm/s. The backing film of the PSA was the 75- μ m PET film. Probe tack (initial adhesion strength) was measured at a constant separation rate of 10 mm/s after initial contact (1 N for 10 s).

2.4. Dynamic Mechanical Analysis. To characterize the viscoelastic property of the UV-patterned and nonpatterned PSAs, we implemented a dynamic mechanical analysis (DMA, Q800, TA Instruments, USA). The flat-shaped test specimens

were prepared by curing the PSA in a flat-shaped silicone mold in order to obtain the PSA into free-standing shape without any substrate. The flat-shaped test specimens were fixed under tension: clamp with 0.1% strain and frequency of 1 Hz. The temperature was increased from 220 K to 380 K at a constant rate (5 K/min). In the case of the UV-patterned PSA, the pattern direction was parallel to the tensile direction of the DMA. All samples were conducted once, not including repeat tests.

2.5. Strain Recovery and Stress Relaxation. The DMA (Q800, TA Instruments, USA), equipped with a tension

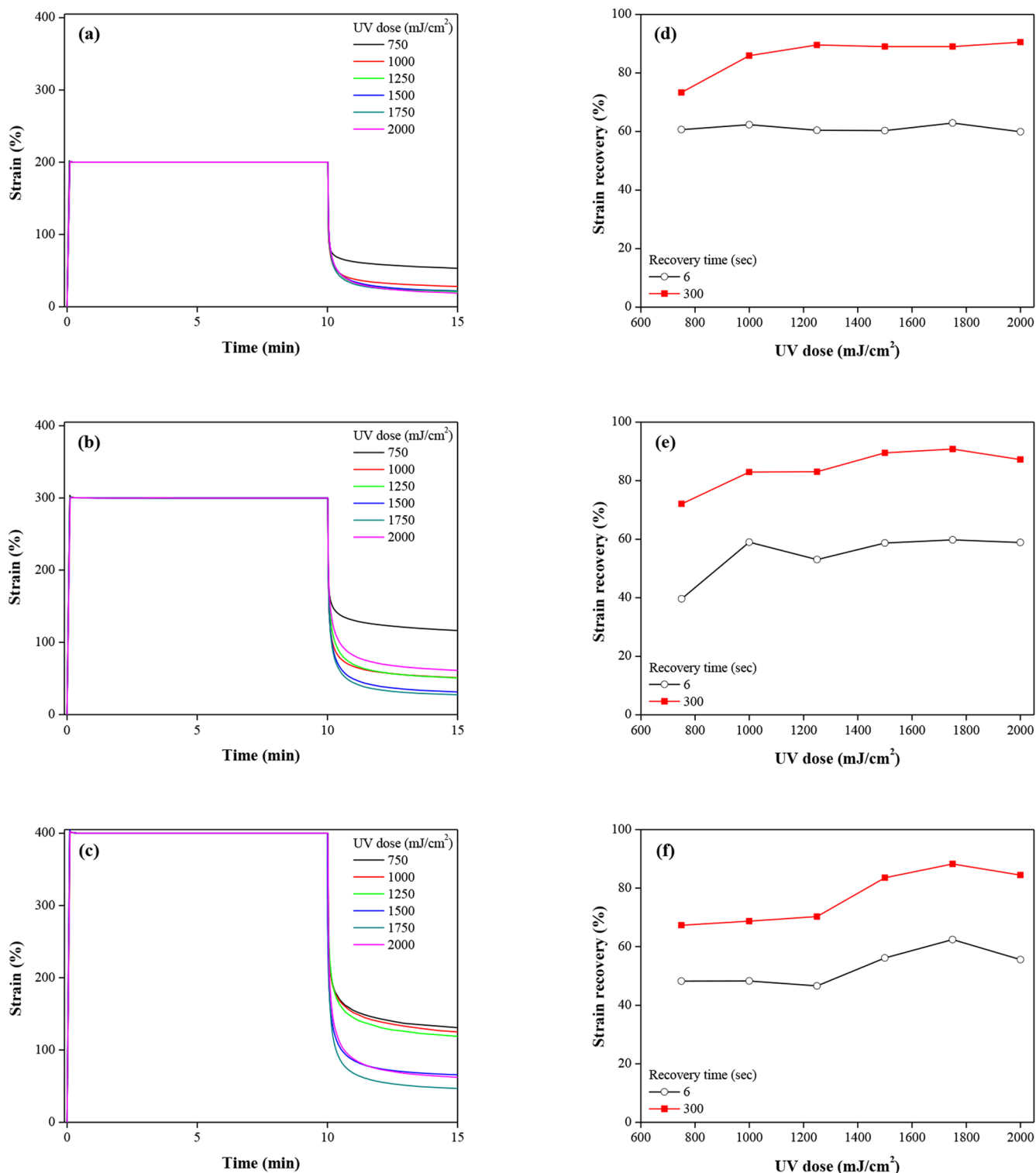


Figure 6. Recovery behavior of PSA: strain = (a) 200%, (b) 300%, and (c) 400%. Strain recovery of PSA: strain = (d) 200%, (e) 300%, and (f) 400%. All samples were conducted once, not including repeat tests.

clamp, was used to evaluate the strain recovery and the stress relaxation of the PSA. As shown in Figure 2a, the test specimen was prepared by using a polycarbonate (PC) substrate (30 mm × 6 mm) and the adhesion area was 20 mm × 6 mm. Strain was applied in the tensile direction so that the PSA experienced shear strain between the polycarbonate substrates. Constant shear strain was applied for 10 min and then, the

force was decreased to zero for 5 min. The shear strain values (200%, 300%, and 400%) were varied for various bending radii of the flexible display module. When the shear strain was increased to 500%, some samples were broken during the experiments so that we selected the shear strain values from 200% to 400%. The strain and recovery time were the same as those used in

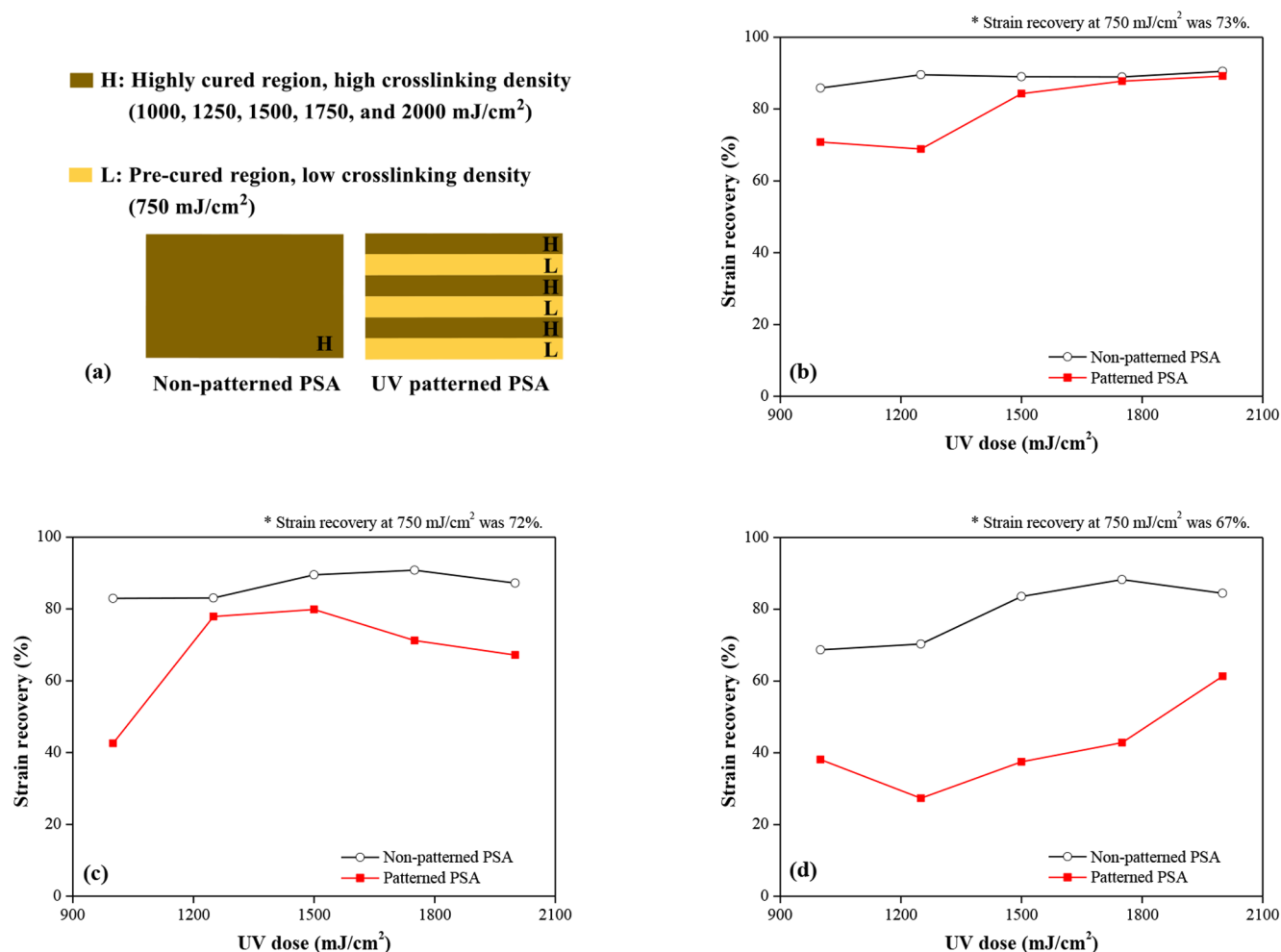


Figure 7. Strain recovery of nonpatterned and UV-patterned PSA: (a) schematic illustration of nonpatterned and UV-patterned PSA, strain = (b) 200%, (c) 300%, and (d) 400%. All samples were conducted once, not including repeat tests.

previous research by our group.²² As shown in Figure 2b, the strain was naturally recovered for 5 min and we calculated the strain recovery using eq 2:

$$\text{strain recovery (\%)} = \frac{S_0 - S_t}{S_0} \times 100 \quad (2)$$

where S_t and S_0 denote the strains at the recovery times of $t = 6$ s and $t = 300$ s, and at the beginning of recovery ($t = 0$ s), respectively.

As shown in Figure 2c, the stress was relaxed during the constant strain condition, and it was calculated using eq 3:

$$\text{stress relaxation (\%)} = \frac{F_i - F_f}{F_i} \times 100 \quad (3)$$

where F_i and F_f denote the initial and final stress under constant strain, respectively. All samples were conducted once, not including repeat tests.

3. RESULTS AND DISCUSSION

3.1. Curing Behavior of PSA. Acrylic PSA was cured by UV irradiation and it was evaluated using FT-IR and gel fraction. In the FT-IR analysis, the intensity of the C=C bond peak (810 cm^{-1}) was weakened as the UV dose was increased, which indicates that the C=C bond reacted with the radical

that was formed by UV irradiation (Figure 3a). In other words, as the UV dose was increased, the conversion, which is the parameter indicating the quantity of the curing reaction, was increased. In addition, the gel fraction of the PSA also increased with UV irradiation (Figure 3b).

From these results, it was determined that the cross-linking degree was easily controlled by changing the UV dose for the PSA. Therefore, by separating the UV penetration area and blocking area, it was possible to prepare the line-patterned PSA, which contained the two regions with different cross-linking densities.

3.2. Lap Shear Strength and Adhesion Performance of PSA. The mechanical strength of acrylic PSA was evaluated through a lap shear test and adhesion test. As the UV dose was increased, the acrylic PSA was cross-linked so that the lap shear strength was enhanced (Figure 4a). Therefore, it is suggested that the highly cured region of UV-patterned acrylic PSA has a higher mechanical strength than that of the precured region of the UV-patterned acrylic PSA (see Scheme 1b).

The peel strength of acrylic PSA was enhanced by UV irradiation, because of the increase in cohesion strength (Figure 4b). The cohesion of acrylic PSA continuously increased with increase in the UV irradiation, which decreased the wetting of PSA to the substrates; this implied that the peel strength of acrylic PSA decreased at a high UV dose. The probe tack also

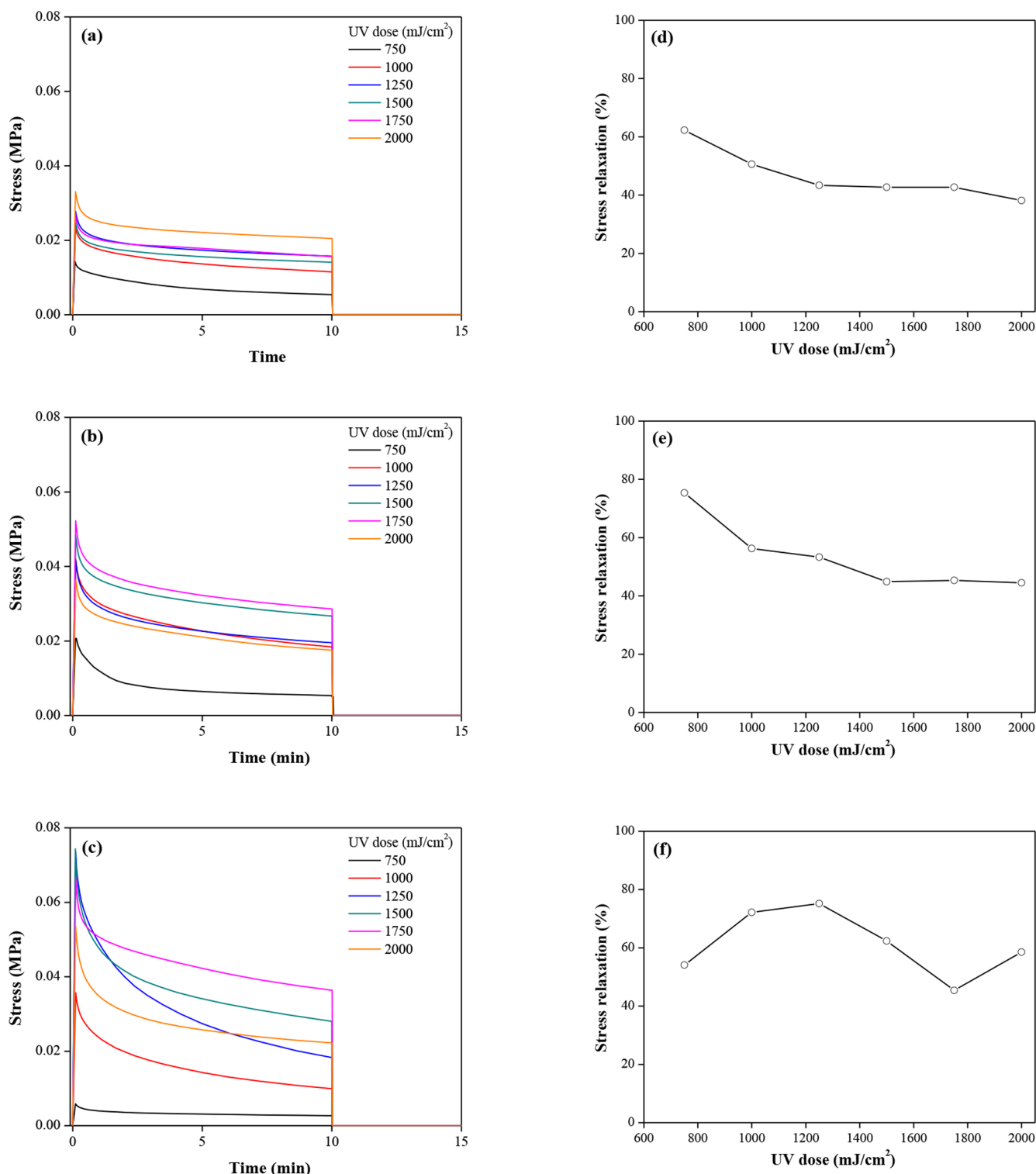


Figure 8. Relaxation behavior of PSA: strain = (a) 200%, (b) 300%, and (c) 400%. Stress relaxation of PSA: strain = (d) 200%, (e) 300%, and (f) 400%. All samples were conducted once, not including repeat tests.

decreased as the UV dose was increased, for the same reason as the decrease in the peel strength.

3.3. Viscoelastic Property of UV-Patterned PSA.

In order to compare the viscoelastic properties of nonpatterned and UV-patterned PSA, the storage modulus and damping factor ($\tan \delta$) were estimated by DMA (Figure 5). As the UV dose was increased, the storage modulus of nonpatterned

PSA was enhanced in the entire temperature range (Figure 5a). There was no significant difference in the peak intensity of the damping factor; however, with UV irradiation, the peak was slightly shifted to the higher temperature region, which indicates increase in the glass-transition temperature (Figure 5b).

The storage modulus and glass-transition temperature of UV-patterned PSA, containing both high and low cross-linking

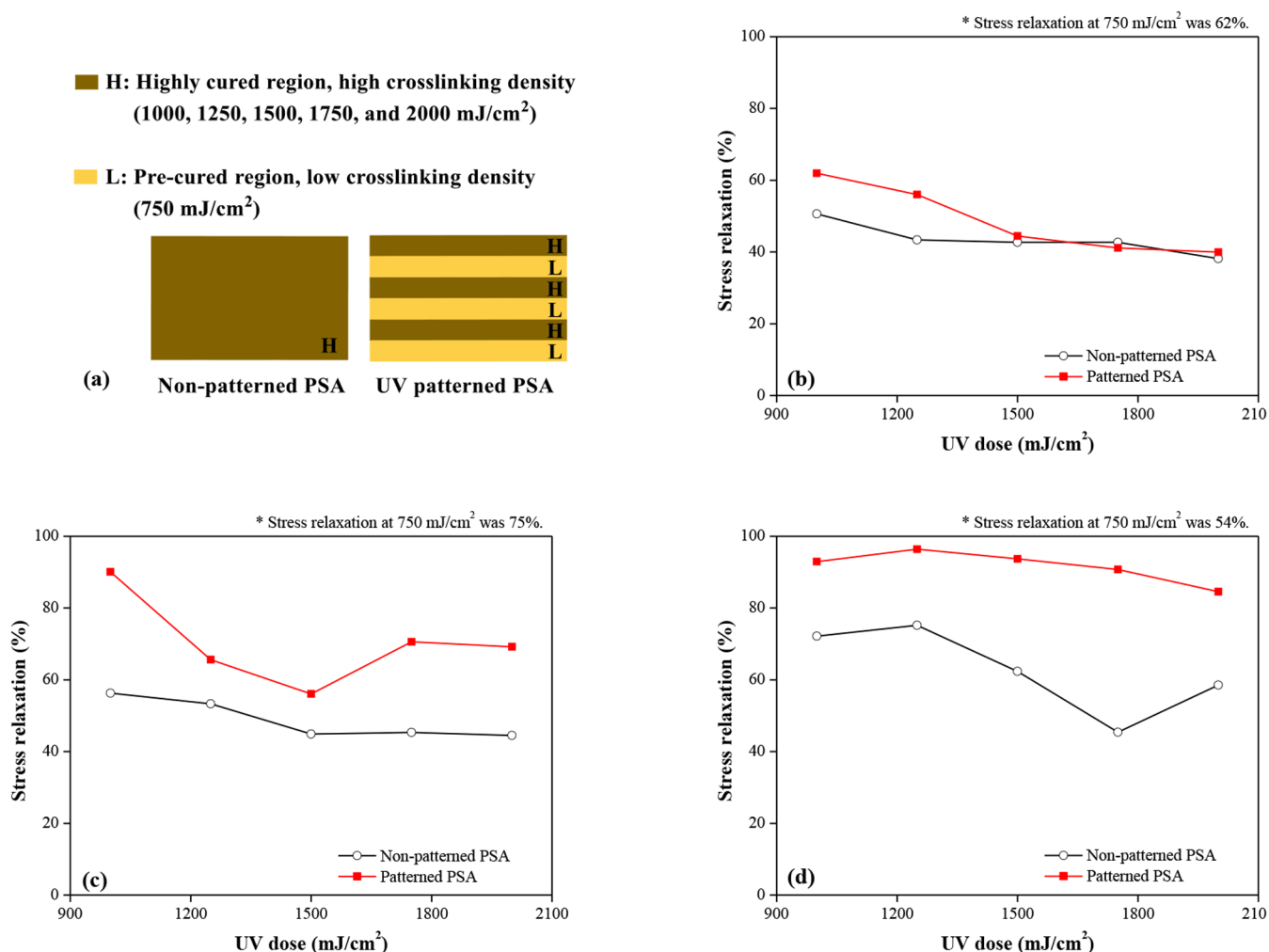


Figure 9. Stress relaxation of nonpatterned and UV-patterned PSA: (a) schematic illustration of nonpatterned and UV-patterned PSA, strain = (b) 200%, (c) 300%, and (d) 400%. All samples were conducted once, not including repeat tests.

density, were also increased with the increase in UV dose of the highly cured region (Figures 5c and 5d). However, especially in the glass state, the storage modulus was lower than that of nonpatterned PSA, because the precured region (UV dose = 750 mJ/cm²), included in the patterned PSA, had the lowest storage modulus.

3.4. Recovery Property of UV-Patterned Acrylic PSA.

PSA is a polymeric material and, hence, it could be naturally recovered after deformation. Figure 6 shows the recovery behavior and strain recovery of nonpatterned PSA. Strain was immediately recovered above 50% for 6 s and gradually recovered until 300 s. The elasticity and recovery property were related to the storage modulus of the polymeric material²⁴ and, hence, the strain recovery of PSA was enhanced by UV irradiation, because of the increase in its storage modulus. In our previous work, strain recovery of PSA was improved by increasing cross-linking density of PSA and it supported the above-mentioned results.^{20,22} As the applied strain was increased from 200% to 400%, strain recovery was decreased, which indicates that high strain resulted in high degree of plastic deformation.

As shown in Figure 7, compared to nonpatterned PSA, UV-patterned PSA had lower strain recovery, because UV-patterned PSA contained the precured region (750 mJ/cm²) that has low storage modulus. However, the strain recovery of

patterned PSA was lower than that of nonpatterned PSA with dose of 750 mJ/cm². This might result from unknown factors such as the interface and Poisson's ratio of two different cross-linking density regions. Poisson's ratios of polymeric materials could be changed by the cross-linking density of the polymer network and under high strain conditions.^{27,28} Under stretched condition, two regions with different Poisson's ratio were differently deformed so that recovery could be disturbed. Therefore, it could be considered that these factors had influence on the recovery behavior of the patterned PSA. Also, as the applied strain was increased from 200% to 400%, the difference of strain recovery between nonpatterned and patterned PSA was widened. When the applied strain was increased to 400%, both nonpatterned and patterned PSA were largely deformed than strain of 200% and 300%. Especially, in the case of the patterned PSA, as the PSA was largely deformed, difference of the deformation behavior between highly cured and precured region was increased because of their different Poisson's ratios. This phenomenon could disturb the recovery of the patterned PSA so that the difference of strain recovery between nonpatterned and patterned PSA was widened at strain of 400%.

3.5. Relaxation Property of UV-Patterned Acrylic PSA.

The stress of viscoelastic materials is relaxed under the constant strain condition, because of their dissipation property

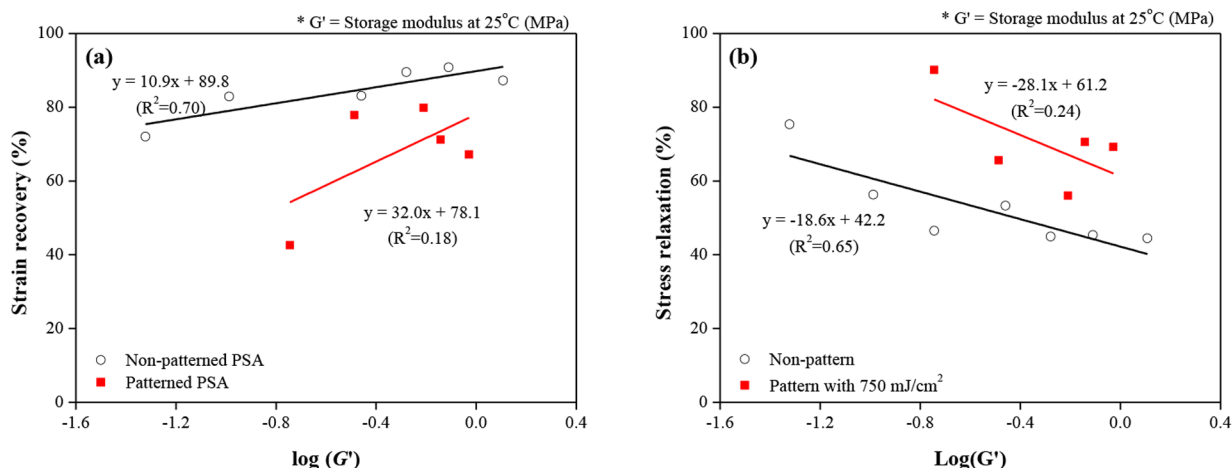


Figure 10. (a) Strain recovery and (b) stress relaxation storage modulus (strain = 300%). All samples were conducted once, not including repeat tests.

that originates from the mobility of the polymer chain. Figure 8 shows the relaxation behavior and stress relaxation of PSA. As a high degree of UV irradiation was applied, stress relaxation was generally decreased. Similarly, in our previous work, stress relaxation of PSA was decreased as the cross-linking density of PSA increased, and it was due to reduction in the mobility of the polymer chain.²² When the applied strain reached 400%, the maximum stress was increased but it was largely relaxed so that the applied strain of 400% had the highest stress relaxation. This suggests that excessively high strain caused enhancement of the dissipation of PSA, because high strain facilitates movement of the polymer chain.

Figure 9 shows the difference of stress relaxation between nonpatterned and UV-patterned PSA. By containing the precured region (750 mJ/cm^2), stress relaxation was significantly improved in patterned PSA than in nonpatterned PSA, because of the high dissipation property of the precured region. As the strain was increased from 200% to 400%, stress relaxation of patterned PSA was higher than that of the low cross-linking density region (750 mJ/cm^2) and the difference of stress relaxation between nonpatterned and patterned PSA was expanded. Similar to strain recovery, high strain promotes the differences of deformation behavior between highly cured and precured region, because of their different Poisson's ratios. While this phenomenon disturbs the strain recovery of the patterned PSA, it significantly improved stress relaxation of the patterned PSA.

According to Figures 7 and 9, by introducing patterning on the PSA, strain recovery was deteriorated but stress relaxation was improved and this effect was enhanced with increase in the applied strain. However, strain recovery was too deteriorated at a strain of 400% and stress relaxation was not enhanced enough at a strain of 200%. It shows that the optimization of the recovery and relaxation was only seen under the 300% strain condition.

As shown in Figure 10, we plotted the strain recovery and stress relaxation as a function of the storage modulus (G'). We found that the strain recovery of PSA has a positive linear relationship with $\log(G')$ and the stress relaxation of PSA has a negative linear relationship with $\log(G')$. In the case of nonpatterned PSA, strain recovery and stress relaxation were changed with the storage modulus but these two properties could not be optimized by controlling the storage modulus. On the other hand, in the case of UV-patterned PSA, the trend lines of strain recovery and stress

relaxation crossed each other, which means that recovery and relaxation could be optimized by UV patterning. Especially, at the range of $\log(G')$ from -0.2 to -0.4 , both of the strain recovery and the stress relaxation were well-optimized above 71%. As this range of the $\log(G')$ is inside of the range of general PSA's $\log(G')$, so-called Dahlquist criterion and Chang's window,^{1,29–31} it supported the possibility of its use in industrial applications.

In addition, a preliminary simulation of the stress distribution was performed, as shown in Figure 11. The type of

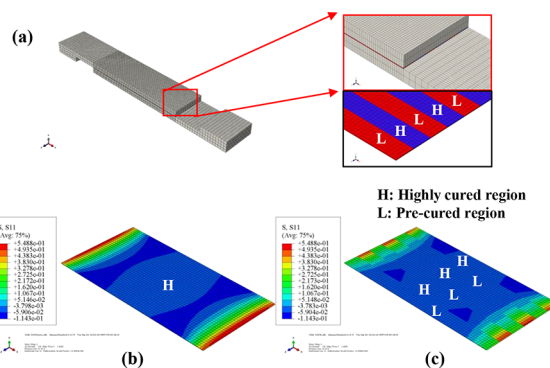


Figure 11. Simulation of stress distribution under the constant strain (300%) condition: (a) schematic illustration for simulation and stress distribution of (b) nonpatterned PSA and (c) UV-patterned PSA.

simulation is finite element analysis, and we set the moduli of H (highly cured region) and L (precured region) as 1 and 0.7 MPa, respectively. Under initial conditions, strain and force were both zero and the strain was applied to 300%. The maximum and overall stress were lower for patterned PSA than for nonpatterned PSA. This observation supports that patterned PSA has better dissipation property than nonpatterned PSA. However, as the simulation did not consider many unknown factors such as interface between the patterned lines and different Poisson's ratio of both lines, it was difficult to precisely explain the above results based on simulation.

4. CONCLUSIONS

We used UV-curable acrylic PSA that was used for the assembly of displays. The curing behavior, mechanical strength, adhesion

performance, viscoelastic, recovery, and relaxation property of the PSA were determined. PSA with high cross-linking density had improved elasticity and recovery but poor dissipation and stress relaxation than PSA with low cross-linking density. We found that the strain recovery of PSA had a positive linear relationship with $\log(G')$ but the stress relaxation had a negative linear relationship with the same. Importantly, we found that the patterned PSA, containing both high and low cross-linking density, had lower strain recovery but higher stress relaxation than nonpatterned PSA. Therefore, while the strain recovery and stress relaxation of nonpatterned PSA could not be optimized by varying the storage modulus, those of the patterned PSA could be well-optimized. Although we suggest patterning of PSA as an optimization method for recovery and relaxation in this paper, there are many other factors that must be considered, including pattern width, strain-pattern angle, chemical composition of PSA, etc. In further research, it is necessary to investigate these factors for better optimization of the recovery and relaxation of PSA.

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Notes

The authors declare no competing financial interest.

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