

# Effect of the Stress Relaxation Property of Acrylic Pressure-Sensitive Adhesive on Light-Leakage Phenomenon of Polarizer in Liquid Crystal Display

Hyunae Chun,<sup>1</sup> Hyun Ah Kim,<sup>2</sup> Gyungsoo Kim,<sup>1</sup> Jaeil Kim,<sup>3</sup> Kyung-Yul Lim<sup>3</sup>

<sup>1</sup>*Environmental and Energy Division, Korea Institute of Industrial Technology, Cheonan, 330-825 Korea*

<sup>2</sup>*Department of Applied Chemical Engineering, Korea University of Technology and Education, Cheonan, 330-708 Korea*

<sup>3</sup>*Technical Development Team, Ace Digitech Ltd., Ochang-Myeon, Chungbuk, 363-883 Korea*

Received 19 September 2006; accepted 3 January 2007

DOI 10.1002/app.26889

Published online 2 August 2007 in Wiley InterScience (www.interscience.wiley.com).

**ABSTRACT:** The acrylic pressure-sensitive adhesive systems with the different stress relaxation abilities are prepared by varying the curing agent from 0.3% to 3.0% and the effect of the stress relaxation ability of pressure-sensitive adhesive system on the durability of LCD polarizer was investigated. The stress relaxation ability of the cross-linked pressure-sensitive adhesive systems was measured by transient tensile test and dynamic viscoelastic test. And the durability was evaluated by the light-leakage test of polarizer coated with the pressure-sensitive adhesive after it was aged under the condition of 60°C and the relative humidity of 95% for 72 h. It was observed that the stress

relaxation ability of pressure-sensitive system is decreased with the crosslinking density, as intended. And the pressure-sensitive adhesive with the lower stress relaxation ability exhibited the lower light-leakage and consequently the higher durability than the other PSA systems. This observation is obviously contradictory to the previous design strategy of PSA system and the reason for this observation was discussed briefly. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 106: 2746–2752, 2007

**Key words:** pressure-sensitive adhesive; stress relaxation; viscoelastic property; light-leakage; polarizer

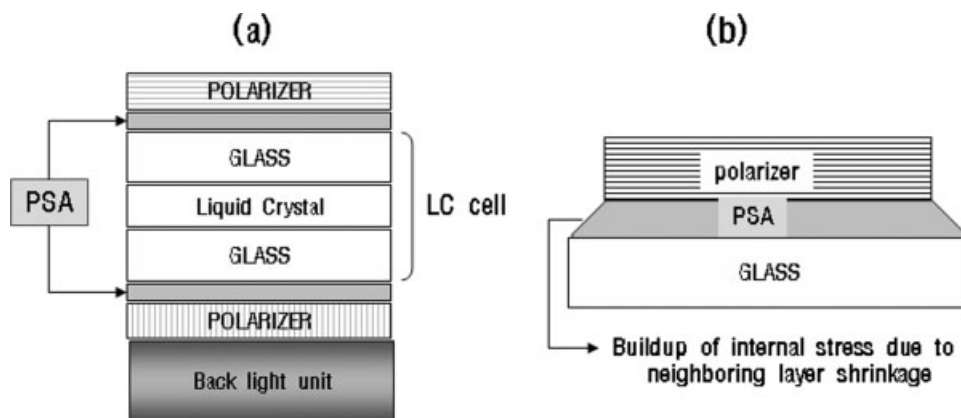
## INTRODUCTION

Pressure-sensitive adhesives (hereinafter abbreviated to PSA) are materials that adhere on contact or by the application of a slight pressure and can be debonded easily through the adhesive failure.<sup>1–7</sup> Because of its inherent advantage such as reworkability, PSA has been used extensively in diverse fields of industries. One of the major application fields of PSA are the optical films in liquid crystal display (LCD). As shown in Figure 1(a), PSA is employed in attaching polarizer on front and rear sides of liquid crystal cell. Among the various PSA systems, the acrylic copolymer-based adhesive has been exclusively used in the packaging of optical films because of its transparency and resistance to heat and humidity.<sup>1,2</sup>

In general, the polarizer employed in LCD consists of 6 ~ 15 optical films, depending on the LCD mode (i.e., TFT, TN, STN) and the required performances. However, basic unit for the current LCD polarizer is simply three layers: Triacetyl cellulose (TAC) film/polyvinyl alcohol (PVA) film/triacetyl cellulose

(TAC) film.<sup>8,9</sup> Here PVA film plays a role of the polarization of the incident light and TAC film is used as a protective layer to keep PVA film from heat and humidity. To attain the high polarization performance, PVA film must have the large optical anisotropy, which can be reached by the stretching of PVA film several times by force during processing. However, the stretched PVA molecules tend to relax to the random state upon the exposure of heat and humidity, which results in the shrinkage of the polarizing film.<sup>9</sup> As shown in Figure 1(b), the deformation of polarizer tends to buildup the stress especially in the PSA layer which is located between the contracted polarizer and glass. This causes the peeling of the polarizer film from the LC cell and the appearance of air bubbles at the glass-adhesive interface. Furthermore, the distortion of the absorption axes of the polarizer because of the shrinkage of PVA molecules results in the deviation of the crossed polarization between the upper and lower polarizer films and finally brings out the light-leakage phenomenon, as shown in Figure 2(a). Here the light-leakage means that the light from the back-light unit of LCD cannot be blocked completely because of the distortion of polarization, which leads to the loss of detail and contrast of display. It has been considered that the light-leakage phenomenon is the

Correspondence to: H. Chun (hachun@kitech.re.kr).



**Figure 1** (a) Schematic of liquid crystal display (LCD) panel. (b) Generation of stress in the pressure-sensitive adhesive layer due to the shrinkage of polarizer when it is aged under the heat and humidity.

most serious problem to be overcome in order to produce the LCD with the high durability.

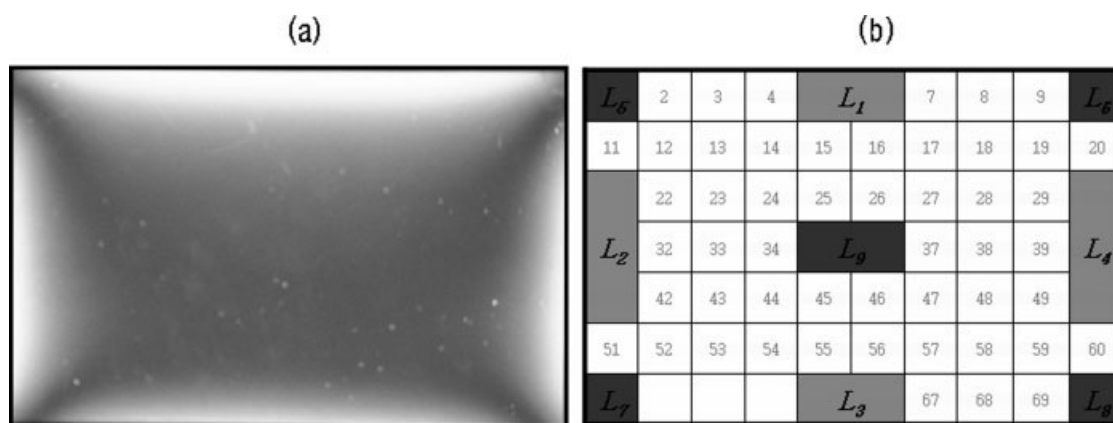
Recently, many attempts have been made to solve the light-leakage problem by the proper design of pressure-sensitive adhesive.<sup>9–13</sup> That is, since the problems were due to stress caused by the deformation of the component films, it has been suggested that the stress relaxation capacity of the adhesive layer has been a key criterion in the design of PSA formulation. To improve the stress relaxation property of PSA, the copolymer with the low glass transition temperature has been prepared. In addition, the composition of PSA system is modified by adding a plasticizer, by mixing an acrylic polymer having a low molecular weight of <30,000 or by decreasing the crosslinking agent.<sup>10–13</sup> However, in spite of the belief that the stress relaxation ability of PSA plays the crucial role in the light-leakage of LCD polarizer, the systematic investigation on the relationship between the stress-relaxation behavior on the light leakage been rarely reported.

In this article, therefore, we will investigate the influence of the stress-relaxation ability of PSA system on the light-leakage phenomenon. Since the variation of the crosslinking density is the very efficient method to control the stress-relaxation property of system, four crosslinked PSA systems were prepared by changing the curing agent from 0.3% to 3.0%. The viscoelastic and mechanical properties of the PSA systems were measured and the results were correlated with the light-leakage phenomenon of the LC panel.

## EXPERIMENTAL

### Materials

The pressure-sensitive adhesive of the current PSA systems consisted of (1) acrylic copolymer, (2) a crosslinking agent, and (3) silane coupling agent. The acrylic copolymer was composed of *n*-butyl acrylate, methyl acrylate, acrylic acid, and 2-hydroxyl-



**Figure 2** (a) Schematic representation of the light-leakage phenomenon of LCD. (b) Designation of the locations where the transmitted intensities were measured by the luminance colorimeter to determine the light-leakage quantitatively.

**TABLE I**  
**Formulations and Gel Content of Acrylic**  
**Pressure Sensitive Adhesives**

Sample	Acrylic copolymer (g)	Isocyanate curing agent (g)	Epoxy curing agent (g)	Silane coupling agent (g)	Gel content (%)
PSA-NC		0	0	0	0.44
PSA-A		0.3			76.2
PSA-B	100	0.8	0.01	0.3	85.0
PSA-C		1.5			91.0
PSA-D		3.0			93.7

ethyl acrylate. It has the weight-average molecular weight of 320,000 and polydispersity of 1.46 determined by Gel Permeation Chromatography (Waters 500). The glass transition temperature of acrylic polymer measured at the heating rate of 10°C/min by the differential scanning calorimetry (TA instrument, Q100) was -32°C. Polymethylenephényl isocyanate as a curing agent and epoxidized soybean oil as a cocuring agent were used together.  $\gamma$ -Methacryloxy propyl trimethoxy silane, the coupling agent was added to increase the adhesion between the PSA layer and the glass. All chemicals used here were kindly supplied by Ace Digitech (Ochang, Korea).

The formulations used in this work were given in Table I. To investigate the effect of crosslinking density on the PSA system, the curing agent was varied from 0.3 to 3.0 wt %. However, within the material systems used in this work, the glass transition temperatures of PSA systems are changed little by the formation of the crosslinks along the polymer backbone. The DSC results showed that all crosslinked acrylic polymers in Table I give rise to the virtually same transition behaviors, which may imply that the molecular weight between crosslinks ( $M_C$ ) of all systems are large enough not to influence the segmental motions of acrylic polymer backbone.

### Sample preparations

For the tensile and rheological tests, 2 mm-thick films were prepared by casting the mixture solution, which was diluted with methyl ethyl ketone to control the solution viscosity. The solvent in the PSA system was first slowly evaporated inside the clean bench at room temperature for 48 h. It was further dried in a vacuum oven at 80°C for 24 h, followed by 100°C for 4 min, and then this film was aged in a thermohygrostat under the conditions of the temperature of 35°C and the relative humidity of 45% for 72 h.

For the measurement of rheological test, the specimen was punched out from the 2 mm-thick film into the disk with the diameter of 25 mm. The dogbone shaped specimen was used for the stress relaxation and the tensile stress-strain measurement. The dimension of stress relaxation specimen was 4 mm in width

and 70 mm in gauge length. For the tensile stress-strain test, the specimen with the dimension of 4 mm in the width and 25 mm in gauge length was used.

For the light-leakage measurement, five layered sample, polarizer/PSA/glass/PSA/polarizer, was prepared as following: The thin PSA film with the thickness of 20  $\mu\text{m}$  was first prepared by bar coating on the Teflon-coated releasing film. This thin PSA film was dried and aged under the same condition as the sample described in the previous paragraph. It was laminated on 185  $\mu\text{m}$ -thick polarizer film, which was subsequently applied onto the both sides of glass. The dimension of glass used here was 235 mm ( $w$ )  $\times$  155 mm ( $h$ )  $\times$  2 mm ( $d$ ). The light absorption axes of both polarizing film were crossed with each other. Before the light-leakage measurement, the sample thus prepared was exposed to the accelerated aging conditions of 60°C and the relative humidity of 95% for 72 h, and then was stored at room temperature for 24 h.

### Gel content

The crosslinking density of pressure-sensitive adhesive was determined by gel content. 0.2 g of the cured sample was immersed in 100 mL of ethyl acetate at room temperature for 24 h. The insoluble fraction (gel content) in polymer was filtered out and dried in a vacuum oven at 85°C for overnight. The gel content was calculated by the eq. (1).

$$\text{gel content (\%)} = \frac{m_d}{m_i} \times 100 \quad (1)$$

where  $m_d$  and  $m_i$  are the dried weight and the initial weight of polymer, respectively.

### Tensile property

The tensile test was carried out at room temperature using the universal testing machine (Instron 4482). For the measurement of tensile stress-strain property, the specimen was deformed at a crosshead speed of 100 mm/min. The values of five measurements were statistically averaged. For tensile stress relaxation measurement, 10% of strain was applied on the sample with the initial crosshead speed of 100 mm/min and the variation of tensile stress was measured as a function of time.

### Viscoelastic property

The viscoelastic properties ( $G'$ ,  $G''$ , and  $\tan \delta$ ) of polymer system were measured in the oscillatory shear deformation mode using dynamic rheometer (Rheometric Scientific, RMS-800). The parallel geometry with the plate diameter of 25 mm and the gap

distance of 1 mm were used. The dynamic measurement was performed at the constant strain of 5% in the frequency range of 0.1–100 rad/s. The testing temperature was room temperature.

### Light leakage

The extent of light leakage of polarizing film was measured by Luminance colorimeter (Minolta, BM-5A) equipped with the digital camera. The sample was irradiated with the LCD backlight unit (Heeyoung Co, Lumimed) and the light-leakage ( $L$ ) through the sample was measured at preassigned positions by luminance colorimeter (BM-5A), as shown in Figure 2(b). The light-leakage ( $L$ ) was quantized by the following equation (2).

$$L = \frac{(L_1 + L_2 + L_3 + L_4)/4}{(L_5 + L_6 + L_7 + L_8 + L_9)/5} \quad (2)$$

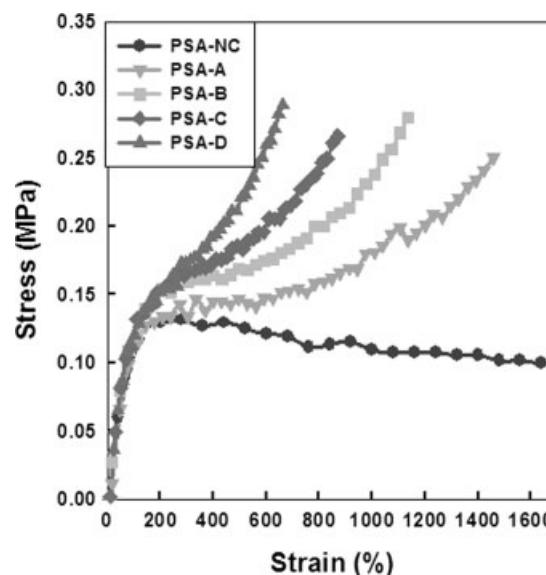
where  $L_n$  is the transmitted light intensity detected at the position denoted as  $n$ . Since the polarizing film generally shows the most serious light leakage at  $L_1, L_2, L_3,$  and  $L_4$  and the least light leakage at  $L_5, L_6, L_7, L_8,$  and  $L_9$ , the values of the numerator and the denominator mean the averages of intensity at the locations where the highest and lowest light-leakage are expected, respectively.

## RESULTS AND DISCUSSION

### Crosslinking density of PSA system

The network density of acrylic copolymer was varied by the increase of the concentration of the curing agent and it was monitored by the gel content. Before crosslinking, the gel content of the polymer used here is negligible. By varying the curing agent from 0.3 to 3.0%, the gel content is increased from 76% to 93.7%, as shown in Table I. In the case of PSA-A system, the sample does not recover its original dimension immediately after the deformation is removed, reflecting its low-crosslinking density and the residual free polymer chain. It takes time for PSA-A system to recover its original dimension after the release of the deformation. In contrast, PSA-D sample with the highest crosslinking density does not show any trace of creep when stretched.

We confirmed, using the tensile test, that this variation of gel content can give rise to the significant difference in the physical property of the PSA. The tensile stress–strain behaviors of PSA-NC, PSA-A to PSA-D were determined and the results are given in Figure 3. (Uncrosslinked PSA-NC does not break within the allowable extension range of our UTM instrument.) It shows that the elongation at break is decreased with the increasing crosslinking density,



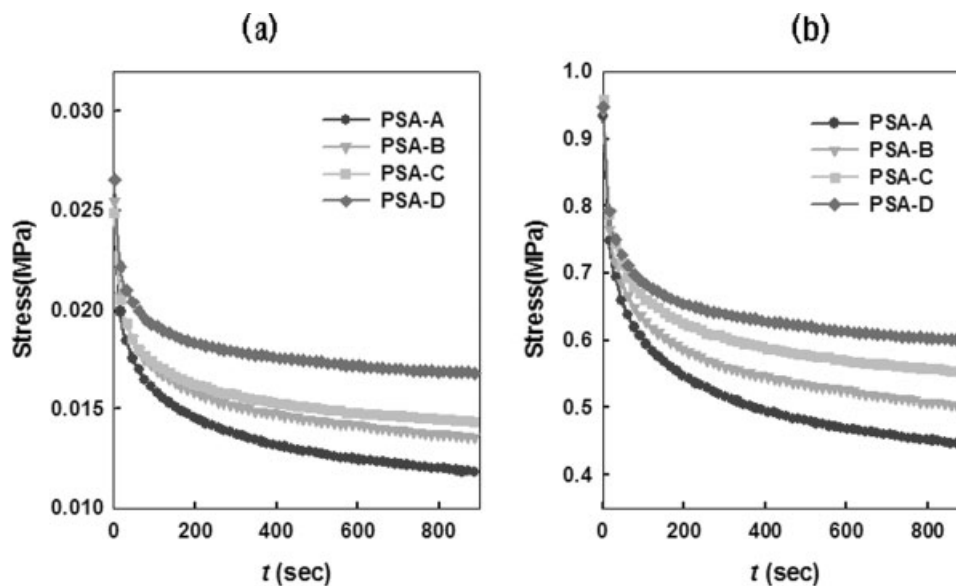
**Figure 3** Tensile stress–strain curves of PSA systems with the various crosslinking density.

as expected. The PSA-D system is broken at the strain of 650%, whereas PSA-A with the lightest crosslinking density is elongated to the strain of 1500% before the break. When the tensile strengths of samples are compared at the same strain of 600%, the tensile strength of PSA-D is 70% higher than that of PSA-A.

In addition to the tensile strength, the crosslink density can affect the adhesion strength of PSA on glass panel. In many cases, not always, the peel strength on the substrate is decreased with the crosslinking density due to the decrease of the viscous energy dissipation.<sup>2,7,14</sup> We also observed that the peel strength of PSA attached on glass substrate is decreased by 20% as the gel content is increase from 80 to 95% in our unpublished work. (In our unpublished work, the peel strength was determined using UTM at the angle of 90° and the deformation rate of 300 mm/min. The thickness of PSA film was 20  $\mu\text{m}$  and the backing material is the 3-layer film (Triacetyl cellulose/polyvinyl alcohol/triacetyl cellulose) with the thickness of 200  $\mu\text{m}$ .) However, in this work, we do not describe the variation of adhesion behavior with the crosslinking density in more details, since the adhesion strength is *not* the decisive factor on the light-leakage behavior that we are focusing in this work. In stead, it is considered that the adhesion strength is more closely related with the peel-off phenomenon of LCD polarizer upon aging.

### Effect of crosslinking on the tensile stress-relaxation of the PSA system

Transient tensile stress,  $\sigma(t)$ , at the constant strain of 10%, is plotted against  $t$  as shown in Figure 4. Ini-



**Figure 4** (a) Tensile stress relaxation curves of PSA systems with the various crosslinking density. (b) Normalized tensile stress relaxation curves ( $\sigma_t$ ) by the initial stresses ( $\sigma_i$ ).

tially  $\sigma(t)$  of all PSA systems fall rapidly due to the viscous energy dissipation arising from the chain movement upon the distortion. Then the relaxation rate is significantly decreased, except the uncrosslinked PSA-NC system. Differently from the highly crosslinked system, all crosslinked PSA systems do not flatten after the initial rapid relaxation and instead they show the slow and continuous relaxation. This second stage relaxation is most significant in the case of PSA-A system which possesses the lowest crosslinking density and the 25% of unattached polymer chain.

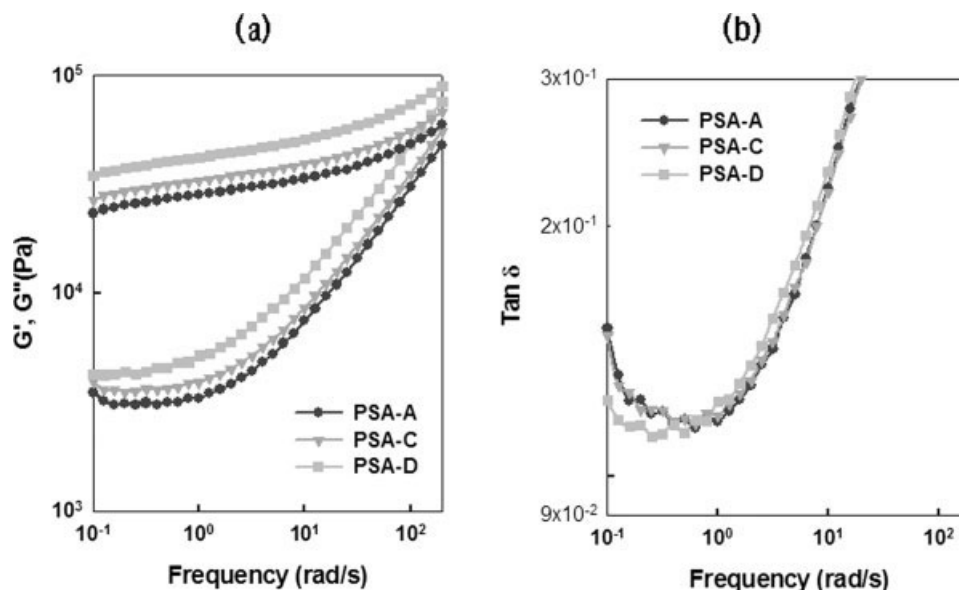
The result in Figure 4 also gives the clue on the crosslinking density of the current PSA systems. For the lightly-crosslinked sample,  $\sigma(t)$  flattens somewhat at a level between the initial fast transition and slow relaxation, which is known to be as  $G_N$ .<sup>15</sup> It is well-established fact that  $G_N$  is related with the average spacing between coupling points. The Figure 4 shows that the  $G_N$  value of PSA system is in the order of PSA-A < PSA-B < PSA-C < PSA-D, reflecting the variation of the crosslinking density.

In the tensile stress relaxation curve, it should be mentioned that the value of  $\sigma(t)$  is related with the stored energy, whereas the decreased value from the initial stress ( $\sigma_i$ ),  $\sigma_i - \sigma(t)$ , means the relaxed energy.<sup>15,16</sup> When the stress values of all crosslinked PSA systems were compared at a given time, the PSA-D with the highest crosslinking density always shows the highest stored energy, whereas PSA-A with the lightest crosslinking density show the highest stress relaxation ability. To figure out the relaxation rate, the  $\sigma(t)$  is normalized by the initial stress ( $\sigma_i$ ) and the result is given in Figure 4(b). This

figure clearly shows that the relaxation rate of lowest crosslinked PSA-A is higher, being compared with the other systems. Since the relaxation time is related to time required for a certain proportion of the polymer chain to respond to the external stress by thermal motion, it is easy to understand that the stress tends to be relieved more easily when the chains are less chemically bonded.

#### Effect of crosslinking on rheological properties of crosslinked PSA

The dynamic storage modulus ( $G'(\omega)$ ) and the loss modulus ( $G''(\omega)$ ) of PSA systems are plotted against frequency in the logarithmic scale in Figure 5(a). It shows that the increase of storage modulus with the frequency is not significant within the measured frequency range at room temperature. This result may be associated with the low glass transition temperatures of the current crosslinked PSA systems: The  $G'(\omega)$  value of the crosslinked polymer becomes more or less flattened below the frequencies where the glass to rubber transition occurs.<sup>15</sup> Loss modulus,  $G''(\omega)$  of PSA system is plotted against frequency together with  $G'(\omega)$  in Figure 5(a). At the frequency region where the flattening of  $G'(\omega)$  is observed, the value of  $G''(\omega)$  is noticeably lower than that of  $G'(\omega)$ . Since the little variation of  $G'(\omega)$  may mean little energy relaxation during deformation and thus may result in the virtually plateau or minimum of  $G''(\omega)$ .<sup>15</sup> However, as the frequency is increased the value of  $G''(\omega)$  is increased since the polymer system approaches to the rubber to glass transition zone. The increase of loss modulus can be manifested by



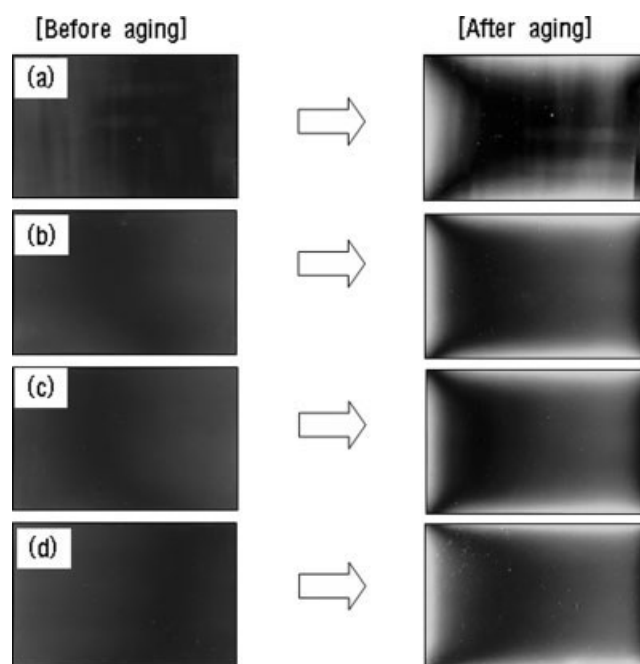
**Figure 5** (a) Storage and loss modulus of PSA systems with the various crosslinking density. (b)  $\tan \delta$  of PSA systems with the various crosslinking density.

the increase of  $\tan \delta$ , as shown in Figure 5(b). The value of  $\tan \delta$  is defined as the ratio of energy lost to energy stored in a cyclic deformation and is closely related with the stress-relaxation ability of the polymer system.<sup>15–17</sup>

The effect of crosslink on the rheological properties of PSA systems can be observed in Figure 5. (We omit the curve of PSA-B system in Figure 5 to provide the clearer figure. When PSA-B is applied the oscillatory shear deformation, it gives rise to the somewhat similar  $G'$  and  $G''$  values as the PSA-C.) The results clearly reveal that the  $G'$  and  $G''$  are increased with the curing agent, which is owing to the increase of the modulus for the polymer at the higher crosslinking density. However, it seems that the effect of crosslinking density on the dynamic moduli is smaller, being compared with that of tensile property. This observation reflects that the crosslinks may give the a little influence on the behavior at the high-frequency end of the plateau zone and in the range of transition zone, even though they give the enormous effect in the terminal zone where the crosslinks can affect the movement of polymeric backbone.<sup>18–20</sup>

In contrast  $G'$  and  $G''$ , the  $\tan \delta$  is decreased with crosslinking density especially at the frequencies below 1 rad/s in Figure 5(b), which implies that the PSA-D system with highest crosslinking density shows the lowest stress-relaxation ability. (If we consider that the PSA systems may be exposed to the more or less slow deformation rate during durability test of LCD, it seems to be reasonable that the stress-relaxation ability at the lower frequency is more important than at the higher frequency.) However, it

should be mentioned that a little influence of crosslink density on  $G'$  and  $G''$  shown in Figure 5(a) leads to the small variation of  $\tan \delta$  with the crosslinking density. In Figure 5(b), it seems that the difference of  $\tan \delta$  value between PSA-A and PSA-C is trivial. In spite of this observation, on the basis of the result in Figures 4 and 5, we may conclude that the PSA-D will show the lower stress-relaxation ability when it



**Figure 6** Variation of light-leakage phenomenon of four PSA systems after aging, measured by CCD camera. (a) PSA-A, (b) PSA-B, (c) PSA-C, and (d) PSA-D.

**TABLE II**  
**Value of Light-Leakage,  $L$  of Pressure-Sensitive Adhesive Systems Used in This Work**

Sample	Light leakage, $L$
PSA-A	1.61
PSA-B	1.38
PSA-C	1.28
PSA-D	1.20

is deformed due to the shrinkage of polarizer film, being compared with the other PSA systems.

### Light-leakage behavior of PSA systems

The light-leakage behaviors of the crosslinked PSA systems are measured by the digital camera and the results are given in Figure 6. It shows that before aging, virtually no light leakage was observed for all PSA systems. But, after the aging, all PSA systems show the light-leakage, especially along the boundary of the polarizing film due to the distortion of the adsorption axis of polarizer.

The extent of light-leakage was quantified by using the eq. (2) and the result is given in Table II. Interestingly, the light-leakage of the current PSA system becomes more serious with the crosslinking density within our sample preparation/measurement conditions. As the gel content of PSA system is increased from 76.2% to 93.7%, the value of light leakage is decreased by ca. 30%. The current result means that the specimen with the higher stress-relaxation ability shows the higher light-leakage behavior and therefore the lower durability. This observation is the conflict with the previous design strategy of PSA system, as described in the previous section.<sup>4-8</sup> Similar observations were found in our unpublished work.

The reason for our observation may suggest that, in addition to the stress relaxation ability, another variable should be considered when the new PSA system is designed. Here we suggest the stronger resistance of the PSA system as a potential variable, based on the current result. That is, even though the lower crosslinked PSA systems can dissipate the applied stress more efficiently, they are more liable to be deformed by the presence of the residual stress and hence it may result in the poor durability. Thus to provide the high durability of PSA system, the high resistance (modulus) towards the residual stress is possibly important in addition to the efficient relaxation ability of the applied stress.

However, it should be mentioned that further research must be accomplished to verify our suggestion. In our laboratory, the research on the light-leak-

age phenomenon and the design of PSA system are currently under investigation and the result will be published soon.

### CONCLUSIONS

Up to date, it has been considered that the higher stress relaxation ability is the one of the most important criterion in the design of the PSA system. However, in our work, it was observed that the PSA-D system with the lowest stress relaxation ability exhibits the lowest light-leakage and consequently the best durability. Within the scope of our work, this result may lead to conclusion that the PSA system with the higher stress-relaxation ability does not always give the lower light-leakage. In addition to the higher stress-relaxation ability, the higher deformation resistance of PSA system against the residual stress may be also important parameter to consider in the design of the PSA system. Finally it should be mentioned that, even though our work contribute to the design strategy of PSA system by correcting one of previous misunderstandings, more researches are still needed to elucidate the light-leakage phenomenon of polarizer of LCD clearly.

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