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Effect of Glass Transition Temperature of Pressure Sensitive Adhesives on Light Leakage in LCD Panel

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Pressure-sensitive adhesives (PSA), which mainly are made of acrylic polymers, have been used to fix optical films to liquid crystal display (LCD) cells to prevent contraction or expansion of the optical film under high temperature and high humidity conditions, while residual stress on the polarizing film is generated and light-leakage results from it in the peripheral area of an LCD panel. In this work, we prepared two different kinds of PSAs and tested light leakage with respect to T_g, verifying that T_g would have an effect on light leakage. We calculated their T_g values using the Fox equation and investigated them by DSC. Rheological properties of the PSAs were also acquired by a rotational rheometer which confirm the relation between T_g and dynamic moduli (G' and G''). It was found that the higher the T_g, the less is an amount of light leaked.

Keywords: Pressure-sensitive adhesive, polarizing film, glass transition temperature, rheology

1 Introduction

Adhesives are generally used to fasten two materials that have different surface properties such as surface tension or surface morphology (1). Since pressure-sensitive adhesives (PSAs), which effectively adhere to various substrates and are relatively easy to re-peel, were developed in the 1990s, they have been widely applied both in the construction field and in the manufacture of household goods because they adhere to a substrate in a short time with only a little pressure. They can also be easily formed to a desired shape. The PSAs are classified into three different types according to the primary material of which they are composed: rubber, acrylic polymer, and silicone. They can also be classified by the solvent being used: organic solvent, waterborne emulsion, and hot-melt (2). Rubber and acrylic polymer based PSAs were widely used in the past, but the amount of waterborne emulsion or hot-melt PSAs has been increasing gradually since the reduction of greenhouse gases became a global priority. Furthermore, ultraviolet cured PSAs or electron-beam cured PSAs have been developed in recent years as a means of coping with global warming.

The PSAs have been widely used for various industrial applications, for example, in the manufacture of automotive and stationary goods. Liquid crystal displays (LCDs), being used in TVs, computer monitors, notebook computers, and cellular phones have gained immense popularity in recent years due to their unique properties: they are thin, consume low power, and display pictures and video at a high resolution with low cost. One of the major components of an LCD is the polarizing film; this film is mainly composed of a polarizer, a compensation film, and PSAs. The basic structure of a polarizing plate and an LCD is illustrated in Figure 1.

The PSAs play an important role in maintaining the reliability of LCDs; further, the efficiency of the LCD manufacturing process is dependent, to a certain extent, on the type of adhesive selected. Therefore, in the LCD manufacturing industry, PSAs are an important component of polarizing films because the films themselves have weak physical properties. A polarizing film is made by unidirectional stretching a polyvinyl alcohol (PVA) film and dyeing with iodine molecules after stretching. Therefore, it can undergo some shrinkage at high temperatures and under high humidity conditions, and this dimensional instability can cause light leakage at the edges of LCDs. Consequently, one of the main functions of the PSAs in the polarizing film is to maintain the original size of the film. Another property of the PSAs in relation to a polarizing film is the rework-ability in the LCD manufacturing process. During the manufacture of the LCDs, a polarizing film is laminated onto the LCD cell and the LCD panel is

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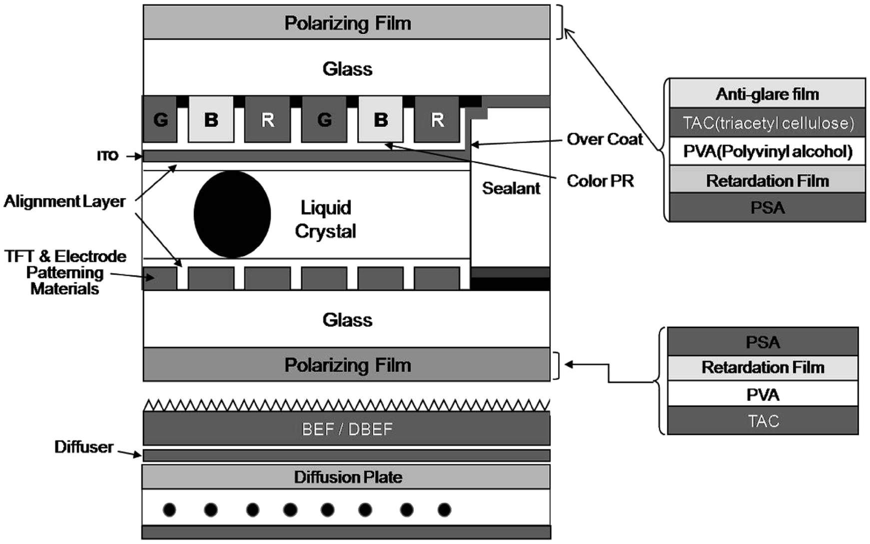

Fig. 1. Basic structure of polarizing film and LCD.

then inspected. However, some problems occur in the lamination process due to insertion of foreign particles between the polarizing film and the LCD cell even though working in a clean room. If foreign particles are present, the polarizing film must be removed from the LCD cell. Therefore, PSAs for a polarizing film must have relatively low adhesion strength between the glass and the polarizing film in order to reuse the polarizing film. In addition, PSAs for a polarizing film must also have good reliability at high temperatures, under high humidity conditions, and at extremely low temperatures in order to ensure durability of the LCDs.

The LCDs require polarizing elements on both sides of the liquid crystal cells because these cells create the images displayed on the screen. Moreover, various optical elements have been added to polarizing films attached to liquid crystal panels in order to improve the display quality. For example, retardation films have been used to prevent discoloration, and other types of films have been used to improve the viewing angle, brightness, and contrast of LCDs. PSAs are also used to attach the films described above to liquid crystal cells. The optical films are attached to liquid crystal cells or other optical films with PSAs in order to reduce light loss. Optical film with pre-applied PSA on one side is often used because the drying step can be skipped when laminating the film to another substrate. This type of optical film easily contracts or expands when exposed to high temperature or humidity. Hence, after this type of optical film is adhered to a liquid crystal panel, it can be easily peeled off. However, this process is imperfect because a liquid crystal cell may become bowed due to the contraction or expansion of the optical film at high temperature and humidity condition. Additionally, light leaks may occur in the peripheral area of liquid crystal panels due to residual stress in the optical film proper.

In order to eliminate the above phenomena, it has been suggested that the composition of PSAs should be changed (3); the inclusion of a plasticizer or an oligomer has been said to effectively soften PSAs used in the LCD manufacturing process. Effects of polymerization conditions such as temperature, time, amount of monomer, molecular weight, molecular weight distribution, and composition of the resultant copolymers have been investigated (4). On the other hand, optical film of the type that is described above is usually punched or slit into pieces of a predetermined size, allowing for the possibility that the pre-applied PSA may be partially removed by the cutting blade or become swelled out from the cutting surface. In this situation, then, the optical film may be contaminated during visual inspection or transport. The optical film must be able to be handled during the fabrication process and issues relating to bowing and light leakage must also be addressed, but improvements with regard to these problems cannot be expected with the use of a PSA composition containing a plasticizer or an oligomer. Furthermore, it is known that the lower the glass transition temperature (T_g), the less amount of light is leaked (3). Nonetheless, the relationship between T_g and light leakage has not been sufficiently investigated. In a typical LCD, linearly polarized light, created by a polarizer, is modulated by the electro-optical properties of the liquid crystal, and an image is created by switching between transmission and non-transmission using another polarizing film. The optical characteristics of the LCD, such as brightness, contrast, and hue, are therefore influenced by the performance of the polarizer.

In this study we will describe the relationship between rheological properties and T_g, and the dimensional stability of PSAs as a means of examining light leakage properties. Additionally, the basic relationship between molecular weight and rheological properties is also considered.

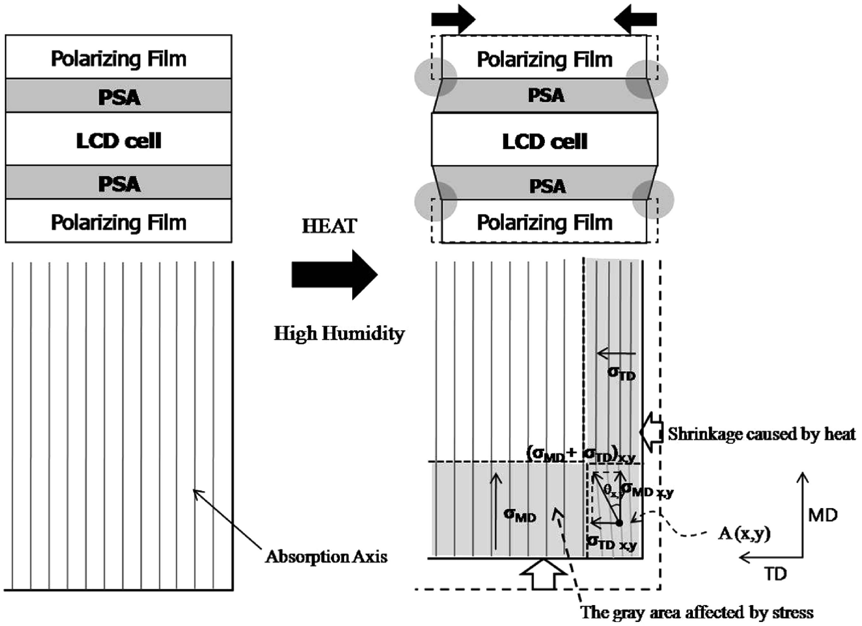

Fig. 2. Schematic diagram to explain the mechanism of light leakage.

2 Experimental

It is well known that LCD panels built with LCD cells, two polarizing plates, and backlight units (BLU) are operated by thin film transistors (TFTs) that act as tiny switches. The LCD cell consists of two pieces of thin glass plates with an LC layer inserted between them. The LCD cell can make the oscillating direction of the incident light change perpendicularly at 90° or 270°. Additionally, two polarizing films cover both sides of the LCD cell and isolate the light to create a "black condition" when a TFT unit turns on. When an LCD panel manufactured in this manner is used for a long time, heat generated from the BLU gradually shrinks the polarizing film and puts a great deal of stress on the edge of the plate. The resultant stress changes the absorbing axis of the polarizing film so that some light leaks despite the black condition. This is called light leakage (Fig. 2). The PSA layer, however, shows a tendency to prevent the polarizing film from shrinking, which in turn causes stress in the film.2

Then, the stress causes the absorption axis of the polarizing film to be bent. Assuming that a bending angle at an arbitrary point A is proportional to the ratio of σ_{MD} and σ_{TD} at that point, it could be calculated by Equation 1, where σ_{MD} and σ_{TD} are stresses in the MD and TD direction, respectively.

$$\theta_{x,y} = \tan^{-1}\left(\frac{|\sigma_{TD}|_{x,y}}{|\sigma_{MD}|_{x,y}}\right) \tag{1}$$

In addition, the intensity of the light passing through the LCD panel is described as given in Equation 2, where I_{in} is the intensity of the incident light, I_{out} is the intensity of the transmitted light, and I_{resi} is the intensity of the light transmitted before the stress occurred.

$$I_{out} = I_{in}\cos^2\left[\frac{\pi}{2} - \theta_{x,y}\right] + I_{resi} \tag{2}$$

Substituting Equation 1 into Equation 2, it can be seen that Equation 3 shows the relationship between the intensity of the transmitted light and the stresses on the polarizing plates.

$$I_{out} = I_{in}\cos^2\left[\frac{\pi}{2} - 2\tan^{-1}\left(\frac{|\sigma_{TD}|_{x,y}}{|\sigma_{MD}|_{x,y}}\right)\right] + I_{resi} \tag{3}$$

Equation 3 shows that the deterioration of polarization takes place caused by the distortion of the absorption axis of the polarizing film and makes light leak in the area affected by the stress. But because Equation 3 has been considered at a point on the LCD panel, we have to expand Equation 3 to an area. Finally, the degree of the light-leakage could be defined as the ratio of the intensity of the light transmitted by small areas at the corner and at the center of the LCD panel, as shown in Equation 4.

$$\begin{aligned} &I_{out.corner\cdot area}\Big/ I_{out.center.area} \\ &= \int_A \left\{\cos^2\left[\frac{\pi}{2} - 2\tan^{-1}\left(\frac{|\sigma_{TD}|_{x,y}}{|\sigma_{MD}|_{x,y}}\right)_{corner.area}\right] + I_{resi}\right\} \\ &\quad \times dA_{corner}\Bigg/\int_A \left\{\cos^2\left[\frac{\pi}{2} - 2\tan^{-1}\left(\frac{|\sigma_{TD}|}{|\sigma_{MD}|}\right)_{center.area}\right]\right. \\ &\quad \left. + I_{resi}\right\} dA_{center} \end{aligned} \tag{4}$$

Table 1. Compositions of PSA series

	BA [wt%]*	*IBA* [wt%]*	*AA* [wt%]*	*HEA* [wt%]*	T_g *(°C) [calculated]*	M_W *[x10^3]*
PSA1	98.5	0.0	0.5	1.0	−53.3	980
PSA2	90.0	8.5	0.5	1.0	−45.0	710
PSA3	80.5	18.0	0.5	1.0	−35.2	830
PSA4	71.5	27.0	0.5	1.0	−25.0	770
PSA5	63.5	35.0	0.5	1.0	−15.0	810

BA*: Butyl acrylate
IBA*: Isobutyl acrylate
AA*: Acrylic acid
HEA*: Hydroxy ethyl acrylate

2.1 Sample Preparation

A nitrogen-introducing tube, a thermometer, and an agitator were set up and butyl acrylate (98.5 wt%), acrylic acid (0.5 wt%), and hydroxy ethyl acrylate (1.0 wt%) were added with toluene into a reaction vessel with a cooling tube. The mixture was subjected to reaction at 60°C in a nitrogen gas stream for 4 h, and further toluene was thereafter added to the reaction liquid to obtain a solution (with a solid content of 30%) containing an acrylic-based polymer with a weight average molecular weight of 980,000 g/mole (measured from GPC). Table 1 shows the composition of the PSA samples.

The 0.15 wt% cross-linker, of a TDI-type isocyanate, was added into the acrylic-based polymer solution mixture. In addition, 0.2 wt% silane coupling agent, relative to 100 wt% solid matter of the acrylic-based polymer solution, was also added as a means of obtaining cross-linked PSA1. Following the same procedure, five other cross-linked PSA samples were prepared as shown in Table 2.

To fabricate optical films coated with cross-linked PSAs, we used a lab-scale applicator in order to control the thickness of the cross-linked PSA layer. A cross-linked PSA layer with a thickness of 25 μm was obtained by coating the cross-linked PSA on a separator consisting of a polyester film surface treated with a silicone-based release agent. The cross-linked PSA layer was transferred onto one side of a polarizing film from the separator on which the cross-linked PSA layer was formed in order to fabricate a pressure sensitive adhesion-type polarizing film.

2.2 Characterization

The T_g of the cross-linked PSAs was measured by differential scanning calorimetry (DSC) (DSC200F3, NETZSCH). All samples were heated to 30°C and rapidly cooled to −100°C at a scanning rate of 5°C/min by liquid nitrogen in order to stabilize the atmosphere during the scanning time. The T_g was obtained in the 2nd heating scan from −100°C to 30°C at 5°C/min.

Dynamic rheological characteristics of the cross-linked PSAs were measured with a rotational rheometer (MCR300, Physica, Germany) using parallel-plate geometry (diameter: 25 mm). Measurements were conducted with 0.5 mm gap distance of parallel-plate and temperature of 25°C. At first, the linear viscoelastic region of PSAs and cross-linked PSAs were determined by amplitude sweep tests which were conducted at constant angular frequency 10 s^{-1}, and then angular frequency sweep tests were performed from 0.5 s^{-1} to 100 s^{-1} with constant strain of 0.1 %.

The fabricated cross-linked PSA layer with a thickness of 25 μm was transferred onto a release film. In order to measure the dynamic rheological characteristics of the cross-linked PSAs, ca. 20 cross-linked PSA layers were laminated to a thickness of 0.5 mm. A dynamic oscillation test was performed in the frequency range of 0.5-500 s^{-1} with a constant strain of 0.1%, which is within the linear viscoelastic region of cross-linked PSAs.

To evaluate the light-leakage the pressure sensitive adhesion-type polarizing films (each size was 100 mm in length and 100 mm in width) were used. The various samples were adhered to both surfaces of a 0.7 mm thick non-alkali glass plate. The composites were then treated in an autoclave at 50°C under 5 atm for 20 min so as to achieve perfect adherence. After the samples were treated at 80°C for 100 h, they were placed on a backlight with 10,000 cd and light leakage was visually observed considering the evaluation criteria described above in Equation 4. As shown

Table 2. Compositions of Cross-linked PSA Series

	BA [wt%]	*IBA [wt%]*	*AA [wt%]*	*HEA [wt%]*	*Cross-linker (TDI-type isocyanate) [wt%]*	*Silane coupling [wt%]*	T_g *(°C) [measured]*
PSA1-C	98.5	0.0	0.5	1.0	0.15	0.2	−42.4
PSA2-C	90.0	8.5	0.5	1.0	0.15	0.2	−37.3
PSA3-C	80.5	18.0	0.5	1.0	0.15	0.2	−34.7
PSA4-C	71.5	27.0	0.5	1.0	0.15	0.2	−21.5
PSA5-C	63.5	35.0	0.5	1.0	0.15	0.2	−11.2

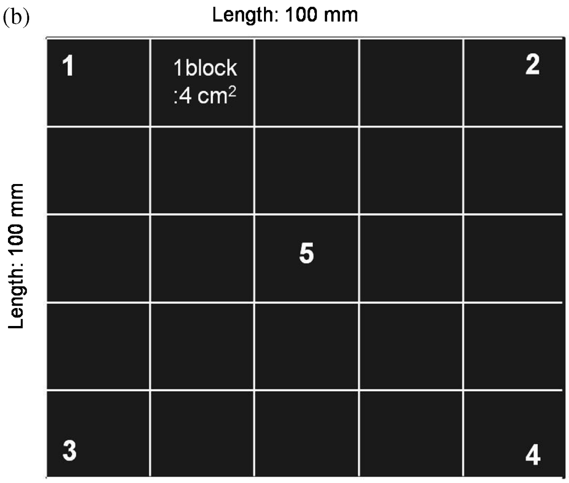

Fig. 3. Apparatus for measuring light leakage.

in Figure 3(a), a CCD camera was located in front of the sample to take full pictures of the samples, and Figure 3(b) represents the measurement for the luminance value from the light leakage through the detecting area of 4 cm^2 at fixed positions on the samples, in which the luminance value is related to the intensity of transmitted light. The luminance value of four side positions (1, 2, 3, and 4) compared with that of standard position 5. By measuring luminance in the center (L_{center}) and that in the corner (L_{corner}) of the samples, we can estimate the light leakage, i.e., how much more light leak was induced in the corners than in the center.

3 Results and Discussion

The PSAs with various glass transition temperature regions, obtained by adding different isobutyl acrylate (IBA) contents, were synthesized in order to investigate the relation between T$_g$ and light-leakage. The T$_g$ of the PSAs was estimated using the Fox equation where the W$_i$ refers to the weight fraction of component i, and T$_{gi}$ is the glass transition temperature of component i (5, 6)

$$\frac{1}{T_g} = \frac{W_1}{T_{g1}} + \frac{W_2}{T_{g2}} \tag{5}$$

The calculated T$_g$ values of our PSAs are listed in Table 1.

Figure 4 and Table 2 show experimental T$_g$ values of the cross-linked PSAs measured via the DSC. Comparing the curves from PSA2-C to PSA5-C with PSA1-C, we find that incorporation of the IBA increased the glass transition temperature. The calculated T$_g$ of the PSAs values from Equation 5 and the experimental T$_g$ of the PSAs values were similar and varied in the same order.

Since the dynamic oscillation response of the PSA gives useful information about basic material characteristics, we examined the complex viscosity of the PSAs as a function of angular frequency, as shown in Figure 5. The dynamic behaviors of the PSAs show typical non-Newtonian viscosity curves (7–10) with a shear thinning behavior in which the complex viscosity decreased linearly with log of the angular frequency. The complex viscosity of the PSA1 was higher than that of other PSA series and the complex viscosity decreased as the IBA amounts increased.

Figure 6 shows the dynamic moduli for the PSAs as a function of angular frequency; the storage modulus (G′) and loss modulus (G″) of the PSAs both decreased with increasing IBA contents at low frequencies. These results indicate that the IBA contribution is one of the main factors affecting the moduli of the PSAs. In addition, the storage moduli (G′) of the PSAs were higher than those of the loss moduli (G″) of the PSAs over the entire angular frequency regions, indicating that the PSAs possessed high elastic properties with high modulus (11–13).

As shown in Figure 7, the complex viscosity of the cross-linked PSA-C showed shear thinning behavior, and the complex viscosity of PSA3-C was higher than the others in the low angular frequency region. Compared to the complex viscosity of the PSA series, the cross-linked PSAs showed a different phenomenon. With an increase of the IBA content, the shear thinning behavior was reduced

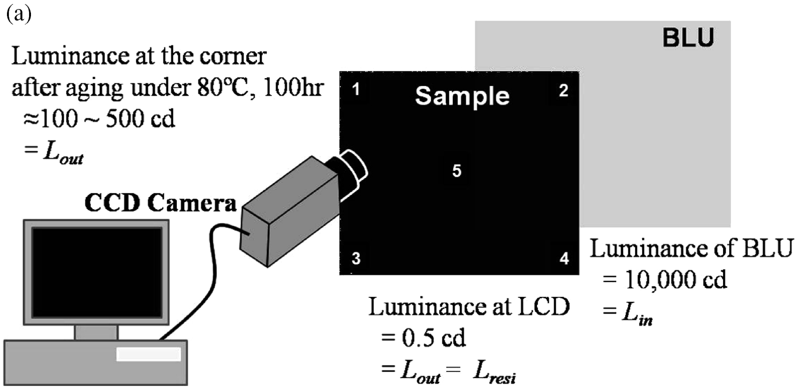

Fig. 4. DSC thermograms in the second heating runs for cross-linked PSAs.

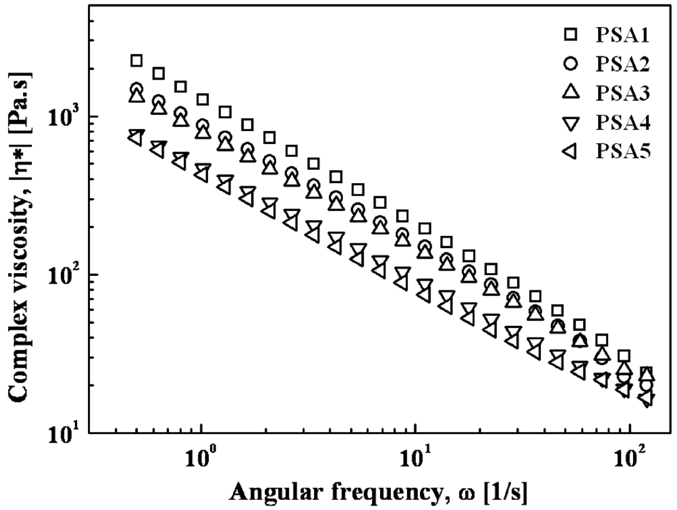

Fig. 5. Complex viscosity ($|\eta^*|$) vs. Angular frequency (ω) for PSA series.

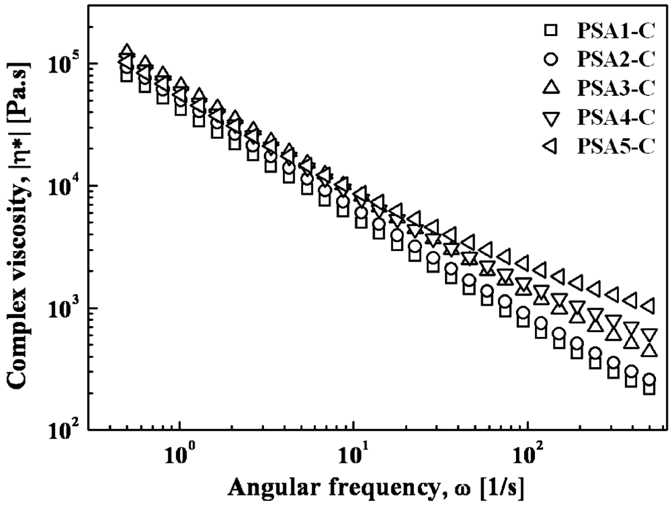

Fig. 7. Complex viscosity ($|\eta^*|$) vs. Angular frequency (ω) for cross-liked PSA series.

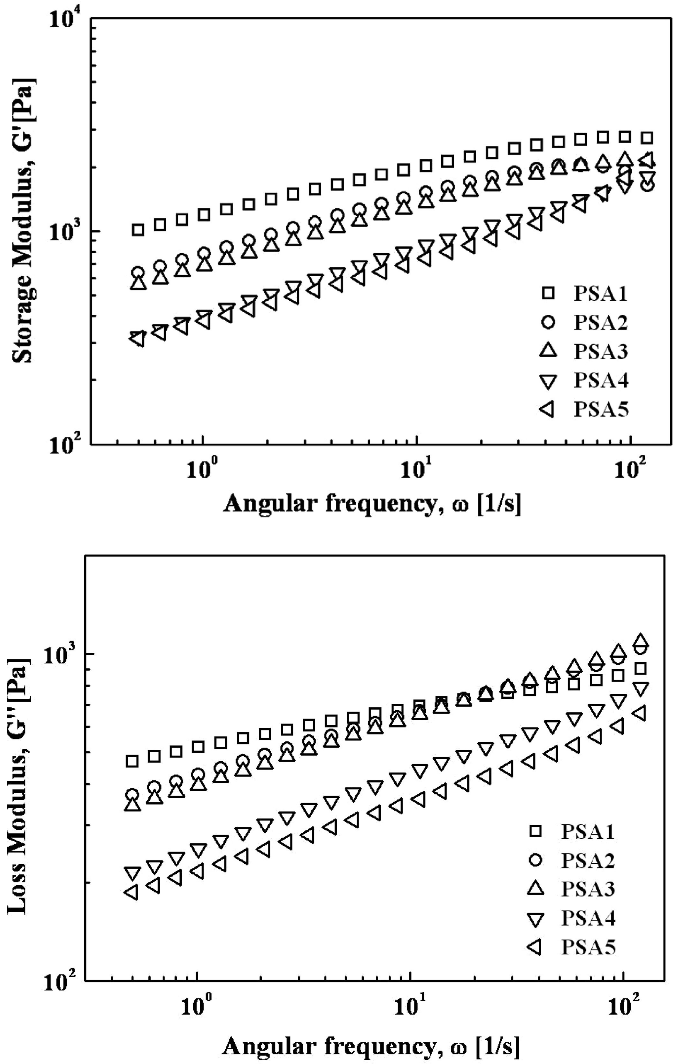

Fig. 6. Storage modulus (G′) and Loss modulus (G″) for PSA series.

above the angular frequency of ca. 10 s^{-1}. This result seems to be dependent on the cross-linked polymer properties and degree of cross-linking, because the cross-linking process enhanced the storage modulus due to formation of strong network structure.

Both G′ and G″ of the cross-linked PSAs, measured by dynamic oscillatory measurements, exhibit a monotonic increase for all frequency regions (Fig. 8), as did cross-linked PSAs as shown in Figure 6, except for G' at high frequency. Transitions of G′ and G″ from the slope change were observed for high IBA content (PSA3-C, PSA4-C, and PSA5-C). Namely, the high frequency response demonstrated a solid-like behavior except PSA1-C and PSA2-C (14–17). In addition, it is also observed that the G′ is higher than the G″ in the entire angular frequency regions, also

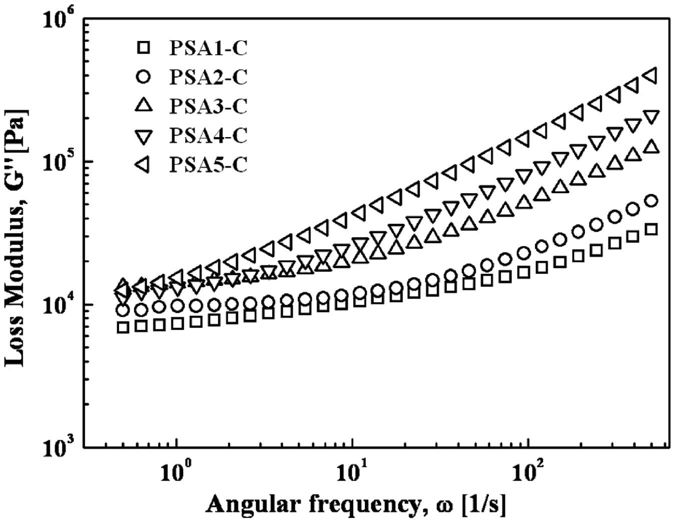

Fig. 8. Storage modulus (G′) and Loss modulus (G″) for cross-linked PSA series.

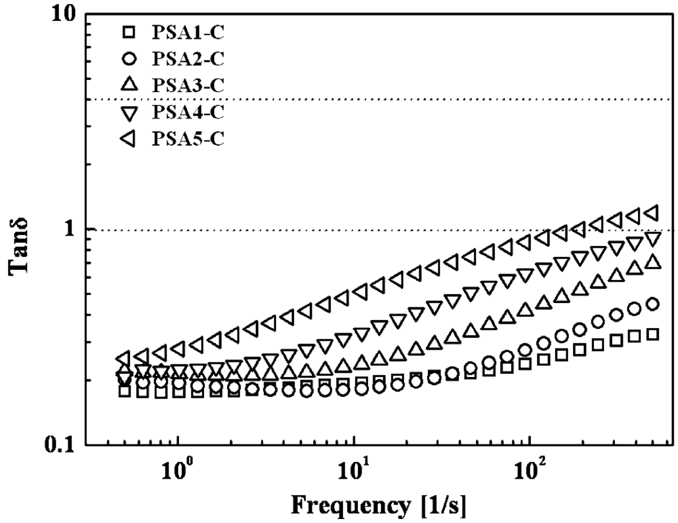

Fig. 9. Tanδ vs. Frequency for cross-linked PSA series.

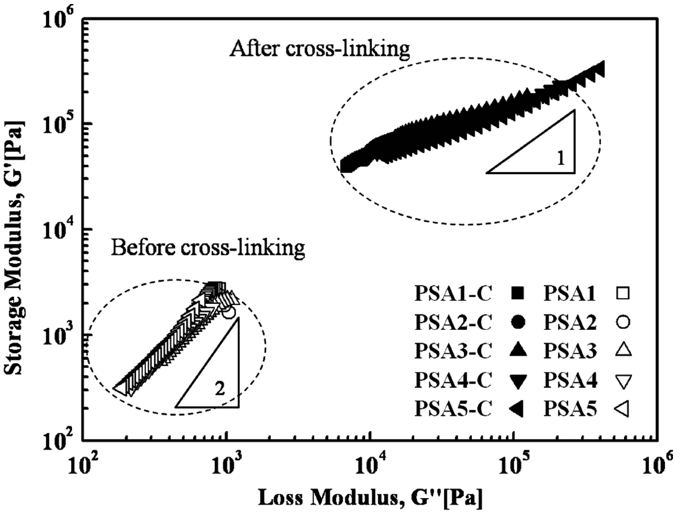

Fig. 10. Cole-Cole plot (G' vs. G'') of PSAs (open symbol) and cross-linked PSA (filled symbol).

suggesting that the cross-linked PSAs became solid-like (18). These solid-like properties can be regarded to be strongly related to IBA properties and degree of cross-linking.

The effect of IBA on degree of cross-linking and physical structure can be interpreted via the frequency-dependent tanδ ($\equiv G''/G'$) as shown in Figure 9. The measured tanδ can be analyzed in terms of its value in certain frequency regions such as non-associated (tan$\delta > 3$), weakly associated ($1 <$ tan$\delta < 3$), and strongly associated (tan$\delta < 1$) (19, 20). In all angular frequency regions, the tanδ of the cross-linked PSAs were found to be frequency-dependent, where the frequency dependence of all dynamic rheological behaviors such as dynamic modulus (storage modulus, G' and loss modulus, G'') and complex viscosity were affected by the degree of cross-linking and added IBA amounts.

Figure 10 represents the G' vs. G'' plots on a log scale for both PSAs and cross-linked PSAs (PSA-C) with different IBA contents. This plot was expressed as a Cole-Cole plot, suggesting that the homogeneous polymer exhibits similar behavior in the Cole-Cole plot (21–24). However, the PSAs and cross-linked PSAs with different IBA contents show different behaviors, having slopes of approximately 2 and 1, respectively. Therefore, it could be concluded that adding a cross-linker to PSAs transforms their internal structures and changes their physical properties.

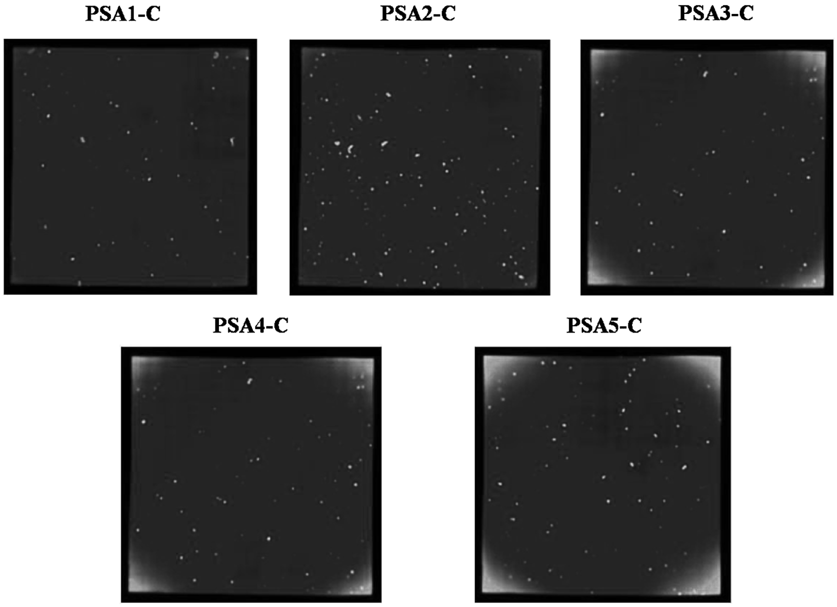

Fig. 11. Pictures of light leakage samples arranged in order of T_g.

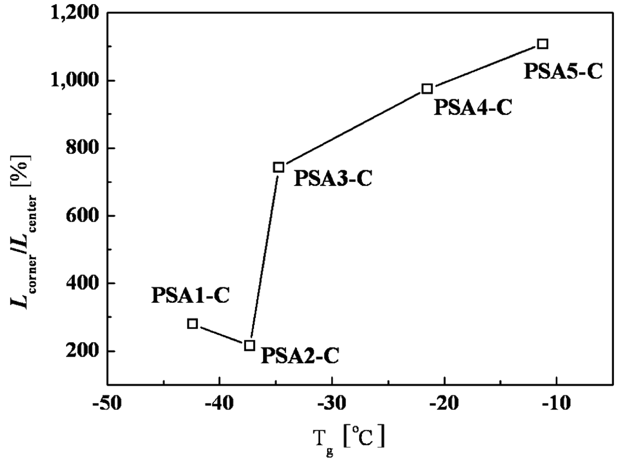

Fig. 12. Luminance difference value vs. T_g of cross-linked PSA series.

As mentioned above, this study was carried out with a hypothesis in mind such that when the T_g increases, the amount of film shrinkage decreases from the heat generated from the backlight unit (BLU), thereby producing less light leakage. We prepared five different samples for light leakage measurements, as shown in Figure 11, and evaluated the amount of light leakage as a relative value, and took pictures of the entire sample at the same time. Figure 11 demonstrates pictures of the samples arranged in the order of increasing T_g of the cross-linked PSAs. It should be noted that as the T_g of the cross-linked PSAs increases, light leakage also increases, with the exception of PSA2-C, whose T_g is –37.3°C (8.5 wt% of IBA). For more precise analysis, the amount of light leakage measured by the apparatus was used to investigate the relationship between light leakage and T_g.

Figure 12 shows a graph plotted with the relative value of luminance difference (=light leakage) with respect to the T_g of the cross-linked PSAs. The relative value of luminance difference was obtained from the average value of four side positions of each PSA as given in Figure 3(b). It was observed that increasing T_g s caused deterioration of light-leakage, with the exception of PSA2-C, and the low value of PSA2-C might be due to its more clear spots than others as shown in Figure 11. Contrary to expectations, light leakage actually increased as the T_g increased. Actually, light leakage decreased somewhat to −37°C and then began to worsen beyond this point.

It is a common fact that as the T_g of cross-linked PSAs increases, the storage modulus (G′) of cross-linked PSAs also increases. In Figure 9, it is observed that tanδ increased as PSA2-C < PSA1-C < PSA3-C < PSA4-C < PSA5-C at around 10 s^{-1} of frequency region, and this is the same order as the light-leakage. Tanδ increase with respect to the T_g means that the PSA with a relatively high T_g of cross-linked PSAs might have poorer resistance for film shrinkage than others with relatively low T_gs.

4 Conclusions

We examined light leakage with respect to T_g for different kinds of the cross-linked PSAs verifying that T_g would have an effect on light leakage. It was found that the higher the T_g of cross-linked PSAs, the more is the amount of light leaked. However, it is important to note that light leakage decreased at some T_gs of the cross-linked PSAs. The light leakage lessened somewhat to −37°C and then began to worsen other temperature region. This is also related to the fact that tanδ increases with respect to the T_g of the cross-linked PSAs. The higher T_g of the cross-linked PSAs may have poorer resistance to film shrinkage than others cross-linked PSAs with a relatively low T_g.

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