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Chapter

Pressure-Sensitive Adhesives for Flexible Display Applications

Tae-Hyung Lee, Ji-Soo Kim, Jung-Hun Lee and Hyun-Joong Kim

Abstract

Pressure-sensitive adhesives (PSA) have been used in electronics for not only attachment of two materials but also shock absorption, thermal and electrical conductivity, electromagnetic shielding, and optical property. Optically clear adhesives (OCA) have been used as a core material for optical performance of display. In addition to basic properties of OCA such as adhesion strength, transmittance, haze, and reliability, it has required dielectric constant, gap filling, and anticorrosion according to a substrate. However, the structural and functional changes of flexible display bring challenges to OCA that protects vulnerable components such as thin-film transistor, OLED, and thin-film encapsulation by stress dispersion and adjustment of a neutral plane. At the same time, flexibility and existing properties are essential. In this chapter, the development of components and performance of OCA, and evaluation methods will be discussed.

Keywords: pressure-sensitive adhesives, PSA, optically clear adhesives, OCA, display, flexible, foldable

1. Introduction

PSA is a material attached by slight pressure and detached easily from the substrate [1]. PSA's semisolid property caused by low glass transition temperature (T_g, generally 25–45°C below usage temperature) at usage temperature makes these features possible [2].

PSA can be classified by type and proportion of components into natural and synthetic rubber, acrylic, silicone, and urethane PSA [3]. The acrylic PSA is widely used in display, mobile phones, and automotive applications due to its high transparency, weather resistance, heat resistance, and high adhesion strength [4–6]. Furthermore, the acrylic PSA can have a wide range of properties because of a variety of acrylic monomers. Although the silicone PSA is not as universal as the acrylic PSA, special applications have been used that require high reliability because of its high resistance to high and low temperatures.

PSA has better processability than liquid-type adhesive because it can adhere without the hardening process and to the three-dimensional substrate. Also in the automobile industry, PSA has been considered as a substitutive process of the traditional mechanical joining method using bolts and screws because of the requirement of weight reduction. In particular, as the application of electronic and automobile industries is expanded, it is required to have not only adhesion strength

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and processability but also complex functions such as electrical and thermal conductivity, high thermal resistance, and reliability of humidity or chemicals.

OCA is used for attachment of layers in the display including cover window, touch panel, polarizers, and the light-emitting layer, which commonly require high transmittance, low haze, and corrosion resistance to ITO (indium tin oxide) film. When OCA is directly bonded to ITO film, the acid component must be excluded for the durability of the ITO film. Prolonged contact with acid can cause touch problems by reacting with metal, which makes surface resistance to increase [7]. It is also important for OCA to minimize air bubbles to reduce defects.

The demand of thin and flexible displays is increasing as interest in small and diverse designs grows [8]. Flexible displays can be distinguished by their intended use and function [9]. Recently, significant progress has been made in achieving active-matrix organic light-emitting diodes with bendable and rollable displays [10–12]. Several PSA properties are additionally required to create these flexible displays. Generally, OCA for flexible displays requires low shear modulus and T_g [13]. The recovery and stress relaxation properties of OCA are important for flexible displays because a high recovery of PSA prevents it from deformation under repeated folding-unfolding conditions [14].

2. Component of PSA

As mentioned above, acrylic PSA has been widely used for not only household or general industrial product but also electronics and automobile industries due to various advantages such as adhesion properties, optical properties, high reliability, and easy modification. Especially in OCA manufacturing, the synthesis and curing by ultraviolet (UV) radiation are used for short process time, high molecular weight, and nonsolvent process. The acrylate is one of the most suitable materials for the UV process. In this chapter, we would like to describe the materials that compose acrylic PSA including OCA.

acrylic PSA is synthesized by selecting several acrylic monomers, and the monomers can be classified into the alkyl (meth)acrylate and functional (meth) acrylate. Also, the alkyl (meth)acrylate is divided into monomers with a low T_g and a high T_g. The acrylic monomer of low T_g has a linear carbon chain that consists of 4–17 atoms, and the monomer of high T_g has a short linear chain (1–3 carbon atoms) or bulky chemical structure such as a cyclic hydrocarbon or aromatic ring. The functional monomer has a hydrophilic functional group such as a carboxyl group and hydroxyl group, and this gives an acrylic polymer reaction site for cross-linking and hydrophilicity to enhance adhesion and cohesion strength. The low T_g monomers are the main component of acrylic PSA, and the high T_g monomers and the functional monomers are added to adjust the characteristics of acrylic PSA. A typical acrylic PSA consists of 50–90% of the low T_g monomer, 10–40% of the high T_g monomer, and 2–20% of the functional monomer [3, 15].

The adhesion properties of PSA are determined by both the viscoelasticity and chemical characteristics. The relationship between the viscoelasticity and the adhesion has been studied, and the range of storage modulus (G') and loss modulus (G'') is suggested as a viscoelastic window [3, 16–19]. The surface free energy can represent the chemical characteristic of PSA. Wettability between PSA and substrate is determined by the relation of surface free energy of PSA and substrate. The immediate adhesion by quick wetting should be demonstrated right after contact with the substrate for effective use of PSA [20]. Kowalski et al. conducted a study about the tack value of acrylic PSA that increased from 300 to 700% depending on

the cross-linking degree when surface free energy of a polymer substrate increased from 20.5 to 42.9 mJ/m^2 [21].

Silicone PSA has extreme resistance at a wide temperature range from −40 to 300°C. In addition, because it can be applied to substrates having various surface energies, it is used for masking tape for printed circuit board plating, electrical insulation tape, and OCA. However, due to its drawbacks such as high price, high process temperature, and high release strength to release film, it has been applied only to specific fields that can withstand harsh environments [22]. This PSA consists of silanol-terminated silicone polymer and silanol-functional siloxane resin. The silicone polymer for PSA is a semisolid gum having a high viscosity and 600–1000 kDa of molecular mass [23].

Many manufacturers of OCA adopt the UV radiation process for the mentioned reasons, and the process is divided into two steps. The first step is a synthesis of an acrylic prepolymer by UV radiation to a mixture of the monomers and photoinitiator. The monomer mixture, except for the photoinitiator, is similar to the composition described above. But, because the acrylic acid, the major functional monomer, cannot be used, other functional monomers are evaluated to enhance adhesion strength. N-vinyl caprolactam (NVC) and N, N-dimethyl acrylamide (DMAA) are the representative monomers [24, 25]. Since nitrogen atom has a high electronegativity, it can improve the cohesion and at the same time prevent the risk of corrosion by acid [26]. In the second step, the synthesized acrylic prepolymer is mixed with multifunctional acrylate as a cross-linking agent and cross-linked by UV radiation. However, the unreacted monomers can be remained after the curing process. Because a drying process is not included in the UV radiation process, the unreacted monomers can be evaporated in the final product [27].

Currently, the development issue facing OCA is flexibility, and this not only prohibits the use of acidic monomers but also limits the selection of other common monomers. The high T_g monomers and functional monomers cause high modulus, and this property restricts the deformation of OCA. 2-Ethylhexyl acrylate (2-EHA) and butyl acrylate (BA) are used as a conventional low T_g monomer, but other monomers with longer carbon chains are considered for flexibility.

3. Development in display and OCA

All kinds of displays have a multilayer structure, and most of these layers are combined with OCA or PSA (**Figure 1**). Without OCA, the light from the backlight
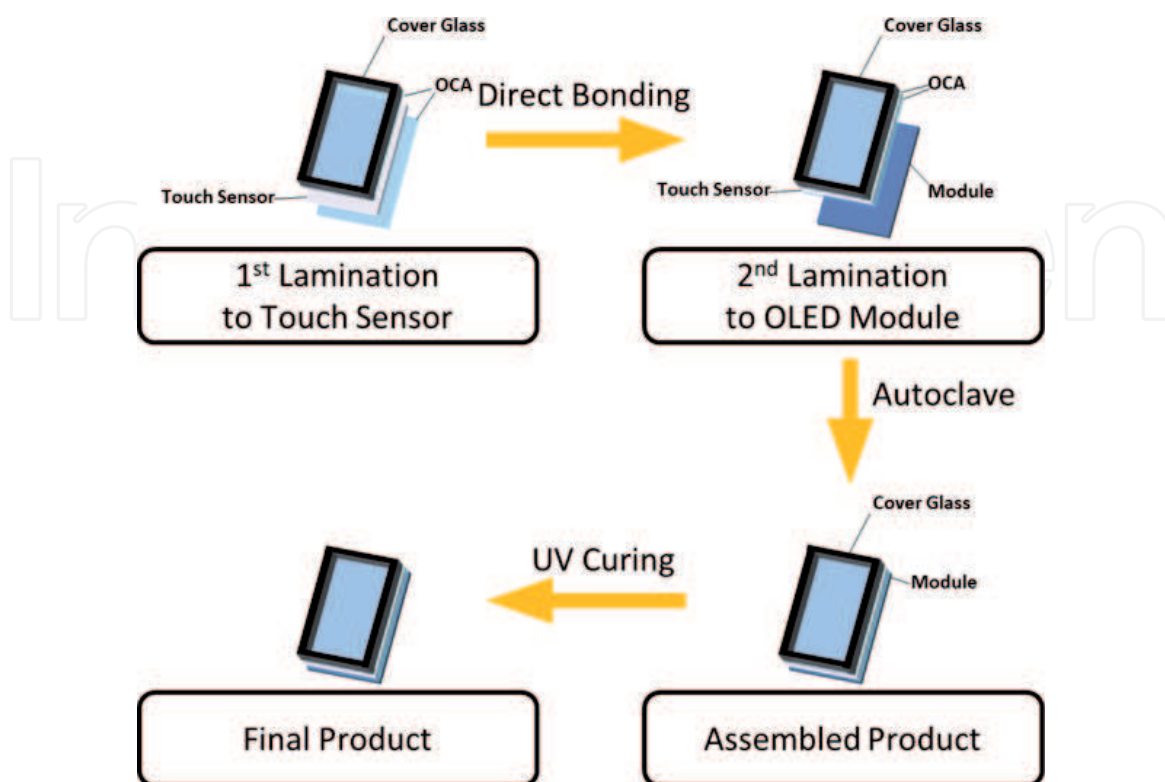

Figure 1.
Multilayer structures of the display. OCA or PSA can be applied to red layers in the structures. A touch sensor can be placed on the encapsulation by OCA or direct deposition.

unit is reflected at the interface between each film and the air due to the differences of the refractive index. Finally, less than 10% of light reach to user's eyes [28]. The presence of OCA affects the clarity of the display screen because OCA between films not only holds the film together but also prevents loss of light. Also, the difference in the refractive index between layers decreases, and a similar refractive index allows the straight progress of light without loss [29]. So, OCA should basically be transparent and have a low haze. It should also be optically isotropic, with less coloring and discoloration in environmental conditions [30].

LCD is the most widely used display to date. As shown in **Figure 1**, because it has to have a more complex structure than OLED display, more PSA layers are used. While the polarizer is used for blocking reflection of external light in OLED, it plays an important role in displaying images in LCD. The most important thing for the polarizer in LCD is to minimize the light leaking [31–34]. According to the study of Ma et al., OCA for polarizer can improve light leaking by reducing the shrinkage stress between a glass substrate and polarizer [35].

OLED is based on pixels that emit light on their own, and it can display pure black by turning off the pixels. The structures between LCD and OLED show stark differences because of their self light-emitting layer and vulnerability to oxygen and moisture [36]. Although there are many technical differences between OLED and LCD, the requirements for OCA are not much different.

A key to next-generation display technology is flexibility. The flexible displays are divided into quasi-flexible and real-flexible. The quasi-flexible display contains curved and bended displays, and Samsung's Galaxy Round is the first smartphone with a curved display. The manufacturing process of the curved display includes the bending process of the OLED panel. When the display panel is bent, spring back force is generated in the plastic film of the module to reverse the elastic deformation [37]. So, OCA of the curved module should have high adhesion property to resist this force. Otherwise, there is a risk of creating air bubbles when OCA is separated away from the glass. In order to achieve high adhesion and filling thick ink step at

Figure 2.
Application process of the 3 M CEF3806.

the same time, 3 M has developed OCA products that have low modulus and good wetting performance with semicured structure (CEF3806, 3 M). Semicured OCA shows optimal adhesion performance through additional UV curing after lamination as shown in **Figure 2**.

The foldable display is a united panel containing a variety of layers and substrates. For a flexible OLED display, it is desirable to have either an OCA that perfectly mechanically decouples the layers from each other in bending (i.e., a material with no modulus) to minimize strain on critical layers or to have OCA that can tune the position of the neutral plane during the folding process [38]. Also, although the thickness of OCA is thinner than that of OCA for a rigid display for reducing strain caused by folding, the strong adhesion, flexibility, and durability to withstand hundreds of thousands of bends are required.

4. Stress dispersion and neutral plane

OLED type of display has been chosen to implement flexible display. Major components of OLED display are thin-film transistors (TFTs), organic light-emitting diodes (OLEDs), and thin-film encapsulation (TFE). However, these layers are easily damaged by minor deformations and lose their functions because they are fragile in common [39]. To avoid this problem, the materials that have low modulus are used to absorb stress, and structural design to place these parts in the neutral plane is applied [40]. When the display composed with multilayers is bent, tensile or compressive deformation occurs depending on the position, but for each design, the layer at a particular position is simply bent without such deformation. This position is called the neutral plane. Masumi Nishimura *et al*. give the following equation for calculating the position of the neutral plane [41]:

$$\lambda = \sum_{i=1}^{n} E_i \left(h_i^2 - h_{i-1}^2 \right) / 2 \sum_{i=1}^{n} E_i t_i \qquad (1)$$

The λ means the distance from the surface of the innermost layer of the multilayer to the neutral plane, while E_i, h_i, and t_i mean the modulus of i-th layer, the distance from the innermost surface to the layer, and the thickness of the layer, respectively. As expressed in the equation, the position of neutral plane is determined by the modulus and thickness of each layer. Because OCA layers are relatively easy to control modulus and thickness without losing function, OCA layers are able to be used for adjustment of the location of the neutral plane.

The studies were conducted to protect several layers by splitting neutral planes within a multilayer. Su et al. utilized PSA layers in flexible piezoelectric mechanical energy harvesters to split the neutral planes, thereby implementing a flexible structure without destroying the components vulnerable to deformation [42]. Based on this research, Nishimura et al. published a study about splitting neutral plane using OCA in a foldable AMOLED structure. The trend of neutral plane formation according to the modulus of adhesive was identified, and the splitting was observed by the use of low modulus adhesive in the system [41].

5. Test methods for foldable OCA

Recently, foldable mobile phone, which is a type of flexible electronics, has been commercialized. So, manufacturers of mobile devices, display, and materials for electronic devices and research institutes have been set evaluation methods for foldable characteristics. The most representative and essential method is the folding

test. Samsung Electronics, a major manufacturer of electronics including mobile phones, released folding test images with the launch of Galaxy Fold in September 2019 [43]. Folding durability not only of the assembled mobile phones but also of single or laminated film used as part of electronics has been evaluated. This test is performed with various environments and test modes such as low or high temperature, high humidity, the radius of curvature, static fold, and dynamic fold for simulation of various usage environments [44].

The folding test can obtain results from a complex interaction between multi-layered components. So, the following various methods have been conducted to evaluate OCA independently.

5.1 Shear strain on folded OCA

When a book is bent, it can be seen that the unbound edge is deformed into an inclined form. Because unlike the book, however, each layer of the foldable display is attached to each other by PSA, each layer of multilayered display is deformed by different type and extent of stress. In this case, as shown in **Figure 3**, the outer layers and inner layers of the neutral plane are subjected to tensile stress and compressive stress, respectively. As such, the stress applied to each layer during folding can be calculated by the following Equation [41]:

$$\sigma = E_i \varepsilon = E i (y - \lambda)/\rho \tag{2}$$

Using a simple equation to calculate the stress (σ) by modulus (E_i) and strain (ε), the stress can be obtained by the positional difference between the target layer and the neutral plane ($y - \lambda$) and the folding radius (ρ) when the modulus of the material is fixed. Since these deviations of strain occur not only between layers but also within a single layer as shown in **Figure 4**, each layer including OCA is subjected to shear stress. So, most tests for evaluating foldable OCA are conducted under shear stress.

Although the shear strain depends on the thickness and placement of other layers, the shear strain of >300% is generally applied to the adhesive layer at a 5 mm radius of curvature. If the radius was reduced to 3 mm, the shear strain is increased up to 500–700% [45]. For the reduction of increase of shear strain by change of radius, the minimization of thickness and optimization of the structural design of multilayers have been studied [46].
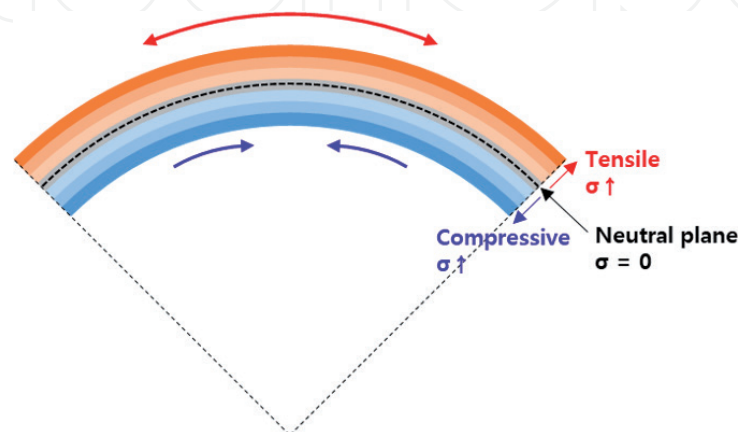
Figure 3.
Schematic diagram of applied stresses of folded multilayered display including neutral plane [41].
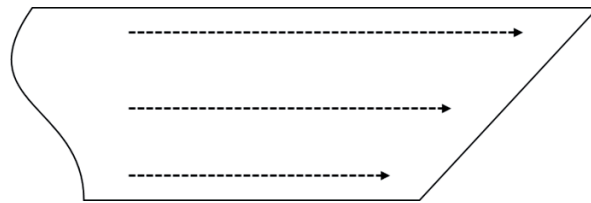
Figure 4.
Schematic diagram of shear deformation of a single layer.

5.2 Adhesion strength

Although the adhesion test is not a specific method for OCA applied to the foldable display, it is an important and fundamental method to evaluate all kinds of PSA. The test mode is selected according to the external force applied between PSA and substrate, and the general methods are peel, tack, lap shear, and pull-off test. The 180° peel test and lap shear test are conducted for foldable OCA (**Figure 5**).

Although the evaluation of adhesion is not a specific test for foldable OCA, this is considered an important factor because of several limits for satisfying adhesion strength. One of these limits is the substitution of the glass cover window to the plastic cover window for foldable properties [47]. The surface energies of several substrates such as plastic, glass, and SUS are listed in **Table 1** [48, 49]. The polyimide (PI) film has been used for the plastic cover window, and the surface energy of plastic substrate including PI is significantly lower than the surface energy of glass and SUS substrates. According to the rule of thumb, the lower surface energy of a substrate is able to decrease adhesion strength between PSA and substrate [50]. It is necessary to increase adhesion strength to the plastic substrate with minimization of rising T_g and modulus of OCA. If sufficient adhesion strength is not developed, it will not be possible to withstand the stress applied between OCA and substrate in continuous folding. The other is the limitation of the usable monomers for increasing adhesion strength as described above.

5.3 Shear modulus

The representative methods to measure shear modulus of PSA are the shear sandwich method by Dynamic Mechanical Analyzer (DMA) and the torsion method by Advanced Rheometric Expansion System (ARES) (**Figure 6**). Shear sandwich and torsion methods are subject to uniaxial and rotational shear stresses, respectively. In both of these methods, the material responds to repetitive deformation, which has a constant frequency, and its viscoelastic property is evaluated by this reaction. The elastic property reacted immediately is represented by the storage modulus, and the viscous property reacted belatedly is represented by the loss modulus [3]. Generally, the shear modulus is the storage modulus and is presented G' separated with the tensile modulus (E'). Since the shear sandwich method can have inaccuracy of pressure in the loading sample, the torsion method has been preferred.

When measuring the shear modulus of foldable OCA, Campbell et al. suggest that the storage modulus should be maintained and has a low T_g in the operating range (−20 to 80°C) [38]. Because, as presented in **Figure 7**, T_g is shown at the primary reduction of storage modulus, lowering T_g can result in modulus reduction below the operating range. Since the temperature affects the movement of molecules, polymers recognize the time or frequency as a different degree
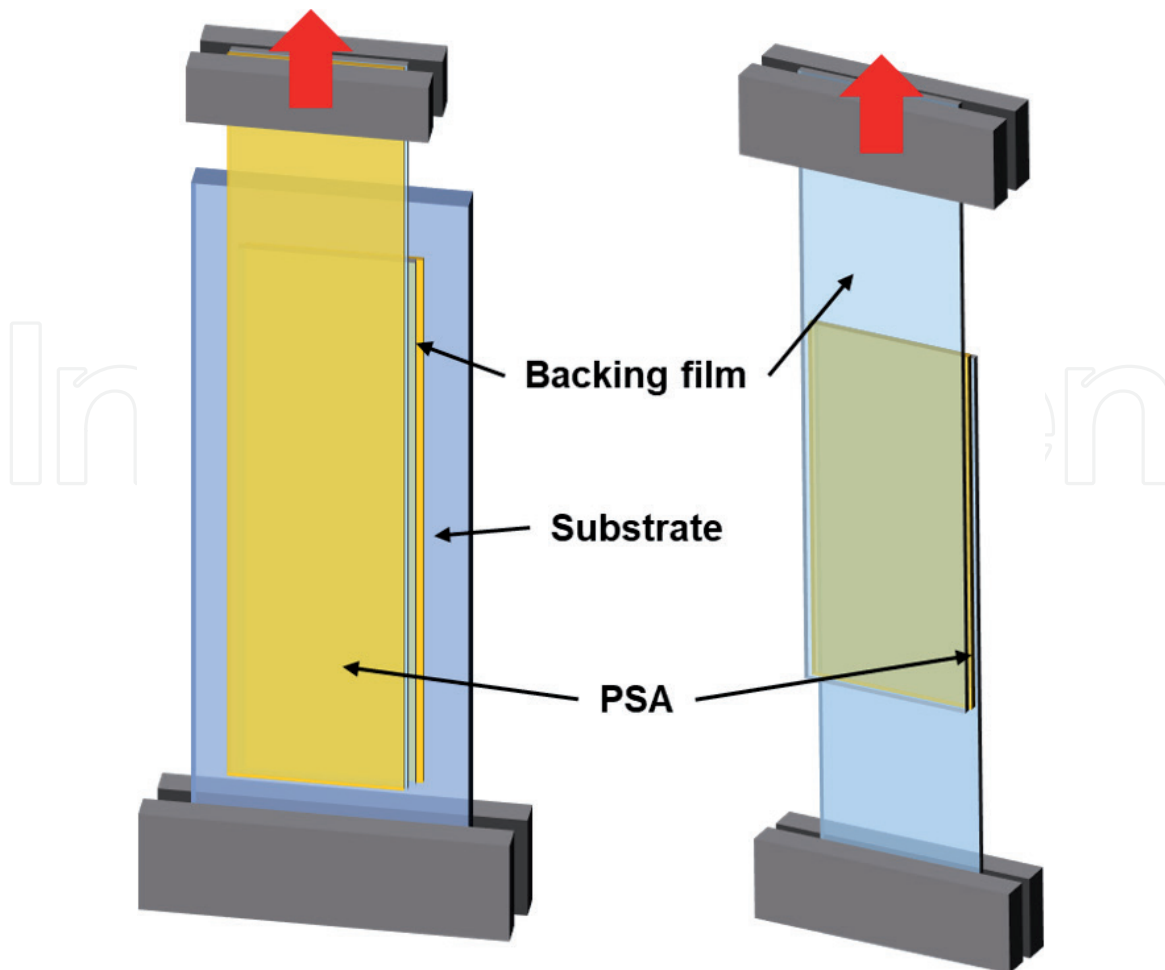
Figure 5.
180° peel and lap shear test for adhesion evaluation.

| Substrate	Polypropylene	Polyester	Polyimide	Glass	SUS
Surface energy (mN/m)	29	43	50	200–300	700–1100

Table 1.
Surface energy of substrates.

according to temperature [51]. Therefore, reducing modulus at low temperatures maintains the flexibility not only in low usage temperature but also in rapid deformation.

The maintenance of the shear modulus ensures adhesion strength [38], strain recovery, and processability at high temperature. If the modulus decreased as the temperature rise, the adhesion strength and strain recovery are able to decrease by cohesion reduction.

## 5.4 Shear creep and stress relaxation

The creep is the strain change with time under constant stress, and the stress relaxation is the stress change with time under a constant strain. Furthermore, the strain recovery after removing stress is an important factor to assess the restoration of PSA after folding. By the folding test, only the functional and macroscopic properties occurred after repetitive or static deformation can be observed, so the flexibility and resilience of a single material can be evaluated through creep and stress relaxation.
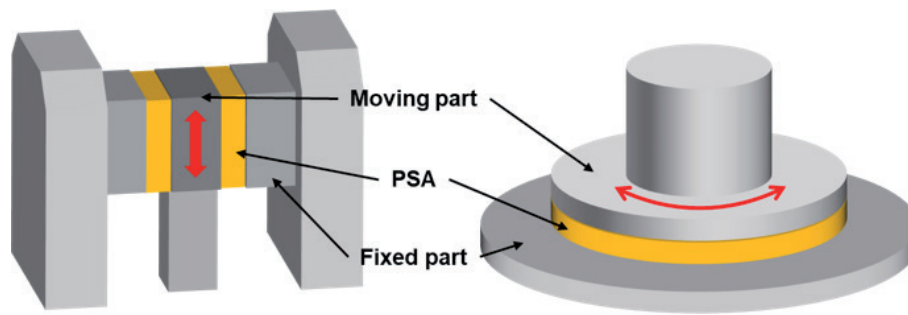
Figure 6.
Shear sandwich and torsion methods for testing shear modulus.
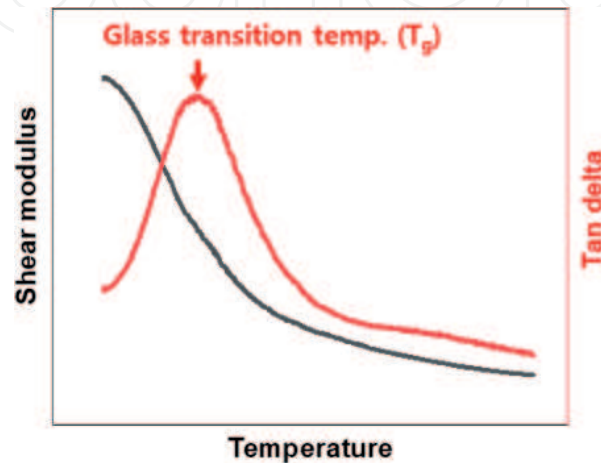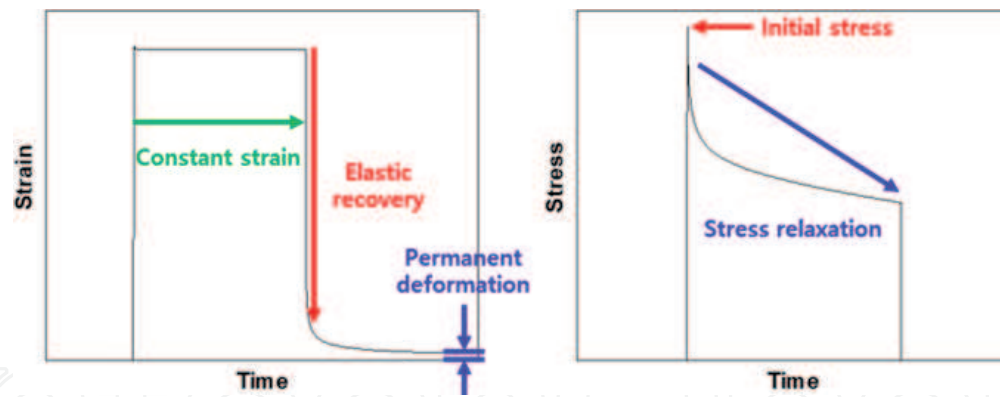

Figure 7.
The graph of shear modulus and tan delta.
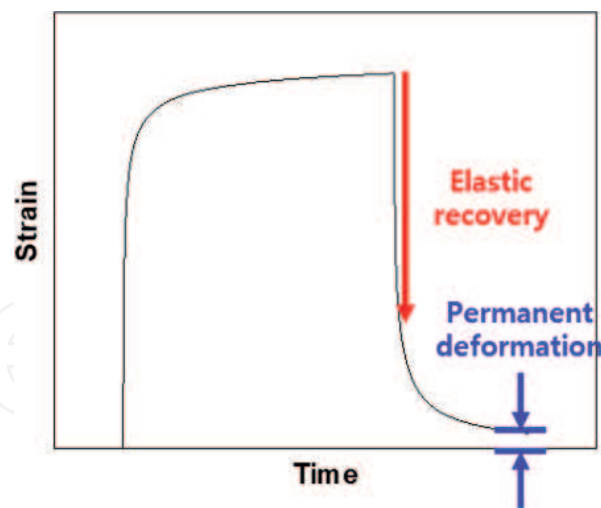
Figure 8.
The creep behavior of OCA.

In the creep test, after immediate deformation of OCA by constant stress, the strain increases continuously because of its viscoelasticity. The elastic property makes the strain to be maintained after the initial deformation, but the viscous property results in an increase of continuous deformation. Lee et al. evaluated creep recovery by dividing it into elastic recovery and residual strain after a specific time (**Figure 8**) [52]. While the elastic deformation is recovered after the removal of stress, the viscous deformation causes permanent deformation that cannot be recovered [53].

Figure 9.
The stress relaxation behavior of OCA.

In the stress relaxation test, the continued decrease of stress is observed after the highest stress appears in response to the initial rapid deformation (**Figure 9**). This reduction is due to molecular movement, bond break, and bond interchange [51]. Energy reduction due to the stretching of molecules and intermolecular motion within a range fixed by cross-linking or entanglement is mostly recoverable after stress removal, but the breakage or interchange of the intermolecular bond and intermolecular slip is irreversible.

Permanent deformation that cannot be recovered after the removal of stress in creep and stress relaxation measurement can cause wrinkles or optical defects during the folding test of the display module or final product. Considering the viscoelasticity of PSA and interaction with adjacent films that have higher modulus, the perfect recovery is not required for OCA. But, high resilience as possible may be advantageous in long-term reliability.

6. Researches of OCA for flexible display

Lee et al. researched several factors that affect the foldable properties of OCA. First is the molecular weight of the prepolymer. The acrylic resins with a molecular weight in the rage of 360,000–690,000 are synthesized, and stress relaxation and recovery properties are evaluated under a constant shear strain of 400%. Although the initial stress of the stress relaxation test increases as the molecular weight increases, the ratio of stress relaxation decreases. The recovery after stress relaxation and creep is also improved [52]. Further, as the content of the cross-linking agent increased, the recovery increases in stress relaxation evaluation. But, it is confirmed that it decreases at over specific content. As the content of the cross-linking agent increases, the cross-link point increases. And the viscous deformation decreases. But, since too many cross-link points restrict the deformation, it results in bond breakage in the strain of 400% and decrease recovery [54]. As such, the entanglement and chemical connection between molecules play a major role in deformation and recovery.

They reported adhesion properties and recovery behavior of cross-linked silicone OCA using a platinum catalyst. The effect of the degree of cross-link is similar to that of acrylic OCA. But, the speed and degree of elastic recovery after stress relaxation is much faster and better than acrylic OCA [55]. Lee et al. also attempted to improve adhesion properties and recovery by connecting a styrene-isoprene-styrene (SIS) elastomer to acrylic OCA. The entanglement of the elastomer shows a positive effect on adhesion and recovery. However, the styrene groups of SIS elastomer cause the low transparency of OCA [56].

Campbell et al. present the shear modulus of standard OCA, foldable OCA, and improved foldable OCA, and results of repetitive tension tests. Compared to standard OCA, the foldable OCA's low modulus is suitable for foldable equipment, and the improved foldable OCA has the modulus of 10–100 kPa even at high temperatures by preventing modulus decrease at high temperature. The decrease of modulus causes poor adhesion and mechanical durability in high temperature. So, they are researching that the foldable OCA has a stable modulus while maintaining a low T_g [38].

Some studies conduct a simulation method to figure out the roles of OCA. Salmon et al. demonstrate the effect of the modulus and elasticity of OCA by modeling of foldable OLED panels, and two types of OCA are used. The first one is 3 M foldable OCA and has lower modulus and higher elasticity, and the other one is 3 M OCA 8180. The softer OCA, 3 M foldable OCA, contributes less bending stress of display panel and less tensile strain of OLED layer than the other OCA. Also, they show buckling due to residual strain after unfolding. The elasticity of 3 M foldable OCA prevents continuous shear deformation and shear creep, and results in lower buckling than 3 M OCA 8180 [57]. Jia et al. presented nonlinear viscoelastic behavior of OCA in a folded display by constitutive model. They show that each layer in a multilayered display has its own neutral plane when OCA exists between the layers and this phenomenon due to the decoupling effect of the soft OCA. Also, they conduct the folding simulation of panel structures with various thicknesses of OCA. The major factor that affects the strain of the first OCA layer laid under a cover window is its own thickness. Consequentially, using thicker first OCA layer improves thermal foldability, because it reduces its own strain and risk of delamination [58].

7. Summary

In the flexible display, OCA is used for protecting TFT, TFE, and OLED in addition to the existing roles in the flat display. The low T_g and fixed modulus at operating temperature range are required to foldable OCA, while it secures high adhesion strength to the plastic substrate. Some high T_g and functional monomers used for the increase of adhesion strength have a limit to be used because of its high T_g and modulus. Also, the low T_g monomers that have long side chains are considered for OCA's high elongation and recovery. The foldable durability can be evaluated by the creep and stress relaxation test, and the strain and recovery of OCA are a combination of the elastic and viscous part. The viscous part can cause an optical defect in the display by permanent deformation. Because the perfect recovery of OCA, viscoelastic material with log T_g, and modulus is difficult, studies to compensate permanent deformation by the interaction with adjacent layers, structural design, and control of component have been conducted.

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