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Adhesion performance and recovery of platinum catalyzed silicone PSAs under various temperature conditions for flexible display applications

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ABSTRACT

We have previously performed stress relaxation tests to confirm the recovery of acrylic pressure-sensitive adhesives (PSAs) for flexible display applications. In this study, we investigate the influence of the crosslinking density of solvent-based silicone PSAs on their adhesion and recovery using a platinum catalyst (Pt) as a crosslinking agent. The peel strength and probe tack of the silicone PSAs increased in direct relation to the Pt content, until the Pt content reached 0.5–1.0 phr. Although these results prompted us to investigate the recovery of crosslinked silicone PSAs, there is a discrepancy between the low release force of silicone PSAs (0–0.5 phr of Pt) and that of the fluorine release film. For this reason, it was very hard to obtain reliable, reproducible results for silicone PSAs. Instead, we chose to investigate the recovery of a silicone PSA crosslinked with 1.0 phr of a crosslinking agent with a high release force. This combination exhibited the best adhesion performance and recovery across various temperature conditions.

1. Introduction

Silicone pressure-sensitive adhesives (PSAs) typically consist of a silanol-terminated silicone polymer and a silanol-functional siloxane resin in a hydrocarbon solvent, most commonly toluene or xylene. Silanol-terminated silicone is a high-molecular-weight polymer with diorganosiloxane repeat units. Silanol-functional siloxane resin is a dispersion of trimethylsiloxy and silsesquioxane units. This "MQ resin" has been shown to be a plasticizer [1]. Lin [2] investigated the adhesive composition and peroxide concentration of silicon PSAs and showed that the adhesion values varied noticeably with the substrate type. In addition, Ho et al. [3] examined fulfilment of the adhesive performance criteria using two silicone adhesives with different tack properties.

A high degree of cohesion is required to support an adhesive against loads and enable clean removal, whereas a low viscosity is required to wet the surface of the substrate and enable close contact. These contradicting demands are balanced by adjusting T_g and the molecular weight distribution, as well as the degree of crosslinking and branching of each copolymer [4–10]. Bu increasing the crosslink density of the polymer, the overall performance of the final product can be improved [11]. Moreover, some additives are highly reactive and increase both the rate and degree of curing. These include acrylate, methacrylate, or maleimide functional additives. Other additives such as allyl-containing cyanurates and isocyanurates only increase the overall degree of curing but not the rate [12,13]. Accordingly, the performance of silicone PSAs can also be controlled by catalysts. Currently, there are two main curing methods for silicone PSAs: peroxide-initiated free-radical cure and platinum-catalyzed addition cure. The platinum method is much more efficient than peroxide curing because it circumvents come of the problems with the latter, such as decreased catalyst efficiency with age; sensitivity to time and temperature; and use at low temperatures [14].

In our previous research, we performed a stress relaxation test to confirm the flexibility of acrylic PSAs [15]. The stress relaxation test is a powerful and reliable method. In addition, the environment is an important factor in the successful application of PSAs in flexible displays. In this study, solvent-based silicone PSAs were crosslinked with various amounts of Pt catalyst. The adhesion performance of the acrylic PSAs was evaluated by performing peel, tack, and lap shear tests using a texture analyzer. A stress relaxation test was performed by DMA to assess the recovery of the crosslinked silicone PSAs under various temperature conditions.

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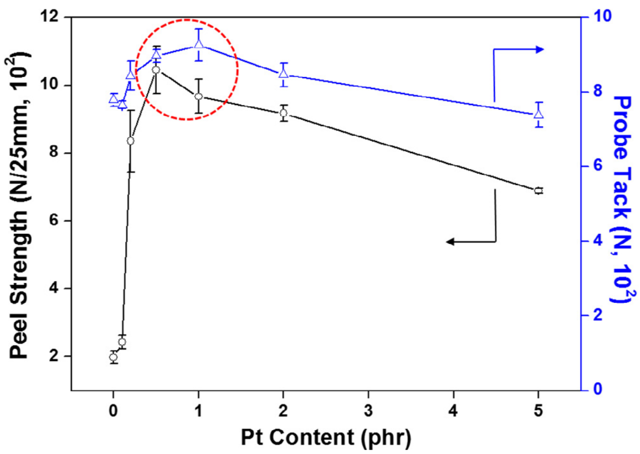

Fig. 1. Peel strength and probe tack of the silicone PSAs as a function of Pt content.

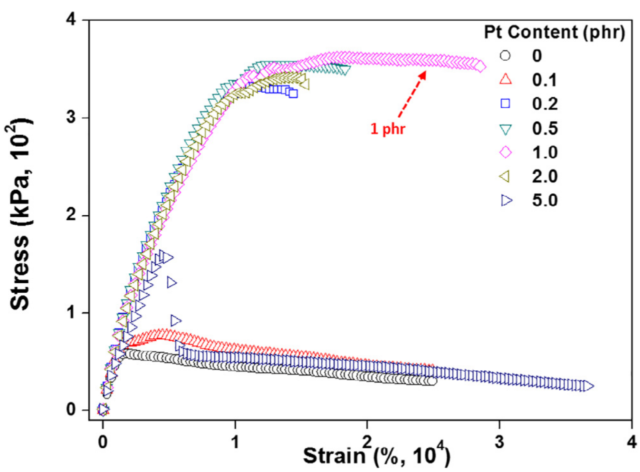

Fig. 2. Stress-strain curve from a lap shear test as a function of Pt content in silicone PSAs.

2. Experimental

The solvent-based silicone PSA (trade name 7657) and Pt catalyst (trade name 4000) were procured from Dow Corning (USA). Crosslinked PSA was prepared by blending 100 wt% silicone PSA with the Pt. The amount of crosslinking agent used was 0, 0.1, 0.2, 0.5, 1.0, 2.0, and 5.0 phr, respectively. The silicone PSAs were coated onto a polyimide film and fluorine release film, to evaluate the adhesion performance and recovery of the PSAs. Actually, the silicone release film is most widely used for acrylic PSA applications. In this work, the fluorine release film was used as liner because of the surface tension between the silicone PSA and the film. If the critical surface tension of the surface is lower than the crucial surface tension of the silicone PSA, the surface acts as a release surface [14]. Then, the PSAs were dried at 80 °C for 10 min and crosslinked at 100 °C for 20 min. Adhesion performance, peel strength, probe tack, and lap shear tests were carried out using a texture analyzer (TA-XT2i, Micro Stable Systems, UK). The results obtained on smooth and rough PSA surfaces are shown to be different indicating the sensitivity of probe tack measurements to the PSA surface. This sensitivity to surface smoothness introduces another variable which may detract from the effects by other more important PSA parameters [14]. The shear strain from the lap shear was calculated using the following equation:

$$\text{Shear strain rate } (\%) = \Delta L/t \times 100$$

where ΔL is the moving distance and t is the thickness of the PSA.

The recovery of the crosslinked silicone PSAs was measured via a stress relaxation test by DMA (Q-800 TA Instruments, USA). Initially, the samples were stabilized for 1 min at 0 N. Subsequently, a 500% strain was applied over 10 min. The measured specimens were maintained for 5 min at 0 N to assess their recovery. The recovery of the crosslinked silicone PSA was assessed as a function of crosslinking density, and temperature.

3. Results and discussion

The effects of Pt concentration on the adhesion performance of the crosslinked silicone PSA samples were investigated according to peel, tack, and lap shear tests. Each of these properties was influenced by the Pt content in the material. Specifically, crosslinking

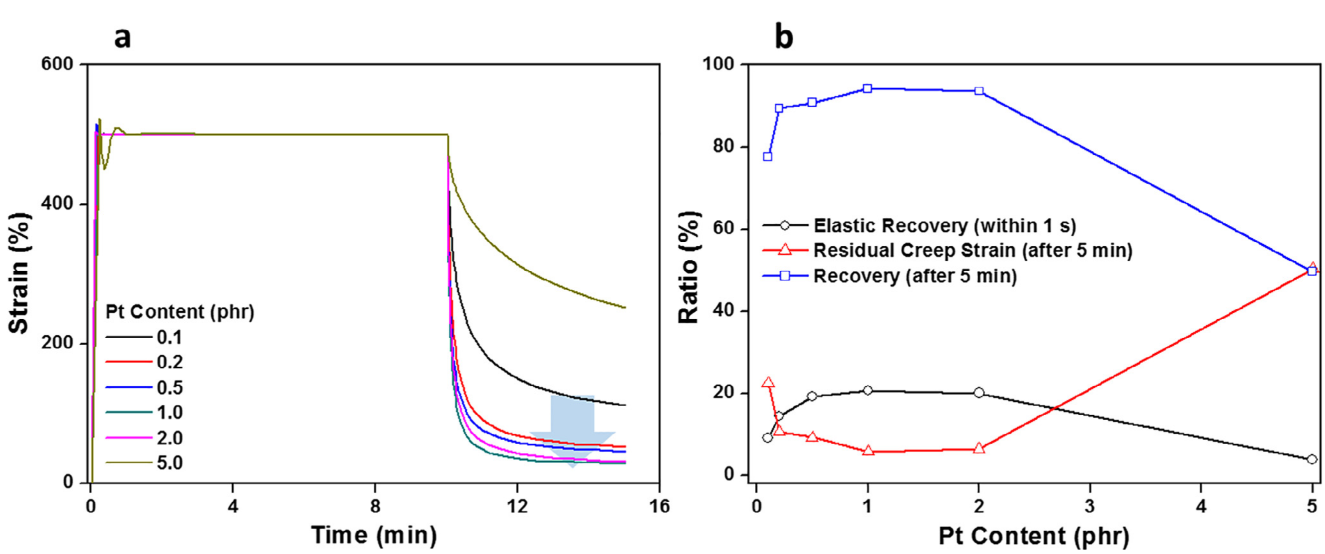

Fig. 3. Stress relaxation results for silicone PSAs as a function of Pt content: (a) strain change and (b) recovery.

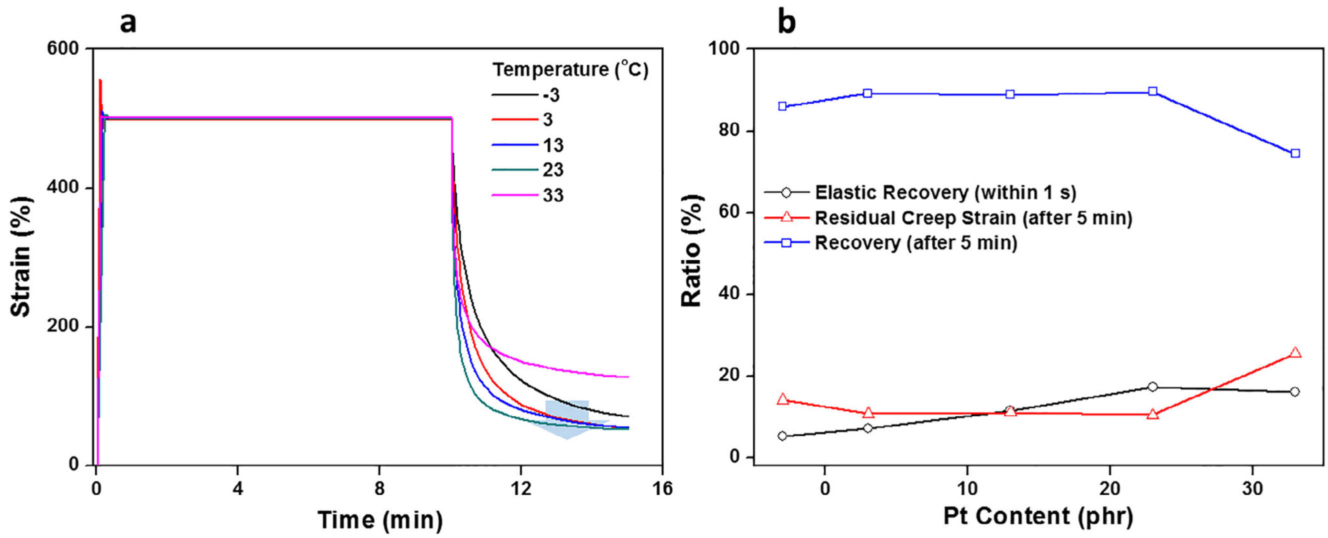

Fig. 4. Stress relaxation results for silicone PSAs as a function of temperature: (a) strain change and (b) recovery.

density is an important variable for controlling adhesion properties [16]. The peel strength and probe tack increased as the Pt increased at the low value (Fig. 1). However, the peel strength considerably decreased for Pt concentrations above 0.5–1.0 phr. Here, cohesive failure occurred when the silicone PSA sample (0–0.2 phr of Pt) was detached from the steel SUS surface.

Conversely, silicone PSAs (0.5–5.0 phr) exhibited interfacial failure while the peel strength decreased with increasing Pt content. The phenomena generated between the surface of the substrates and their adhesives, as well as their peel strength, suggested that a critical concentration of the crosslinking agent was required for achieving the optimal adhesion properties. As the Pt content increased, both the peel strength and probe tack of the crosslinked silicone PSAs decreased. This was attributed to the increased crosslinking density and the expected decrease in the compliance of the crosslinked PSAs [2].

The maximum stress of the crosslinked silicone PSAs increased with the Pt content (Fig. 2). The strain also consistently increased at Pt contents ranging from 0 to 1.0 phr, without any failure. However, both the stress and strain at maximum stress decreased as a function of Pt loading above 1.0 phr. Ultimately, the stress of a PSA with 5.0 phr Pt decreased, approaching that of neat PSA. However, this sample endured shear stress with a very high strain value, which can be attributed to crosslinking. Fig. 3 shows the strain change in the crosslinked silicone PSAs during a stress relaxation test, as a function of the Pt concentration. The elastic recovery of the crosslinked silicone PSAs increased in tandem with the concentration of the catalyst. In particular, the elastic recovery of the silicone PSAs crosslinked with 1.0 phr of Pt was the highest. On the other hand, the residual creep strain decreased with the crosslinking system. However, the residual creep strain increased with Pt concentrations above 1.0 phr.

To investigate the recovery of the silicone PSAs under various temperature conditions, a crosslinked PSA with 1.0 phr of Pt was selected according to its adhesion performance and stress relaxation results. As can be seen in Fig. 4, the recovery increased with increasing temperature, reaching a maximum at 23 °C, because of the increased relaxation of polymer chains within the PSA. However, the recovery began to decrease at 33 °C, which caused the sample to lose its elasticity, because PSAs typically have a low glass transition temperature.

4. Conclusions

In this research, silicone PSAs were mixed with Pt to prepare crosslinked silicone PSAs. The adhesion performance and recovery of the crosslinked PSAs were measured in relation to the effects of crosslinking density. The crosslinked silicone PSA with 1.0 phr of Pt was investigated as a function of temperature. The results of peel strength, probe tack, and lap shear tests showed that the adhesion properties of the PSAs improved as the Pt increased at the low value, whereas the adhesion considerably decreased as the concentration of the crosslinking agent increased beyond 0.5–1.0 phr. The recovery from the stress relaxation test consistently improved when the Pt content reached 1.0 phr. Moreover, the recovery of the crosslinked silicone PSA with 1 phr of Pt improved when the temperature was increased from -3 to 23 °C.

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