

# Fabrication of Transparent Conducting Electrode Pattern by Metal Mesh Method on Stretchable Film Substrate

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## Abstract

Smart phones have become one of major electronic devices for human being. The easy and rapid exchange of information between man and machine is carried out by the touch screen panel (TSP) installed in the smart phones. Currently the smart phones are widely spread out while displays for automobiles are in the initial stage of developments. Since the automobile displays should fit the curved area of indoor space of the vehicle, the TSPs for automobiles need to be fabricated on the flexible and stretchable substrate film. In this study, we selected the thermoplastic polyurethane film as the substrate film and examined the materials and process for fabrication of metal mesh type transparent conducting electrode films suitable for TSPs of automobile displays. The optimum UV resin system was established by the evaluation of various UV resin formulations in the UV imprinting process to make  $3 \times 3.5$   $\mu\text{m}$  trench patterns on the metal mesh type electrode film utilizing the embossed metal mask plate. The Ag paste has also been prepared by synthesis of Ag nanoparticles and formulation with the binder polymers. The inlay (filling) of Ag paste in the trench layer of transparent electrode film showed that the variation of electrical resistance was within 10% upon bending up to 100,000 times to radius of 3 mm.

## Keywords

Touch Screen Panel, Silver Paste, Stretchable Substrate Film, UV Imprinting,

## 1. Introduction

The automobile displays are becoming an important part of display devices owing to the connected car issue as shown in recent consumer electronics show (CES) and mobile world congress (MWC). Some luxury cars have multiple displays including central information display (CID), rear seat entertainment and head-up displays [1] [2]. Among these, CID is the key display for automobile industry. The touch screen panel (TSP) is widely employed in the CID and its size is gradually increasing [3]. In this study a 12-inch TSP has been fabricated utilizing a flexible and stretchable substrate film. As the size of TSP increases, the indium tin oxide (ITO) film should be replaced with a metal mesh type conducting electrode film due to the high electrical resistance of ITO film [4]. We selected thermoplastic polyurethane film as the substrate film of flexible TSP since the fabrication of TSP module requires curved surface to fit in the CID panel of the automobile. Although the TSPs fabricated on the rigid transparent film like polyethyleneterephthate (PET) films have been widely used, those on transparent flexible and stretchable films have not been reported in depth, especially for automobile displays [5] [6]. The formulation of photosensitive UV resin and silver paste are key materials for the low electrical resistance and high transparency of the TSP for automobile application. The preparation of the two important materials and the application processes have been examined.

## 2. Experiments

The transparent films tested as substrate for metal mesh type conducting electrode films include polyimide (PI-100), thermoplastic polyurethane (TPU-100 and TPU-150), another polyurethane (PU-100) and nylon (Nylon-100), obtained from Kolon Industries, Korea. The UV oligomer and monomers for formulation of photosensitive UV resins are shown in **Figure 3**, obtained from CCTech. Co., Korea. The binder polymers for the preparation of silver pastes were as following: polyurethane (Ebecryl 8411, Allnex, Sydney, Australia),  $M_w = 1000$ ; Polyester-1 (ES-365, SK Chemical Co., Korea),  $M_w = 40,000$ ,  $T_g = 16^\circ\text{C}$ ; Polyester-2 (ES-215, SK Chemical Co., Korea),  $M_w = 35,000$ ,  $T_g = -10^\circ\text{C}$ ; Epoxy resin (KDN-255, Kukdo Chemical Co., Korea),  $M_w \leq 3000$ ,  $EEW = 450 - 500$ .

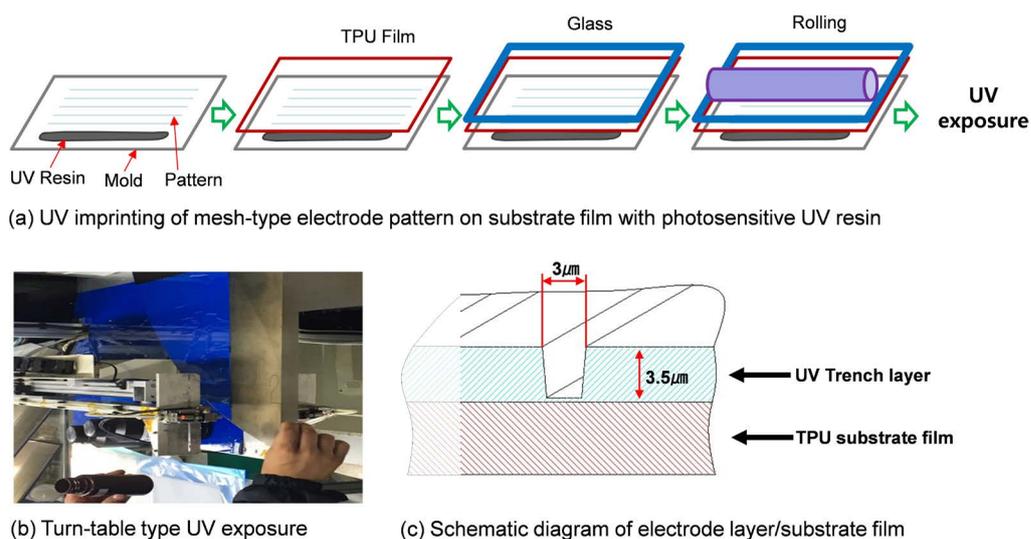
The optical properties of the substrate films and UV imprinted trench layer on substrate film were measured with HR-100 optical instrument. The mechanical properties of the films were measured with Universal Testing Machine (AGX-100N, Shimadzu Co., Japan) instrument. The bending and electrical resistance test of transparent electrode films were conducted with Flexible Endurance Tester (DLDM 111LHA, Yuasa Co., Japan) and Resistance Meter (K-9634FT, MIK21 Co., Korea). The inlay (filling) of the silver paste was carried out with scratching equipment.

In order to make a TSP for automobile application a metal mesh type electrode film should be fabricated. The process of imprinting the mesh type electrode pattern, UV exposure equipment and schematic diagram of the electrode pattern layer on substrate film are shown in **Figures 1(a)-(c)**, respectively. The photosensitive UV resin is coated on the embossed metal mold and a flexible TPU film is placed on top of the mold. Then a transparent glass is put on the TPU film, followed by rolling of UV resin, UV exposure and demolding.

### 3. Result and Discussion

#### 3.1. Substrate Films and Properties

The mechanical and optical properties of the substrate films are important subject of TSP for application to automobile industry. We have chosen five different transparent films including polyimide (PI-100) film, thermoplastic polyurethane films (TPU-100 and TPU-150 the numbers indicate thickness of the film in  $\mu\text{m}$ ), another polyurethane film (PU-100) and nylon (Nylon-100) film as candidates. The optical properties of the five films are shown in **Figure 2** and **Table 1**. The optical properties of the five films were similar and all films satisfied the specifications for



**Figure 1.** UV imprinting process of mesh type electrode pattern on the TPU film.

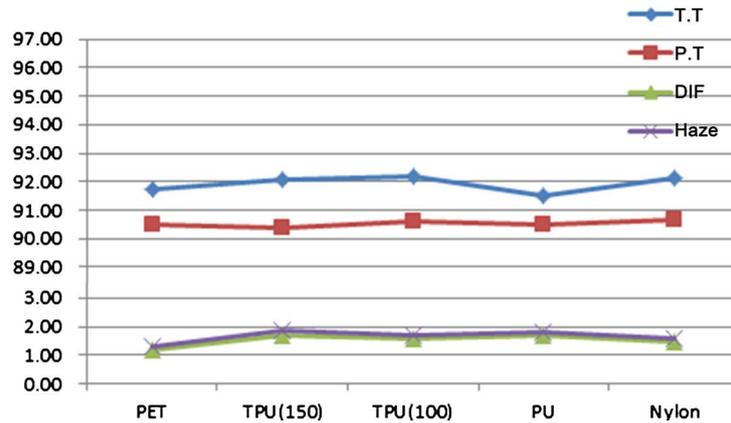
**Table 1.** The optical (a) and mechanical properties (b) of substrate films.

(a)

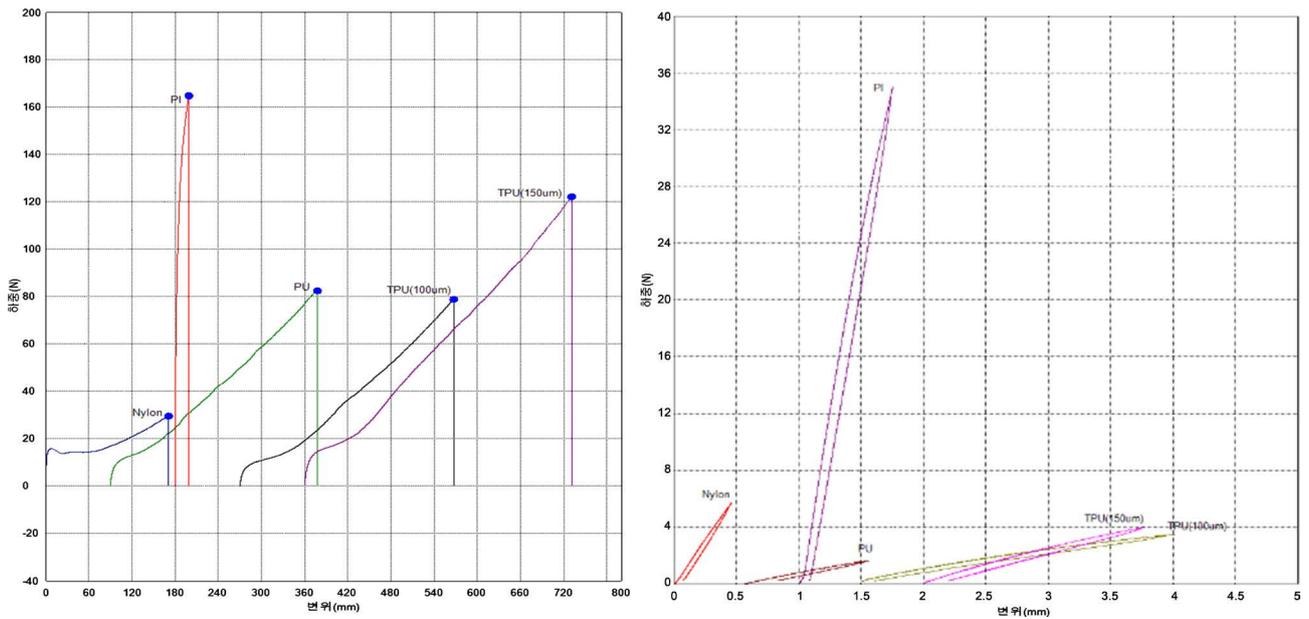
Film thickness ( $\mu\text{m}$ )	Transmittance (360 ~ 740 nm)	Yellow index ( $b^*$ )	Haze
Target value	89	2.5	3.2
PI (120)	91.72	0.83	1.30
Nylon (120)	92.11	0.51	1.57
TPU (150)	92.09	0.38	1.86
TPU (100)	92.16	0.37	1.68
PU (120)	91.49	0.3	1.81

(b)

Film thickness ( $\mu\text{m}$ )	Max. stress	Elongation (0.2% strain)	Stress (0.2% strain)	Elastic recovery
Unit	$\text{N}/\text{mm}^2$	mm	Newton (N)	%
PI (120)	231.8	1.27	53.9	80.8
Nylon (120)	74.0	1.27	9.1	84.8
TPU (150)	122.1	2.12	5.6	98.5
TPU (100)	78.8	2.72	4.3	89.2
PU (120)	82.6	1.35	3.4	97.3



(a)



(b)

**Figure 2.** The optical (a) and mechanical property (b) curves of substrate films.

use in TSP fabrication.

The mechanical properties of the films were quite different as shown in **Figure 2(b)** and **Table 1**. Although the mechanical strength of the Nylon and PI films were higher than the PU or TPU films, the elastic recovery of the PU and

TPU films were better than those of the Nylon and PI films. The TPU-150 film was chosen as the substrate film for flexible and stretchable TSP since it had both high elastomeric recovery (98.6%) and high Young's modulus of 5.63, suitable for the in-mold fabrication process of automobile TSPs.

### 3.2. UV Resin Formulation and Properties

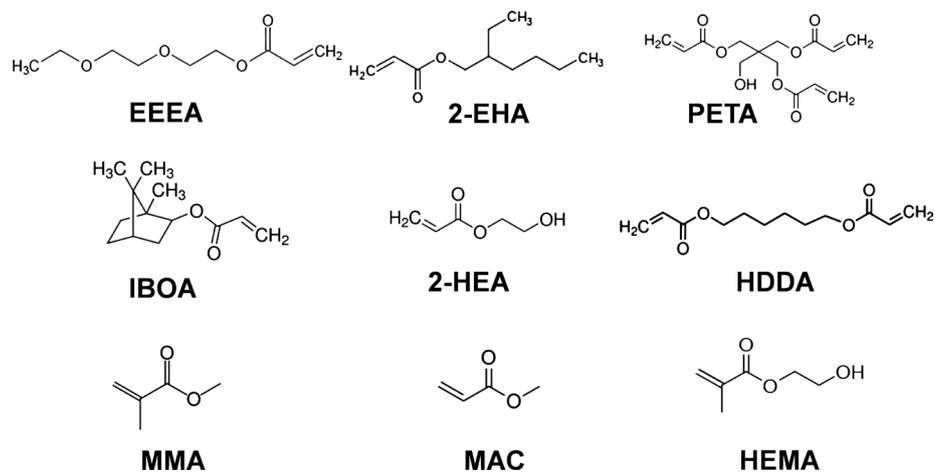
The formulation of UV resin is important in the UV imprinting process due to two reasons. First the UV resin affects the demolding of the substrate film on which fine mesh electrode patterns were formed after UV curing off the embossed metal mold.

Second, the optical and mechanical properties of the substrate film with UV imprinted electrode pattern layer are also dependent on the formulation of photosensitive UV resin. The structures of UV monomers used in the formulation are shown in **Figure 3**.

In order to check the basic demolding and optical properties of the mesh electrode patterned film, a simple UV resin formulation was prepared which were composed of F-150 polyurethane oligomer, IBOA, 2-EHA and PI-TPO photoinitiator dissolved in 2-HEMA monomer (25 wt%) as shown in **Table 2(a)**. Here F-150 UV oligomer was included to have a good adhesion to the TPU-150 substrate film. IBOA and 2-EHA monomers were employed to achieve the balance of hard and soft monomers which affect the demolding of electrode patterned TPU film off the metal mold. The test films were obtained in three different ways 1) UV resin only, 2) unpatterned UV resin layer/TPU film and 3) patterned UV resin layer/TPU film by using UV imprinting method as shown in **Table 2(b)**.

From **Table 2(b)** the two-layer film fabricated with UV imprinted electrode pattern on substrate film exhibited improved optical property compared to the film made with flat (without pattern) UV resin layer on TPU film, especially the optical properties like haze and  $b^*$  value. This may be due to the index matching effect of two-layer film and interference of light in the patterned two-layer film.

After confirming that the two-layer films consisted of patterned mesh electrode



**Figure 3.** The chemical structures and abbreviations of UV monomers.

layer on TPU-150 substrate film have good optical property for touch panel use, the mechanical and other process properties of the composite film were examined. Several issues are involved in the relationship among the UV resin formulation and mechanical/process properties of the two-layer film. The improvements of UV resin formulation are explained in **Table 3**.

**Table 2.** UV resin formulation (a) and optical properties of films (b) made with 1) UV resin only 2) Unpatterned UV resin layer/TPU film and 3) patterned UV resin layer/TPU film.

(a)						
Sample	UV oligomer		UV monomer		Photoinitiator	Total (%)
	F130	IBOA	2-EHA	PI-TPO		
				(25 wt%, 2-HEMA)		
UVX-3	60	35			5	100
UVX-4	50		45		5	100
UVX-5	50	15	30		5	100

(b)					
Optical Properties		Transmittance	b*	Haze	
Target value		89.0	3.20	2.50	
Preparation of films for examination of optical properties:					
1) Film made with UV resin only		UVX-3 (1)	93.2	2.63	2.83
2) Unpatterned UV resin layer/TPU film		UVX-3 (2)	92.7	1.93	2.08
3) Patterned UV resin layer/TPU film		UVX-3 (3)	92.6	1.99	2.15
		UVX-4 (1)	93.2	4.88	5.23
		UVX-4 (2)	92.8	2.23	2.41
		UVX-4 (3)	92.8	1.92	2.07
		UVX-5 (1)	93.5	4.44	4.74
		UVX-5 (2)	92.6	2.60	2.90
		UVX-5 (3)	92.7	2.26	2.44

**Table 3.** The improvement of UV resin formulation for mesh electrode imprinting.

Sample	Improvement of UV resin formulation	UV oligomer			UV monomer						Photoinitiator	Total (%)		
		F150	EEEE	MMA	MAC	PETA	IBOA	HDDA	2-HEA	2-EHA	PI-TPO			
											(25wt% in 2-HEA)			
UVX-6	a) Adhesion, stretchability	50								35	10		5	100
UVX-7		40	15							15	5	20	5	100
UVX-8		20	15							25	5	30	5	100
UVX-9	b) Stickiness, bad smell, demolding	40		20						10	5	20	5	100
UVX-10		40	5		10					15		20	10	100
UVX-11		15	2.5			5	37.5	20				10	10	100

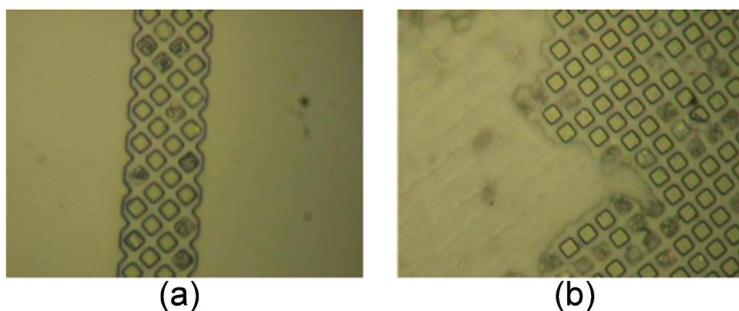
In UVX-6, 7, 8 resin formulations, the adhesion of electrode mesh layer on the substrate film was good enough upto 200% elongation of the double layer film. But the stickiness of the surface of electrode mesh layer was too high after UV curing. This was due to the high amount of soft monomers (sum of EEEA and 2-EHA monomers) which was in the range of 35 wt% to 45 wt%. So we included a hard monomer, methylmethacrylate (MMA) as shown in UVX-9, but the UV resin containing MMA was found not to be cured by UV exposure upto 150 mJ. This may be due to the steric hindrance effect of methyl group in the MMA monomer during the UV curing process. When the MMA monomer was replaced with methylacrylate (MAC) (UVX-10), the UV curing was found to proceed under a normal UV dose of 100 mJ. However, the adoption of MAC monomer in the UV formulation caused the environmental problem of bad smell. So we reformulated the UV resin without MAC monomer as shown in UVX-11.

The next important task of UV resin formulation was to solve the clean demolding of the two-layer film off the embossed metal mold after UV curing process. The UV resin formulations for this purpose are shown in **Table 4**. The UV formulation (UVX-11) exhibited good electrode pattern with dimension of  $5 \times 5.5$  (width  $\times$  depth)  $\mu\text{m}$ . When we used the UVX-11 resin formulation to pattern the mesh electrode layer with dimension of  $3 \times 3.5 \mu\text{m}$ , the electrode pattern after demolding showed some defect as shown in **Figure 4**.

The difficulty of demolding  $3 \times 3.5 \mu\text{m}$  mesh electrode layer on TPU double layer film off the embossed metal mold seemed to be due to the increased interfacial

**Table 4.** UV resin formulations for improving the demolding of the metal mold with the electrode pattern size of  $3 \times 3.5 \mu\text{m}$ .

Sample	Improvement of UV resin formulation	UV oligomer			UV monomer				Photoinitiator		Total (%)
		F150	EEEA	PETA	IBOA	HDDA	2-HEA	2-EHA	PI-TPO (25wt% in 2-HEA) (25wt% in EEEA)		
UVX-11	a) UV exposure	15	2.5	5	37.5	20		10	10		100
UVX-12		20	7.5	5	27.5	20			10	10	100
UVX-13	b) Demolding	20	7.5	5	22.5	20	5		10	10	100
UVX-14	(trench size; $3 \times 3.5 \mu\text{m}$ )	20	7.5	5	10	20	17.5		10	10	100
UVX-15		20	7.5	5		25	22.5		10	10	100



**Figure 4.** The engraved electrode pattern (a) narrow bezel part (b) wide bezel part obtained by demolding after UV curing process.

surface area between the patterned electrode layer and the fine metal mold.

So we first increased the amount of the photoinitiator as shown in UV resin formulation in UVX-12. The demolding of the two-layer film became easier as the amount of photoinitiator was increased, but the complete demolding of clean two-layer film off the metal mold was not achieved. This seemed to be caused by higher amount of hard UV monomer, IBOA in the formulation. So we replaced part of the IBOA hard monomer with neutral monomer 2-hydroethylacrylate (2-HEA) while keeping the amount of the soft monomer (EEEE) 15 wt% constant. When the amount of IBOA hard monomer was decreased to 10 wt%, the demolding of the two-layer film off the metal mold became clean and complete as shown in **Figure 5**.

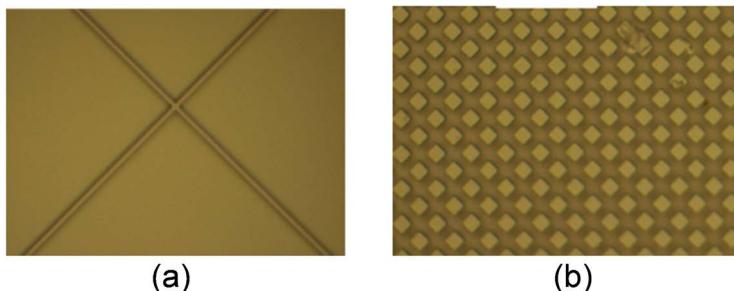
We further improved the UV formulation (UVX-15) by completely removing IBOA monomer and increasing the amount of HDDA cross-linking monomer to 25 wt%. The increase of the photoinitiator and HDDA crosslinking monomer could lead to the complete participation of diacrylate monomers in the crosslinking reaction thus increasing the strength of the double layer film during the demolding process.

### 3.3. Ag Paste, Inlay and Electrode Performance

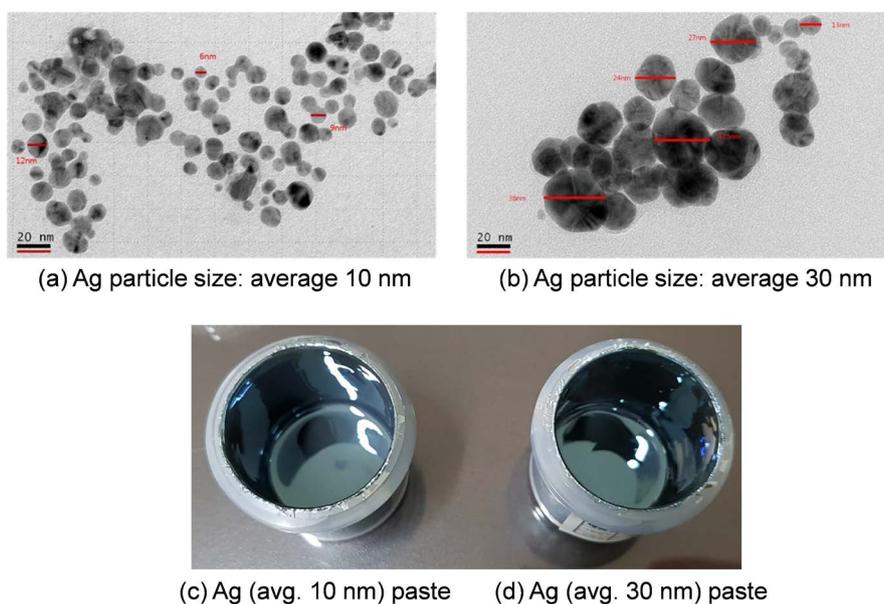
After establishing the optimum condition for patterning of trench type mesh electrodes, the formulation of Ag paste and its inlay (filling) into electrode trench (3  $\mu\text{m}$  width) and the properties of the completed transparent conducting electrode film were examined.

First silver nanopowder was synthesized by using aliphatic amine, carboxylic acid as source of surfactant, silver acetate as precursor of Ag nanoparticle and hydrazine as reducing agent of silver ion. After synthesis of the Ag nanoparticles the product suspension was washed with 2-(2-ethoxyethoxy)ethyl acetate (ECA) solvent, and then separated by centrifuge to obtain Ag nanoparticles in a powder form. The TEM images of Ag nanoparticles (a, b) and the Ag pastes (c, d) made with the Ag nanoparticles are shown in **Figure 6**.

The Ag nanoparticles were used to prepare stretchable Ag paste samples by using three different binder polymers (polyurethane, polyester, epoxy resin) and BYK dispersant in ECA/r-butylolactone (r-BTL) = 50:50 wt% mixture solvent as shown in **Table 5**.



**Figure 5.** The engraved electrode pattern (a) sensor part and (b) bezel part of electrode pattern obtained by demolding after UV curing process.



**Figure 6.** TEM images of Ag nanoparticles and camera images of Ag pastes.

**Table 5.** Formulation of Ag paste for selection of binder polymer.<sup>1</sup>

Ag Ink	Ag nanoparticles		Binder polymer <sup>2</sup>	Solvent	Additive
	Ag (10 nm)	Ag (30 nm)		ECA/r-BTL <sup>3</sup> = 50/50	BYK-180 <sup>4</sup>
<b>AgP-1</b>	8.8	70.2	Polyurethane: 4.0	16.0	1.0
<b>AgP-2</b>	8.8	70.2	Polyester-1: 4.0	16.0	1.0
<b>AgP-3</b>	8.8	70.2	Polyester-2: 4.0	16.0	1.0
<b>AgP-4</b>	8.8	79.2	Epoxy resin: 4.0	7.0	1.0

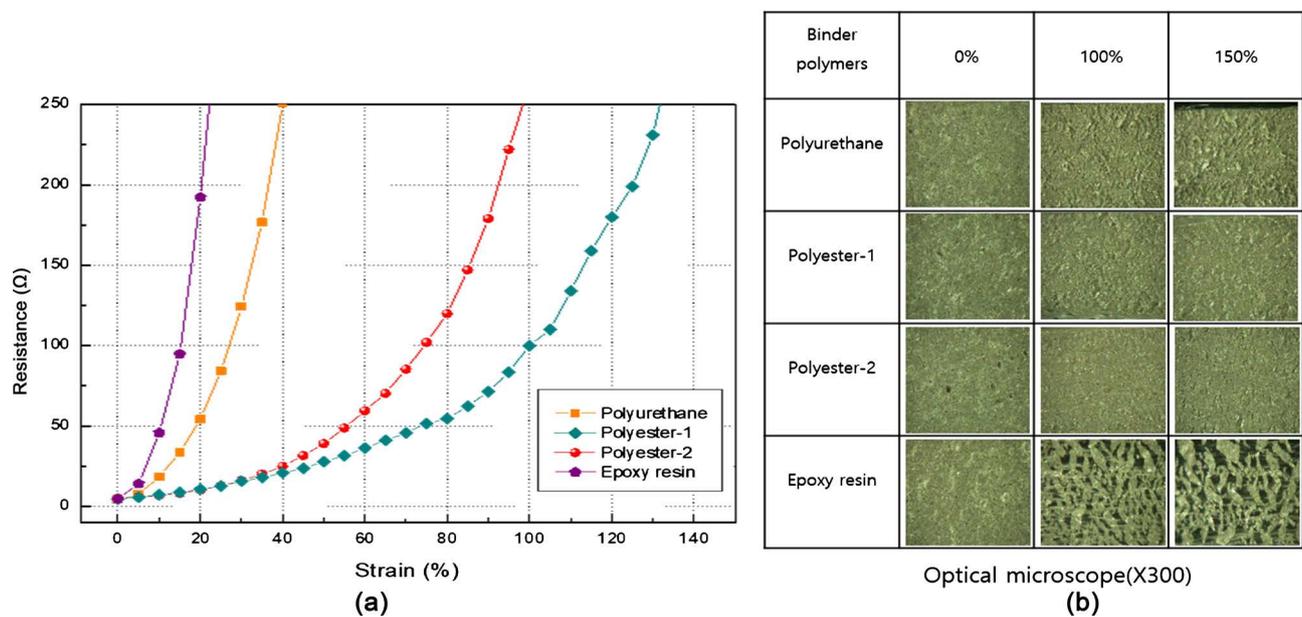
<sup>1</sup>Formulation of Ag paste is based on wt%. <sup>2</sup>Binder polymers: polyurethane (Ebecvyl 8411, Allnex, Sydney, Australia),  $M_w = 1,000$ ; Polyester-1 (ES-365, SK Chemical Co., Korea),  $M_w = 40,000$ ,  $T_g = 16^\circ\text{C}$ ; Polyester-2 (ES-215, SK Chemical Co., Korea),  $M_w = 35,000$ ,  $T_g = -10^\circ\text{C}$ ; Epoxy resin (KDN-255, Kukdo Chemical Co., Korea),  $M_w \leq 3000$ ,  $EEW = 450 - 500$ . <sup>3</sup>Solvent: r-BTL (r-butylzollactone). <sup>4</sup>Additive: BYK-180 (BYK Co., Germany).

The Ag pastes in **Table 5** were tested by screen printing on the newly made stretchable polyurethane film of thickness 150  $\mu\text{m}$  (utilizing the Clear Flex 30 A and B resin obtained from Smmoth-on Inc., U.S.A.) on which crosslinked polyester masking layer was coated to a thickness of 3  $\mu\text{m}$  according to the author's previous work [7]. After screen printing the polyurethane film with polyester masking layer, the electrode pattern was cured thermally at 130 $^\circ\text{C}$  for 30 min in a convection oven to give dry Ag electrode pattern with thickness of 30  $\mu\text{m}$ . The electrical resistance vs. the strain plots of Ag electrode pattern (30  $\mu\text{m}$ ) screen printed on the polyester masking layer (3  $\mu\text{m}$ )/Clear Flex polyurethane (150  $\mu\text{m}$ ) double-layer film is shown in **Figure 7(a)**. The Ag paste with polyester as binder polymer showed moderate increase of electrical resistance upto 150% strain while the Ag pastes with epoxy or polyurethane as binder polymer showed very high resistance values. The optical microscopic images of screen printed Ag pastes with epoxy or polyurethane as binder polymer exhibited micro-cracks

over 100% strain while Ag pastes with polyester binders showed no cracks upto 150% strain (**Figure 7(b)**) [8].

The Ag paste with polyester-1 binder polymer (AgP-2) showed wider strain range than AgP-3 paste with polyester-2 from **Figure 7**. However, the adhesion of Ag paste with polyester-2 binder polymer (AgP-3) to the substrate film was better than that of Ag paste with polyester-1 binder, thus polyester-2 was selected as binder polymer of Ag paste.

Having selected the polyester-2 as binder polymer we further modified the formulation of Ag pastes for the low electrical resistance and good adhesion to mesh electrode layer. The good adhesion of Ag paste to the trench layer made of UVX-15 resin is important because the depth of Ag paste filled in the trench layer is less than 3.5  $\mu\text{m}$  which is labile to the strain deformation. The modification of Ag pastes is shown in **Table 6**. Here the Ag pastes (Ag T-1 and Ag T-2) showed lower electrical resistance than Ag T-3 paste. The electrical resistance usually decreases with the increasing Ag nanoparticles and decreasing binder polymer which is a non-conductor. It was noted that the effect of decreasing the



**Figure 7.** The surface resistance vs. strain curves (a) and optical microscope images (b) of the screen printed Ag pastes made with formulations in **Table 5**.

**Table 6.** Silver paste formulations and evaluation for low electrical resistance and high adhesion to the trench layer surface.

Ag Ink Pastes*	Ag nanoparticles		Binder Polymer: Polyester-2	Solvent ECA/r-BTL = 50/50	Additive BYK-180	Evaluation	
	Ag (d = 10 $\mu\text{m}$ )	Ag (d = 20 $\mu\text{m}$ )				Electrical Resistance ( $\Omega/\text{square}$ )	Adhesion
Ag T-1	8.8	70.2	2.0	18	1.0	4-5	X
Ag T-2	8.8	70.2	3.0	7	1.0	4-5	O
Ag T-3	8.8	79.2	4.0	7	1.0	8-10	O

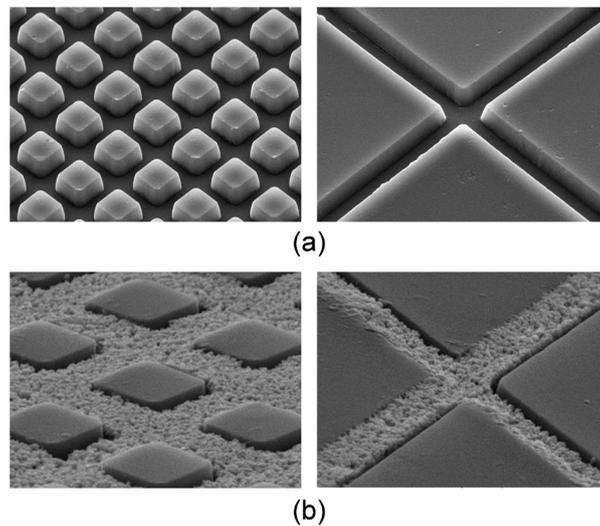
\*The amount of components are expressed in wt% unit.

binder polymer was stronger than increasing the Ag nanoparticles from **Table 6**. The comparison between AgT-1 and AgT-2 pastes also indicated that there is a limit to the decrease of binder polymer which could not guarantee the adhesion to the trench layer surface.

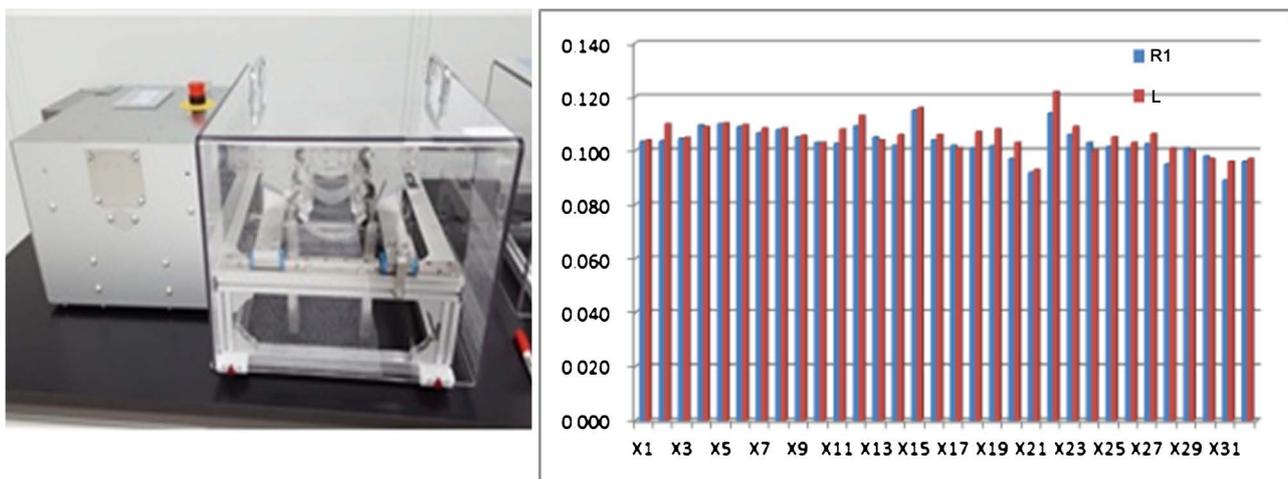
After optimizing the Ag paste formulation for inlay (filling) into the trench layer, the Ag paste (AgT-2) was filled in the trench layer by using scratching machine. **Figure 8** shows that the Ag paste was filled well in the trench layer both in the sensor part and bezel part.

The variation of electrical resistance of the Ag paste (AgT-2) filled electrode film upon bending to 3 mm radius upto 100,000 times was 8.8% (the specification 10.0%) as shown in **Figure 9**.

This basic research on the materials and process of metal mesh type TSP fabrication will be a sound foundation for use in the automobile display industry.



**Figure 8.** The SEM images of empty trench layer pattern (a) and filling (inlay) of Ag paste in the trench layer (b).



**Figure 9.** Bending test equipment and the variation of electrical resistance of Ag paste filled electrode film after 100,000 times, 3 mm radius bending test.

## 4. Conclusion

Touch screen panels (TSPs) for automobile application require a flexible and stretchable substrate film. In this work a thermoplastic polyurethane film was selected as a substrate film for fabrication of mesh type electrode patterns by using photosensitive UV resin. In the mesh electrode patterning the balance of hard and soft monomer in addition to the balance of multifunctional monomer and monofunctional monomer were found important for the optical and mechanical properties of UV resin formulation, especially the demolding process after UV curing off the finely patterned metal mold. It was also noted that the formulation of silver pastes is very important for the inlay process and balance between the low electrical resistance and bending test up to 100,000 times with radius of 3 mm. The nano-sized silver particles were found to give the low electrical resistance of 4 - 5  $\Omega$ /square in combination with the polyester binder polymer with high molecular weight ( $M_w = 35,000$  g/mol) and low glass transition temperature ( $T_g = 16^\circ\text{C}$ ).

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## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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