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Transparent Conductive Multilayer Films with Optically Clear Adhesive Interlayer for Touch Panel Devices

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Abstract: We synthesized Optically Clear Adhesive (OCA) polymers and tested as an interlayer between two polyethylene terephthalate (PET) films on top of which transparent Indium Tin Oxide (ITO) was deposited. The polymers of OCA was produced from free radical polymerizations by using 2-ethylhexyl acrylate (2-EHA), methyl methacrylate (MMA), Butyl Acrylate (BA), Acrylic Acid (AA) and 2-hydroxy ethyl methacrylate (2-HEMA). Some of fundamental OCA characteristics including T_g, Probe Tack and Peel strength were investigated with the change of MMA and 2-HEMA monomers. The characteristics of ITO coated three layer film were also investigated on the change of RF powder, deposit pressure and heat treatment. These results suggest that the multilayer film using synthesized OCA have the potential to be used in the fabrication of touch panel devices.

Key words: Touch panel devices, OCA, ITO, PET, multilayer film

INTRODUCTION

The touch screen interface is being used in a wide variety of applications to improve human-computer interaction (Stetson, 2006; Nichols, 2007; Lee *et al.*, 2007). As the technology advances, people may be able to operate computers without mice and keyboards. Because of its convenience, touch screen technology solutions has been applied more and more to industries, applications, products and services. The touch panels themselves are based around four basic screen technologies: resistive, capacitive, infrared (IR) and Surface Acoustical Wave (SAW) (Chang and Holguin, 2005).

Each of those designs has distinct advantages and disadvantages. As the human body is also a conductor, touching the surface of the screen results in a distortion of the body's electrostatic field, measurable as a change in capacitance. Different technologies may be used to determine the location of the touch. The advantage is that capacitive technology transmits almost 90% of the light from the screen. The superior efficiency gives capacitive better than resistive technology. The base substrates of capacitive touch panel consist of an insulator such as Optically Clear Adhesive (OCA) and film coated with a transparent conductor such as Indium Tin Oxide (ITO) (Potter *et al.*, 1988; Sears *et al.*, 1992).

Among different base polymers used in making OCA, alkyl acrylates, such as poly (n-butyl acrylate), poly (2 ethyl-hexyl acrylate) and poly (iso-octyl acrylate), was frequently used for the fastest growth in commercial applications. Their popularity is mainly attributable to optical clarity, oxidative and ultraviolet resistance, migration resistance, low toxicity and low cost.

In this study, Optically Clear Adhesive (OCA) resin were synthesized and tested as an interlayer between two polyethylene terephthalate (PET) film on top of which thin transparent Indium Tin Oxide (ITO) conductive film was deposited (He and Gu, 2003). These multilayer type ITO-PET films were used in the fabrication of touch panel devices. In a further study, the synthesized OCA films with colorless, high thermal stability and flexibility were applied to the touch panel. Some of fundamental OCA performances were also investigated.

MATERIALS AND METHODS

Materials and purification: 2-ethylhexyl acrylate (2-EHA, Aldrich Chemical Co.), methyl methacrylate (MMA, Aldrich Chemical Co.), butyl acrylate (BA, Jusei Chemical Co.), acrylic acid (AA, Aldrich Chemical Co.), 2-hydroxy ethyl methacrylate (2-HEMA, Aldrich Chemical Co.) were purified by distillation under reduced pressure. 2,2'-azobisisobutyronitrile (AIBN, Junsei Chemical Co.),

isoporon diisocyanate(IPDI, Aldrich Chemical Co.) were used as received. Ethyl acetate (Samchun Pure Chemical Co.) was purified by distillation under reduced pressure and stored over 4-Å molecular sieves. The acrylate monomers used in OCA were shown in the Table 1 (Demartea *et al.*, 1996; Leong *et al.*, 2002; Do *et al.*, 2006).

Synthesis of Optically Clear Adhesive (OCA): The polymers of OCA were synthesized using 2-EHA, BA, AA, 2-HEMA and MMA through solution polymerization initiated by AIBN in ethyl acetate. The polymerization was performed in a 500 mL, four-necked, round-bottomed flask equipped with a thermometer, condenser, dropping funnel and mechanical stirrer. The typical synthetic method was as follows. The flask was charged with a monomer and solvent mixture and then polymerization was started at 60°C in a heating mantle for 3 h. After polymerization, the remaining initiator was added and the flask was kept at 60°C for 3 h (Gower and Shanks, 2004). Table 2 shows the composition of polymerization.

Table 1: Characteristics of monomers used in OCA

Monomer	Abbreviation	Tg*(°C)	Function
2-Ethylhexyl acrylate	2-EHA	-60	Soft
Butyl acrylate	BA	-45	Soft
Acrylic acid	AA	90	Functional
2-Hydroxyethyl methacrylate	2-HEMA	55	Functional
Methyl methacrylate	MMA	105	Hard

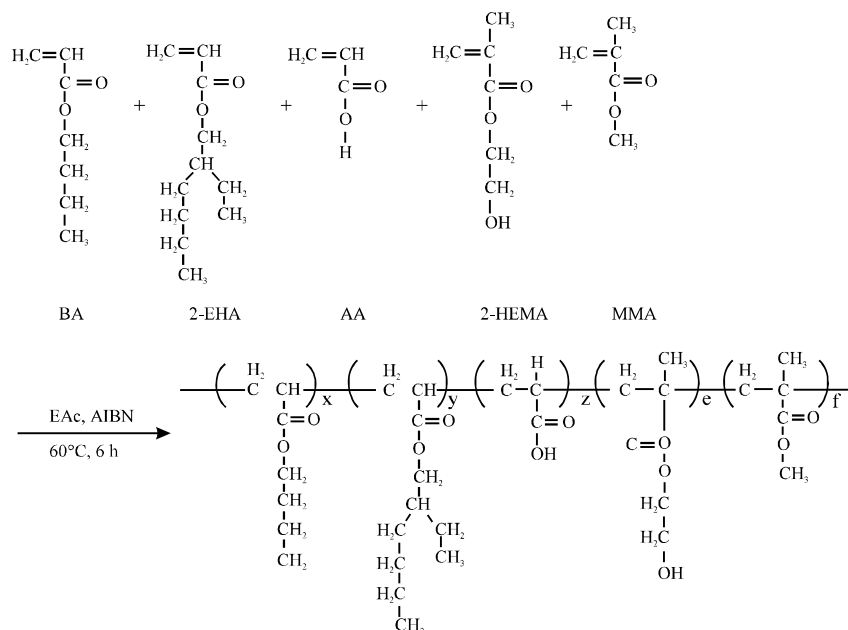
*After polymerization

We used dropping funnel in order to increase reproducibility and control exactly molecular weight and viscosity. We checked both temperature and viscosity change and maintained agitation speed of 50 rpm because of exothermic reaction and decrease of viscosity. Scheme 1 showed the procedure for the formation of OCA.

ITO Deposition of OCA multilayer films: The ITO films were prepared by RF magnetron sputtering of an oxide ceramic target ($\text{In}_2\text{O}_3/\text{SnO}_2 = 90/10$ wt.%) at room temperature. The temperature of substrate rose up to 60°C during sputtering. The distance from target to substrate was 100 mm. A base pressure below 5.0×10^{-6} Torr was maintained prior to the deposition. High purity Ar was

Table 2: Polymerization recipe of OCA resin

Components	OCA resins (%)				
	OCA-1	OCA-2	OCA-3	OCA-4	OCA-5
Initial solvent					
EAc	15.0	15.0	15.0	15.0	15.0
Dropping 1					
2-EHA	28.0	23.5	19.0	28.0	28.0
BA	8.2	8.2	8.2	8.2	8.2
2-HEMA	0.3	0.3	0.3	0.4	0.5
AA	3.2	3.2	3.2	3.2	3.2
MMA	0.0	4.5	9.0	0.0	0.0
Dropping 2					
AIBN	0.1	0.1	0.1	0.1	0.1
EAc	4.3	4.3	4.3	4.3	4.3
Dilution solvent					
EAc	40.0	40.0	40.0	40.0	40.0



Scheme 1: Synthesis of Optically Clear Adhesive (OCA)

Table 3: Deposition conditions of ITO films on laminated film

Target	(90%)In ₂ O ₃ : (10%)SnO ₂
Base pressure	8×10 ⁻⁶ torr
Working pressure	1 mTorr
Power	1400 W
Speed	Mid
Repeat time	1
Ar	30 sccm
O ₂	0.2 sccm

used as the sputtering gas and the process pressure was kept at 2.1×10^{-3} Torr. Table 3 showed Deposition conditions of ITO.

Measurements: Fourier transformed infrared (FT-IR) spectra of the OCA were obtained with a JASCO FT-IR 620 instrument from 700 to 4000 cm⁻¹ on KBr pellets. Differential Scanning Calorimetry (DSC) was performed with a Dupont 2000 differential scanning calorimeter. Samples of approximately 5 ~ 6 mg in weight were sealed in hermetic aluminum pans and scanned in the calorimeter with heating rate of 10°C min⁻¹ in the range of 30~500°C under nitrogen atmosphere and the melting point (T_m) and the glass transition temperature (T_g) values were taken as the change of the specific heat in the heat flow curves and the peak baseline was determined from the horizontal straight method. For dynamic scanning, calibration of the calorimeter was conducted for the heating rate using an indium standard. Mechanical properties of the films were measured on an Instron 5566 tensile tester with a 5 kg load cell. Ultraviolet-visible (UV-VIS) spectra of the polymer films were recorded on a UV-visible spectrometer (Jasco V-650). The SEM and AFM micrographs were obtained with a Hitachi S-4300 and a NanoScope III system, respectively.

RESULTS AND DISCUSSION

The formation of OCA was confirmed by IR spectroscopy as shown in Fig. 1. The IR spectrum of the OCA showed characteristic absorption bands at 2960 and 2870 cm⁻¹ attributed to aliphatic (C-H) stretchings, respectively; the absorption band at 1450 cm⁻¹ to CH₂ deformation, 1750 cm⁻¹ attributed to ester C=O stretching and 3450 cm⁻¹ attributed to O-H stretching.

Examination of the molecular weight data presented in Table 4 shows the decrease in molecular weight as MMA increases. The OCA polymers increased 2-HEMA show higher molecular weight. The polydispersity of OCA polymers were uniform about 1.3, 1.4 regardless of MMA and 2-HEMA content (Leong *et al.*, 2002).

The viscosity of OCA polymers is important factor for affecting wetting and coating properties of substrate. The viscosity of OCA polymers was about 450 ~ 1200 cps

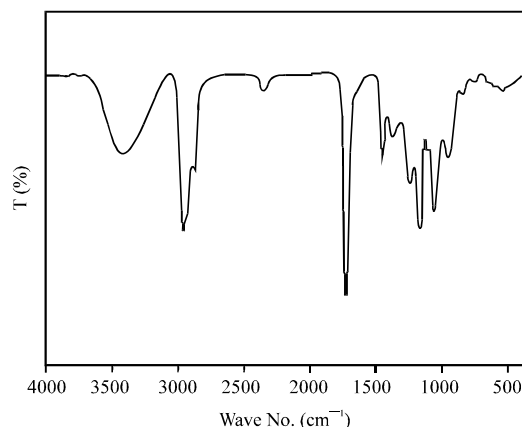


Fig. 1: FT-IR spectrum of OCA-1

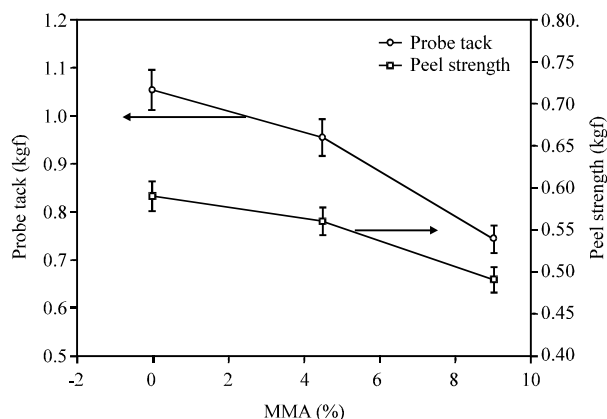


Fig. 2: Adhesion properties: probe tack and peel strength of OCA with variation in MMA content

Table 4: Synthetic data and viscosity of OCA resins

OCA resins	Mn	Mw	PDI	T _g (°C)	Viscosity (cps, 25°C)
OCA-1	77,000	105,000	1.3	-55	450
OCA-2	65,000	93,000	1.4	-46	900
OCA-3	61,000	89,000	1.4	-33	1200
OCA-4	83,000	111,000	1.3	-53	780
OCA-5	91,000	119,000	1.3	-49	940

and increased as MMA and 2-HEMA content increase. This reason might high T_g of PMMA and hydrogen bond of 2-HEMA.

Figure 2 and 3 showed the probe tack and peel strength of OCA polymers. The probe tack and peel strength of OCA polymers decreased for decrease of flexibility and softness as MMA increase. The peel strength of OCA polymers increased for increase of cohesiveness and stiffness as 2-HEMA increase. The molecular weight and entanglement increase induced increase of cohesiveness and stiffness due to 2-HEMA (Fig. 4).

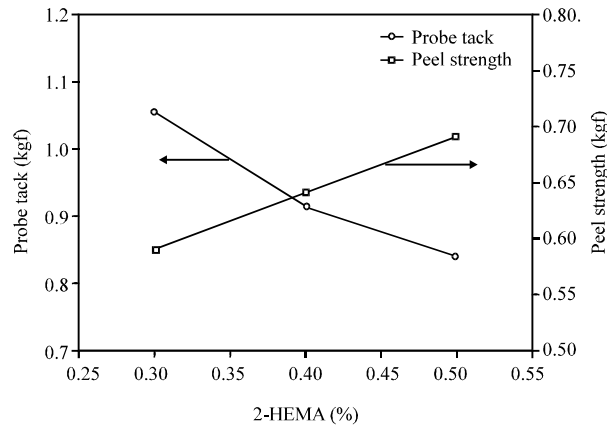


Fig. 3: Adhesion properties: probe tack and peel strength of OCA with 2-HEMA content

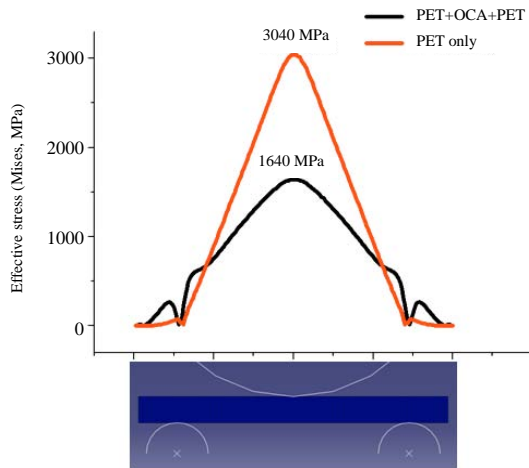


Fig. 4: Analysis results of stress distribution on single and laminated type

Table 5: Each material's elastic modulus and Poisson's ratio

Analysis	PET	ITO	OCA
Elastic modulus (GPa)	2.5	92.00	0.30
Poisson's ratio	0.3	0.33	0.45

We simulated stress distribution by using ABAQUS software of finite element analysis to interpret correlation of cushion effect of OCA layer-stress distribution of ITO layer. Table 5 showed elastic modulus and Poisson's Ratio for each material.

Figure 4 and 5 showed the distribution and value of stress as pushed by the same force. Multilayer film had broader distribution and smaller effective stress than single layer.

The characteristic of ITO affect the durability of touch panel. ITO layer with high degree of crystallinity through heat treatment had excellent durability and

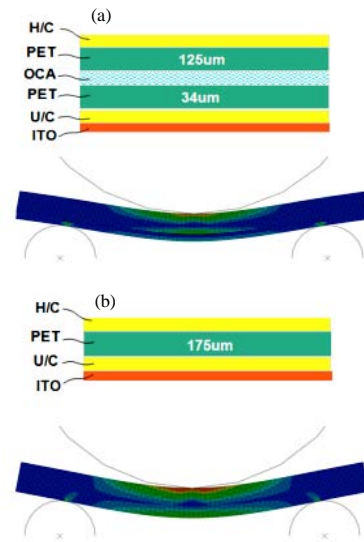


Fig. 5: Stress distribution simulation of (a) single layer and (b) multi-layer

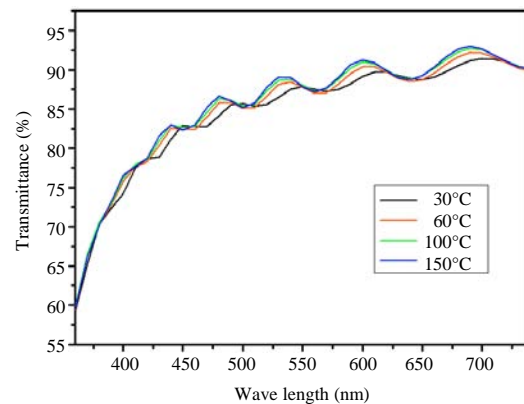


Fig. 6: Changes in transmittance after annealing at various temperatures

transmittance. The ITO-coated films were prepared at room temperature and annealed by thermal methods. The effect of annealing on the sheet resistance, microstructure and transmittance of the ITO coated films were investigated. The transmittance and sheet resistance of the annealed ITO coated films, as a function of annealing temperature, is plotted in Fig. 6 and 7.

The transmittance of the ITO coated films was increased as annealing temperature was increased. The sheet resistance of the annealed ITO films sharply decreased from 480 to 390 $\Omega \text{ cm}^{-2}$ with increasing the annealing temperature. The ITO was crystallized uniformly and the crystalline ITO was enlarged at high annealing temperature. As a result, the sheet resistance of ITO

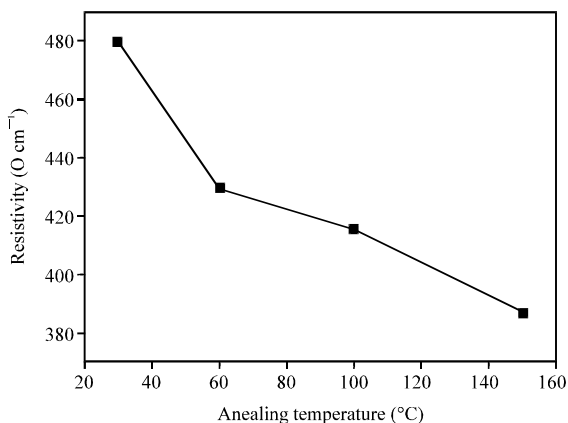


Fig. 7: Changes in sheet resistance after annealing at various temperatures

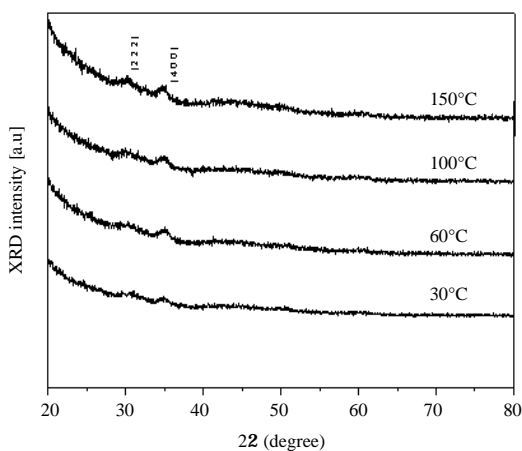


Fig. 8: XRD of ITO films with annealing temperature

films decreased in correlation with the growth of crystalline (Nisha *et al.*, 2005).

Hence, through the X-ray diffraction patterns of ITO films which were annealed were analyzed and are provided in Fig. 8. The diffraction peaks of thermal annealed ITO films were appeared that correspond to the refractions of 222 and 400 (Bae *et al.*, 1993).

These results correspond with the growth of crystallites and the intensity ratio of 400 to 222 planes of the ITO films by thermal annealing.

In observing the changing crystalline in ITO thin films, surface morphology of the ITO films were investigated with AFM images are shown in Fig. 9a-d. According to AFM analysis of the ITO films, the Root-Mean-Square (RMS) surface roughness of the non-annealed ITO thin films was 1.3 nm. The roughness values of the ITO films decreased after annealing and the RMS values of at 150°C annealed ITO thin films was

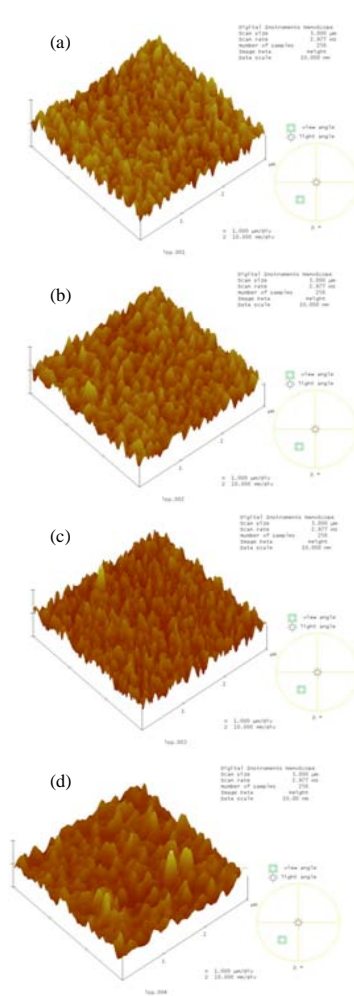


Fig. 9: AFM image of ITO film with annealing temperature, (a) 30, (b) 60, (c) 100 and (d) 150°C

1.1 nm, respectively. This showed that roughness of annealed ITO films is more uniform than that of the non-annealed ones.

CONCLUSIONS

The optically clear adhesive (OCA) polymers were prepared by free radical polymerizations by using 2-ethylhexyl acrylate (2-EHA), methyl methacrylate (MMA), butyl acrylate (BA), acrylic acid(AA), 2-hydroxy ethyl methacrylate (2-HEMA) as comonomers. The OCA polymers showed that their Tg increased as more MMA and 2-HEMA monomers were incorporated in the polymers. The increase of Tg with MMA or 2-HEMA may be explained by the hindered rotation due to the presence of methyl substituent in MMA and 2-HEMA units. The probe tack and peel strength of OCA polymer decreased

for decrease of flexibility and softness as MMA increase. The peel strength of OCA polymers increased for increase of cohesiveness and stiffness as 2-HEMA increase. The transmittance was increased and sheet resistance was decreased as annealing temperature of ITO coated films was increased, The ITO coated PET/OCA/PET films showed no curl or bubble formation after the heat treatment (150°, 1 h). These results suggest that the multilayer films using synthesized OCA polymer can be expected to be applied to the touch panel.

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