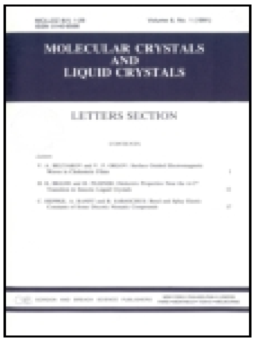

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Environment-Friendly Acrylic Pressure Sensitive Adhesives (PSAs) with Improved Antistatic Property

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A series of acrylic PSAs were synthesized by pre-emulsion addition method and characterized by FT-IR spectroscopies. Electrical and mechanical properties of antistatic waterborne acrylic PSAs containing electrically conductive fillers were investigated in this paper depending on the component of monomers, reactive emulsifiers, and the type of conductive fillers. PSA synthesized by reactive emulsifier showed higher physical properties and its composite with 1wt% of conductive fillers showed good dispersity with optimal peel strength.

Keywords Pressure sensitive adhesives (PSAs); Reactive emulsifier; Conductive fillers; Acrylic polymer; Pre-emulsion addition method; Antistatic property

Introduction

Pressure sensitive adhesives (PSAs) adhere instantly and firmly to nearly any surface under the application of light pressure, without covalent bonding or activation[1]. A growing interest is observed in antistatic pressure sensitive adhesives (PSAs) as electrical/electronic components in many areas for both industry and household[2, 3]. PSAs are increasingly used for demanding applications, such as interconnects in electronic assemblies[4], protective tapes for electronics. Protective tapes for electronics should have electrostatic discharge properties in order to prevent the damage on the electronics by the electrostatic discharge that occurs upon tape removal. Therefore the pressure sensitive adhesive (PSA) used for preparation of the protective film should combine typical PSA characteristics with electrical conductivity. Conductive fillers such as metal powders or carbon-based fillers, for example a special grade of graphite or carbon black, are used to improve antistatic properties. Although filler content should be greater than several precent, unfortunately, certain level of filler content can damage important PSA characteristics such as tackiness[5].

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Antistatic waterborne arylic PSAs are not commercially available on the market. The development of these PSAs requires special suitable self-adhesive polymers and their modification through adding of electrical conductive fillers[5]. In this study, antistatic waterborne acrylic PSAs were synthesized from various components of monomers, reactive emulsifiers by pre-emulsion addition method and mixed with various conductive fillers. This paper reports the synthesis of environment-friendly antistatic waterborne acrylic PSAs by evaluating the effect of conductive fillers on waterborne acrylic PSA characteristics and suggests a novel method that can effectively reduce electrical resistivity with minimal loss of peel strength[6, 7].

Experimental

It has been generally known that alkyl(meth)acrylate with 4∼17 carbons are suitable for the synthesis of PSA, and 2-ethylhexyl acrylate(2-EHA), butyl acrylate(BA), and isooctyl acrylate(IOA) are typically used as main monomers. However, due to their low glass transition temperature(Tg) and cohesiveness, they were often copolymerized with other functional monomers, leading to higher Tg and better cohesiveness. These functional monomers and emulsifiers were obtained from Aldrich Co.(USA), LG Chem Co.(Korea), Wuzhou Co.(China), and Nippon Zeon Co.(Japan) and used without purification. All the other chemicals used in the present experiment were analytically pure.

Conductive fillers(JISTAT 1000, JISTAT 2000 NT) were obtained from Joongil Chem Co.(Korea) and used as received. The polymerization procedure was typically followed pre-emulsion technique which was described in our previous paper[7].

Various kinds of formulations with emulsifier compositions were used to obtain the optimal PSA for the improvement of antistatic property. First, acrylic PSAs with solid content of 56% and theoretical Tg of -54°C based on BA/2-EHA as main monomers, MMA as a cohesive monomer, acrylic acid(AAc) as a functional monomer for bonding strength, were synthesized with various NP(nonyl phenol) free reactive nonionic emulsifiers, such as S-1, S-2, S-3, and reactive anionic emulsifiers, such as S-5, S-6, and reactive nonionic/anionic emulsifiers, such as S-1/S-5, S-2/S-6. Second, new acrylic PSAs were evaluated their peel strengths, such as initial, moisture and heat resistances.

Similar to the preparation of pristine acrylic PSA, the antistatic acrylic PSAs with conductive fillers were fabricated through an in situ polymerization method by mixing for 3h at room temperature.

The structures of synthesized acrylic PSAs were identified using JASCO FT-IR 6200 specrophotometer with a wave range of 4,000∼400 cm^{-1}. Polyethylene terephthalate (PET) film and SUS 304 were used as substrates to evaluate the physical properties of PSAs as follows. First, PSA coated PET film with 100 μm/wet thickness was dried at 100°C for 4 min and cut in 25×150 mm^2 size, followed by the adhesion to SUS 304 substrate by pressing 3 times with 2 kgf pressure using a hand roller. The universal testing machine (GT-A1 70006 or AI 700S) at cross-head speed of 50 mm/min was used to evaluate various types of peel strengths, such as initial, moisture, and heat resistant peel strengths. Initial peel strength was measured after the fabricated sample specimens were kept for 3 min at 25°C. The fabricated specimens first stabilized at 25°C for 72 h were kept at 80°C for 72 h for heat resistant peel strength, and at 40°C under 95% relative humidity for 72 h prior to the evaluation of moisture resistant peel strength[8].

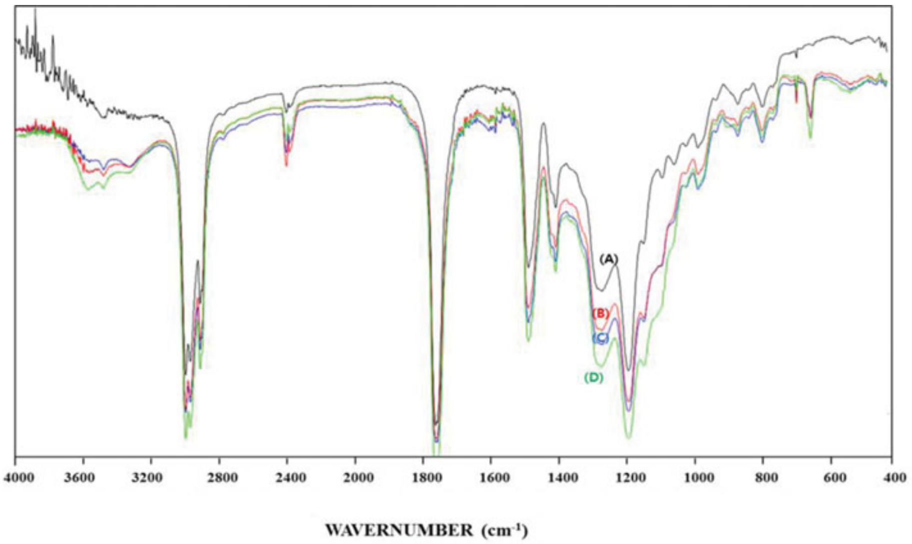

Figure 1. FT-IR spectra of acrylic PSAs with various emulsifiers and conductive fillers. (A) Conventional emulsifier(C-E), (B) Reactive emulsifier(NA-2), and conductive filler (1wt%) (C) JISTAT 1000, (D) JISTAT 2000NT.

The surface resistivity of the acrylic PSA surface on the film was measured with a SIMCO ST-3 surface resistivity tester. The morphology of conductive fillers dispersed in acrylic PSA was observed with a SEM(Supra 35, Carl zeiss, Japan).

Results and Discussion

The FT-IR spectra(Figure 1) of acrylic PSAs and antistatic waterborne acrylic PSAs with reactive/conventional emulsifier, and conductive fillers indicated characteristic absorption peaks at 2900 ~ 2850 cm^{-1} (stretching vibration of aliphatic C—H), 1730 cm^{-1} (stretching vibration of C=O), 1450 cm^{-1} (CH_2 scissoring) with the concurrent disappearance of vinyl group peak at 1680 cm^{-1} (stretching vibration of vinyl C=C).

As shown in Table 1, PSAs based on reactive nonionic/anionic emulsifiers (NA-1, NA-2) had generally higher physical properties compared to those based on reactive nonionic(N-1, N-2, N-3) or anionic(A-1, A-2) emulsifiers. It had been widely accepted that anionic emulsifiers could provide repulsive force between two similarly charged micelle particles[9]. However, it is thought that PSAs based on reactive anionic emulsifiers may have anionic center at one end of the polymer chain, leading to the decrease in physical properties of PSAs. This is why we have used mixed nonionic/anionic emulsifiers to optimize the content of anionic center of PSA. Therefore, it was decided to choose PSA based on NA-2 for further formulation with conductive filler because it had higher physical properties as well as higher tack property with Tg of −54°C.

As shown in Table 2, viscosity, surface resistivity, and peel strength of PSA matrix with the conductive filler decreased with increasing conductive filler content.

In order to evaluate the dispersion of conductive fillers in PSA matrix, PSA(NA-2) with JISTAT 1000(1wt%) and JISTAT 2000NT(1wt%) were evaluated by SEM images, which showed that the conductive fillers are dispersed in the PSA matrix(Figure 2). In addition,

Table 1. Formulation and physical properties for acrylic PSAs based on emulsifiers[a]

		N-1	N-2	N-3	A-1	A-2	NA-1	NA-2	C-E[b]
Formulations (g)	2-EHA	68	68	68	68	68	68	68	68
	BA	21	21	21	21	21	21	21	21
	AAc	2	2	2	2	2	2	2	2
	MMA	9	9	9	9	9	9	9	9
	S-1	4	—	—	—	—	2	—	—
	S-2	—	4	—	—	—	—	2	—
	S-3	—	—	4	—	—	—	—	—
	S-4	—	—	—	—	—	—	—	2
	S-5	—	—	—	4	—	2	—	—
	S-6	—	—	—	—	4	—	2	—
	S-7	—	—	—	—	—	—	—	2
Conversion (%)		94.6	92.9	91.1	99.5	98.7	98.9	99.8	99.8
Initial peel strength (kgf/25 mm)		0.6	0.7	0.7	1.1	1.0	1.2	1.3	1.0
Moisture resistant peel strength (kgf/25 mm)		0.8	0.9	0.8	1.2	1.2	1.3	1.2	0.9
Heat resistant peel strength (kgf/25 mm)		1.0	0.8	1.0	1.3	1.3	1.4	1.4	1.2

Reaction Conditions : initiator (KPS, potassium persulfate), 0.3 g; $NaHCO_3$, 0.2 g; H_2O, 80 g; theoretical solid content, 56%; theoretical Tg, −54°C; reaction temp., 85°C; reaction time, 8 h.

[a] Emulsifiers : S-1, $CH_2{=}CHCH_2OCH_2CH(CH_2OR)[(OX)_nOH]$; S-2, $CH_2{=}CHCH_2OCH_2CH(CH_2OR)[OY)_nOH]$;

S-3, $CH_2{=}CHCH_2OCH_2CH(CH_2OR)[(OZ)_nOH]$; S-4, $ROCH_2(OCH_2CH_2)_nOH$;

S-5, $CH_2{=}CHCH_2OCH_2CH(CH_2OR)[(OX)_nOSO_3NH_4]$; S-6, $CH_2{=}CHCH_2OCH_2CH(CH_2OR)[OY)_nOSO_3NH_4]$;

S-7, $ROCH_2CH(OCH_2CH_2)_nOSO_3NH_4$

[b] C-E : Conventional Emulsifier (S-4, S-7)

Table 2. Surface resistivity and peel strength of acrylic PSAs

Sample		Viscosity (CPS/25°C)	Surface resistivity (Ω/square)	180° peel strength (kgf/25 mm)
NA-2		49000	10.5	2.257
JISTAT 1000(wt%)	0.5	16500	9.9	2.224
	1.0	13000	9.4	2.204
	2.0	7500	6.6	1.948
	3.0	3500	5.7	1.453
	4.0	1900	5.2	—
JISTAT 2000NT(wt%)	0.5	16000	9.7	1.862
	1.0	13000	9.2	1.858
	2.0	6000	6.3	1.770
	3.0	3500	5.2	1.208
	4.0	1500	4.9	—

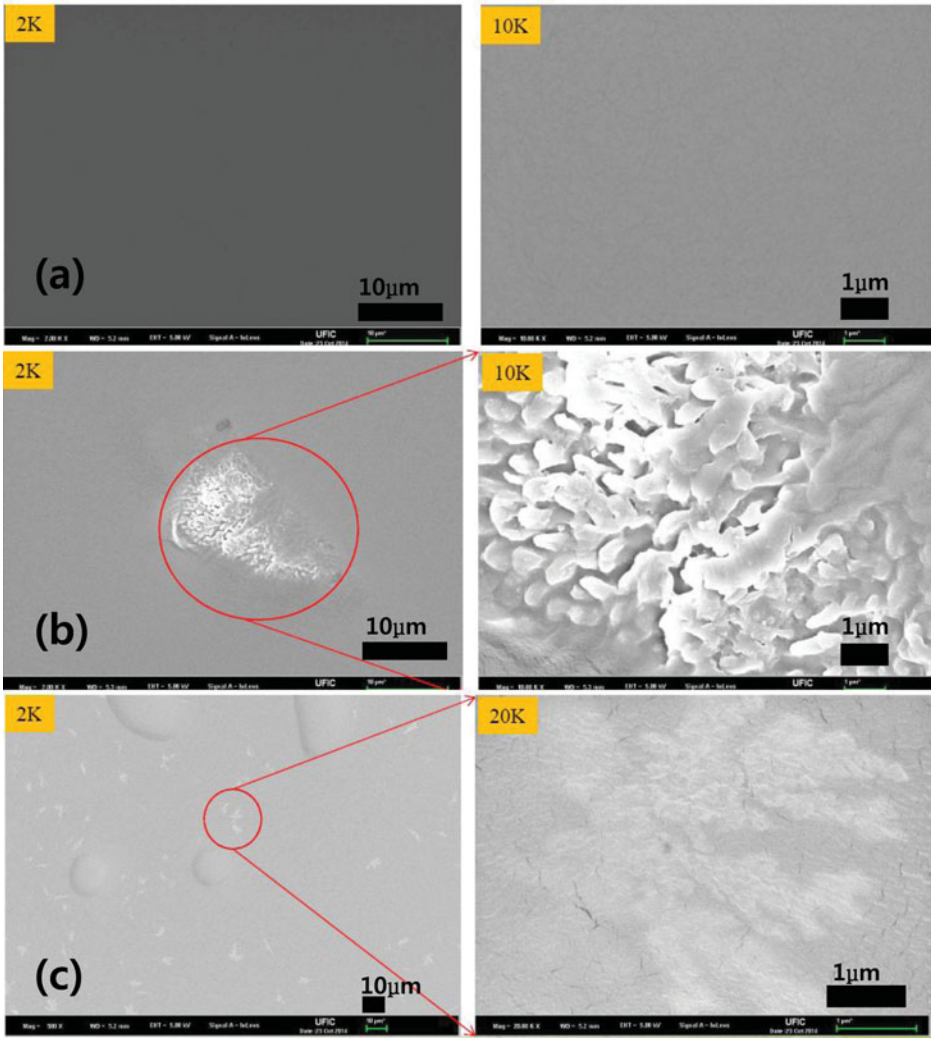

Figure 2. SEM images of acrylic PSAs with conductive filler. (a) NA-2, (b) JISTAT 1000(1wt%), (c) JISTAT 2000NT(1wt%).

the dispersion of conductive filler, JISTAT 2000NT was more homogeneous than JISTAT 1000, meaning that the dispersion of conductive filler in the PSA matrix improved antistatic property.

Conclusions

Various kinds of PSAs with different reactive emulsifiers were applied for the potential use as adhesives to improve antistatic property in electronic assemblies. Among them, PSA based on both reactive nonionic/anionic emulsifier(NA-2) exhibited the optimal physical properties such as moisture and heat resistances with experimental Tg relatively close to its theoretical value and its composite with conductive filler(JISTAT 2000NT) showed improved antistatic property due to its better dispersion of filler in PSA matrix. Continuous work involves the practical application to electronic devices and the evaluation of environmental friendly properties.

Acknowledgments

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