Synthesis of Silicone–Acrylic Resins and Their Applications to Superweatherable Coatings

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ABSTRACT: Silicone–acrylic resins were synthesized to prepare superweatherable paints for building materials. The raw materials used were *n*-butyl acrylate, methyl methacrylate, and *n*-butyl methacrylate as acrylic monomers and 3-methacryloxypropyltrimethoxysilane (MPTS) as a silicone monomer reactive with the acrylic monomers. Acrylic copolymers were synthesized such that their glass-transition temperatures were adjusted to 30°C and their MPTS contents were varied to 10, 20, and 30 wt %. As the content of silicone and MPTS increased, average molecular weight and viscosity increased, and thermal stability at high temperatures improved. When we tested the properties of coatings by blending the synthesized silicone–acrylic resins with a white pigment, adhesion was superior with various substrates, and their properties were suitable on the whole. Weatherability was tested by an outdoor exposure test with a weather-ometer and an accelerated weathering tester, and their results showed that silicone–acrylic resin composed of 30 wt % MPTS was a superweatherable coating. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 81: 1614–1623, 2001

Key words: superweatherable coatings; silicone–acrylic resin; weatherability test

INTRODUCTION

Weatherability is generally defined as the property of a material of being able to resist a wide range of environments. When coatings are exposed to the outdoors, synthesized resins, the main components of coatings, are oxidized by such things as sunlight, heat, water, oxygen, and air-polluting materials. Therefore, weatherability, especially for coatings, is defined as the property of being resistant to gloss loss and chalk, which are highly affected by UV.[1,2]

As the prosperity of national life impels demands for a more environmentally friendly and pleasant life, weatherable coatings have become widely used in construction and building materials that are exterior decorations. These types of coating are highly crosslinked, making them resistant to various environments.

Until the late 1980s fluororesins represented by fluoroethylene–alkyl vinyl ether copolymers were the main stream used in weatherable coatings.[3] Introducing a higher alkyl group into the molecules improved their flexibility and solubility. In addition, bringing in a hydroxyl group as a

functional group allowed the design of two varieties of these copolymers: the heart-dried and two-component types, both of which were crosslinkable with polyisocyanate at room temperature. However, these fluororesins were expensive and easily polluted the atmosphere. Moreover, their hardness was very low, and working conditions were not favorable. Therefore, a new type of weatherable coatings was needed. At present, special attention is being paid to inorganic ceramic coatings and silicone–acrylic coatings for use as new interior and exterior paints in the architectural field. Inorganic ceramic coatings possess nonflammability and antipollution characteristics because inorganic pigments are composed of pure ceramic components. But they tend to crack easily and need to be heated at 180–200°C for 30 min.

On the other hand, silicone–acrylic coatings can easily be applied to various materials because their film hardness can be easily controlled. In addition, they can be made into one-component curable coatings whose curing is initiated by moisture at room temperature. These coatings are cheaper than fluoro-based coatings. Though two-component waterborne silicone–acrylic coatings have been studied by several authors,[4] there have been few reports on the synthesis and weatherability comparison of silicone–acrylic resins that can be cured by moisture at room temperature.

To invesetigate silicone–acrylic coatings Rao and Babu[5] studied the solubility, molecular weight, and thermal behavior of vinyltriacetoxysilane-bromo methacrylate copolymers, claiming that lactone formation was delayed with an increasing content of vinyltriacetoxysilane. Yasuyuki et al.[6] synthesized and then grafted silicone- and acrylic emulsions. Their TEM results on phase separation of the grafted elastomers indicated that the degree of phase separation was affected by the synthesis method and by the composition of the silicone–acrylic resins.

In this study we synthesized four-component silicone–acrylic resins by free-radical copolymerization of three acrylic monomers and a silicone monomer. The monomers and the reason they were chosen are: methyl methacrylate, responsible for the hardness and weatherability of coatings; *n*-butyl acrylate, for flexibility of coatings; *n*-butyl methacrylate, for moisture resistance; and 3-methacryloxypropyltrimethoxysilane, for reactivity with acrylic and metacrylic monomers and weatherability. The reaction products were obtained either as mill-base resins or as letdown resins. White coatings were formulated according to the formulation chart for general-purpose architectural coatings, and the mill-based silicone–acrylic resins were blended with the letdown resins in a 3:7 ratio. We studied the effect of silicone content on the properties and weatherablity of coatings in order to develop superweatherable coatings. Weatherablity was tested using the outdoor exposure and accelerated weatherablity tests.

EXPERIMENTAL

Chemicals

All the chemicals were reagent grade and were used as received. The monomers were *n*-butyl acrylate (BA; Tokyo Kasei Kogyo Co.), methyl methacrylate (MMA; Tokyo Kasei Kogyo Co.), and *n*-butyl acrylate (BMA; Junsei Chemical Co.), with 3-methacryloxypropyltrimethoxysilane (MPTS; Sigma Chemical Co.) as a reactive silicone monomer. 2,2-Azobisisobutyronitrile (AIBN; Wako Pure Chemical Co.) was used as an initiator, and methyl trimethoxysilane (MTS; Sigma Chemical Co.) and trimethyl orthoformate (TMO; Junsei Chemical Co.) were used as drying agents. Tinuvin-292 (HALS; Ciba-Geigy Co.) and Tinuvin-384 (a benzotriazole derivative; Ciba-Geigy Co.) were used as the UV stabilizer and the UV absorber, respectively. Di-*n*-butyltin dilaurate (DBTDL; Songwon Industry Corp.) was used as a curing catalyst, TiO_2 as a white pigment, and CAB-551-0.01 (Eastman Kodak Co.) as a leveling agent.

Synthesis of Silicone–Acrylic Resins for Dispersion of Pigment

Introduced into a 1-L four-necked flask were 140 mL of xylene and 120 g of toluene, and the compositions of KMB-30 listed in Table I were added to the solvent mixture under a N_2 stream. Into this mixture was dropped another solution mixture, of 2.52 g of AIBN and 5.4 g of MTS, at 82°C for 120 min and aged for another 120 min. Then a solution of 0.36 g of AIBN that had been dissolved in 3.6 g of xylene was added four times in this sequence: right after aging, after 30 min, after 60 min, and during heating to 90°C for 30 min. Finally, the reaction mixture was aged at 105°C for 30 min. The endpoint of the reaction was determined by measuring its solid content, and the reaction was terminated at a solid content of 97%.

Table I Polymerization Conditions for and Physical Properties of Silicone–Acrylic Resins

Exp. No.	Materials				T_g (°C)		Viscosity (Stoke)	Si content (%)		Nonvolatile (%)	Conversion (%)
	BA[a] (g/mol)	MMA[b] (g/mol)	BMA[c] (g/mol)	MPTS[d] (g/mol)	Calculation	by DSC		Calculation	by AA		
KMB-30	47.5 (0.37)	121.7 (1.22)	172.8 (1.22)	18.0 (0.07)	30	33	10.9	0.52	0.50	49.4	88.8
KLD-31	44.0 (0.34)	112.5 (1.13)	159.8 (1.13)	43.7 (0.18)	30	29	5.9	1.25	1.23	49.8	89.0
KLD-32	36.8 (0.29)	94.2 (0.94)	133.8 (0.94)	95.2 (0.38)	30	26	4.2	2.72	2.71	49.4	88.8
KLD-33	29.7 (0.23)	75.9 (0.76)	107.8 (0.76)	146.6 (0.59)	30	31	2.3	4.20	4.16	49.2	88.4

[a] BA: n-butyl acrylate
[b] MMA: methyl methacrylate
[c] BMA: n-butyl methacrylate
[d] MPTS: 3-methacryloxypropyltrimethoxysilane

At the termination step 50% of the solid content of the reaction product was obtained by charging 12.6 g of MTS and 100 g of xylene. Unreacted monomers were removed by precipitating the solid product in excess n-hexane. The precipitated product was vacuum-dried at 50°C and 5 mmHg, producing a transparent viscous copolymer—KMB-30.

Synthesis of Letdown Silicone–Acrylic Resins

Into a 1-L four-necked flask were introduced 260 g of xylene and a monomer solution—KLD-31, whose compositions are indicated in Table I—along with 2.52 g of AIBN and 2.32 g of TMO. The reaction conditions and procedures were the same as illustrated in the previous section. At the completion of the reaction the addition of 5.4 g of TMO and 100 g of xylene resulted in the reaction product having a 50% solid content. Purification was done in the same way as described in the previous section, producing a transparent viscous copolymer with 10 wt % MPTS, KLD-31. The same synthesis and purification procedures resulted in KLD-32 and KLD-33, copolymers with 20 wt % and 30 wt % MPTS, respectively.

Instrumental Analysis

FTIR (Bio-Rad, FTS-40) and ^{1}H-NMR (Varian, Unity-300) were used for the structural analysis of silicone–acrylic resins. Their molecular weights and distributions were determined by gel permeation chromatography (GPC) with a Waters R-410. Thermal gravimetric analysis (TGA) was done by a Shimadzu TGA-50H under air and by differential scanning calorimetry (DSC) with a Thermold DSC 4000) under N_2. Silicon content was determined according to ASTM D-3733 with an PerkinElmer 5200 atomic absorption spectroscope.

Measurement of Kinematic Viscosity and Solid Content

Kinematic viscosity was measured by a transparent solution test according to KS M 5000-2121. The solid content was obtained by measuring the residual weight of a 1 g-sample that had been stored in an oven at 105±2°C for 3 h.

Formulation of Coatings

The coatings were formulated with TiO_2 by blending the mill-base resin and the letdown resin at a ratio of 3:7. The details of the formulation compo-

Table II   Preparation of White Enamel for Architectural Coatings

Types	Materials	Weight (wt %)
Mill base	mill-base silicone–acrylic resin	21.6
	TiO$_2$ (rutile)	24.0
Letdown	letdown silicone– acrylic resin	50.4
	leveling agent	0.1
	UV absorber	0.2
	UV stabilizer	0.1
	Xylene	3.6
Mill base/Letdown: 3/7		

sitions are listed in Table II. Coatings with MPTS contents of 10, 20, and 30 wt % were designated as SA-3010, SA-3020 and SA-3030, respectively.

Measurement of Physical Properties

For tests of physical properties thin films were applied to substrates such as tin plate (KS D 3516), steel plate (KS D 3512), glass (KS M 5000-1121) and aluminum sheet (KS D 6701). Measurements were done in accordance with the conditions listed in Table III.

Weatherability Test

The outdoor exposure test was done according to KS M 5000-3241. For the accelerated weatherability test a Sunshine weather-Ometer (WOM; Atlas Electric Device Co., Ci65A) and QUV accelerated weathering tester (QUV, Q-Panel Co.) were employed following the KS M 5000-3231 method. Gloss retention, yellowness index difference, lightness index difference and color difference were determined at exposure times of 500, 1000, 2000, 3000 and 4000 h.

RESULTS AND DISCUSSION

Acrylic monomers used for the synthesis of silicone–acrylic resins are classified into acrylates and methacrylates. In the case of acrylates an increased carbon number results in improved flexibility but a decline in hardness and antipollution characteristics. On the other hand, because polymers of methacrylates have a higher glass-transition temperatures than their acrylate counterparts, methacrylates lead to better hardness,

thermal stability, chemical resistance, and weatherability but have poor plasticity. As a result, blending is required in order to tailor the physical properties of resins.[7]
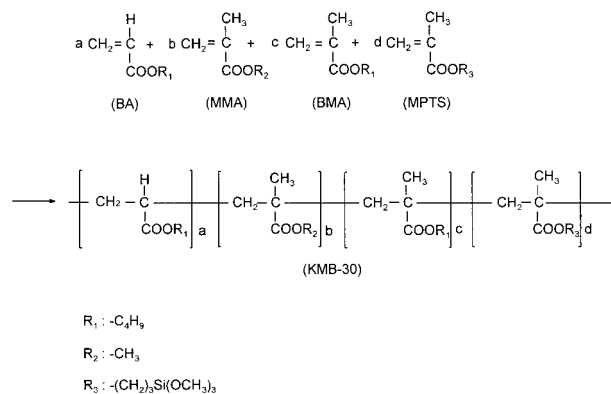
The glass-transition temperatures (T_g) of acrylic resins were calculated using the Fox equation.[8] In general, higher T_g resins have higher viscosity and faster dryness but poor mechanical properties such as flexural, impact strength, and adhesion properties. Therefore, T_g was controlled at 30°C, which is suitable for architectural coatings.

Analysis of Mill-Base Resins

The molecular structure of a mill-base silicone–acrylic resin (KMB-30) is shown in Scheme 1, and

Table III   Test Methods and Conditions of Physical Properties

Types of Tests	Instrument and Specifications
Viscosity (KU)	Krebs-Stormer viscometer Pacific Scientific Co., serial 80328 KS M 5000-2122
Specific gravity	KS M 5000-2131
Fineness of grind	Braive Instruments Co., type 2020 KS M 5000-2141
Drying time	Set-to-touch, dry-hard, dry-through method KS M 5000-2512
Hardness	Yasuda Seiki Seisakusho, serial 4664 JIS K 5400 (8.4.1)
Flexibility	Mandrel: Pacific Scientific Co., conical KS M 5000-3331
Impact resistance	DuPont impact tester type 552 Ureshima Seisakusho JIS K 5400 (8.3.2)
60° Specular gloss	Glossmeter Pacific Scientific Co., Glossgard II KS M 5000-3312
Crosshatch adhesion	ISO 2409
Abrasion resistance	Abrasion tester Toyo Seiki Seisakusho, Taber FS 141C-6192.1
Contrast ratio	KS M 5000-3111
Salt exposure test	ASTM B-117
Storage stability	KS M 5000-2031

Scheme 1

R$_1$: -C$_4$H$_9$

R$_2$: -CH$_3$

R$_3$: -(CH$_2$)$_3$Si(OCH$_3$)$_3$

its physical properties are tabulated in Table I along with its polymerization conditions. Optimization of the feed compositions was already described in a previous work.[9]

The FTIR spectrum of KMB-30, shown in Figure 1(a), confirms the structure of KMB-30 by showing Si—O—CH$_3$ at 845 cm^{-1} and stretching vibrations of C=O and C—O at 1740 cm^{-1} and 1150 cm^{-1}, respectively. From the latter two peaks it can be inferred that there is an ester group present in KMB-30. Figure 2(a) presents the ^{1}H-NMR spectrum and also confirms the structure of KMB-30 by the following chemical shifts: CH$_3$—C at 1.0 ppm, C—CH$_2$—C at 1.4 ppm, C—H at 1.6 ppm, C—CH$_2$—CO— at 2.4 ppm, CH—CO— at 2.7 ppm, and CH$_3$—O—/Si—O—CO$_3$— at 3.6 ppm. The molecular weight and distribution of KMB-30 were determined from the GPC elution curve (Fig. 3). The calculated average molecular weights were M_n = 31,200, M_w
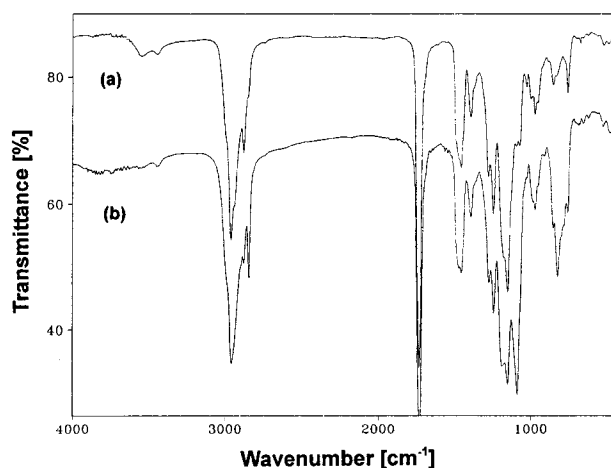

Figure 1   FTIR spectra of (a) KMB-30 and (b) KLD-33.
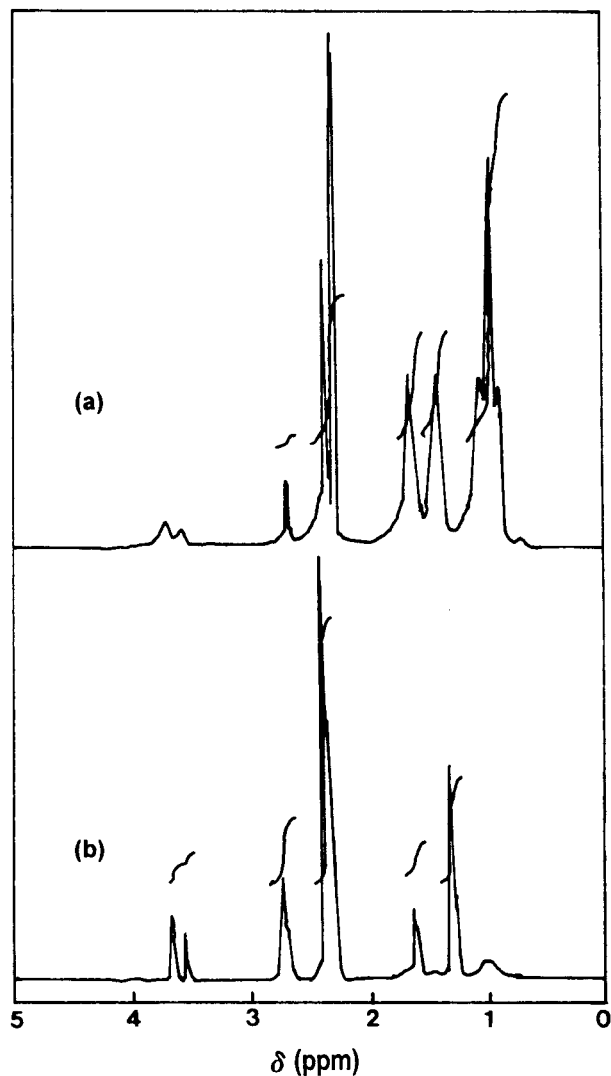

Figure 2 ^{1}H-NMR spectra of (a) KMB-30 and (b) KLD-33.

= 59,400, and M_z = 94,500, and the polydispersity was 1.90, indicative of a fairly narrow distribution.

Analysis of Letdown Resins

The synthesis conditions and physical properties of letdown silicone–acrylic resins (KLD) are listed in Table I. The conversion of KLD resins did not show much difference, but viscosity decreased with an increasing content of MPTS. This may be related to the improved solubility of MPTS in hydrocarbon.

Figure 1(b) shows the FTIR spectrum of KLD-33, which is very similar to the Figure 1(a). A higher content of MPTS in KLD-33 led to increased intensities of Si—O—CH$_3$, and the Si–O
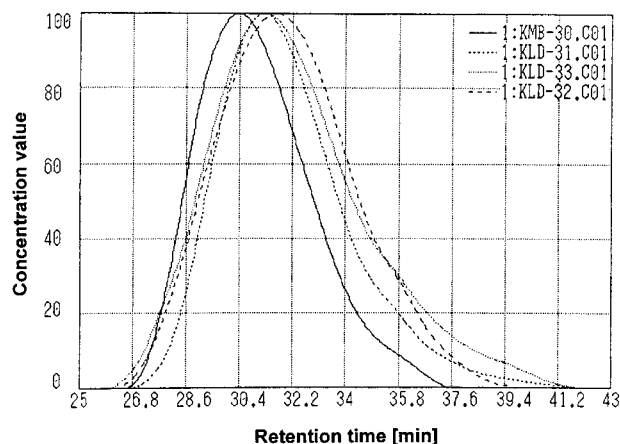
Figure 3 GPC curves for KMB-30, KLD-31, KLD-32, and KLD-33.

peaks at 820 cm^{-1} and 1090 cm^{-1} were increased. The structure of KLD-33 was also shown by a 1H-NMR spectrum [Fig. 2(b)]: CH$_3$—C at 1.0 ppm, C—CH$_2$—C at 1.3 ppm, C—H at 1.6 ppm, C—CH$_2$—CO— at 2.3 ppm, CH—CO— at 2.7 ppm, and CH$_3$—O—/Si—O—CO$_3$— at 3.6 ppm. The GPC results for the three KLD resins are as follows: for KLD-31—M_n = 17,800, M_w = 41,400, M_z = 70,800, and polydispersity = 2.32; for KLD-32—M_n = 18,300, M_w = 43,900, M_z = 87,900, and polydispersity = 2.40; and for KLD-33—M_n = 14,500, M_w = 45,700, M_z = 97,800, and polydispersity = 3.15. When compared with the results of KMB-30, it can be seen that the broader molecular weight distributions of the KLD resins resulted from the increased MPTS composition of higher molecular weights.

Thermal Behavior of Silicone–Acrylic Resins

Paul's[10] TGA study on ethyl acrylate (EA)–MMA copolymers indicated that activation energy and thermal stability increased with an increasing content of EA. Finzel[11] reported, through the measurement of mass change by WOM accelerated test (4300 h), that alkyd resin–based coatings lost 54% of their original mass but silicone–acrylic reins modified with 30% silicone lost only 20.3%.

Figure 4 shows the TGA thermograms of blended KMB-30 and KLD whose weight ratios were 3:7. It also shows the effect of MPTS content on the thermograms of the silicone–acrylic resins. As the MPTS content increased, thermal stability increased in this order: ML-3030 > ML-3020 > ML-3010. This result is consistent with Finzel's result, which said that the increased silicone content improved thermal stability.

Physical Properties of Silicone–Acrylic Resins

The physical properties of silicone–acrylic paints are tabulated in Table IV. Because the glass-transition temperature of resin was adjusted using the Flory–Fox equation, the viscosity of all the coatings was low enough to be processed. The fineness-of-grind was satisfactory in spite of quick (60 min) dispersion, and the contrast ratio was in the acceptable range of 0.942–0.951, depending on the content of the white pigment. We believe that the these characteristics were the result of the excellent performance of TiO$_2$, which we used as white pigment in our formulation. It is generally known that the coloring and hiding powers of TiO$_2$ as a white pigment are superior to ZnS, lithophone, Sb$_2$O$_3$, and ZnO. The hardness of coatings was H, suitable for the architectural coatings and was lowered as the MPTS content increased. This phenomenon can be understood in terms of the reduction of viscosity because of the increase of MPTS content, as shown in Table I. The gloss was a little low compared to the standard value, and the drying time was less than 70 min without addition of any curing catalysts, which indicates the fast dryness of silicone–acrylic resins. Abrasion resistance was in the range of 0.35–1.07 mg and was not affected by the content of MPTS. Flexibility was good because of the *n*-butyl acrylate used, and heat resistance increased with the MPTS content because of the inherent heat resistance of a silicone compound. Impact resistance was good for all the direct sides of the samples
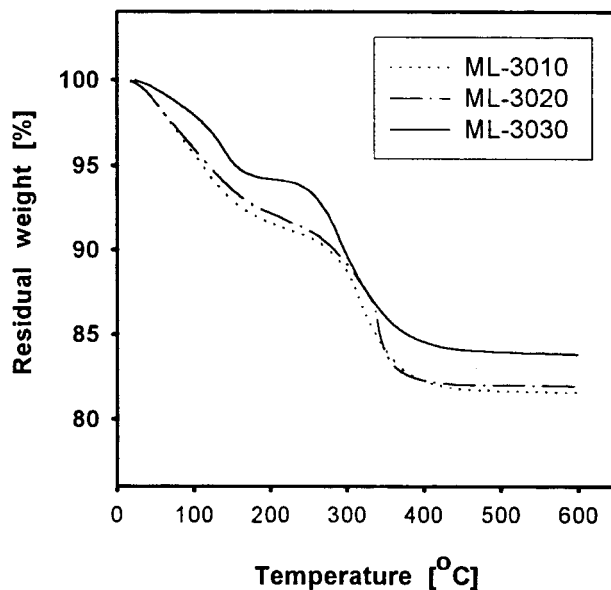

Figure 4 TGA thermograms of silicone–acrylic resins—ML-3010: KMB-30/KLD-31 = 3/7; ML-3020: KMB-30/KLD-32 = 3/7; ML-3010: KMB-30/KLD-33 = 3/7;.

Table IV   Film Properties of Silicone–Acrylic Resin Coatings

Type of Test	Name of Sample	SA-3010	SA-3020	SA-3030
Viscosity (KU)		90	106	70
Fineness of grind		7^+	7^+	7^+
Contrast ratio		0.951	0.944	0.942
Pencil hardness (7 day)		H	H	F~H
60° Specular gloss		85.7	82.0	85.2
Drying time (min)	Set-to-touch	3	2	3
	Dry-hard	57	33	55
	Dry-through	67	43	70
Storage stability (60°C × 10 days)		good	good	good
Abrasion resistance (mg loss/1000 cycle)		0.35	1.07	0.60
Flexibility (1/8")		good	good	good
Heat resistance (150°C × 1 hr)	Gloss retention (%)	92	94	96
	Color difference (ΔE)	0.39	0.26	0.64
Impact resistance (500 g/30, 50 cm)	Direct 30 cm	good	good	good
	50 cm	good	good	good
	Reverse 30 cm	good	poor	poor
	50 cm	poor	poor	poor
Crosshatch Adhesion (%)	Steel plate	100	100	100
	Tin plate	100	100	100
	Aluminum	100	100	100
	PET	100	100	100
	Brass	100	100	100
	Tile	100	100	100

but was poor for the reverse sides of some samples. Crosshatch adhesion was tested on six substrates, and the adhesion was perfect for all substrates, which resulted mainly from the excellent adhesion performance of acrylic resins, which indicates the versatile applicability of our resins.

Salt Exposure Test

The samples were prepared into X shaped scribe area according to the ASTM D 1654-2 (scraping). Rusting and blistering were estimated on both scribed and inscribed areas according to ASTM D 610 and ASTM D 714, respectively. Degree of rusting was divided into 10 grades and was designated as 10 and 0^F for the rust formation of less than 0.01% and 100%, respectively. Degree of blistering was divided into 4, depending on the diameter of blister. The change in the surface condition during the salt exposure test was observed at 100, 200, 300, and 400 h (Table V). As the exposure time and MPTS content increased, negligible changes occurred in the grade number. All the resins were highly resistant to salt.

Outdoor Exposure Test

The outdoor exposure test was done by measuring gloss retention, yellowness index difference, color

Table V   Results of Salt Exposure Test

Test	Scribe Area								Anscribe Area							
Time (h)	Rusting				Blistering				Rusting				Blistering			
Name of sample	100	200	300	400	100	200	300	400	100	200	300	400	100	200	300	400
SA-3010	10	10	10	9F	10	10	9F	9D	10	10	10	10	10	10	10	10
SA-3020	10	10	10	9F	10	10	9F	9D	10	10	10	10	10	10	10	10
SA-3030	10	10	10	9F	10	10	9F	9MD	10	10	10	10	10	10	10	10

F: few; M: medium; MD: middle dense; D: dense; No 9: 0.1–0.4 mm; No 8: 0.5 mm.
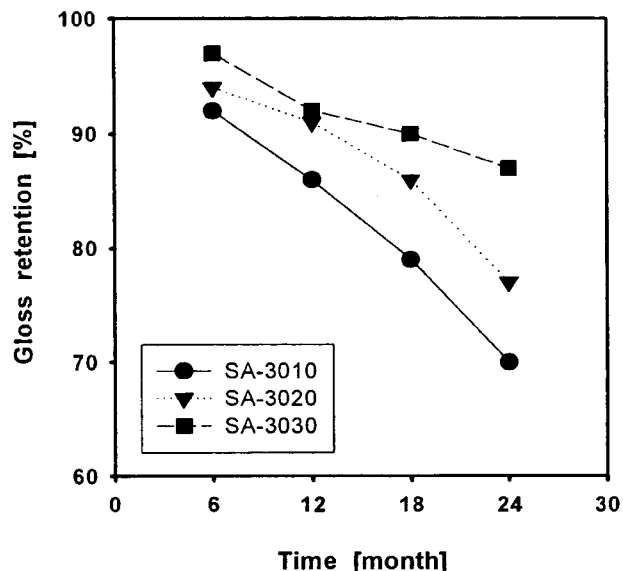
Figure 5 Effects of outdoor exposure time on gloss retention of silicone–acrylic resin coatings.
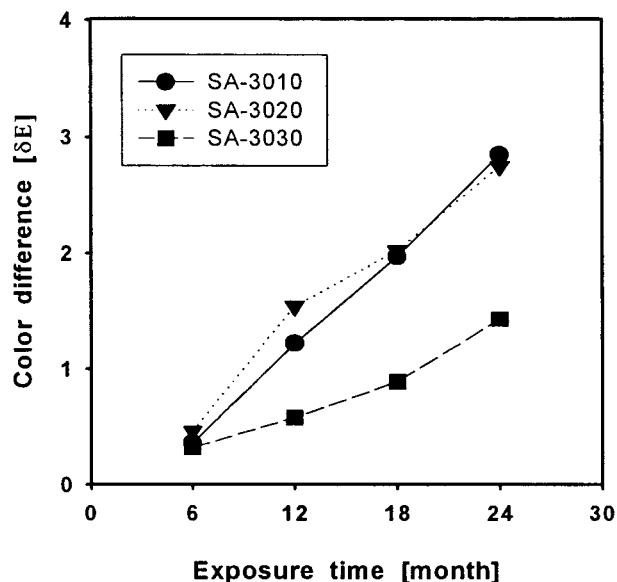

Figure 7 Effects of outdoor exposure time on color difference of silicone–acrylic resin coatings.

difference, and lightness index difference after-outdoor exposures of 6, 12, 16 and 24 months.

Figure 5 shows gloss retention as a function of time. Gloss retention increased with MPTS content. For SA-3030 gloss retention remained at 87%, even after 24 months (30 wt % of MPTS). Figure 6 compares the yellowness index differences and the index differences, which were less than 0.5 after 24 months of exposure. The indexes were quite dependent on the MPTS content, and
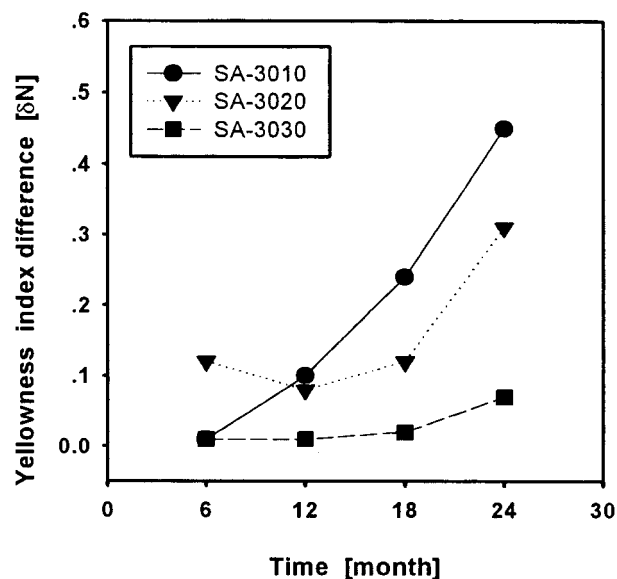
Figure 6 Effects of outdoor exposure time on yellowness index difference of silicone–acrylic resin coatings.

SA-3030 (MPTS 30 wt %) did not show any yellowness phenomenon. When it was exposed for 24 months, the index difference was only 0.1, indicative of its excellent weatherability. The indexes abruptly increased after 12 months. Figure 7 displays color difference as a function of time. Color differences were less than 3.0 for all the samples after 24 months' exposure. The MPTS content also had an effect on the results, with SA-3030 garnering a value of 1.4, a difference not even discernable with the naked eye. But SA-3010 and SA-3020 reached a value of 3.0, which is slightly discernable with the naked eye. Figure 8 illustrates the lightness index difference. On 24 months' exposure all the values were below 1.4, and the lightness had a tendency to darken. The lightness index difference for SA-3030 were less than 0.6 and became a little dark, indicating no chalk phenomenon.

Accelerated Weatherability Test

The accelerated test was done by WOM and QUV for 1000, 2000, 3000, and 4000 h. Figure 9 represents gloss retention at various times. It was found that retention increased with time. When SA-3030 with an MPTS content of 30 wt % was exposed for 4000 h, gloss retention was retained at 90% and 82% according to the results from the WOM and QUV tests, respectively. When these values are compared with the standard for weatherability, SA-3030 has shown to be a superweath-
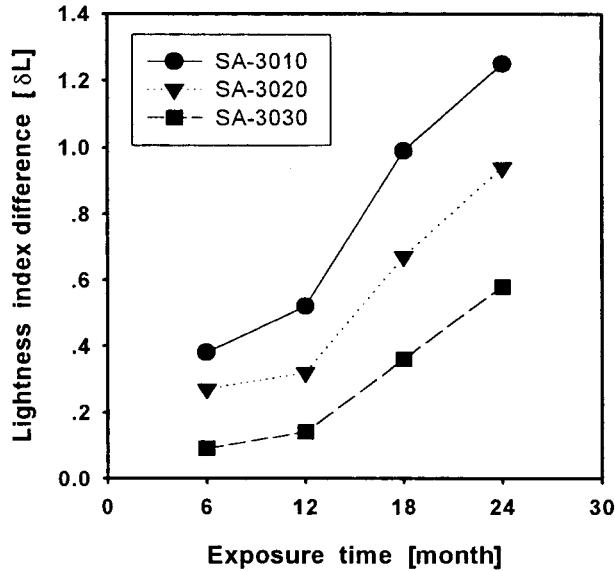
Figure 8 Effects of outdoor exposure time on lightness index difference of silicone–acrylic resin coatings.
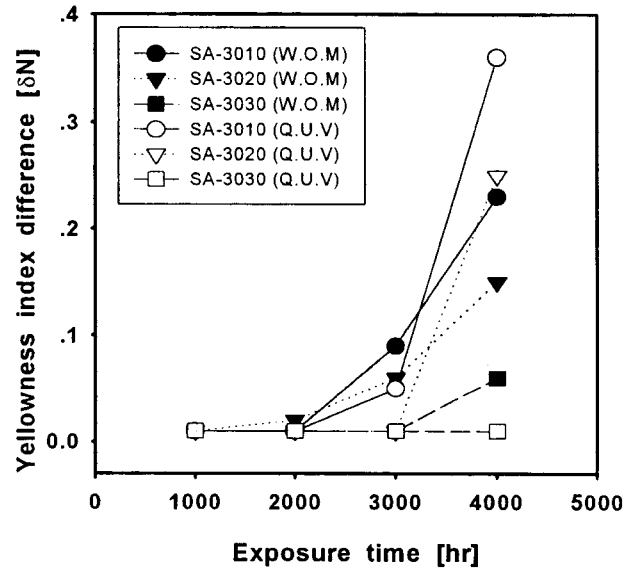

Figure 10 Effects of exposure time on yellowness index difference of silicone–acrylic resin coatings in accelerated weather test.

erable coating. Figure 10 shows the effect of exposure time on the yellowness index difference. The index difference after 4000 h of exposure was below 0.4 for all coatings tested. The accelerated test results were much lower than those obtained in the outdoor exposure test. In the case of SA-3030 the index difference was less than 0.1, indicating a negligible yellowness phenomenon. The yellowness had a tendency to abruptly increase after 3000 h. Figure 11 shows the color difference

results obtained by the accelerated tests, which were below 4.0 for all paints after 4000 h of exposure. These results were inferior to the results obtained by the outdoor exposure test (24 months). The effect of MPTS content on color difference was as follows: 3.0 for SA-3010, which was detectable with the naked eye; 2.7 for SA-3020, a little detectable; below 2.0 for SA-3030, hardly detectable. These results indicate that SA-
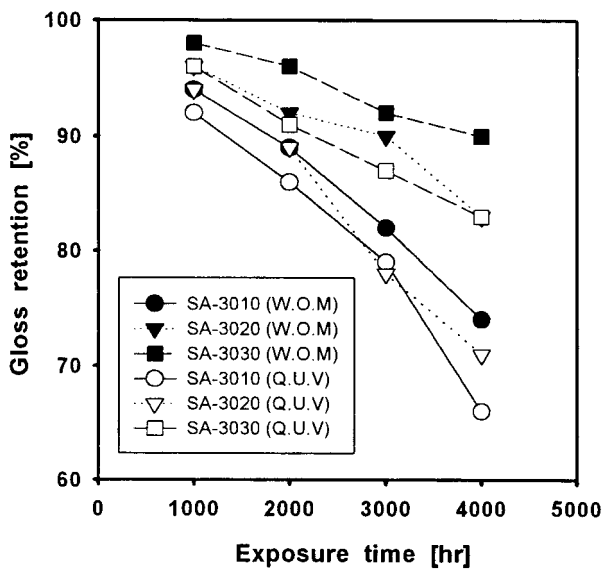

Figure 9 Effects of exposure time on gloss retention of silicone–acrylic resin coatings in accelerated weather test.
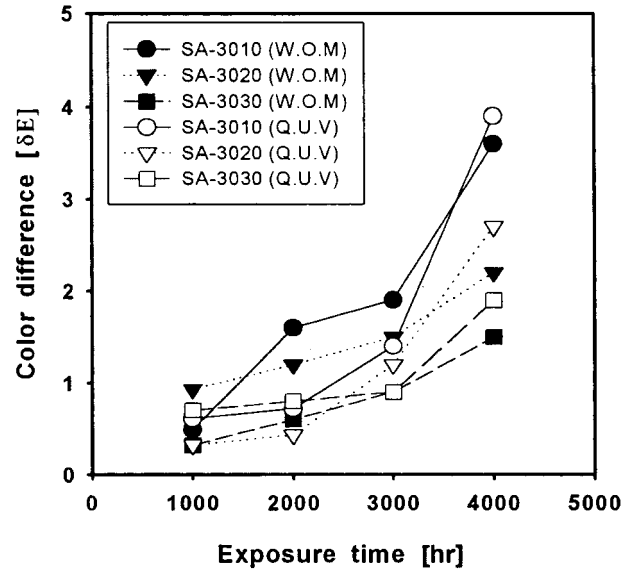

Figure 11 Effects of exposure time on color difference of silicone–acrylic resin coatings in accelerated weather test.
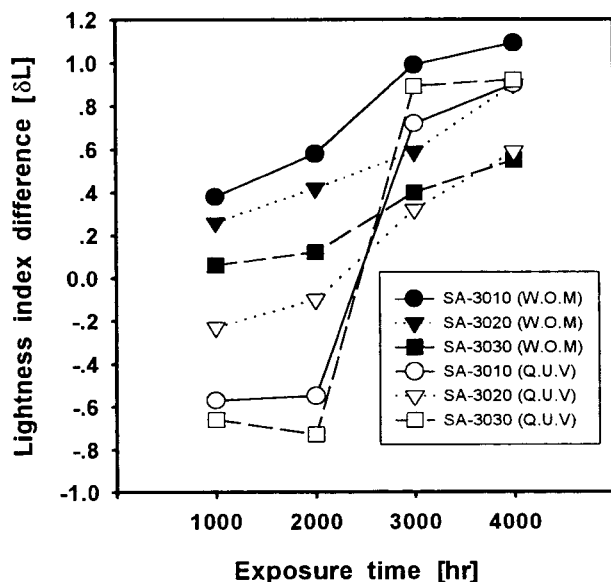
Figure 12 Effects of exposure time on lightness index difference of silicone–acrylic resin coatings in accelerated weather test.

3030 did not show any color degradation after a long period of time. Figure 12 illustrates the lightness index difference. After 4000 h of exposure all values were below 1.2, and the lightness had a tendency to darken. The lightness index difference for SA-3030 was less than 1.0, an indication of no chalk phenomenon.

On the basis of the results of the weatherability tests, increased weatherability can be attributed to an increasing content of MPTS. As mentioned in the introduction, improved weatherability of fluoro-resin coatings and silicone resin coatings is believed to result from the large bonding energy between the atoms in these materials and its prevention of degradation.[12] The present results support this reasoning.

CONCLUSIONS

Silicone–acrylic resins (KLD) were synthesized from *n*-butyl acrylate, methyl methacrylate and *n*-butyl methacrylate and 3-methacryloxypropyl-trimethoxysilane (MPTS). Then white coatings were formulated, and their film properties and weatherability were tested.

KLD had an M_n of 14,500–17,800, polydispersity of 2.33–3.15, viscosity of 2.3–5.9 stoke, and conversion of 88.4–89.0%. In the synthesis of KLD, average molecular weight and viscosity increased but thermal stability improved with increasing silicone component of MPTS. The formulated coatings had excellent adhesion with various substrates, and other physical properties were favorable. Both the outdoor exposure test and the accelerated test indicated that gloss retention, yellowness index difference, color difference, and lightness index difference were satisfactory. In particular, SA-3030, which has 30 wt % MPTS, proved to be a category of super-weatherable coatings.

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