Cure Behavior of Polyester-Acrylate Hybrid Powder Coatings

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INTRODUCTION

Increasingly stringent health, safety, and environmental regulations within the industrial finishing industry have caused coatings formulators to focus on the development of technologies that meet the dual requirements of environmental compliance and high performance. As a result, powder coatings are enjoying high growth as industrial finishers look for ways to meet these challenges. Carboxylic acid functional polyester resins for powder coatings in particular have experienced increased acceptance in the market place. Their ability to be cured by a wide range of epoxide curatives illustrates the versatility of this technology.

Triglycidyl isocyanurate (TGIC) is commonly used to cure these polyesters. Since the early 1980s, TGIC-cured polyester powder coatings have been very successful in gaining market share, particularly in applications requiring excellent exterior durability, such as automotive trim, patio furniture, and lawn and garden equipment. From 1985 to 1995, U.S. thermosetting powder coatings market share of polyester TGIC powder coatings increased from approximately 11 to 19.5%.[1,2]

Legislation recently passed by the Technical Progress Committee of the European Commission indicates that powders containing TGIC have to be labeled according to the provisions of the Hazardous Materials Directive.[3] *Table* 1 illustrates a few curing agent alternatives that have been introduced in the marketplace in attempts to address this issue.

Acrylate-Cured Polyester Powder Coatings

Glycidyl methacrylate (GMA) acrylic resins provide the polymeric backbone to powder coatings now being evaluated for the future topcoat/clearcoat of automobiles.[4] These resins have excellent resistance to environmental hazards such as ultraviolet light and acid rain. Also, because of its highly polar nature, polymers based on GMA have excellent adhesion to metal substrates.[5] Therefore, they have been chosen as candidates for use as crosslinkers in polyester powder coatings requiring high durability.

Volatile-free, two-component powder coatings have recently been developed that offer the performance prop-

Interest in finding alternatives to TGIC-based powder coatings have driven development of alternate binders for the exterior durable powder coating market. Thermoset powder coatings utilizing carboxylic acid functional polyester resins cured with epoxide-functional acrylate polymers have been evaluated and characterized. Modification of the composition and molecular weight of either component influences the physical properties of formulated powder coating. The behavior of these coatings during the cure process has been investigated and compared with polyester/TGIC systems.

erties of TGIC-cured polyester coatings.[6] These curing systems are based on epoxide-functional acrylate resins, utilizing GMA.

Development of a polyester/acrylate curing system necessitates the optimization of both the polyester and acrylate resins. As is typical in powder coatings, an imbalance or weakness in one of the binder components' properties usually will detrimentally affect the entire coating system. For example, if either component has a glass transition temperature (T_g) below 40°C, the storage stability of the powder paint will usually be poor.

A balance of appropriate coating viscosity during the curing process (chemorheology) and reactivity is necessary to produce a highly crosslinked film with adequate open time to flow, the result being a smooth visual appearance with good mechanical and chemical properties. Comparison of the melt viscosity behavior of the polyester/acrylate system and other common commercial powder coatings (*Figure* 1), indicates that the rate of viscosity increase occurs more rapidly in the polyester/acrylate system.

Table 1—TGIC-Free Alternatives

Current Alternatives	Advantages	Disadvantages
ß-Hydroxyalkyl amide (HAAM)	Good mechanicals Low toxicity[a]	Volatiles, yellowing, hydrolytic stability[a]
Tetramethoxymethyl glycoluril (TMMGU)	Good mechanicals[b]	Volatiles, storage stability[b]

(a) Wood, K. and Hammerton, D., "Hydroxylalkylamide Crosslinkers for Powder Coatings," *Mod. Paint Coat.*, 81 (6), pp. 60 & 62 (June 1991). Supplied as Primid® by EMS Chemie, Inc.

(b) Jacobs, W. and Sansur, S., "Powderlink® 1174 Crosslinker for Powder Coatings," R.G. Lees, *Proc. 1992 Waterborne, Higher Solids, and Powder Coatings Symposium*, pp. 196-204.

EXPERIMENTAL

Powder Coating Formulation and Processing

White powder coatings were formulated on a 1:1 epoxide:acid equivalence basis, using a 0.5/1.0 pigment/binder ratio. The general formulation is shown in *Table* 2.

Premixing of each formulation was performed in a Henschel FM-10 high-intensity mixer for two minutes. Extrusion was carried out with a Werner & Pfeiderer ZSK-30 twin screw extruder operated at 250 rpm, with Zone 1 set @ 100°C, and Zone 2 set @ 80°C. The molten extrudate was cooled by means of Strand chill roll. The resulting chips were ground with a Brinkmann ZM-1 centrifugal mill at 15,000 rpm. Powder was classified with a 200 mesh sieve, utilizing a Powder Process System reverse air sieve, and applied electrostatically onto cold-rolled steel panels with a Nordson Versa-Spray corona charging gun. The applied voltage was 80 kV. Film thickness of the coated panels ranged from 50-63 μm, and the cure schedule for each coating was 15 min at 200°C.

Coating Characterization Methods

GEL TIME REACTIVITY TEST (Powder Coating Institute (PCI) Recommended Procedure #6, Powder Coating Institute Test Method Manual, Powder Coating Institute, 1800 Diagonal Road, Suite 370, Alexandria, VA 22314.): A Thermo-Electric Co. cure plate, set at 200°C, was utilized for measuring each powder coating's gel time.

INCLINED PLATE FLOW (PCI Recommended Procedure #7): Powder pellets were made using a Parr pellet press and the pellets were placed on 25 mm × 75 mm × 1 mm microscope slides. The slides were then transferred to a 62.5° angled steel plate maintained at 180°C. After a 15-min contact time, the distance of pellet flow was recorded.

GEL FRACTION ANALYSIS: Powder coatings were electrostatically applied to aluminum plates, each plate containing a release agent. After curing the powders at 200°C for 10 min, the films were removed from the aluminum substrate and weighed on a Mettler AE-200 analytical scale, then dipped in acetone for 24 hr. The specimens were then allowed to dry for one hour in a 100°C oven and weighed again. The fraction of film retained by weight was recorded.

MELT VISCOMETRY (ASTM D 4287-88): The melt viscosity of each resin was measured with an ICI Cone &

Plate Viscometer, Model VR-4510 (BYK-Gardner, Inc.) using a VR-4200 cone with a cone angle of 2°C and a nominal diameter of 0.56 in. The shear rate was 3,600 second[-1].

DIFFERENTIAL SCANNING CALORIMETRY (**DSC**): The samples were sealed in hermetic aluminum pans and scanned with the TA Instruments (DSC 2920 by heating from 0° to 280°C at 10°C/min. The glass transition and fusion (T_f) temperatures were determined from the midpoint of the respective endothermic transitions. The onset of the cure temperature (T_{oc}) was determined from the cure exotherm transition. The Arrhenius parameters, pre-exponential factor ($\log(Z)$), and activation energy (E_a) as well as the heat of reaction (ΔH) were calculated using the Borchardt and Daniels Kinetics Data Analysis Program, version 4.0.

CHEMORHEOLOGY: The flow and cure properties of the powder coatings[9,10] were recorded on a TA Instruments CSL² 500 Stress Rheometer equipped with an ETM high temperature unit. The CSL²500 was operated at 1% strain for all samples using a parallel plate geometry with a 0.5 mm gap. The temperature was ramped from 30° to 250°C at 5°C/min heating rate for all samples, and the frequency of the experiments was set at 1.0 Hz. Stress sweep measurements showed no signs of non-linearity under these conditions.

A sequence of tests was also performed to simulate a practical oven cure schedule of 10 min at 180°C. This was carried out by heating each sample from 30° to 180°C at a rate of 30°/min. The samples were then held at this peak temperature for an additional five minutes. The frequency and strain were set at 1.0 Hz and one percent, respectively.

By applying a defined stress and measuring the magnitude and phase angle of the resulting displacement, the complex modulus (G^*) can be determined. From this parameter, the complex viscosity (η^*) and storage modulus (G') were calculated. The Arrhenius kinetic crosslinking parameters, $\log(k_\infty)$ and E_k, were determined
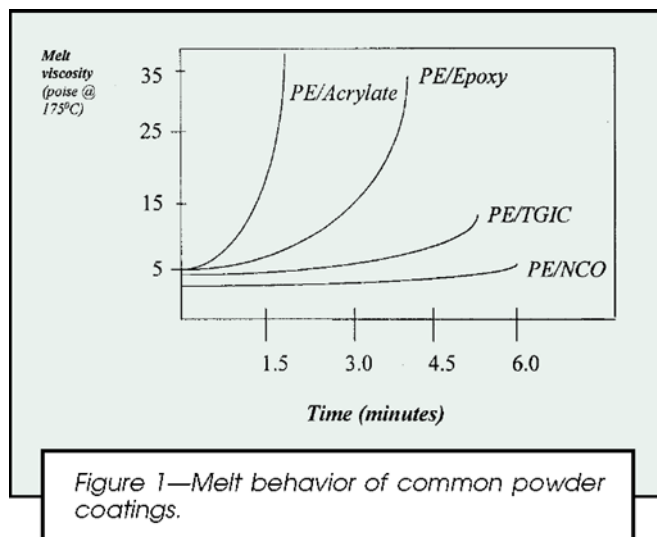

Figure 1—Melt behavior of common powder coatings.

Table 2—Polyester/Acrylate Experimental Formulations

	Parts By Weight
Polyester + GMA acrylic	100
Flow agent[a]	1
Benzoin	0.5
TiO$_2$[b]	50

(a) Modarez MFP Powder, Synthron Specialty Chemicals, Inc.
(b) TiPure R-960, DuPont White Pigments & Minerals Products, Inc.

by fitting the cure portion of the complex viscosity data. Other parameters, such as the minimum complex viscosity (η^*_{min}) and temperature for onset of rapid cure (T_{rc}) were obtained directly from the chemorheological scans.

TRANSMISSION ELECTRON MICROSCOPY (TEM): Specimens of powder coatings consisting of approximately 90 wt% polyester resin and 10 wt% acrylic resin were prepared for TEM analysis in Teflon molds at three different temperature/time conditions: 105°C for 20 min, 200°C for 4 min, and 200°C for 10 min. To obtain electron-transparent specimens for TEM, the resultant films (measuring ca. 1 mm thick) were sectioned in a Reichert-Jung Ultracut-S cryoultramicrotome maintained at –100°C. Phase contrast between the polyester and acrylic resins was enhanced through preferential heavy metal staining with the vapor of one percent aqueous ruthenium tetroxide (RuO$_4$) for seven minutes. The stained sections (nominally measuring 100 nm in thickness) were subsequently examined on a Zeiss EM902 (transmission) electron spectroscopic microscope operated at 80 kV and 0 eV energy loss (to eliminate the contribution of inelastically scattered electrons to image formation).

RESULTS AND DISCUSSION

Polyester

Carboxylic acid equivalent weight is one of the most important factors in determining the performance of a polyester-based coating. Therefore, a range of acid values and molecular weights (M$_n$) were chosen for study (*Table* 3). PE1 and PE2 were initially developed for cure with linear aromatic di-epoxides. The T$_g$ of these resins was held relatively constant to maintain physical stability of the powder coating.

Initial polyesters contained exclusively terephthalic acid as the acid component. However, accelerated weathering studies indicated premature gloss loss and coating discoloration with this type of polyester. Polyester resins with proprietary modifications made to the polymer backbone displayed improved exterior durability. Mechanisms of polymer photo-oxidation for these types of polymers have been described by Belder and Koldijk.[7]

Table 4 shows some key results of using PE1 and PE2 when cured with a GMA acrylic. The low equivalent weight polyester has good impact resistance but poor flow and very fast gel time, while increasing the equivalent weight (PE2) results in poor impact properties with better flow.
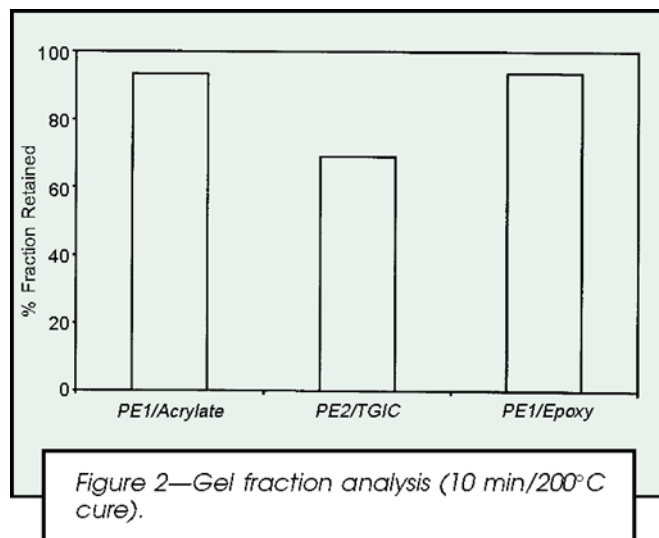

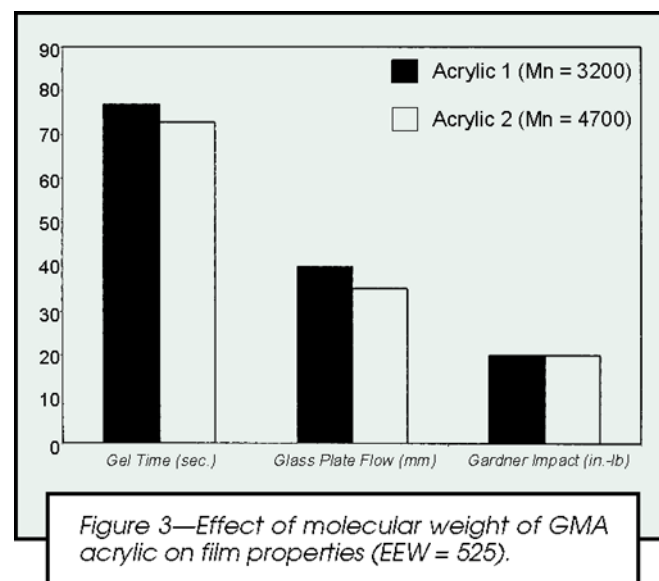Figure 2—Gel fraction analysis (10 min/200°C cure).

Figure 3—Effect of molecular weight of GMA acrylic on film properties (EEW = 525).
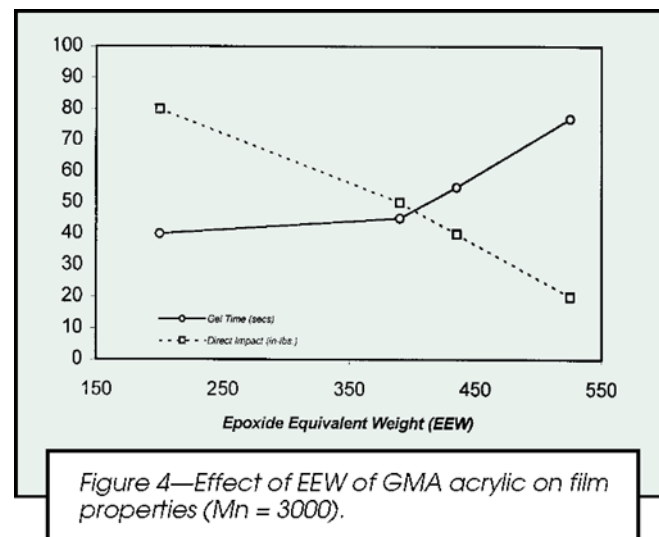

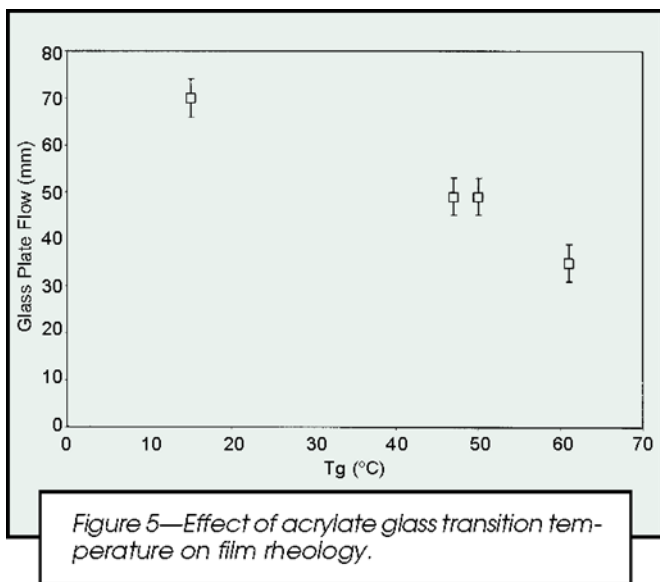Figure 4—Effect of EEW of GMA acrylic on film properties (Mn = 3000).

Figure 5—Effect of acrylate glass transition temperature on film rheology.

Gel fraction analysis (*Figure* 2) indicates a high extent of reaction under typical cure conditions.

This data suggests two key conditions: (1) acid functionality must be limited to allow proper melt viscosity reduction to occur before the onset of gelation, for good flow and leveling, and (2) a minimum of crosslinking sites should be maintained to achieve high crosslink density and good impact resistance. Based on these results, an alternate high equivalent weight polyester (PE3) was synthesized and used for subsequent work.

GMA Acrylic

Fine-Clad® A-229-30-A GMA acrylic resin (GMA1) was selected as the initial crosslinker for this study because of its ability to produce smooth, glossy films in multiple component systems, i.e., coating binders with bi-functional polyester resins crosslinked with isocyanate prepolymers and GMA acrylic polymers.[8]

Other GMA resins were synthesized, varying the epoxide equivalent weight (EEW), molecular weight, and T_g. An important consideration in acrylate polymer design is the balance of exterior durability and reactivity. *Table* 5 displays the key properties of the crosslinkers. The GMA acrylic crosslinkers were all designed to have a higher functionality than TGIC. This is due to the observation that higher overall functionality is required for the GMA acrylic-cured systems to obtain similar mechanical and chemical performance. GMA7 was developed as a solution, and was studied alongside the solid resins. The GMA crosslinkers were evaluated using PE3.

Table 3—Polyester Resin Properties

Resin	Acid Value (mg/g KOH)	Mn	T_g (°C)	Viscosity (P, 200°C)
PE1[a]	55	1700	64	50
PE2[b]	35	3000	65	65
PE3	30	3200	60	60

(a) Fine-Clad® M-8841, Reichhold, Inc.
(b) Fine-Clad® M-8930, Reichhold, Inc.

Table 4—Powder Coating Cure and Flow Properties

Polyester Resin	PE1	PE2
Polyester/Acrylate[a] (wt ratio)	66/34	75/25
Gel time (sec)	15 ± 2	45 ± 4
Inclined plate flow (mm)	15 ± 2	50 ± 4
Impact resistance, direct (in.-lb)	50 ± 10	10 ± 2

(a) Fine-Clad® A-229-30-A, Reichhold, Inc.

The following three graphs illustrate some of the influence of the GMA acrylic. *Figure* 3 shows that variation in molecular weight has only small influence on key film properties in the two-component system.

Increasing epoxide functionality, however, has a direct effect on performance. *Figure* 4 illustrates the increase in impact resistance, with increasing reactivity (short gel time) as functionality increases. Decreasing the T_g of the acrylate resin results in more flow (*Figure* 5) and, hence, better coating appearance.

These results indicate that the optimized candidate would have a high level of functionality, but still have low T_g for good film formation and appearance. Unfortunately, obtaining good reactivity often necessitates sacrificing film appearance. Powder coatings based on GMA7, however, have a balance of these two competing properties. Characterization on this polyester/acrylate hybrid powder coating has revealed intriguing similarities and differences compared to TGIC-cured polyesters.

Thermal Analysis

Comparison of the DSC data illustrates the three main thermal events[11] that occur upon heating: (1) the transition of the powders from glassy to rubbery state (T_g); (2) powder fusion at the melt onset temperature (T_f); and (3) exothermic peaks indicating crosslinking, characterized by the temperature for onset of cure (T_{oc}) and the peak cure temperature. *Figure* 6 shows these events. While both powder coating systems have similar fusion temperatures and exothermic peak temperatures, the acry-
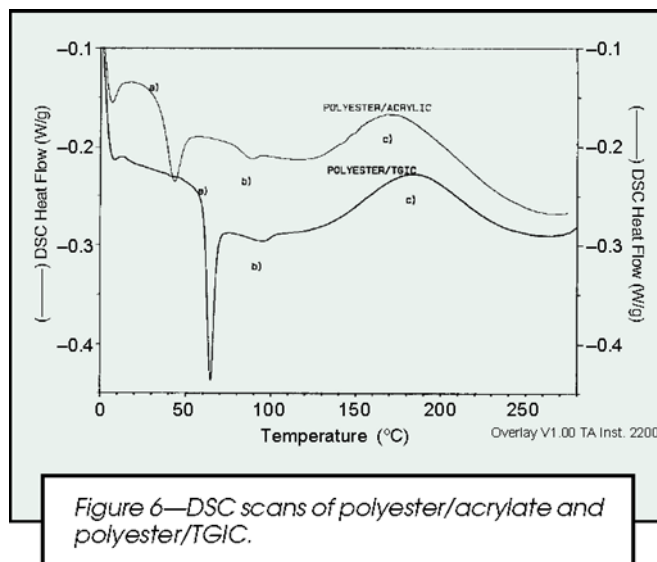

Figure 6—DSC scans of polyester/acrylate and polyester/TGIC.

Table 5—Crosslinker Properties

Resin	Mn	EEW	Epoxide/Chain	T_g (°C)	Viscosity (P, 175 C)
GMA1[a]	3200	525	6.1	56	80
GMA2	3200	525	6.1	49	50
GMA3	4700	525	9.0	61	>100
GMA4	2200	490	4.5	47	46
GMA5	2900	435	6.7	50	70
GMA6	2000	390	5.1	50	39
GMA7	3000	200	15.0	15	<1
TGIC[b]	297	99	3.0	—[c]	<1

(a) Fine-Clad® A-229-30-A, Reichhold, Inc.
(b) Araldite PT-810, Ciba Inc.
(c) Melt Point = 95°C.

late-cured polymer has significantly lower T_g (37°C vs. 60°C).

DSC data indicates a faster cure rate for the polyester/acrylate system. Of particular interest to the end-user of powder coatings is the cure rate in the temperature range of 175° to 200°C. This range is the most commonly used for curing of general industrial powder coatings. The calculated rate constant is approximately 70 to 90% higher in this region. The Borchardt and Daniels kinetic parameters are given in *Table* 6, and indicate that the faster cure rate for the acrylate-cured system is due to a higher pre-exponential factor. The energy of activation is actually higher for the acrylate-cured system, but with a higher overall heat of reaction and reaction order.

Chemorheology

Figure 7 displays the melt and cure behavior of both epoxide crosslinked systems as the powders undergo a temperature ramp from 30° to 250°C.

The melt portion of the curves show the minimum viscosity is achieved by polyester/acrylate much sooner—nearly 35°C lower than polyester/TGIC (*Table* 7). The slope of the acrylate cure curve is much steeper, indicating a much faster reactivity. The acrylate system reaches its maximum melt viscosity at approximately 180°C, while the TGIC-based powder still undergoes an increase in viscosity at 200°C. The final storage modulus (G') is higher for the acrylate-cured system.

Isothermal chemorheology studies shown in *Figure* 8 also show the difference in cure behavior of these two systems, with the polyester/acrylate quickly achieving maximum viscosity within the five-minute test period. The calculated kinetic parameters[12] correlate well with those obtained from DSC.

Microscopy

Shown in *Figure* 9 are TEM micrographs collected from the PE3/GMA7 based coatings prepared at various time/temperature conditions. Each micrograph clearly reveals a bi-phase microstructure composed of electron-opaque (dark) dispersions of acrylate preferentially stained by RuO_4 in an unstained or lightly stained polyester matrix (gray). The numerous light regions correspond to holes that formed during film preparation, most likely due to incomplete film coalescence. Coating

Table 6—Thermal Analysis Data

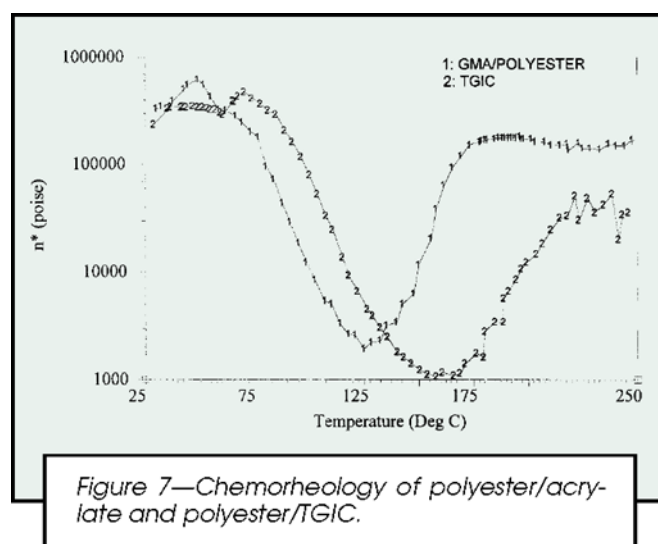|	Polyester/Acrylate	Polyester/TGIC
T_g (°C)	37	60
T_f (°C)	91	95
T_{oo} (°C)	128	134
Log (Z) (sec⁻¹)	18.2 ± 2.3	14.1 ± 0.8
E_a (kJ/mole)	155 ± 18	125 ± 6
ΔH (J/g)	15.8 ± 3.5	11.9 ± 1.2
Reaction order	3.9 ± 0.3	2.8 ± 0.3

Figure 7—Chemorheology of polyester/acrylate and polyester/TGIC.

Table 7—Chemorheological Data

	Polyester/Acrylate	Polyester/TGIC
η^*_{min} (Poise/temp.)	1926/125°C	1063/158°C
η^* (Poise at 180°C)	166100	2785
η^* (Poise at 200°C)	170900	14470
T_{rc} (°C)	146	173
G' (200°C)	1.06 x 10⁶	0.87 x 10⁵
Log (k_w) (P⁻¹)	25.5 ± 3.0	18.6 ± 1.2
E_k (kJ/mole)	173 ± 11	131 ± 5

Table 8—Polyester/Acrylate and Polyester/TGIC Powder Coating Formulations

	Polyester/Acrylate	Polyester/TGIC
	Parts by Weight	Parts by Weight
PE2[a]	—	93.0
PE3	92.0	—
TGIC[b]	—	7.0
GMA7	8.0	—
Flow agent[c]	1.0	1.0
Benzoin	0.5	0.5
TiO$_2$[d]	50.0	50.0

(a) Fine-Clad® M-8930, Reichhold, Inc.
(b) Araldite PT-810, Ciba, Inc.
(c) Modarez MFP Powder, Synthron Specialty Chemicals, Inc.
(d) TiPure R-960, DuPont White Pigments & Mineral Products, Inc.

Figure 8—Isothermal chemorheology (180°C).
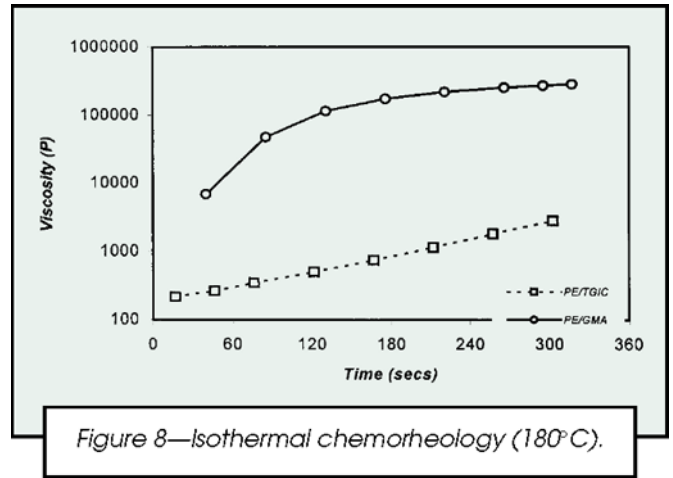
porosity may be attributed to the chemical crosslinking of the polyester and acrylate resins. These micrographs illustrate the inherent difficulty in maintaining uniform dispersion in polyester/acrylate coatings.

The acrylate-rich domains exhibit a broad size distribution ranging in diameter from about 80 to 800 nm, with many of the large domains appearing as aggregates of small domains (*Figure* 9a). Aggregation indicates that coalescence during phase separation between the polyester and acrylate domains was hindered by the initiation of concurrent polyester-acrylate crosslinking. Increasing in cure temperature, but reducing cure time, induces large-scale phase separation (*Figure* 9b), since very few small domains (measuring on the order of 80 nm in diameter) are visible. Longer cure times appear to result in a slight improvement in phase mixing (*Figure* 9c). However, a comparison of the micrographs reveals that the low-temperature, long-time heat cycle yields the greatest population of small (<100 nm in diameter) acrylate domains in the coatings examined here.

These phase separations indicate the strong influence of the GMA acrylic physical properties upon the glass transition and melt temperature of the coating. The in-

herent incompatibility of polyester and acrylate may also be reflected in the higher activation energy required to initiate crosslinking, and may help explain why higher functional acrylate polymers are necessary to obtain good mechanical properties. This could be due to some of the epoxide functional groups remaining within the acrylate domain during the coalescence phase and not being accessible for crosslinking. Hence, maintaining an optimized dispersion of acrylate within the polyester is critical to enhanced film performance.

Although system storage stability currently is not optimized, developments such as higher T_g polyesters are being investigated to solve this shortcoming. Additionally, other processing techniques are being evaluated to more effectively mix the liquid acrylate resin with the solid components of the powder coatings.

CONCLUSIONS

Thermal analysis and chemorheology have shown that a polyester resin cured with an appropriate GMA acrylic

Table 9—Film Properties of Polyester/Acrylate and Polyester/TGIC

	Polyester/Acrylate	Polyester/TGIC	Test Method
Cure schedule	15 min, 200°C	15 mins, 200°C	
Film thickness, mils	2.0–2.5	2.0–2.5	ASTM D 1186
Gardner impact resistance Direct/reverse (in.-lb)	80/80	80/80	ASTM D 2794
Pencil hardness	H-2H	H-2H	ASTM D 3363
Knoop hardness	12.4	15.1	ASTM D 1474
MEK resistance	Slight softening	Slight softening	ASTM D 4752
Storage stability, 7 days @ 35°C	No change	No change	PCI Test #1
Salt spray resistance, 1000 hr, B-1000 panels	3.2 mm Creep from scribe, no adhesion loss	2.8 mm Creep from scribe, no adhesion loss	ASTM B 117
% Non-volatiles	99.2	99.1	PCI Test #9
Film Appearance			
Long wave rating	65	40	—[a]
Short wave rating	40	30	—[a]
PCI smoothness rating	4	4-5	—[b]
60°/20° Gloss	92/75	95/83	ASTM D 523

(a) BYK-Gardner wave scan rating.
(b) PCI Visual Smoothness Standards (1-10, 10 = best).

has a faster cure rate than the TGIC. This is due to a higher Arrhenius pre-exponential factor, despite a higher energy of activation. TEM studies show a bi-phase microstructure in the polyester/acrylate film, which may explain the differences in cure rate and suggests processing of the powder may be more critical for this technology.

Table 8 displays a prototype powder coating formulation based on PE3 and GMA7. The improved film properties of PE3/GMA7 are shown in *Table 9*, with comparison to the properties of the polyester/TGIC powder coating. *Figure* 10 shows accelerated weathering comparisons for both systems. The key properties of impact, UV resistance, volatile emission, and chemical resistance are nearly identical in these two technologies.

The GMA acrylic-cured polyester hybrid technology illustrated here is well suited for "general metal" end users, particularly as a replacement for TGIC-cured polyester powders. Powder coating formulations utilizing high GMA functionality, low T_g crosslinkers with carboxylic acid functional polyester resins have been developed whose film properties mirror those of TGIC-cured powders. Future developments in this technology will focus on providing even smoother films, lower cure temperatures, and UV durability to rival that of automotive topcoat-grade coatings. These properties should be provided as further enhancements to this binder system are made, including improvements in handling and storage stability.
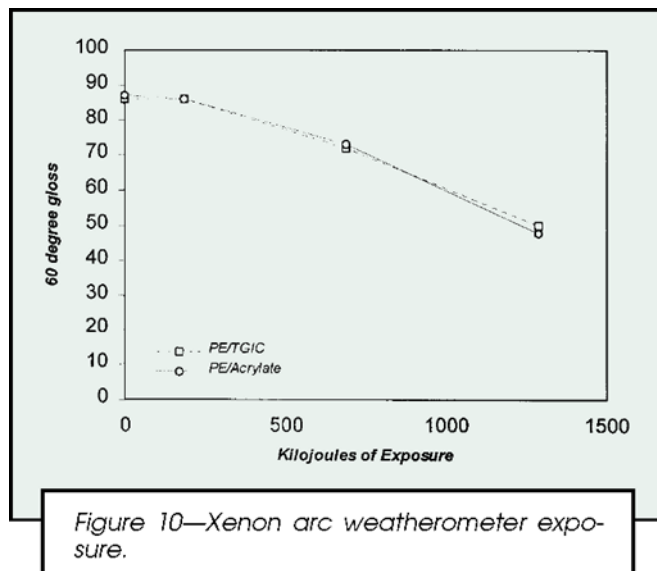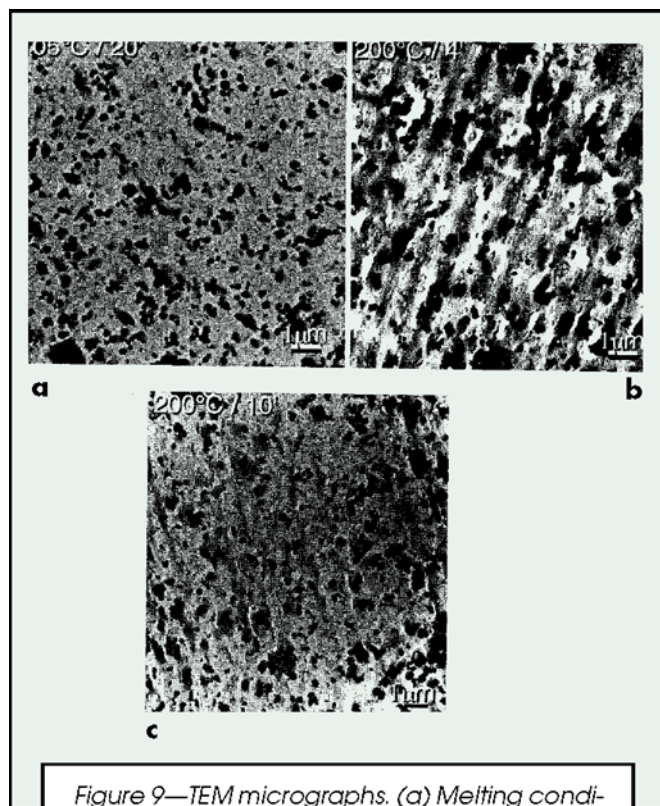

Figure 9—TEM micrographs. (a) Melting condition; (b) initial cure state; and (c) full cure cycle condition.

Figure 10—Xenon arc weatherometer exposure.

ACKNOWLEDGMENTS

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