

Synthesis, Characterization, and Performance Evaluation of Hard, Anticorrosive Coating Materials Derived from Diglycidyl Ether of Bisphenol A Acrylates and Methacrylates

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ABSTRACT: Diglycidyl ether of bisphenol A acrylate (DAC) and diglycidyl ether of bisphenol A methacrylate (DMAC) were synthesized by the reaction of an epoxy [diglycidyl ether of bisphenol A (DGEBA)] with acrylic acid and methacrylic acid, respectively. The synthesized resins were characterized by determination of the acid, hydroxyl, and saponification values. Structure elucidation was done by gel permeation chromatography, Fourier transform infrared spectroscopy, ¹H-NMR spectroscopy, and ¹³C-NMR spectroscopy. DACs were cured with melamine formaldehyde resin at low pH values. The pH of the resin systems was adjusted with phosphoric acid. The coatings of these systems were formed on mild steel specimens for physico-

mechanical and chemical/corrosion-resistance performance. The coatings of DAC and DMAC showed excellent scratch hardness and good impact-resistance performance. The coatings of DMAC showed better performance than DAC with respect to chemical and corrosion resistance. Thermogravimetric analysis and differential scanning calorimetry were used to investigate the thermal stability and curing behavior of these systems. DAC showed a little higher glass-transition temperature than DMAC and also showed a higher thermal resistivity. © 2004 Wiley Periodicals, Inc. *J Appl Polym Sci* 95: 494–501, 2005

Key words: surfaces; coatings; toughness

INTRODUCTION

Epoxy resins are an important class of thermosetting polymers that are chiefly used as adhesives, laminates, boards, molds for casting, and composite materials in the aerospace and aircraft industries.^{1–4} In addition to these applications, these resins find substantial application in high-performance surface coatings.^{5,6} Coatings from epoxies have been found to show marked resistance to chemicals and corrosive environments. However, these coatings fail to give satisfactory performance under strained conditions.^{7,8} Recently, vigorous interest has been shown in the modification of epoxies to overcome shortcomings such as low toughness, poor weathering resistance, low thermal stabil-

ity, low pigment-holding ability, and yellowing.⁹ These drawbacks of epoxy resins restrict their wide application in paints and coatings. The epoxy resins have been modified by other oligomers/polymers to improve these properties. The most effective method for the modification of epoxy resins has been their blending with other polymers.¹⁰ The toughening of epoxy resins has generally been done by blending with thermoplastic polymers.^{11,12} However, such blending increases the viscosity of the resins and leads to the requirement of a larger amount of solvent for surface coating. The evaporation of excess solvent during and after curing causes shrinkage of the coating and produces internal stresses.¹³ The performance of coatings in these conditions is also affected because of the presence of residual solvents. A dilute solution of resins is, therefore, not recommended for paints and coatings.¹⁴ Phenolic epoxy resins, however, have been reported for excellent thermal resistivity and high scratch hardness, but these system cure at quite high temperatures.¹⁵ Silicone-polymer-modified epoxy resins have recently been used to achieve better performance, but the phase separation and low surface energy of silicone-modified epoxy resins hamper their use in surface coatings.¹⁶ The unique chemistry, outstanding properties, and wide array of monomer availability are the driving force for tailor-making

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products from acrylics.¹⁷ Very often, acrylics are used to modify and improve the performance of other resins commonly used in the paint industry, such as alkyds, epoxies, and polyesters. The modification of epoxy resins through blending by acrylate is known to improve adhesion, flexibility, nonyellowing, resistance against scratch, and resistance to chemicals.^{9,18,19} The high polarity of the resin improves the adhesion and pigment-holding properties. As a curing agent, melamine formaldehyde (MF) resins show superior performance in terms of reactivity, storage stability, and cost effectiveness and are mainly used to crosslink the hydroxyl groups present in the alkyds, acrylics, polyesters, or epoxies.²⁰ In this study, we have used MF to cure diglycidyl ether of bisphenol A acrylate (DAC) and diglycidyl ether of bisphenol A methacrylate (DMAC) to obtain high-performance coating materials and to incorporate the excellent characteristic of its components. The physicomechanical and chemical/corrosion-resistance performance and the thermal stability of the developed coating materials were tested. We have observed that most authors who have worked on the epoxy acrylate (EAC) blending or MF curing of acrylates and alkyds have not studied the coating characteristics of the synthesized resins.^{18–20}

EXPERIMENTAL

Materials

The epoxy resin of diglycidyl ether of bisphenol A (DGEBA; epoxy equivalent = 190) was procured from Ciba Specialty Chemicals (New Delhi, India). MF was obtained from S.D. Chemicals (New Delhi, India). Triphenyl phosphine was purchased from Himedia (Mumbai, India). Acrylic acid and methacrylic acid were procured from Merck (Darmstadt, Germany). Potassium hydroxide, methanol, and ethylene glycol monomethyl ether were analytical grade and were procured from Merck (Mumbai, India).

Synthesis of DAC

DAC was synthesized from the reaction of DGEBA with acrylic acid in a four-necked, round-bottom flask equipped with a mechanical stirrer, condenser, dropping funnel, and thermometer. Ethylene glycol monomethyl ether (100 mL), DGEBA (90.0 g, 0.25 mol), acrylic acid (43.2 g, 0.6 mol), triphenylphosphine (0.45 g, 0.5 wt % of DGEBA), and hydroquinone (0.0043 g, 0.01 wt % of acrylic acid) were added to the flask. The last material was added to protect the thermal polymerization of the double bond. The reaction mixture was heated at 80°C for 3.5 h. The acid value of the reaction was determined at regular intervals to monitor the progress of the reaction.²¹

Synthesis of DMAC

The reaction flask was fitted with the accessories as was done for the synthesis of DAC. DMAC was synthesized by the reaction of 90 g (0.25 mol) of DGEBA dissolved in 100 mL of ethylene glycol monomethyl ether with 51.6 g (0.6 mol) of methacrylic acid, 0.45 g (0.5 wt % of DGEBA) of triphenylphosphine, and 0.0052 g (0.01 wt % of methacrylic acid) of hydroquinone. The reaction mixture was heated at 90°C for 4.0 h. The determination of the acid value at regular intervals was used to monitor the progress of the reaction.²¹

After the completion of the reaction, the reaction mixture was cooled to room temperature and washed with deionized water. Excess solvents and water were removed in a high-vacuum rotary evaporator (Tanco, Pvt. Ltd., Delhi, India) and by further heating in a vacuum oven at 105°C for 12 h.

Characterization of the resins

DAC and DMAC were characterized by gel permeation chromatography (GPC), Fourier transform infrared (FTIR) spectroscopy, ¹H-NMR, ¹³C-NMR, differential scanning calorimetry (DSC), and thermogravimetric analysis (TGA). FTIR spectra of the films of these polymers on a NaCl cell were recorded on a PerkinElmer (RX-1) spectrophotometer. ¹H-NMR and ¹³C-NMR spectra of the polymer were taken on a Jeol (Peabody, MA) (JNM FX-100) 300-MHz spectrometer with CDCl₃ as a solvent and tetramethylsilane as an internal standard. DSC measurements of the uncured DAC/MF and DMAC/MF were conducted on a DuPont 900 C (T.A. Instrument, Inc., New Castle, DE) differential scanning calorimeter in aluminum pans at a heating rate of 10°C/min and with a sample size of 10.0 mg. TGA (TA Instrument, Inc., New Castle, DE) was done in a N₂ atmosphere at a heating rate of 10°C/min to study the thermal stability of the cured films. GPC was performed with a Waters 510 (Water Corp., Milford, MA) gel permeation chromatograph with a refractive index detector and ultrastyrigel 10 Å, 500 Å, 100 Å columns and with tetrahydrofuran as the mobile phase. The flow rate was maintained at 1 mL/min. Various physicochemical properties, including specific gravity, refractive index, color value, and hydroxyl value,²² of these polymeric resins were also determined per standard methods (Table I).

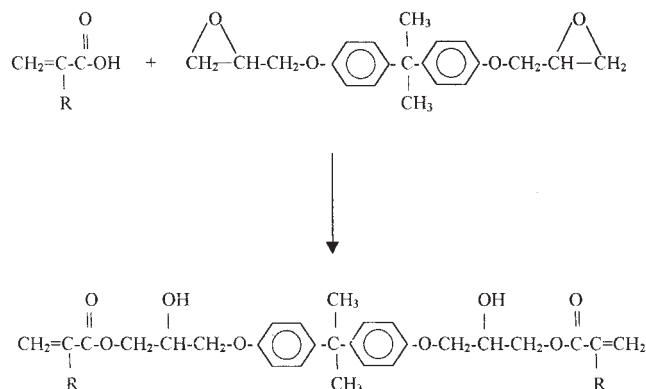
Preparation and testing of the coatings

The coating systems were prepared by the blending of the DAC and DMAC with MF in varying ratios (Table II) by a solution method²³ in different reaction vessels under vigorous stirring at room temperature. The coatings of these systems were applied on commer-

TABLE I
Physicochemical Characterizations of EAC and EMAC

Property	EAC	EMAC
Specific gravity	1.32	1.34
Refractive index	1.43	1.39
Inherent viscosity (dL/g)	0.852	0.896
Color value (No.)	2	2
Hydroxyl value (mg KOH)	7.12	6.52
Iodine value	70.0	66.0
Saponification value	201.26	192.78

cially available mild steel strips $30 \times 10 \times 1$ mm in size for the chemical resistance test and on a $70 \times 25 \times 1$ -mm aluminum panel for the determination of specular gloss at 45° with a glossmeter (model RSTP-20, Digital Instruments, Santa Barbara, CA), scratch hardness test (BS 3900), bending test (ASTM D 3281-84), and impact resistance test (IS:10 part 5/s 3) (Sheen Instruments Ltd., UK). The coated samples were baked for 10–30 min in an oven at different temperatures (110 – 140°C) to determine the optimum baking time and temperature. The optimum baking time and temperatures were 130°C and 20 min. Coating thickness was measured by an Elcometer (model 345, Elcometer Instruments, Ltd., Manchester, United Kingdom). The thickness of these coatings was about $60 \pm 5 \mu\text{m}$. The chemical/corrosion resistance tests were performed in water, acids (5 wt % HCl and 2 wt % H_2SO_4), and alkali (5 wt % NaOH) by placement of the samples in 3-in. diameter porcelain dishes and the dipping of the coated samples in the aforementioned media. Periodic examination was conducted until the coatings showed evidence of softening or deterioration (Table II). Salt-spray tests (ASTM B 117-94) were carried out in a salt mist chamber. The weathering resistance test (ASTM G 53) was also carried out by exposure of the coated panels to ultraviolet (UV) radiation (wavelength = 285 – 315 nm) and high-humid-



(1) R = H (acrylic acid) (2) R = CH_3 (methacrylic acid)

Figure 1 Synthesis of EAC and EMAC.

ity conditions in a QUV accelerated weatherometer (Sheen Instruments Ltd., UK).

RESULTS AND DISCUSSION

Product characterization

Figure 1 shows the reaction scheme of DGEBA with acrylic acid and methacrylic acid to form epoxy acrylate (EAC) and epoxy methacrylate (EMAC). The oxirane group of DGEBA reacted with a carboxylic group to form ester without scissoring the polyester molecular chain.⁹ The progressive decrease in acid value confirmed the formation of ester linkages, which was further confirmed by FTIR, $^1\text{H-NMR}$, and $^{13}\text{C-NMR}$ spectroscopy.

IR spectroscopy showed (Table III) characteristic bands at 3400 – 3428 cm^{-1} (DAC) and 3423 cm^{-1} (DMAC) for OH, bands at 1718.7 (DAC) and 1722.8

TABLE II
Film Properties of the EAC/MF and EMAC/MF Resin Systems

Resin code ^a	Scratch hardness (kg)	Impact resistance (lbs/in.)	Gloss at 45°	Bending (mm/diameter)	Humid resistance (h)	Salt spray (h; 3.5 wt % at 35.5°C)	QUV weatherometer (h)	Chemical resistance test ^b		
								HCl (5 wt %; 10 days)	H_2SO_4 (2 wt %; 10 days)	NaOH (5 wt %; 10 days)
EAC/MF-10	2.7	150	60	5.2	360	180	12	A	B	C
EAC/MF-20	3.6	150	60	6.8	480	192	14	A	B	C
EAC/MF-30	4.8	150	64	7.1	600	216	14	A	A	A
EAC/MF-40	5.0	150	64	9.6	612	204	14	B	B	A
EMAC/MF-10	2.9	175	60	4.4	504	192	13	A	B	C
EMAC/MF-20	3.78	200	70	5.0	614	216	15	A	A	B
EMAC/MF-30	5.0	200	70	5.6	658	228	17	A	A	A
EMAC/MF-40	5.0	200	70	6.0	652	222	15	A	A	B

^a The last digit of the resin code indicates the weight percentage of MF.

^b A = unaffected; B = loss in gloss; C = loss in gloss; D = film cracked at partially removed.

TABLE III
IR Absorption Bands of EAC and EMAC

Assignment	Band positions (cm ⁻¹)	
	EAC	EMAC
ν (O-H)	3402	3423
ν (Ar-H)	3029	3016
ν (CH, CH ₂ , CH ₃)	2930	2865-2955
ν (C=O)	1718.7	1722.8
ν (C=C)	1636.2	1636.4
ν (C=C) phenyl	1609.8, 1510.5	1610.6, 1510
δ (CH, CH ₂ , CH ₃)	1456.5	1458.0
ν (C-O)	1297.6	1296.9
δ (Ar-H)	1249.2	1248.9

(DMAC) for C=O, bands at 1297.6 (DAC) and 1296.9 cm⁻¹ (DMAC) for ester groups, which confirmed the formation of DAC and DMAC. ¹H-NMR spectra (Figs. 2 and 3) showed a doublet at $\delta = 4.2$ –4.3 and 4.0–4.2 ppm for the presence of a —O—CH₂ group in DAC and DMAC, respectively, whereas peaks of methyl groups attached to hydroxy appeared at $\delta = 3.8$ and 3.9 ppm as a doublet. The additional peak for the

methyl group of DMAC appeared at $\delta = 1.8$ ppm. ¹³C-NMR spectra (Figs. 4 and 5) showed peaks at $\delta = 165.2$ and 168.0 ppm for the carbonyl of ester in DAC and DMAC, respectively, whereas the peaks for the carbon attached to a hydroxyl group appeared at 67.2 ppm (DAC) and 69.8 ppm (DMAC).

The number-average molecular weight of DAC and DMAC as determined from GPC were 550 and 573, respectively, with polydispersity values of 1.08 and 1.05. The expected theoretical values were 524 and 552 for the complete conversion of epoxy groups and the absence of a secondary reaction. The nearly the same difference in the experimental and theoretical values of the molar masses of DAC and DMAC (26 and 21 units, respectively) appeared to be experimental error. The saponification values for DAC and DMAC were 201.26 and 192.78 mg of KOH/g, respectively. These values showed good agreement with the theoretical values of 213.74 mg of KOH/g for DAC and 202.8 mg of KOH/g for DMAC, which were calculated with the assumption of complete conversions during synthesis. The negligible difference between the experimental

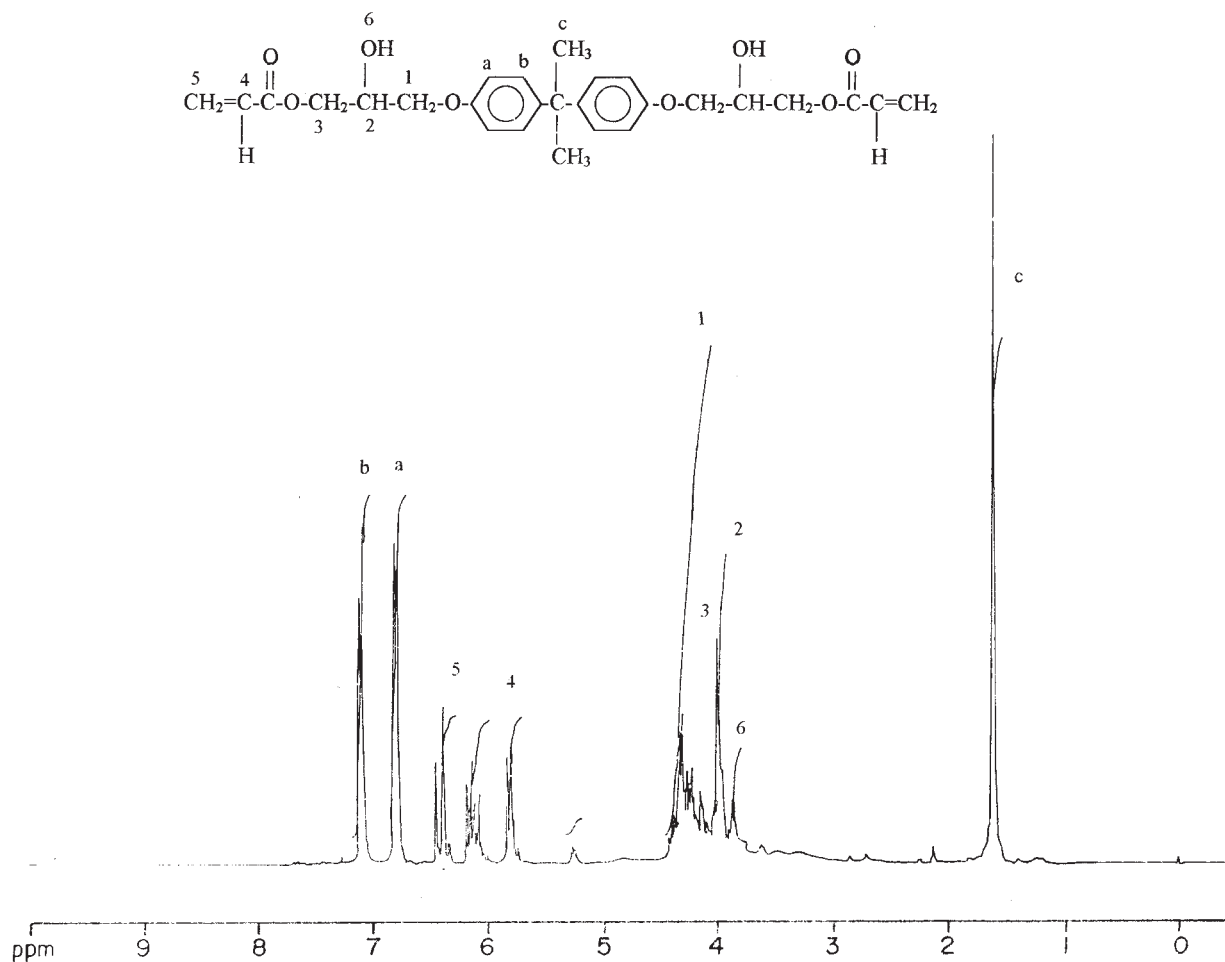


Figure 2 ¹H-NMR spectra of EAC.

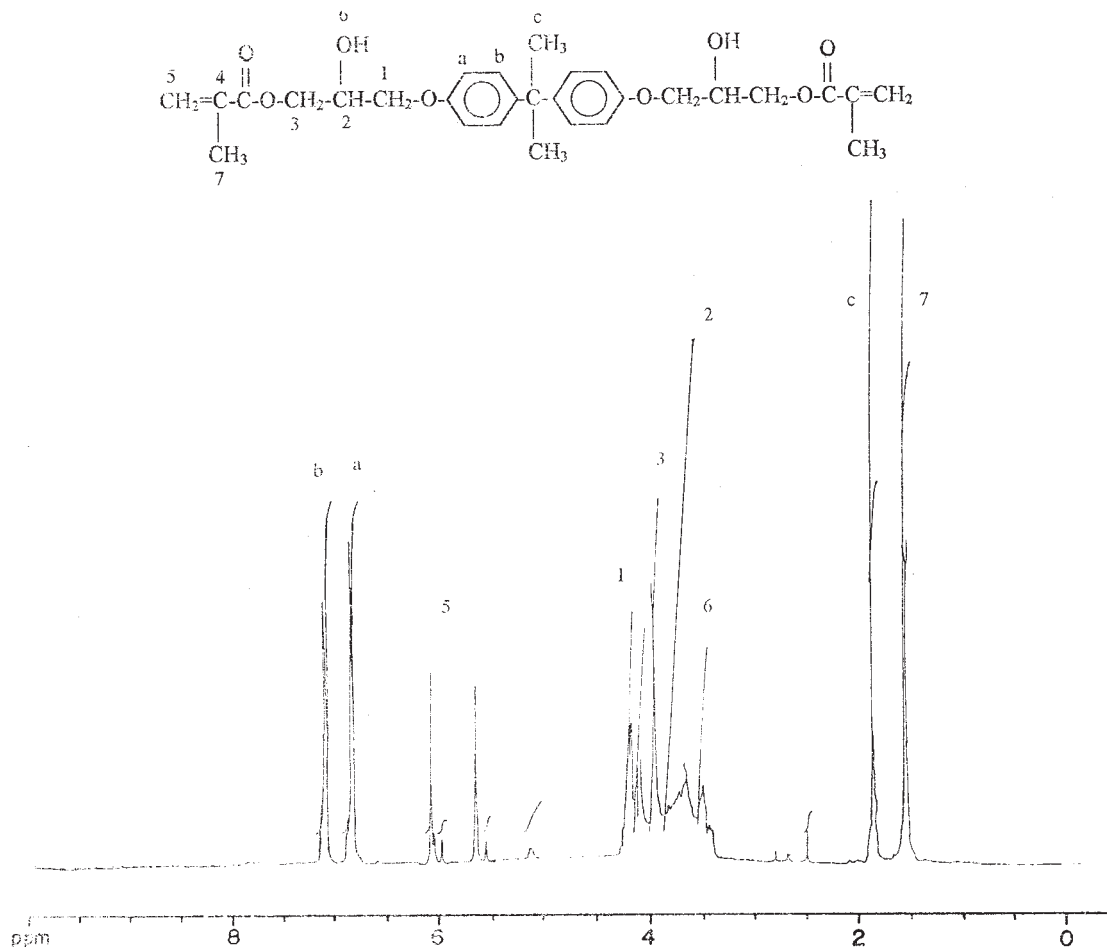


Figure 3 $^1\text{H-NMR}$ spectra of EMAC

and calculated saponification values ruled out the presence of any secondary reaction.

Thermal analysis

The DSC thermograms of DAC/MF and DMAC/MF (Fig. 6) showed an endothermic peak followed by an exothermic peak with a shoulder. The endothermic peaks in the thermograms were attributed to the comelting of DAC/MF and DMAC/MF and were observed at 103 and 96°C, respectively. The slightly higher melting temperature of DAC/MF may have been due to an enhanced interaction of constituent units in this polymer.²⁴ The appearance of a broad exothermic peak was assigned to the crosslinking reaction of DAC/MF and DMAC/MF. The starting temperature of the exotherm was higher in DMAC/MF than DAC/MF because of the higher reactivity of the DAC/MF system than DMAC/MF system. Curing started through the nucleophilic substitution reaction of the hydroxyl groups of DAC and DMAC with the MF. The lower reactivity of DMAC toward MF as compared to DAC could be attributed to an electronic effect. The methyl group (electron-

releasing group) attached to carbon increased the electron density at the carbon attached to a hydroxyl, which caused a reduction in its reactivity toward the MF; this was also responsible for the thermal polymerization of the acrylic end group.

The TGA thermograms of the cured films (Fig. 7) showed that the thermal degradation of the DMAC/MF and DAC/MF thermosets started at 270 and 310°C, respectively. The starting temperature of thermal degradation in the two thermosets was nearly the same. Both thermosets showed similar behavior versus temperature. Fifty percent weight loss occurred at 460 and 470°C, respectively. The complete decomposition of DAC/MF and DMAC/MF occurred above 600°C. From these observations, we concluded that coatings of DAC/MF and DMAC/MF showed high thermal resistivity, as they underwent almost no weight loss up to a temperature of 250°C.

Coating properties

The coatings of DAC/MF and DMAC/MF were applied on mild steel strips. The pH values of the coating

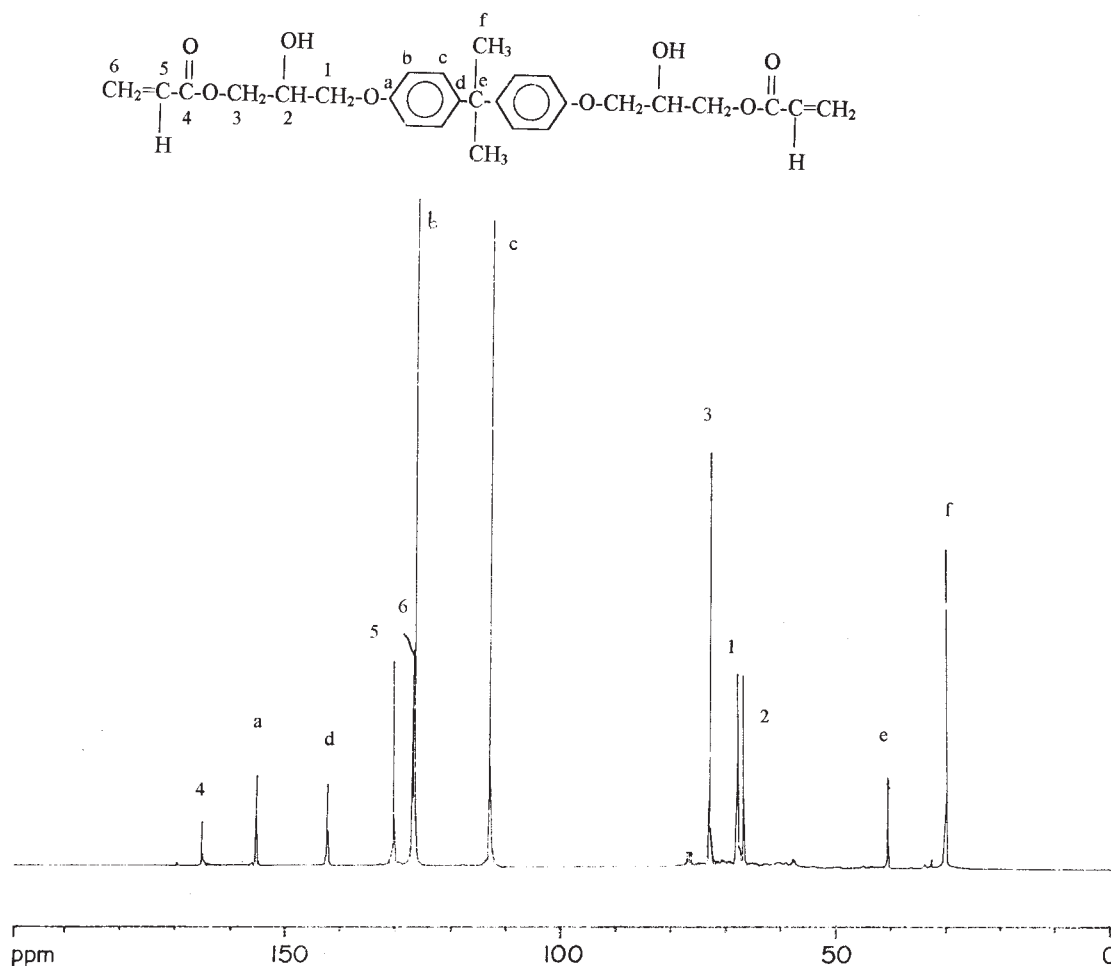


Figure 4 ^{13}C -NMR spectra of EAC.

solutions of the two systems were adjusted to 3.6 ± 0.3 with the help of phosphoric acid. It has been reported that use of the acid increases the curing rate and that the rate of initiation of the reaction is higher at lower pH values.²⁵ The selection of the acid is also important as it has significant influence on the corrosion protective ability of the coatings. The significant improvement brought about by phosphoric acid could be explained by the *in situ* phosphatizing mechanism.²⁶ Phosphoric acid can act as a metal phosphate film adhering to the metal, which is known to provide a corrosion protection barrier.^{26,27} The crosslinking reaction is considered to proceed by a nucleophilic substitution reaction of the hydroxyl group of DAC and DMAC with MF. The thermal polymerization of the acrylic end group further enhanced the toughness of the polymeric film.

On increasing the loading of MF, a progressive increase in scratch hardness was observed to a loading of 30 wt %, after which no significant improvement was observed. The coatings were made with 10, 20, 30, and 40 wt % MF in DAC and DMAC. As the loading of MF in the acrylate increased, almost all of the physi-

comechanical properties increased. We found (Table II) that DAC with 30 wt % MF (DAC/MF-30) showed the best physicomechanical properties. The coatings of this system showed a scratch hardness of 4.8 kg, an impact resistance of 150 lbs/in., and a gloss value of 64 at 45° and passed a bending test at a 7.1-mm diameter cone. Only the bending test was on the lower side, which indicated a less flexible system.

DMAC/MF coatings with 30 wt % MF (DMAC/MF-30), like DAC/MF-30, gave the best physicomechanical performance. It passed the scratch hardness test at 5.0 kg and the impact resistance test at 200 lb/in., had a gloss value of 70 at 45° , and passed the bending test at a 5.6-mm diameter cone. The coatings of this system, therefore, gave high mechanical performance and good accelerated weathering performance and also showed good resistance against attacks from 5 wt % HCl, 2 wt % H_2SO_4 , and 5 wt % NaOH. This data also showed that coatings of the DMAC/MF-30 resin showed better performance than those of the DAC/MF-30 resin.

The performances of the coated samples were tested in a humid and salty environment in a salt mist cham-

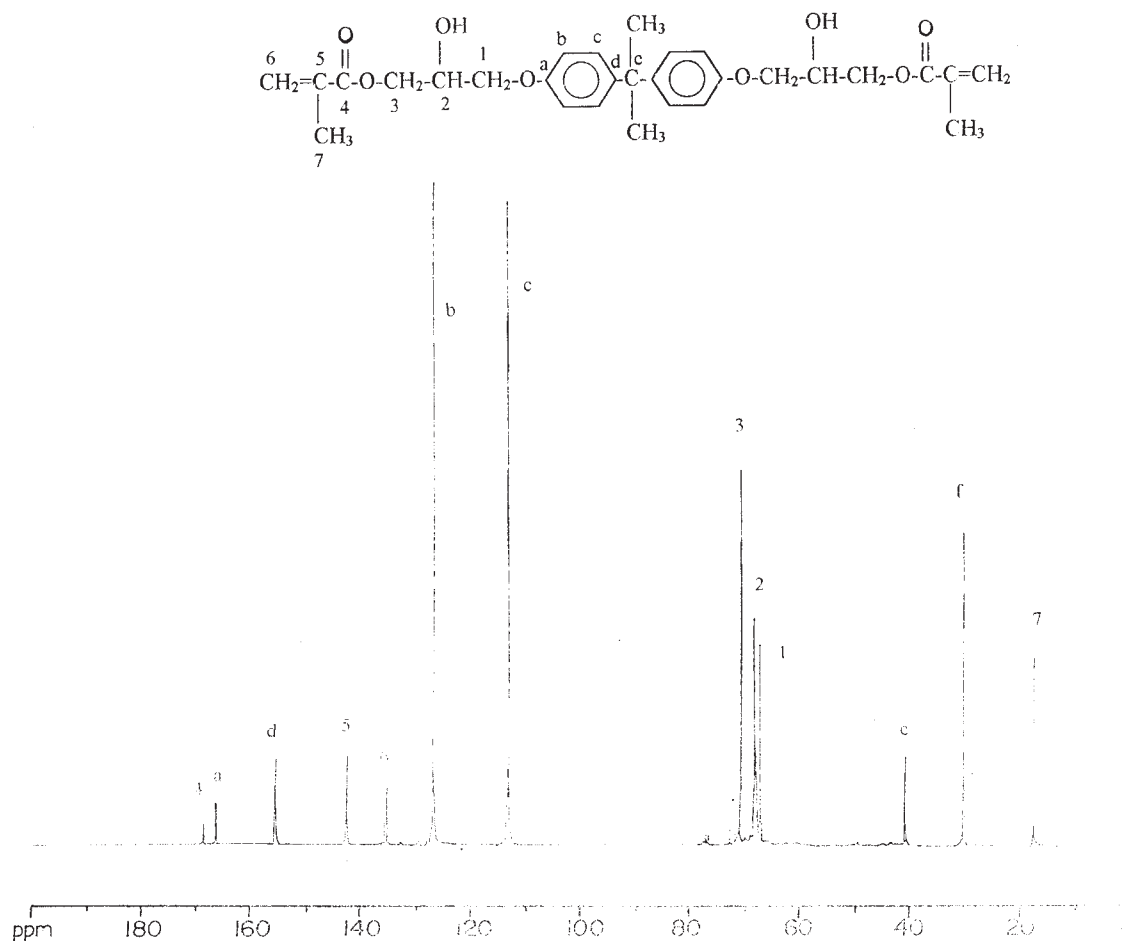


Figure 5 ^{13}C -NMR spectra of EMAC.

ber for 650 h and 228 h, respectively. No significant weight loss was noticed in either of the tests; only a slight loss in gloss was observed in the DAC/MF-30 system. This may have been due to an ingress of water molecules in the film of the DAC/MF system, whereas

in DMAC/MF, this effect was negligible because of the hydrophobic nature of the methyl group. It has been also reported that methacrylate has a poorer electron-withdrawing nature than acrylate.²⁸ The weathering resistance of the coated samples was ex-

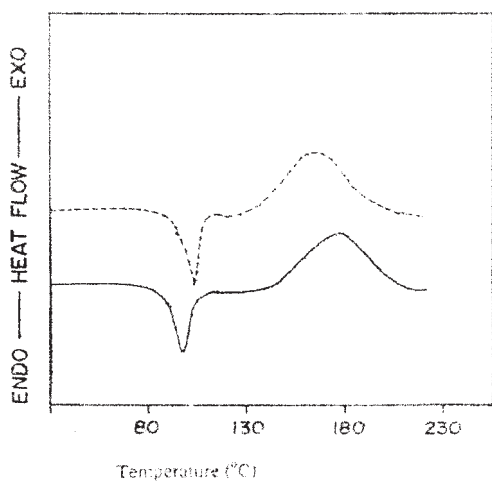


Figure 6 DSC of (· · ·) EAC/MF and (—) EMAC/MF.

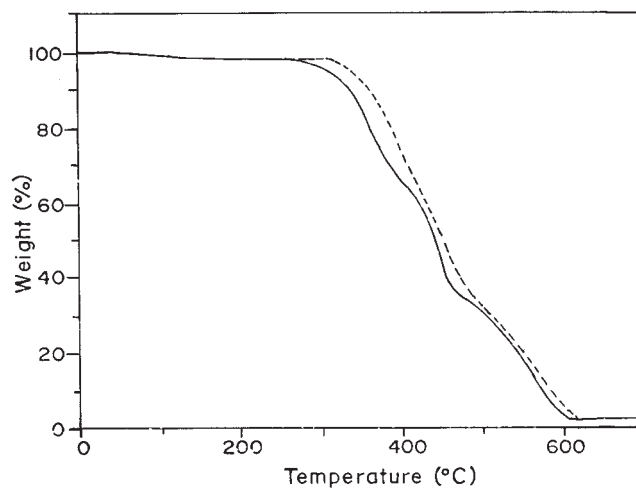


Figure 7 TGA of (· · ·) EAC/MF and (—) EMAC/MF.

aminated by exposure to UV radiation (wavelength = 285–315 nm) in high humidity conditions in an UV-accelerated weatherometer. DMAC/MF-30-coated samples showed no significant change and retained their gloss even after 17 h of continuous exposure. The DAC/MF-coated samples started to show yellowing in 14 h, and their gloss decreased on further exposure. However, this effect became visible in the DMAC/MF-coated samples in 18 h. The greater resistance of the DMAC/MF coatings to UV exposure may have been due to the nonavailability of tertiary hydrogen at the α position of the carbonyl group in DMAC. Coatings of DAC/MF and DMAC/MF were also tested for chemical resistance by soaking of the coated specimens in 5.0 wt % HCl, 2.0 wt % H₂SO₄, and 5.0 wt % NaOH solutions and distilled water (Table II). Coatings of DMAC/MF showed improved performance in solutions of acids and water because of the presence of a pendant methyl group. The presence of ester linkage in the polymer chain in DAC/MF coatings was considered to cause poor performance in the alkali solution. Coatings of DMAC/MF with ester linkages also showed similar susceptibility to alkali; however, they showed alkali resistance for a longer duration than those of DAC/MF. This was attributed to the presence of a pendant methyl group, which not only imparted better hydrophobicity but also shielded the ester linkages from hydrolysis.²⁹

CONCLUSIONS

DAC and DMAC obtained by the reaction of DGEBA with acrylic and methacrylic acid and curing by MF showed superior physicomechanical and anticorrosive properties, which conclusively overcame the brittleness of the pure epoxy resins. Also, the DAC/MF and DMAC/MF resin systems showed the highest values of impact resistance, scratch hardness, gloss, and related parameters for anticorrosive properties at 30 wt % MF in DAC and DMAC. Out of the DAC/MF and DMAC/MF, the latter showed the highest values of these parameters. These systems at 30 wt % MF

showed the optimum bending and flexibility characteristics.

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