

# Coemulsion and Electrodeposition Properties of Mixtures of Cationic Epoxy Resin and Cationic Acrylic Resin Containing Blocked-Isocyanate Groups

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## SYNOPSIS

2-Ethylhexanol-half-blocked-toluene diisocyanate (2EH-half-blocked TDI) was first reacted with 2-hydroxyethyl methacrylate (HEMA) to prepare HEMA-TDI-2EH monomer containing blocked-isocyanate groups. This monomer was reacted with butyl acrylate, styrene, and *N,N*-dimethylaminoethyl methacrylate to prepare an acrylic copolymer **III'** containing blocked-isocyanate groups and tertiary amine groups. The acrylic copolymer **III'** can be mixed with an epoxy-amine adduct **IV'**, acetic acid, and deionized (D.I.) water to form an electrodepositable coemulsion. The electrophoretic codeposition of the coemulsion and physical and chemical properties of the codeposited film were investigated. The resin composition of film deposited from coemulsion was determined by Fourier transform infrared (FTIR) quantitative analysis to study the coemulsion and electrophoretic codeposition behavior. The applicability of this two-component coemulsion in primer-surface (primer) electrodepositable paint was also discussed. The results indicate that the deposition yield of cationic acrylic copolymer **III** is greater than that of cationic epoxy resin **IV**, i.e., the deposition velocity of **III** is faster than that of **IV**. However, resins **III** and **IV** can be well codispersed in D.I. water to form stable coemulsions; thus, the resin composition of deposited is almost equal to the coemulsion resin composition. Moreover, the throwing power of coemulsion is almost equal to that of **IV** emulsion but greater than that of **III** emulsion. The optimum resin compositions of coemulsions for obtaining better gel content of deposited films are between 0.65 and 0.8 (**III/III + IV**, by weight). © 1994 John Wiley & Sons, Inc.

## INTRODUCTION

Electrodeposition has become increasingly important in the coating industry because it offers higher paint utilization, stronger throwing power, better outstanding corrosion protection, lower environmental contamination, definitely smoother coverage, and more susceptibility to the automatic process when compared with nonelectrophoretic coatings.

Since the Ford Motor Co. first commercially applied anionic electrodeposition, the process has been used almost universally in such highly valued coating operations as priming motor cars, electrical appliances, and many other commercial articles. The most frequently used cationic resins included acrylic

copolymers,<sup>1,2</sup> epoxy resins and their copolymers,<sup>3-6</sup> polyurethanes,<sup>7</sup> and polyimides.<sup>8-10</sup> Epoxy resins and their copolymers offer higher levels of corrosion resistance and, hence, have been adopted to provide the first or prime coat to a variety of products such as automotive bodies. Acrylic copolymers provide better weather resistance and outstanding gloss, suitable for applications as topcoats.

In our previous study,<sup>11</sup> two-component electrodepositable resins containing a cationic main resin and a nonionic cross-linker were prepared; in the mean time, the resin composition of their electrodeposited films was determined by FTIR quantitative analysis to investigate their electrophoretic behavior. The results indicate that under the conditions providing stable coemulsion, the resin composition deposited is almost equal to the coemulsion resin composition. However, the electrophoretic codeposition properties of mixtures of a

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cationic epoxy resin and a cationic acrylic resin are not well understood. In this study, a cationic acrylic resin containing blocked-isocyanate groups and a cationic epoxy resin were mixed and their coemulsion was electrodeposited to obtain a primer-surface paint providing higher levels of corrosion resistance and weather resistance. Furthermore, the composition of electrodeposited film was determined by FTIR to discuss the codeposition and coemulsion behavior.

## EXPERIMENTAL

### Materials

Monomers used in the preparation of cationic acrylic copolymer **III** including butyl acrylate, styrene, *N,N*-dimethylaminoethyl methacrylate, 2-hydroxyethyl methacrylate (HEMA, Wako first grade), 2-ethylhexanol (2EH, Wako extrapure grade), and toluene-2,4-diisocyanate (TDI).

Epon 1004 (diglycidyl ether of bisphenol A, commercially available from Shell Chemical Co. and possessing an epoxy equivalent weight of about 950), and diethylamine (Hanawa extrapure grade) were used in the preparation of epoxy-amine adduct **IV**. Ethylene glycol mono-*n*-butyl ether (butyl cello-solve, BCS) (Wako extrapure grade) and acetic acid were used in the preparation of the electrodeposition emulsion.

### Instruments

An electrodeposition apparatus<sup>12</sup> and a Jasco-7000 FTIR were employed. The electrodeposition apparatus included a glass reaction vessel with a cover in which the copper electrode (used as anode) and aluminum plate (cathode) with a surface area about 30 cm<sup>2</sup> were hung. Approximately 100 mL of emulsion was placed in the glass vessel of the electrodeposition apparatus. The electrodes were immersed in the emulsion and electrodeposition was carried out at 50 V for 3 min. After deposition, the aluminum plate was rinsed with distilled water and dried for 30 min in an air-circulating oven at 100°C, then reweighed to obtain the deposition yield of deposited film (mg/cm<sup>2</sup>).

### Synthesis

#### Cationic Acrylic Copolymer **III'**

In a three-necked reaction vessel, a half-blocked TDI (**I**) was prepared by slowly adding 2EH (26 g, 0.2

mol) to TDI (34.8 g, 0.2 mol) over a period of about 40 min below 10°C. At the completion of addition, the batch was kept below 20°C for an additional 40 min and then heated to 40°C and held for 1 h. HEMA (26 g, 0.2 mol) was then added into the flask. The whole mixture was kept at 40°C for about 2 h until no free isocyanate groups (at IR spectrum 2250 cm<sup>-1</sup>) were evident to give the monomer **II** containing blocked-TDI.

*n*-Butanol (50 g) was charged into another reactor equipped with a condenser, stirrer, thermometer, dropping funnel, and an inert nitrogen blanket. The monomer **II**, styrene (20.8 g, 0.2 mol), butyl acrylate (76.8 g, 0.6 mol), *N,N*-dimethylaminoethyl methacrylate (31.4 g, 0.2 mol), 50 g butanol, and 2,2'-azobisisobutyronitrile (AIBN) (3.24 g, about 1.5 wt % of the total amount of monomers) were well mixed in a flask. The reactor charged *n*-butanol was heated to 110°C, and then the whole mixture of monomers was added by means of a dropping funnel over a 3 h period. At the completion of addition, the reaction was allowed to continue at 110°C. During the reaction, the conversion and viscosity of the intermediate was tested at 1 h intervals. The conversion of the intermediate was determined by removing *n*-butanol and unreacted monomers from the sample at 80°C under reduced pressure. Over 9–10 h (including the time of adding monomers) was required to make the resultant product's conversion approach 100%. After the reaction, *n*-butanol was removed under reduced pressure to give an acrylic copolymer **III'** containing blocked-isocyanate and tertiary amine groups.

#### Epoxy-Amine Adduct **IV'**

An epoxy resin (Epon 1004) (250 g, 0.132 mol) and toluene (150 mL) were charged into a three-necked flask. The mixture was heated to 80°C to dissolve the epoxy resin. Diethylamine (19.25 g, 0.263 mol) was then added into the flask. The whole mixture was kept at that temperature for about 2–3 h until no free epoxy groups (at IR spectrum 910 cm<sup>-1</sup>) were evident to give the resin solution of the epoxy-amine adduct **IV'** in toluene. Finally, toluene was distilled off under reduced pressure. When cooled to room temperature, a golden, brittle solid of epoxy-amine adduct **IV'** was obtained.

#### Preparation of Electrodeposition Emulsion

The general process for the preparation of coemulsion of the **III'**/**IV'** mixture involved first the dilution of the mixed resins (**III'** 5 g, **IV'** 5 g) with 4

g BCS at 60°C to a solution with 70 wt % solid content, and the solution was treated with 0.45 mL HOAc (about 90 mol % of the total theoretical neutralization) and dispersed in 86 g D.I. water to form a coemulsion of 10 wt % solid content.

### Electrodeposition

Approximately 300 mL of emulsion was placed in the glass vessel of the electrodeposition apparatus. The electrodes were immersed in the emulsion and electrodeposition was carried out at 50 V for 3 min. The cathode was removed from the emulsion, washed with water, and successively heated to 100°C to obtain the deposition yield.

### FTIR Quantitative Analysis

#### Calibration Curve

III' and IV' resins with various weight ratios were mixed and well dissolved in THF, then coated on potassium bromide (KBr) windows and dried. The dried films were analyzed in the absorbance mode and at a resolution of 4 cm<sup>-1</sup> by signal averaging 16 scans. The absorbance areas of carbonyl group [C=O] (at approximately 1900–1650 cm<sup>-1</sup>) and hydrocarbon [CH] stretching vibration (at approximately 3000–2700 cm<sup>-1</sup>) were calculated and recorded. The [C=O]/[CH] absorbance area ratio (abs. ratio) were plotted against the III'/(III' + IV') weight ratios to derive the calibration curve.

#### Quantitative Analysis

The electrodeposited film was dissolved in THF applied to a KBr window and then dried. The sample was analyzed at the same testing conditions as described above to obtain the [C=O]/[CH] abs. ratio. From the [C=O]/[CH] abs. ratio and the calibration curve, we can obtain the resin composition of the electrodeposited film.

#### Gel Content

A 40 wt % solid content solution, prepared by dissolving and mixing III' and IV' in THF, was spread on aluminum foil and baked at 190°C for 40 min in an air oven to form a cross-linked film. The cross-linked film was immersed in acetone to reflux for 24 h. After drying, the gel content was calculated by dividing the residual weight by the initial weight of cross-linked film.

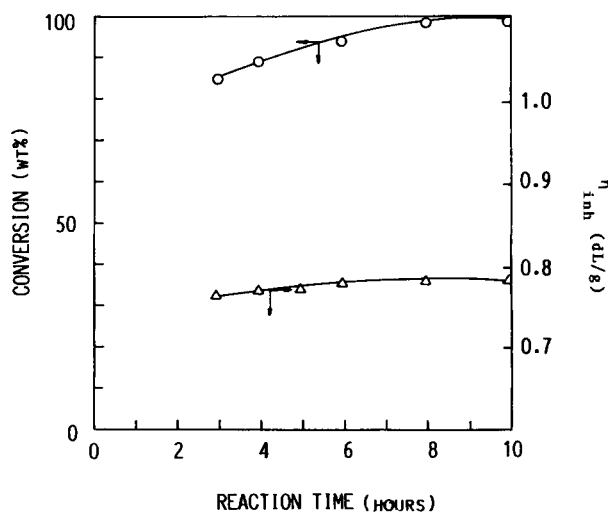
### Throwing Power and Rupture Voltage

The throwing power of emulsion was determined by the tube penetration test method. The apparatus and testing procedures of throwing power and rupture voltage were described in the previous publication.<sup>12</sup>

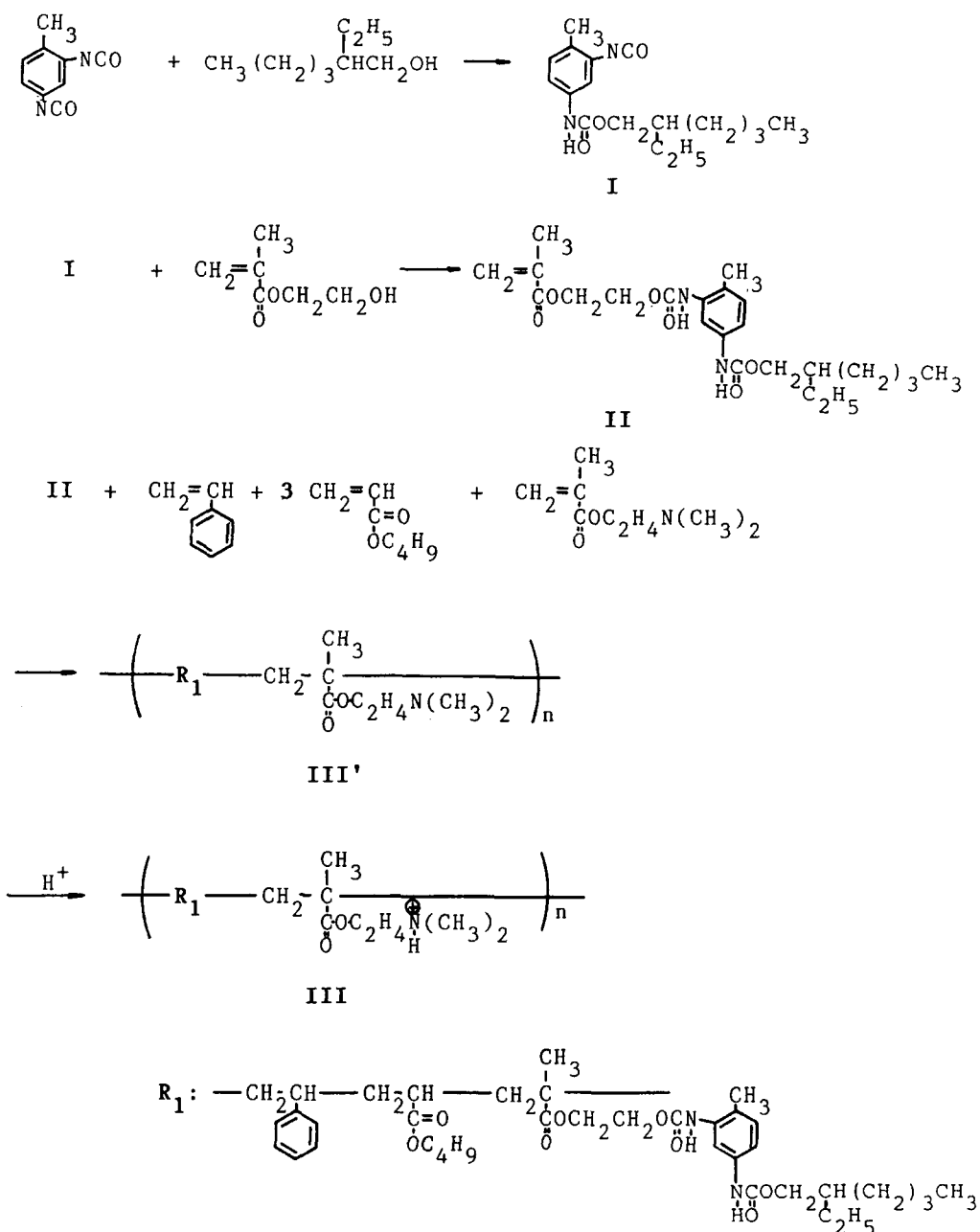
## RESULTS AND DISCUSSION

### Synthesis of Cationic Acrylic Copolymer III and Epoxy-Amine Adduct IV

The monomer II containing blocked-TDI was first prepared by reacting TDI with 2EH and HEMA. Then, monomer II, styrene, butyl acrylate, *N,N*-dimethylaminoethyl methacrylate, and AIBN were well mixed and charged into the reactor by a dropping funnel over a period of 3 h at 110°C to prepare an acrylic copolymer III'. During the reaction, the conversion and viscosity of the intermediate were tested at 1 h intervals. The effects of reaction time on viscosity and conversion of the intermediate are shown in Figure 1. Over 9–10 h was required to make the conversion approach 100%. The viscosity of the intermediate was increased by increasing the initial reaction time and reached a limit after 9 h. The acrylic copolymer III' and the epoxy-amine adduct IV' can be mixed with acetic acid to form the cationic acrylic resin III and the cationic epoxy resin IV. The synthetic procedures of cationic resins III and IV are shown in Scheme 1.



**Figure 1** Conversion and viscosity of acrylic copolymer vs. reaction time during copolymerization. Reaction temperature: 110°C; initial 3 h: adding time of monomers mixture.



Scheme 1

### Deposition Yields of "Resin III" and "Resin IV" Emulsions

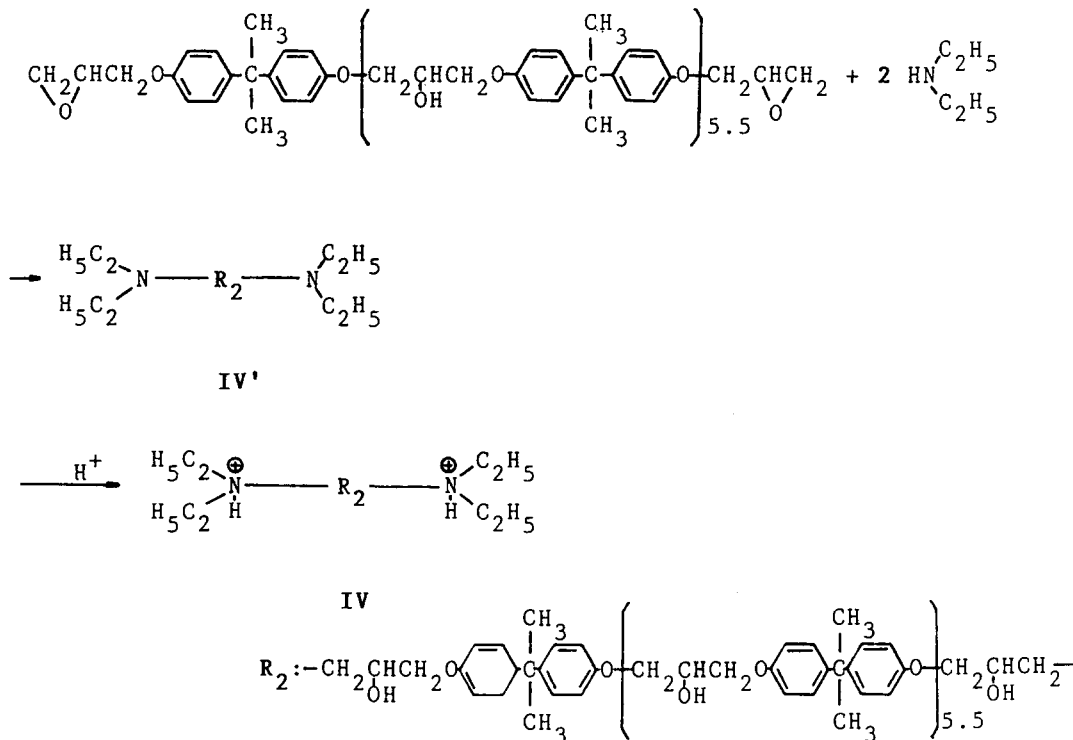
The relationships between deposition time, applied voltage, and deposition yield of resins **III** and **IV** are shown in Figure 2 and 3. For resin **IV**, the deposition yield increased with the deposition time in the initial 2–3 min and reached a limit when an insulating film formed. For resin **III**, the deposition yield cannot reach a limit within 4 min. From Figures 2 and 3, the deposition yield of **III** is greater

than that of **IV**, i.e., the deposition velocity of **III** is faster than that of **IV**.

### Properties of Resins III/IV Mixture

#### Gel Content

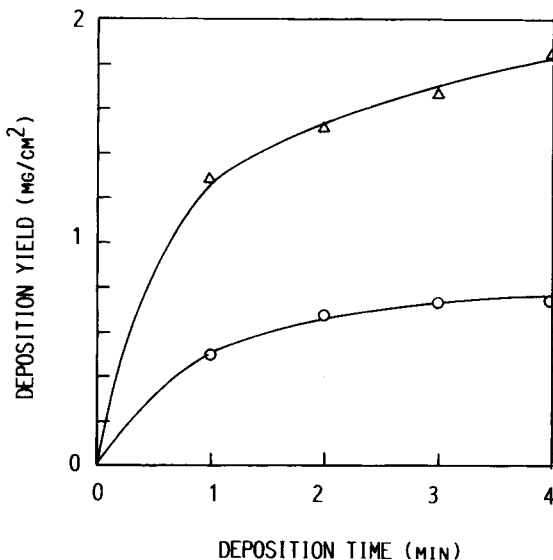
The **III**/**IV**' mixed film was cured by baking at 190°C for 40 min. The mechanism is the formation of urethane linkage between interpolymers by de-blocking (i.e., elimination of 2EH) the blocked-TDI



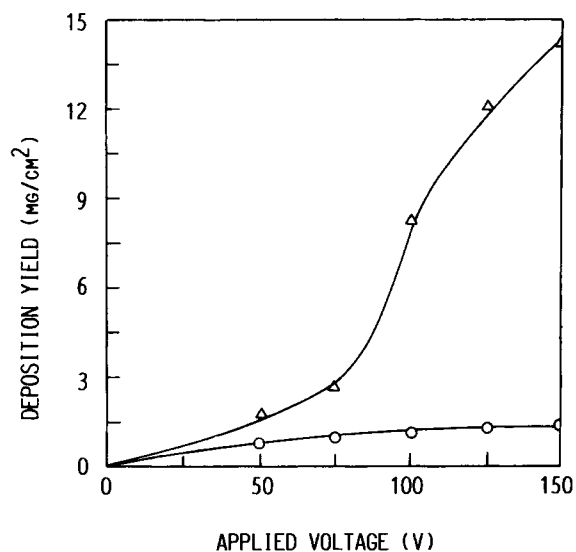
Scheme 1 (Continued)

groups of resin **III'** and subsequently being reacted with the hydroxyl groups pendant on the polymer **IV'** backbones. Figure 4 shows the effect of composition of mixed film on its gel content after ther-

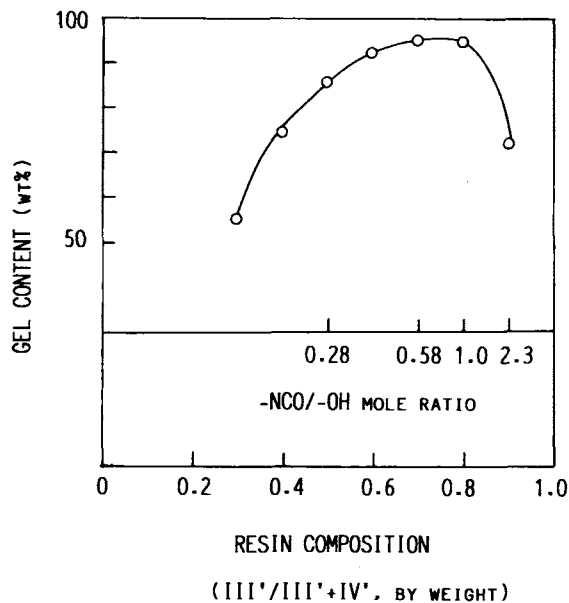
mal curing. The optimum composition (**III'/III'IV'**, by weight) of the mixed film is at 0.65–0.8 (i.e., the molar ratio of free —NCO and —OH during curing is at about 0.6–1.0). The result suggests that



**Figure 2** Effects of deposition time on deposition yield of resin **III** emulsion and resin **IV** emulsion. Resin concentration: 10 wt %; BCS content: 4 wt %; applied voltage: 50 V; ( $\Delta$ ) resin **III** emulsion, pH 4.94; ( $\circ$ ) resin **IV** emulsion, pH 5.28.



**Figure 3** Effects of applied voltage on deposition yields of resin **III** emulsion and resin **IV** emulsion. Resin concentration: 10 wt %; BCS content: 4 wt %; deposition time: 3 min; ( $\Delta$ ) resin **III** emulsion, pH 5.0; ( $\circ$ ) resin **IV** emulsion, pH 5.20.

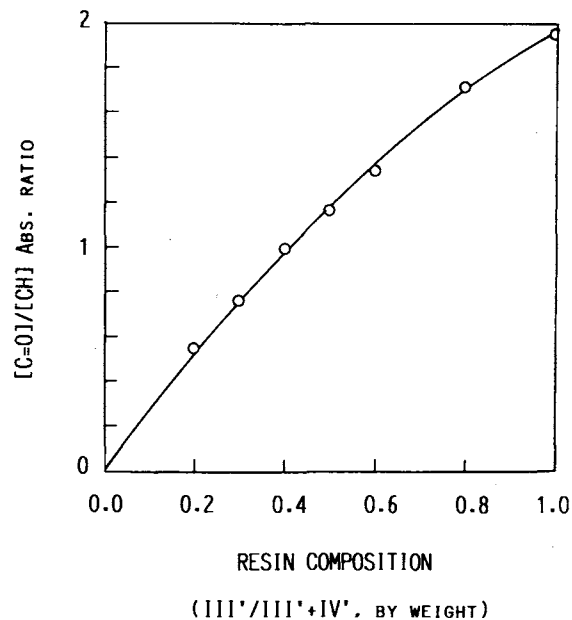


**Figure 4** Effects of resin composition on gel content. Curing temperature: 190°C; curing time: 40 min.

at the  $\text{—NCO/—OH}$  molar ratio of 0.6 the cured film would have adequate cross-linking density to obtain higher levels of gel content. Increasing the  $\text{—NCO/—OH}$  molar ratio increases cross-linking density and keeps the gel content more than 95%. However, when the  $\text{—NCO/—OH}$  molar ratio is greater than 1, the excess of resin **IV'** would be dissolved out in acetone and decreases the gel content.

#### Calibration Curve

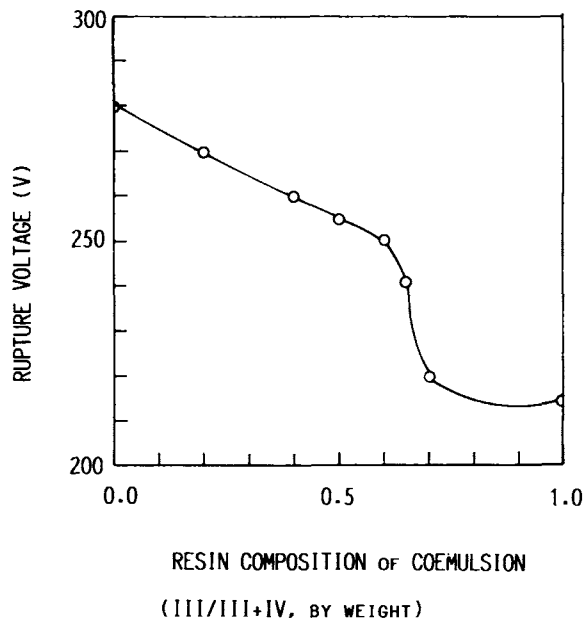
The resins of two-component emulsions containing the cationic acrylic resin **III'** have the IR absorption of carbonyl  $[\text{C}=\text{O}]$  and hydrocarbon  $[\text{CH}]$  groups and those containing the cationic epoxy resin **IV'** have  $[\text{CH}]$  absorption. The strong  $[\text{C}=\text{O}]$  peak appears at  $1700\text{ cm}^{-1}$  and the peak of  $[\text{CH}]$  stretching vibration appears at  $2900\text{ cm}^{-1}$ . Resins **III'** and **IV'** with various weight ratios were well mixed and dissolved in THF, then spread on KBr windows and dried. The samples were analyzed by IR spectroscopy. The absorbance areas of the  $[\text{C}=\text{O}]$  stretching band (at approximately  $1900\text{--}1650\text{ cm}^{-1}$ ) and the  $[\text{CH}]$  stretching vibration band (at approximately  $3000\text{--}2700\text{ cm}^{-1}$ ) were calculated. The  $[\text{C}=\text{O}]/[\text{CH}]$  abs. ratios were plotted against **III'**/**(III' + IV')** weight ratios to give the calibration curve, as shown in Figure 5. It shows that the  $[\text{C}=\text{O}]/[\text{CH}]$  abs. ratio increases with increasing resin **III'** content.



**Figure 5** Calibration curve of **III'**/**IV'** mixed film.

#### Rupture Voltage

Effects of coemulsion resin composition on rupture voltage are shown in Figure 6. The rupture voltages of resin **III** emulsion and resin **IV** emulsion are 215 and 280 V. From Figure 6, the rupture voltage is not proportional to the coemulsion resin composition.



**Figure 6** Effects of resin composition on rupture voltage. Resin concentration: 10 wt %; BCS content: 4 wt %.

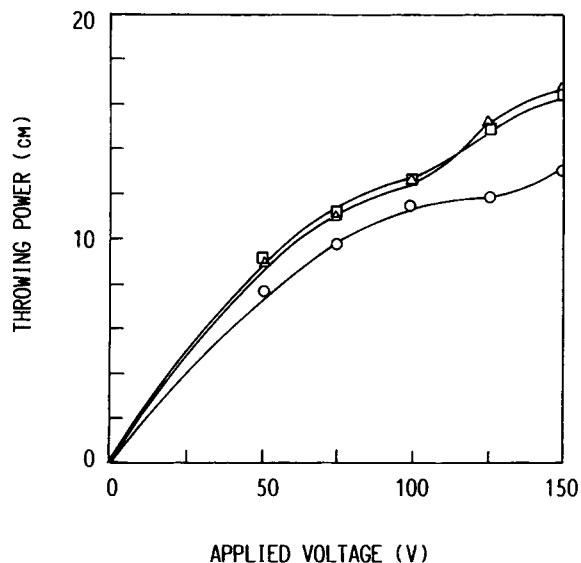
At the "III" composition higher than 40%, the rupture voltage of coemulsion can be kept at a higher level. Thus, the applied voltage can be improved by blending a proper amount of cationic epoxy resin into the acrylic resin.

### Throwing Power

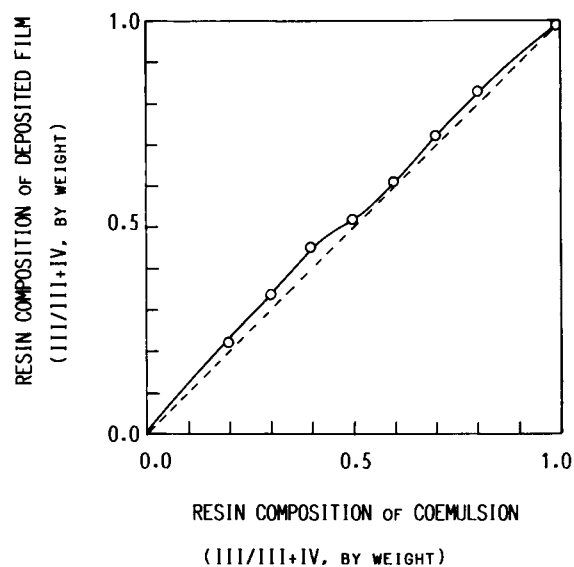
Effects of applied voltage on the throwing power of "III" and "IV" emulsions and "III/IV" coemulsion are shown in Figure 7. The throwing power has a tendency to increase with increasing voltage. This can be readily rationalized by taking into account the higher potential drop leading to a deeper penetration of the cationic resin into the shielded portions. The throwing power of emulsions are  $IV \text{ III}/IV > III$ . It might be explained by the fact that the throwing power of the codispersed emulsion was affected more significantly by cationic epoxy resin IV.

### Effects of Coemulsion Resin Composition on the Composition of Deposited Film

The effects of coemulsion resin composition on the resin composition of deposited film in a bath of 10 wt % of resin concentration are shown in Figure 8. The "III" content in deposited film increases with increasing "III" in the coemulsion. Moreover, the



**Figure 7** Effects of applied voltage on throwing power. Resin concentration: 10 wt %; BCS content: 4 wt %; deposition time: 2 min; (O) resin III emulsion, pH 5.0; ( $\Delta$ ) resin IV emulsion, pH = 5.28; ( $\square$ ) III/IV coemulsion, pH 5.18; III/(III + IV) weight ratio = 0.65.

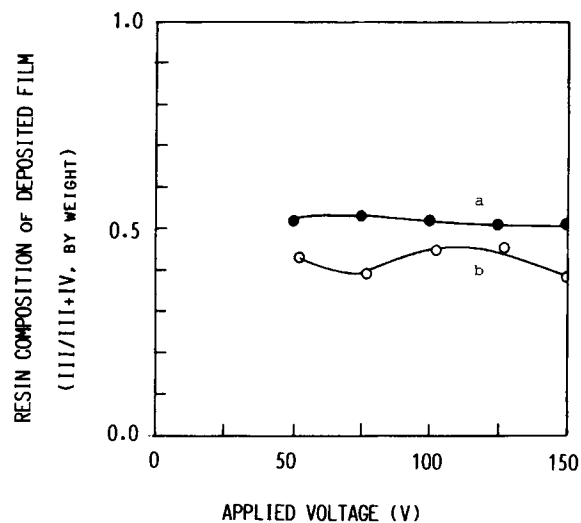


**Figure 8** Effects of coemulsion resin composition on resin composition of deposited film. Resin concentration: 10 wt %; BCS content: 4 wt %; pH 4.94–5.15.

"III" content in the deposited film is close to the value of ideal blending (the diagonal line of Fig. 8, assuming that the coemulsion resin composition equals the resin composition of deposited film). It indicates that at every coemulsion resin composition these two resins can be well blended to codisperse in D.I. water to form stable coemulsions. This may be due to the fact that the molecule backbone of acrylic resin III' is so flexible that it can easily entangle with epoxy resin IV' to give uniform and stable coemulsions. From Figures 8 and 4, to obtain the better thermal curing properties of deposited film, the resin composition must be kept between 0.65 and 0.8.

### Effects of Applied Voltage and the Emulsifying Method on the Resin Composition of Deposited Film

The effects of applied voltage and the emulsifying method on the resin composition of deposited film are shown in Figure 9. The codispersing of resins III and IV in D.I. water takes two forms: dispersing mixed III/IV resinous solution in D.I. water and mixing the aqueous dispersed III emulsion and the IV emulsion. At various voltages, the resin compositions of deposited films of the former are plotted as line "a" and those of the latter are plotted as line "b." At various voltages, resin compositions of deposited films of line "a" are almost the same and



**Figure 9** Effects of applied voltage on resin composition of deposited film. Resin concentration: 10 wt %; BCS content: 4 wt %; deposition time: 2 min; III/(III + IV) weight ratio = 0.5; (●) coemulsion prepared by dispersing mixed III/IV in D.I. water; (○) coemulsion prepared by mixing III emulsion and IV emulsion.

very close to the coemulsion resin composition; however, the values of line "b" are different from each other and lower than those of line "a." Nevertheless, coemulsions from the latter codispersion

were not stable. Some precipitation occurred with the latter coemulsions after 3–4 h. The results indicate that the coemulsions of line "a" can make two resins well entangled and form a stable codispersion, but the coemulsions of line "b" make two resins separated in the aqueous dispersion. Thus, the deposition velocities of III and IV and precipitation of coemulsion are the factors affecting resin composition of deposited film of line b.

### Physical and Chemical Properties of Cured Codeposited Films

Physical and chemical properties of the cured deposited films are shown in Table I. All cured deposited films showed excellent adhesive strength, good hardness up to 1–3 H, and high acidic and alkali resistance. However, the cured deposited film derived from cationic acrylic resin alone showed less salt-spray resistance and the cured film derived from the cationic epoxy resin alone appeared less weather-resistant when both were compared to those derived from the mixture of cationic acrylic resin III and cationic epoxy IV. The results indicate that the coemulsion derived from the mixture of cationic acrylic resin III and cationic epoxy resin IV can give a superior primer–surface coating with higher levels of corrosion and weather resistance.

**Table I** Physical and Chemical Properties of the Cured Deposited Films

Properties	1 <sup>a</sup>	2 <sup>b</sup>	3 <sup>c</sup>	4 <sup>d</sup>
Adhesive strength <sup>e</sup>	100/100	100/100	100/100	100/100
Pencil hardness (H) <sup>f</sup>	1	3	2	2
Salt-spray resistance <sup>g</sup>	> 3 mm	< 1 mm	< 1 mm	< 1 mm
Alkali resistance <sup>h</sup>	99.0	99.2	99.6	99.2
Acidic resistance <sup>i</sup>	99.3	99.1	99.3	99.4
Weather resistance <sup>j</sup>	○	△	○	○

<sup>a</sup> A cationic acrylic resin, which was prepared by the copolymerization of butyl acrylate (BA), styrene (ST), *N,N*-dimethylaminoethyl methacrylate (DMAEMA), and *N*-(*n*-butoxymethyl)acrylamide (NBMA), and the reaction molar ratio of BA/ST/DMAEMA/NBMA was 3/1/1/1.

<sup>b</sup> A self-curable cationic Epon 1004–diethylamine adduct with pendant 2-ethylhexanol-blocked isocyanate groups.

<sup>c</sup> Mixture of cationic III and IV; resin composition of coemulsion: III/III + IV weight ratio = 0.6.

<sup>d</sup> Mixture of cationic III and IV; resin composition of coemulsion: III/III + IV weight ratio = 0.7.

<sup>e</sup> Determined by the standard method of JIS D-0202 (1 mm<sup>2</sup> × 100).

<sup>f</sup> Measured with Mitsubishi pencils by using a Bravive-Instruments B-4000.

<sup>g</sup> Evaluated by the standard method of JIS Z-2371, sprayed with 5% aqueous NaOH for 240 h.

<sup>h</sup> Tested by dipping in 0.1*N* aqueous NaOH for 240 h; expressed as the weight percentage of the weight of cured film after dipping corresponding to the one before dipping.

<sup>i</sup> Tested by dipping in 0.2*N* aqueous H<sub>2</sub>SO<sub>4</sub> for 48 h; the value recorded as the wt % is described in the preceding footnote.

<sup>j</sup> The level of weather resistance was judged by the gloss maintenance percentage of film after 500 h UV radiation corresponding to the one before radiation. The gloss of film was tested by the ProGloss at 85° (Models Pro-3, supplied by Hunter Associates Lab, Inc.). When the gloss maintenance % were > 90%, 50–89%, and < 49%, the level of weather resistance were presented as ○, △, and X.



## CONCLUSIONS

1. At the same conditions, the deposition yield of cationic acrylic resin **III** is greater than that of cationic epoxy resin **IV**, i.e., the deposition velocity of **III** is faster than that of **IV**.
2. The mixed resins **III** and **IV** at various weight ratios can all blend well and codisperse in D.I. water to form stable coemulsions; thus, the resin compositions of electrodeposited films are close to the coemulsion resin compositions.
3. To obtain better thermal curing properties of deposited film, the coemulsion resin composition (**III/III + IV**, by weight) must be kept between 0.65 and 0.8.
4. The throwing power of the codispersed emulsion is affected more significantly by cationic resin **IV**; thus, the order of the throwing power of emulsions is  $\text{IV} = \text{III/IV} > \text{III}$ .
5. The coemulsion derived from the mixture of cationic acrylic resin **III** and cationic epoxy resin **IV** can give a superior primer-surface coating with higher levels of corrosion and weather resistance.

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