

Preparation and Properties of UV-Autocurable BTDA-Based Epoxy-Multiacrylate Resins. Effects of the Degree of Polymerization and the Epoxy Type

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SYNOPSIS

A series of UV-autocurable epoxy-multiacrylate resins was synthesized, and the effects of degree of polymerization (DP) and epoxy type on their properties were investigated. These autocurable multiacrylate resins possess good pot life and are cured rapidly when exposed to ultraviolet (UV) without the addition of photoinitiator or photosensitizer. The curing rate of the autocurable resins was probably dependent on the number of abstractable hydrogen in epoxy resins. Stress-strain, differential scanning calorimetry (DSC), and dynamic mechanical analysis (DMA) were used to characterize the properties of cured multiacrylate resins. Increased crosslinking density of cured films improved tensile properties. Increasing the molar ratio of epoxy resin in the multiacrylate resins was found to decrease the effective acrylate concentration of resins and to depress crosslinking density of cured resins, which also resulted in an increased elongation at break but a decreased Young's modulus and breaking strength. Furthermore, the different structures of epoxy resins were used to give wide range properties of cured epoxy-multiacrylate resins with a glass transition temperature (T_g) range from 74 to 102°C. The film properties of the multiacrylate resins coated on steel plates were also investigated.

INTRODUCTION

During the last 20 years, an ever-growing number of resins that polymerize readily under UV irradiations, especially the reactive multifunctional acrylates (MFAs),¹⁻⁴ have appeared on the market when the advantages⁵⁻⁷ of this new technology became apparent. Besides the faster cure and lower energy consumption, as compared to conventional surface coating, the major reason for the commercial breakthrough of UV curing is its cost-effectiveness, together with space saving, reduced pollution, and product quality. Since most of the acrylic monomers and oligomers do not produce initiating species with a sufficiently high yield upon UV exposure, it is necessary to introduce a photoinitiator that will initiate the polymerization. A typical UV-curable for-

mulation,^{8,9} therefore, will contain two basic components: (1) a photoinitiator¹⁰ that produces initiating species with a high efficiency and (2) a monomer and an oligomer^{11,12} bearing at least two unsaturations that generate the polymer network.

Curing of acrylic oligomers by UV radiation is increasing in industrial importance, in particular, for coating of wood, paper, plastics, and tin plate and for curing ink. Different types of chemical structures can be used for the prepolymer backbone, most notably polyurethanes, epoxy derivatives, polyesters, and polyethers. The relationship of film properties and type of oligomer is relatively well understood. For instance, a bisphenol A type epoxy acrylate oligomer¹³ is known to give a hard film with excellent adhesion to metal substrates, whereas an aliphatic urethane acrylate oligomer^{14,15} is anticipated to give a tough and flexible film with good weatherability. Therefore, the molecular design of radiation-sensitive oligomers has been a major concern in recent years.

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To obtain UV resins with fast and efficient curing, relatively large amounts of the photoinitiator must be included within the system. In most cases such a high level of photoinitiator will not dissolve or mix homogeneously with the remainder of the system. Even in the systems wherein these large amounts of photoinitiators are compatible, most of the initiator will remain chemically unbonded in the final cured composition and produce deleterious effects on the properties of cured materials. To overcome these disadvantages, many patents¹⁶⁻¹⁹ related to acrylic oligomers and monomers having built-in photoinitiators, which are autopolymerizable through UV radiation, have been reported.

Recently, we reported the syntheses of UV-autocurable polyurethane and polyester multiacrylate oligomers²⁰⁻²² and the effects of chemical structure, molecular weight, and acrylic functionality on their properties. The relationships between the structure and properties of cured films, obtained by UV polymerization of a semicrystalline urethane-multiacrylate oligomer,²³ were also described. All these autocurable oligomers possessed good pot life and were cured readily by UV in the absence of photoinitiator.

In this study a series of UV-autocurable epoxy-multiacrylate resins, whose structures are varied by changing the degree of polymerization of resins and the chemical structure of epoxy resins, is synthesized. The effects of epoxy type and degree of poly-

merization on their curing rate and properties are investigated. Finally, the properties of cured films coated on steel plates are also described.

EXPERIMENTAL

Materials

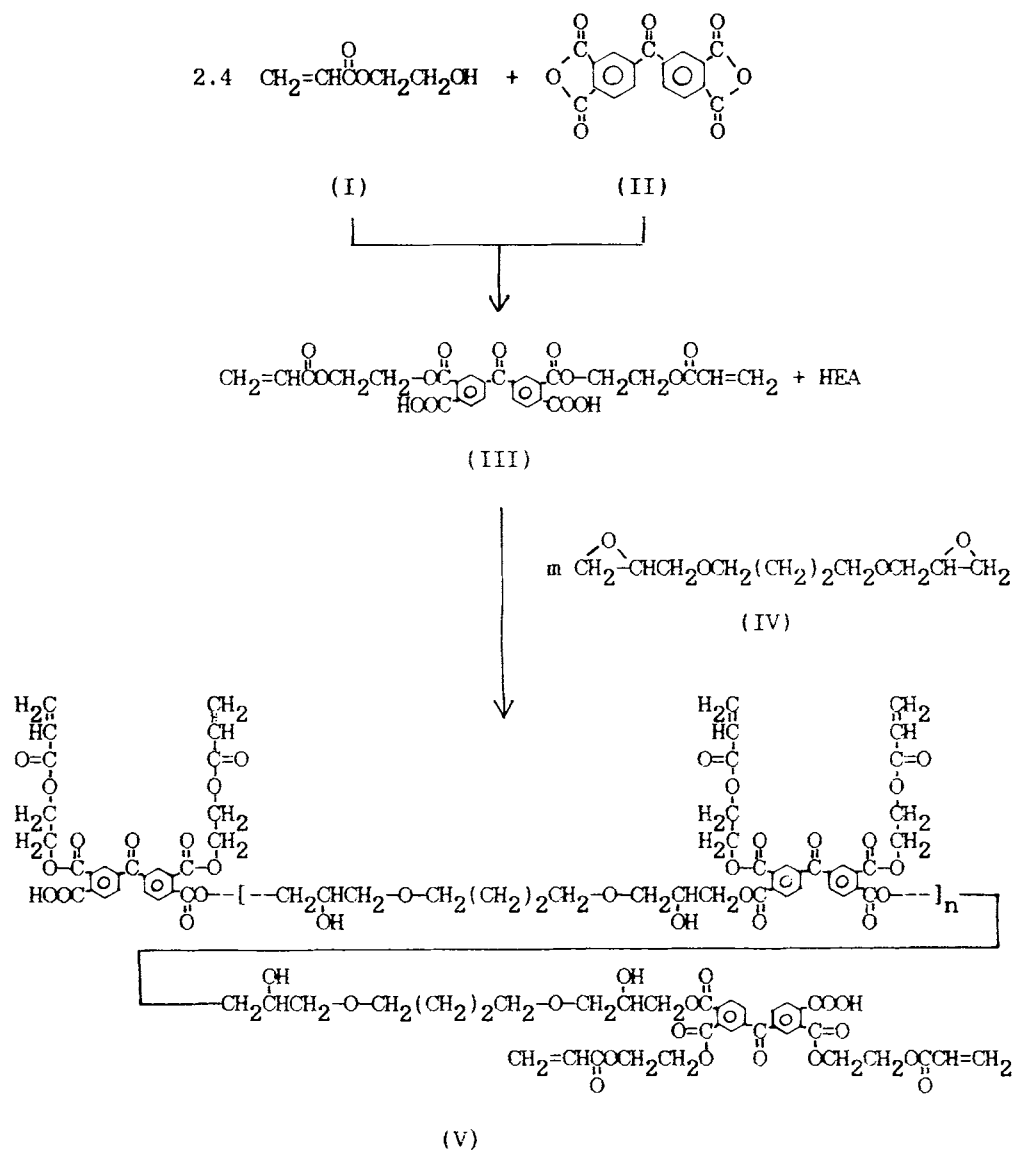
3,3', 4,4'-Benzophenone tetracarboxylic dianhydride (BTDA) and 2-hydroxyethyl acrylate (HEA) were extra pure grade from Tokyo Kasei, Japan. A list of epoxy resins used in this study is given in Table I.

Syntheses of UV-Autocurable Epoxy-Multiacrylate Resins

The autocurable epoxy-multiacrylate resins (V) were synthesized as shown in Scheme 1. 0.2 mol of BTDA and 0.48 mol of HEA were charged into a 300-mL four-neck flask. A magnetic stirrer was used to achieve good mixing in the reaction flask. A small amount of thermal inhibitor (0.6% hydroquinone based on the weight of reactants) was added into the reaction mixture, and the temperature was raised to 95°C. A dry nitrogen purge was added to assure a moisture-free environment. The reaction was kept at 95°C for 4 h to obtain half ester (BTDA-HEA) intermediate containing pendant acid groups. After

Table I The Structure and Properties of Various Epoxy Resins

	Chemical Type	Structure	Viscosity (cps at 25°C)	Weight per Epoxide	Supplier
WC67	Diglycidyl ether of 1,4-butanediol		14-18	120-130	Wilmington Chem. Corporation
WC68	Diglycidyl ether of neopentyl glycol		14-18	130-140	Wilmington Chem.
WC69	Diglycidyl ether of resorcinol		300-500	120-135	Wilmington Chem.
MK107	Diglycidyl ether of cyclohexane dimethanol		55-75	155-165	Wilmington Chem.
EP828	Diglycidyl ether of bisphenol -A		9000-14000	182-194	Shell Chem. Corporation



Scheme 1 Syntheses of autocurable epoxy-multiacrylate resins: I, 2-Hydroxyethyl acrylate (HEA); II, BTDA; III, Half ester intermediate (BTDA-HEA); IV, Epoxy resin (CW67, diglycidyl ether of 1,4-butanediol); V, Autocurable epoxy-multiacrylate resin (CW67-MFAs).

the reaction mixture was cooled to 60°C, 0.1 mol of epoxy resin (such as diglycidyl ether of 1,4-butanediol) and 1.5 wt % of catalyst were added. The reaction mixture was then heated to 105°C for 3 h to complete the ring-opening reaction of epoxide and acid groups. Caution was exercised to avoid thermal polymerization of acrylate groups by keeping temperatures between 100 and 105°C.

The chemical structures of the epoxy-multiacrylate resins were varied by changing the types and the reaction moles of epoxy resin. All epoxy resins used in this study are listed in Table I.

MEASUREMENT OF VARIOUS PROPERTIES

Characterization methods and curing process were performed according to a previous study.²⁰ The viscosity of resins was measured at 25°C using a Brookfield viscometer. The molecular weight of multiacrylate resins was determined by gel permeation chromatography (GPC). The DuPont 910 differential scanning calorimeter (DSC) was used to investigate exotherm of crosslinking reactions and to obtain initial curing temperature. The initial

curing temperature is taken as the onset of the exothermic reaction.

Prepared epoxy-multiacrylate resins were cast between two Mylar films and irradiated by a high-pressure mercury lamp (Model USH-500D manufactured by Ushio Electric Co., Japan) to obtain cured films of about 0.2 mm in thickness. Tensile testing was run at 25°C using an Instron Testing Machine (Model 1130) with a crosshead speed of 5 cm/min and a gauge length of 20 mm. Dynamic mechanical data were collected in the resonant model from the DuPont 9900 data station connected to 983 Dynamic Mechanical Analyzer.

Coating Properties

Hardness

Pencil hardness was determined as a measure of surface hardness according to JIS K 5651.

Flexibility

Flexibility was assessed by the indenter. A 2-mm ϕ diameter of mandrel was used to fall on the wedge-shaped specimen in order to examine visually the cracking caused.

Impact Resistance

The impact resistance of a paint film was assessed by using DuPont Impact Tester. A rounded block was allowed to fall from a 500-mm height on the specimen. Visual examination was made for the surface damage.

Extensibility

The resistance of a surface coating to deformation stress was tested by using an Erichsen Extensibility Tester. A ball-ended rod (5 mm ϕ) was pressed into the uncoated side of a painted test panel to a 4-mm depth and was visually examined to find out whether there was any surface crack or not.

Adhesion

The adhesion was determined by using the cellophane tape cross-cut test (10 lines by 10 lines cut in 1 cm²) at 25°C.

Chemical Resistance

HCl, NaOH, and NaCl were each diluted to 5% while acetic acid was diluted to 3%. The specimen with an engraved cross was immersed in each solution

for 72 h to explore whether stripping around the cross took place.

RESULTS AND DISCUSSION

Preparation and Properties of UV-Autocurable Epoxy-Multiacrylate Resins

Scheme 1 summarizes the synthesis reaction of UV-autocurable epoxy-multiacrylate resin and Figure 1 shows the infrared (IR) spectra of epoxy-multiacrylate resin and its preparation intermediates. The half ester intermediate (BTDA-HEA) with pendant acid group was the addition reaction product of benzophenone tetracarboxylic dianhydride (BTDA) and hydroxyethyl acrylate (HEA) with the molar ratio of 1 : 2.4. The IR spectrum of product mixture showed that the anhydride characteristic absorption peaks of BTDA at 1860 and 1760 cm⁻¹ completely disappeared, indicating the reaction was completed. The acid value of the BTDA-HEA mixture was 138.7 mg KOH/g and the viscosity at 25°C was 12

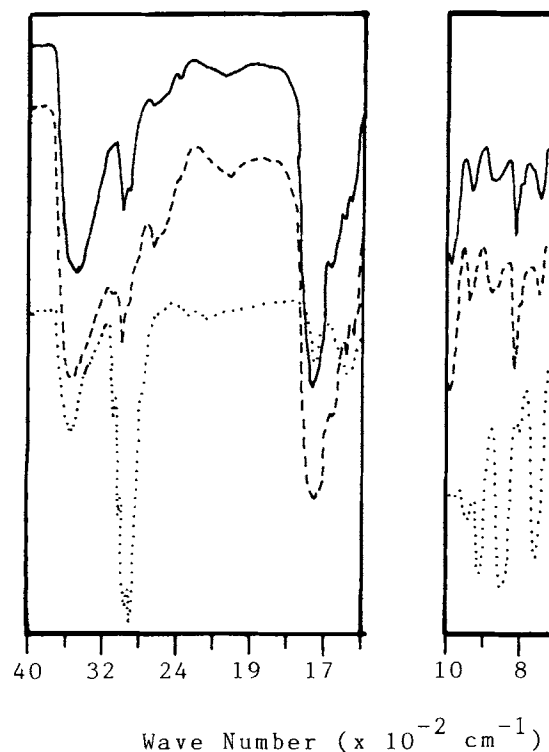


Figure 1 IR spectra of UV-autocurable epoxy-multiacrylate resin (WC67-MFAs, —), half ester intermediate (BTDA-HEA, ---), and diglycidyl ether of 1,4-butane-diol (WC67, ····).

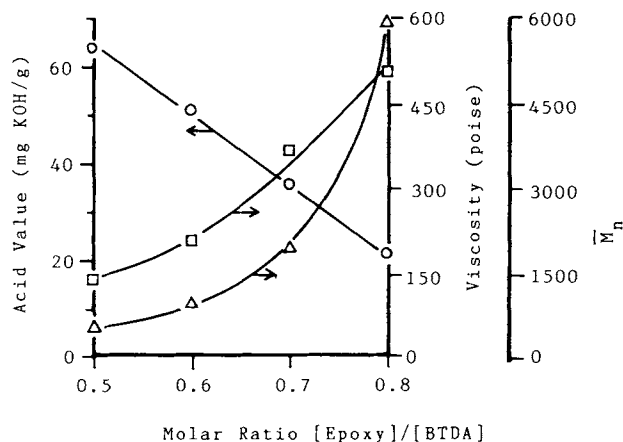


Figure 2 The molecular weight, viscosity, and acid value of multiacrylate resins (WC67-MFAs) as function of the molar ratio of epoxy/BTDA. (○) acid value, (△) viscosity, (□) molecular weight.

P. Then, the half-equimolar epoxy resin (WC-67) based on the mole of BTDA was added into the BTDA-HEA mixture to synthesize the UV-autocurable epoxy-multiacrylate resin (WC67-MFAs) via the ring-opening reaction of epoxide and acid group. The excess of HEA monomer played the role of reactive diluent during the reaction without isolation. After the reaction time of 3 h at 105°C in the presence of triethylbenzylammonium chloride as a catalyst, the IR spectrum of multiacrylate resin showed that the absorption peak of epoxide group at 910 cm^{-1} completely disappeared. The acid value of resin was reduced to 61.7 mg KOH/g and the viscosity at 25°C was increased to 56.5 P. From these phenomena it is evident that the reaction of epoxide and acid group was completed and the epoxy-mul-

tiacrylate resin with number-average molecular weight of 1425 was obtained.

In order to obtain a series of autocurable epoxy-multiacrylate resins (WC67-MFAs) with a different degree of polymerization (DP), a change in the molar ratio of epoxy resin to BTDA was carried out. As shown in Figure 2, increasing the molar ratio from 0.5 to 0.8 results in an increase in the viscosity at 25°C from 56.5 to 595 P and number-average molecular weight from 1425 to 5000, but a decrease in acid value from 61.7 to 22.1 mg KOH/g.

Several epoxy-multiacrylate resins with a different epoxy type were also obtained by using different epoxy resins in the molar ratio of epoxy resin to BTDA at 0.7. The chemical structures of various epoxy resins are collected in Table I, and the fundamental properties of corresponding epoxy-multiacrylate resins are listed in Table II.

Influence of the Degree of Polymerization on Pot Life and Curing Rate of Epoxy-Multiacrylate Resins

Autocurable resins must be activated and cured rapidly by UV radiation. At the same time they also must be inactive at ambient temperature during storage and handling. To evaluate their thermal stability, differential scanning calorimetry (DSC) was used to study the thermal curing reaction of those multiacrylate resins. As shown in Figure 3, all initial curing temperatures of resins are above 220°C as heat is liberated by crosslinking reaction. Therefore, autocurable epoxy-multiacrylate resins prepared by this work possess good storage and handling stability at ambient temperature.

The epoxy-multiacrylate resins were cured directly by UV irradiation without addition of any

Table II The Effect of Epoxy Type on Fundamental Properties of Autocurable Epoxy-Multiacrylates

Properties	WC68	WC69	MK107	EP828
\bar{M}_n	3615	3605	3660	3990
\bar{M}_w	8675	8110	8970	9975
\bar{M}_w/\bar{M}_n (MWD)	2.40	2.25	2.45	2.50
η (cps) ^a	12	33	22	56
Acid value (mg KOH/g)	36.8	33.7	29.5	28.7
Solubility	(Dissolves in acetone, chloroform, dioxane, THF, DMF, DMSO, and DMAc; insoluble in ether, hexane and ethyl acetate)			

^a Viscosity is measured in a viscous of 50% oligomer and 50% acetone.

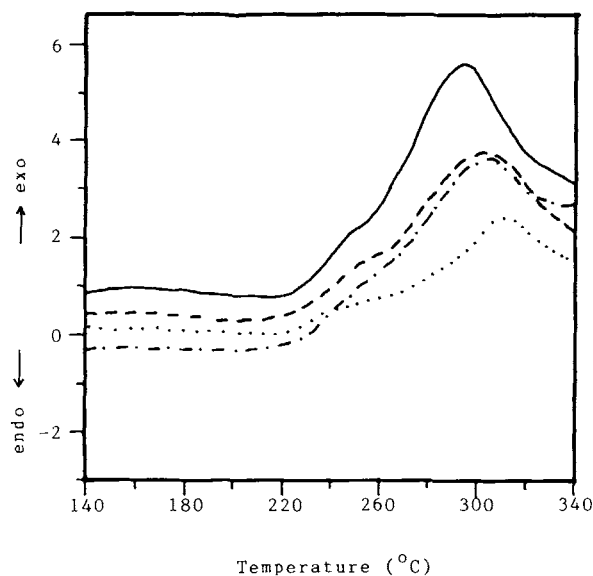


Figure 3 DSC thermograms of WC67-MFAs resins with various molar ratios of epoxy/BTDA. molar ratio: (—) 0.5; (---) 0.6; (- · - · -) 0.7; (· · · · ·) 0.8.

photoinitiators. The curing process was carried through a transparent Mylar film to prevent the inhibition of oxygen on the curing reaction. As shown in Figure 4, the curing reactions of all multiacrylate resins were completed by the irradiation time of 10 s and their gel contents were above 80%. When effective acrylate concentration in the multiacrylate resin was reduced by increasing the molar ratio of epoxy resin to BTDA from 0.5 to 0.8, it would have been expected that a proportionate decrease in cure

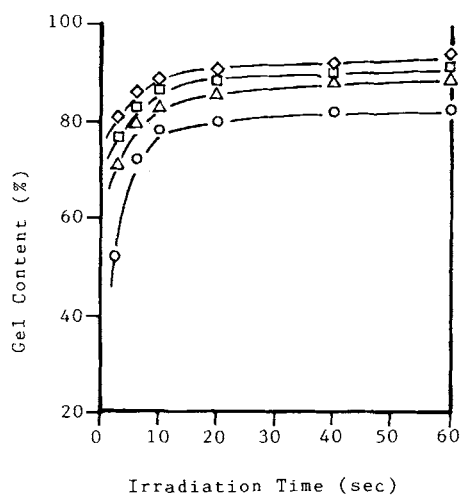


Figure 4 Gel content of WC67-MFAs resins with various molar ratios of epoxy/BTDA for UV process, molar ratio: (○) 0.5; (△) 0.6; (□) 0.7; (◇) 0.8.

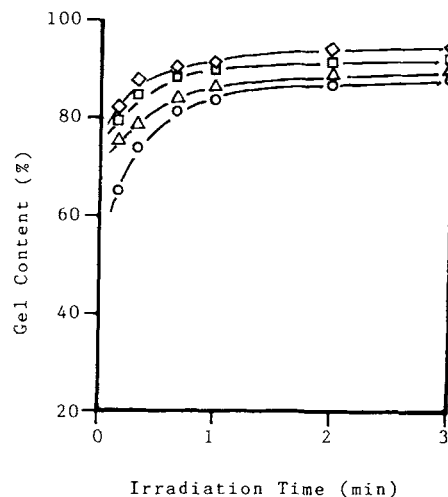


Figure 5 Gel content of WC67-MFAs resins with various molar ratios of epoxy/BTDA for sunlight process, molar ratio: (○) 0.5; (△) 0.6; (□) 0.7; (◇) 0.8.

speed should have occurred. On the contrary, the curing rate was increased steadily as the molar ratio of epoxy was increased.

A plausible explanation may be obtained from photopolymer theory and the efficiency of hydrogen abstraction from activated photosensitizers. Studies²⁴ on benzophenone/hydrogen donor systems have indicated that particularly reactive hydrogen donors are those in which the active hydrogen is positioned alpha to a nitrogen or oxygen, as in amines, ethers, or alcohols. Diglycidyl ether of 1,4-butanediol possesses four active hydrogen donor

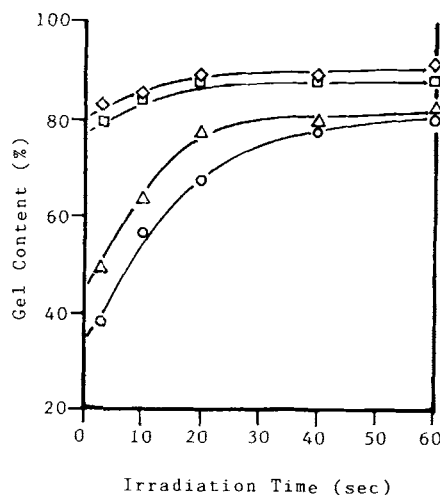


Figure 6 Gel content of epoxy-multiacrylate resins with various epoxy type for UV process, epoxy resin: (○) EP828; (△) WC69; (□) WC68; (◇) MK107.

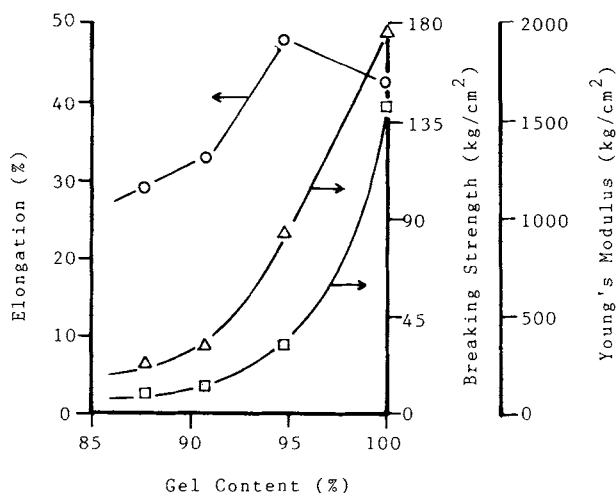


Figure 7 The relationship between gel content and tensile properties of cured WC67-MFAs resins. (○) elongation at break; (△) breaking strength; (□) Young's modulus.

sites per mole. Therefore, the enhanced reactivity by an increase in the molar ratio of epoxy resin to BTDA is probably due to the formation of more radicals in epoxy-multiacrylate resin and subsequent direct addition polymerization and/or chain transfer polymerization across acrylate double bonds of the HEA monomer. Another explanation may be attributed to probability since an increase in degree of polymerization leads to an increase in acrylic functionality of each prepolymer chain and the op-

portunity of crosslinking taking place in the UV curing reaction.

All autocurable epoxy-multiacrylate resins were also very sensitive to sunlight in the oxygen-free environment. As shown in Figure 5, the curing reaction of resins was completed by irradiation time of 1 m, and their gel content reached more than 90% after irradiation time of 3 m in sunlight. Moreover, the cured films of gel content above 99% could be obtained by prolonging the irradiation time for sunlight to 30 min. Therefore, the curing rate of resin irradiated by sunlight also increased with increasing the degree of polymerization of resin.

As shown in Figure 6, the UV curing rate of epoxy-multiacrylate resins with a comparable degree of polymerization was affected by the chemical structure of epoxy resin. On a relative reactivity basis, the following order (fastest to slowest) was observed: MK107-MFAs > WC68-MFAs > WC69-MFAs > EP828-MFAs. This may possibly be due to the greater number of abstractable hydrogens in diglycidyl ethers of neopentyl glycol and cyclohexane dimethanol, and steric effects of bulky phenyl group in diglycidyl ethers of bisphenol A and resorcinol.

Influence of Gel Content and Degree of Polymerization on the Mechanical Properties of Cured Resin

For epoxy-multiacrylate resin prepared from the molar ratio of epoxy resin to BTDA at 0.8, as shown

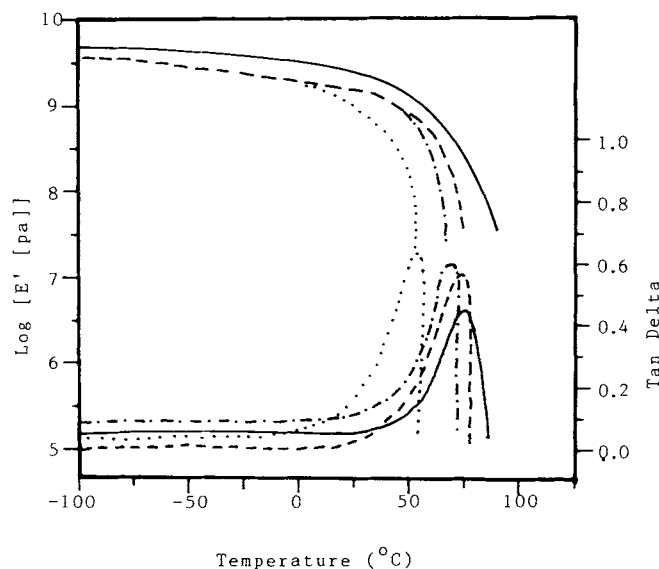


Figure 8 Dynamic mechanical spectra for cured WC67-MFAs resins with various gel contents. (—) 99.6%; (---) 94.8%; (- · - · -) 90.8%; (· · · · ·) 87.7%.

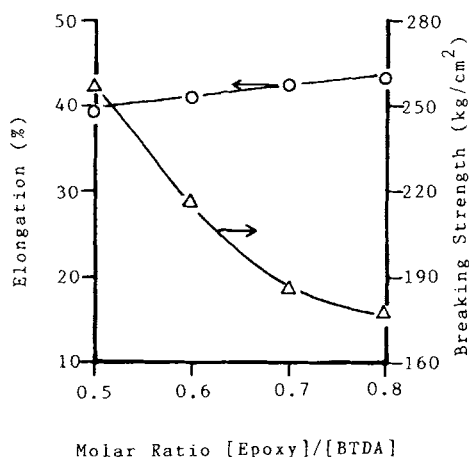


Figure 9 Breaking strength and elongation at break of cured WC67-MFAs resins as function of the molar ratio of epoxy/BTDA.

in Figure 7, both breaking strength and Young's modulus increase with crosslinking as the gel content increases from 88 to 99.6%, while elongation at break has a maximum value at gel content of 95%. Therefore, the crosslinking process enhances the toughness of cured film at gel content below 95%. When the gel content is above 95%, further crosslinking causes the cured film to be more brittle and less elongation at break.

The storage modulus (E') and loss factor ($\tan \delta$) of cured resins with a different gel content were determined using a DMA in the resonant model, and glass transition temperature (T_g) was recorded at $\tan \delta_{\max}$. Figure 8 shows the temperature dependence of storage modulus and loss factor for cured resins as a function of gel content. The storage modulus

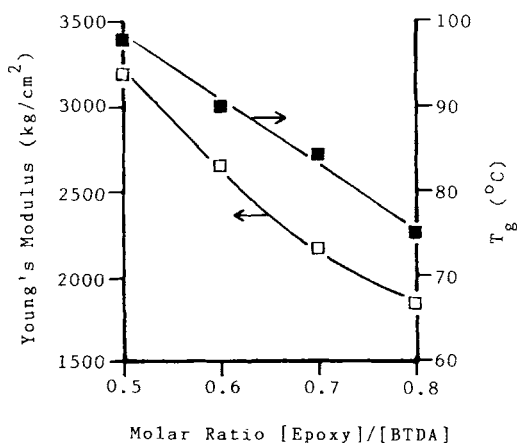


Figure 10 Young's modulus and T_g of cured WC67-MFAs resins as function of the molar ratio of epoxy/BTDA.

in the temperature range of -100 to 100°C is affected by crosslinking density, which increases with increasing the gel content. Also, an increase in gel content from 88 to 99.6% corresponds to a shift in T_g from 52.2 to 75°C .

As shown in Figures 9 and 10, the tensile properties and T_g of cured films, whose gel content was fixed at nearly 99.4%, are also affected by the degree of polymerization of resin. Increasing the molar ratio of epoxy resin to BTDA leads to a decrease in the effective acrylate concentration, which causes a lower crosslinking density in the cured film. Therefore, increasing the molar ratio from 0.5 to 0.8 results in a drop in breaking strength but an increase in elongation at break, while Young's modulus and T_g decrease.

Influence of Epoxy Type on the Mechanical Properties of Cured Films

The difference in storage modulus and T_g resulting from the difference in chemical structure of epoxy resins in the cured films is shown in Figure 11. The storage modulus and T_g are affected by stiffness or flexibility of the polymer chain. The cured EP829-MFAs resin has the highest storage modulus followed by WC69-MFAs, MK107-MFAs, and the WC68-MFAs resin in the temperature range of -100 to 100°C . The increase of modulus corresponds to the shift of T_g to higher temperature. The T_g values of cured WC68, MK107, WC69, and EP828 multiacrylate (MFAs) resins are 74, 85, 88, and 102°C , respectively.

The effects of the chemical structure of epoxy resins on the tensile properties of cured films are listed in Table III. Among all cured multiacrylate resins, the cured EP828-MFAs resin shows the highest Young's modulus and breaking strength but the lowest elongation at break. The trend of tensile properties are in agreement with that of the glass transition temperature because a decrease in T_g

Table III Effect of Epoxy Type on Tensile Properties of Cured Multiacrylate Resins^a

Oligomer	Gel (%)	TS (kg/cm ²)	YM (kg/cm ²)	E (%)
WC68	99.5	165	1840	53
WC69	99.5	240	2580	38
MK107	99.4	216	2112	50
EP828	99.4	361	7657	33

^a YM = Young's modulus; TS = breaking strength; E = elongation at break.

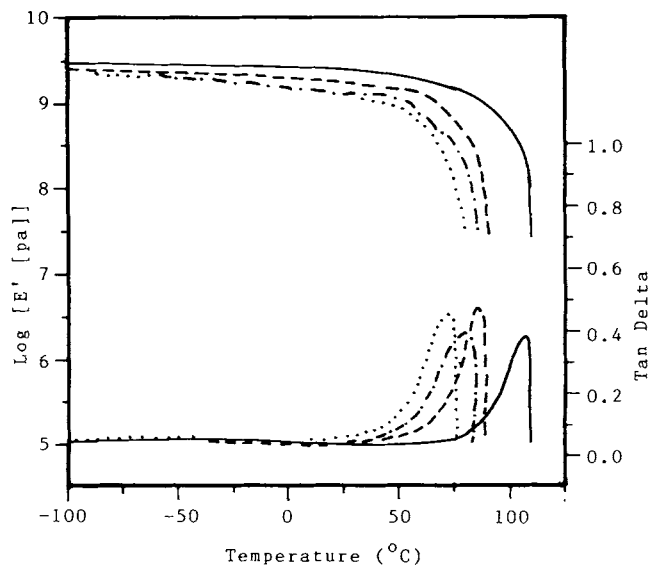


Figure 11 Dynamic mechanic spectra for cured epoxy-multiacrylate resins with various epoxy types. (—) EP828; (---) WC69; (-·-·-) MK107; (· · · · ·) WC68.

shows a more flexible molecular chain in the cured film and leads to a drop in Young's modulus and breaking strength but an increase in elongation at break.

Coating Properties of Epoxy-Multiacrylate Resins

The values of performance properties of epoxy-multiacrylate resin coated on steel plates are collected in Tables IV and V. The cured coating shows the excellent results, except the resistance of alkali corrosion. The poor resistance of coatings toward alkali is due to hydrolysis of ester groups in the resin. Film

hardness decreases when epoxy resin, which decreases T_g of the cured film, was used, but the other properties of the film are practically unchanged.

CONCLUSION

The obtained UV-autocurable epoxy-multiacrylate resins show excellent pot life and are rapidly cured by UV or sunlight irradiation without addition of any photoinitiators. The reactivity and the extent of photopolymerization of epoxy-multiacrylate resins depend largely on their chemical structure and

Table IV The Effect of Molar Ratio of Epoxy Resin/BTDA on Coating Properties of Cured Epoxy (WC-67) Multiacrylates

	Molar Ratio of Epoxy Resin/BTDA			
	0.5	0.6	0.7	0.8
Adhesion	100/100	100/100	100/100	100/100
Hardness	HB	HB	HB	HB
Impact test ($\frac{1}{2}\phi \times 1 \text{ kg} \times 500 \text{ mm}$)	OK	OK	OK	OK
Flexibility	OK	OK	OK	OK
Extensibility (mm)	4	4	4	4
Acid corrosion test	OK	OK	OK	OK
5% HCL, 72 h				
3% Acetic acid, 72 h	OK	OK	OK	OK
Salt corrosion test	OK	OK	OK	OK
5% NaCl, 72 h				
Alkali corrosion test	Fail	Fail	Fail	Fail
5% NaOH, 72 h				

Table V The Effect of Epoxy Type on Coating Properties of Cured Epoxy Multiacrylates

	WC68	WC69	MK107	EP828
Adhesion	100/100	100/100	100/100	100/100
Hardness	HB	H	HB	3H
Impact test ($\frac{1}{2}\phi \times 1 \text{ kg} \times 500 \text{ mm}$)	OK	OK	OK	OK
Flexibility	OK	OK	OK	OK
Extensibility (mm)	4	4	4	4
Acid corrosion test	OK	OK	OK	OK
5% HCl, 72 h				
3% Acetic acid, 72 h	OK	OK	OK	OK
Salt corrosion test	OK	OK	OK	OK
5% NaCl, 72 h				
Alkali corrosion test 5% NaOH, 72 h	Fail	Fail	Fail	Fail

the number of abstractable hydrogens in epoxy resin. It can be said that the chemical structure and the acrylic concentration of the resins are the major factors to determine the properties of cured film. Increasing the degree of polymerization of resins leads to a decrease in the effective acrylic concentration of resins and the crosslinking density of cured films. These results are reflected in a decreased storage modulus, Young's modulus, and breaking strength but an increased elongation at break, and the shift of T_g to lower temperature. The wide range properties of the cured epoxy-multiacrylate resins can be obtained by changing the chemical structure of epoxy resins. Among prepared multiacrylate resins, for instance, the cured bisphenol A type epoxy-multiacrylate resin (EP828-MFAs) has the highest values of storage and Young's modulus, breaking strength, and T_g . Film hardness of the resins coated on steel plates decreases when epoxy resin, which decreases T_g of the cured film was used, but the other coating properties are practically unchanged.

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