

## Novel Thermally Stable and Chemically Resistant Epoxy Polymers Derived from Phenylene Oxides

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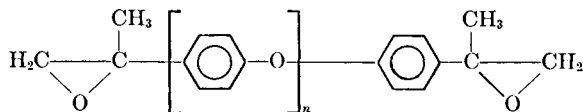
### Synopsis

Thermogravimetric and differential thermal analysis data are presented for three novel bisepoxy monomers and formulated coatings derived therefrom. The coatings were insensitive to impact in liquid oxygen and showed excellent resistance to liquid nitrogen tetroxide, concentrated nitric acid, and a variety of organic solvents. All coatings showed excellent adhesion to metals and oxidation resistance superior to any known commercial epoxy polymer. Thermal resistance was excellent at 500°F. for periods of at least 500 hr. in air. Other data on these novel epoxy polymers are also presented.

The versatility of epoxy resins as coating materials, adhesives, electrical potting compounds, and structural plastics has been amply demonstrated by their widespread application in almost all facets of industry. Over 90% of commercially available epoxy resins are glycidyl ethers of readily available di-, tri-, or polyfunctional phenolic compounds. Suitably polymerized by treating with Lewis acids ( $\text{BF}_3 \cdot \text{Et}_2\text{O}$ , pyromellitic dianhydride, etc.), these epoxy resins yield polymers that will withstand temperatures to about 200–250°C. for several hours in air without serious oxidative or thermal degradation. Above 250°C., however, oxidative and degradative problems become very serious, and commercially available epoxy polymers cannot be reliably employed for extended periods.<sup>1,2</sup>

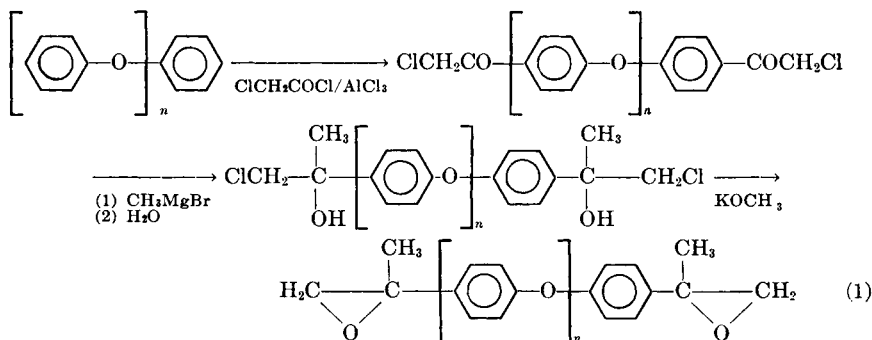
For many aerospace applications, it is desirable to retain the ease of handling of the epoxy resins. For this reason the present investigation was undertaken to improve the thermal and oxidative stability of epoxy polymers. This report describes the structure, properties, and polymerization of a novel series of difunctional epoxy monomers derived from phenyl ether and *p*-phenylene oxides, in which the aliphatic epoxy moiety has been reduced to its shortest length (i.e., epoxyethyl), and the reactive "benzylic" position has been blocked by methyl substitution.

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These structures ( $n = 1, 2, 3$ ) were developed as the result of a theoretical analysis of the molecular configuration of all known epoxy resins possessing thermal stability. Bonds known to be thermally or oxidatively unstable were eliminated. Phenyl ether and *p*-phenylene oxide chains were chosen because it is well known that they are thermally stable and exhibit outstanding oxidative resistance.<sup>3</sup>

Monomers were prepared in three successive steps. Phenyl ether (or the *p*-phenylene oxide) was treated under Friedel-Crafts conditions with chloroacetyl chloride and aluminum chloride to produce the 4,4'-bis(chloroacetyl) derivative in 90% yield. The bis(chloroacetyl) derivative was then treated with methylmagnesium bromide to form the bis(chlorohydrin) quantitatively. The bisepoxide was prepared in 90–98% yield by treating the bis(chlorohydrin) with potassium methoxide. Details of the synthesis are reported elsewhere.<sup>4</sup> The overall reaction scheme is shown in eq. (1).



The bisepoxides (I, II, and III) discussed in this report are white microcrystalline solids.\* Bisepoxide I melts at 51.5°C., and II and III melt at 103.5 and 119.0°C., respectively.

## RESULTS AND DISCUSSION

### Polymerization Studies

Initial polymerization studies on bisepoxide (I) employed boron trifluoride-ethylamine complex as the Lewis acid catalyst. The temperature was

\* A report of work on the bisepoxide I was presented by Chen.<sup>5</sup> The tan-colored material of Chen (melting range 40–43°C.) was shown by our investigation to be contaminated with KOH, KCl, CH<sub>3</sub>OH, and gummy by-products (possibly vinyl derivatives). We here present data on carefully purified, white bisepoxide I, which melts sharply at 51.5°C.

increased from ambient, stepwise, and the cure was terminated by heating at 260°C. for 22 hr. Although this polymerization procedure demonstrated the thermal and oxidative stability of the polymer, the practical limitations it imposed precluded its general application. As is well known, the ethylamine complex of boron trifluoride is employed specifically to reduce the catalytic activity of boron trifluoride at ambient temperatures.<sup>6a</sup> Therefore, our initial attempt to develop a room-temperature cure involved the study of less stable boron trifluoride adducts. It was quickly discovered that the use of ketones as solvents (chosen to provide practical systems for spraying coatings), completely masked any differences in the reactivity of various aliphatic amine complexes with boron trifluoride. A possible explanation was that the boron trifluoride selectively coordinated with the ketonic carbonyl groups, and was not released until approximately the dissociation temperature of the amine complex. A minimum of 1 hr. at 232°C. was required to form a coherent polymeric coating with BF<sub>3</sub>-alkylamine complexes in ketonic solvents.

In solvents that coordinate less readily with boron trifluoride (e.g., tetrahydrofuran, dichloromethane) the difference in reactivity was clearly evident. Thus, BF<sub>3</sub>·Et<sub>2</sub>O reacted with bisepoxide I at -59°C. in dichloromethane solution to produce a pale yellow precipitate, which became a rigid foam on warming to 50-60°C. The foam melted at 155°C. and solidified on heating at 250°C. for several hours. The precipitate obtained at -59°C. probably represented the product of coordination of at least one BF<sub>3</sub> molecule with the oxirane oxygen atoms. Subsequent reactions indicated that the BF<sub>3</sub> catalyst may have simultaneously effected the well-known rearrangement of the oxirane rings to a carbonyl compound, since BF<sub>3</sub>·Et<sub>2</sub>O had been shown to cure bisepoxide I to a hard polymer in 10 min. at 232°C.<sup>7</sup>

In addition to boron trifluoride-alkylamine catalysts, the following organic Lewis acids and bases were employed as curing agents for bisepoxide I: methyl nadic anhydride, pyromellitic dianhydride, benzophenone tetracarboxylic acid dianhydride, *m*-phenylenediamine, and diethylenetriamine. Stoichiometric amounts of each curing agent were added to bisepoxide I, and the mixture was cured for 1, 2, and 3 hr. at 177°C. In no case was a completely cured polymer, suitable for use as a coating, obtained.

Four inorganic Lewis acid curing agents were unsuccessful in effecting a complete cure of bisepoxide I, namely, silicon tetrachloride, thionyl chloride, phosphorus oxychloride, and phosphorus trichloride.

Bisepoxide I could be successfully cured by using *p*-toluenesulfonic acid, with which the oxirane rings of the monomer reacted without the formation of a stable intermediate complex. Because resistance to strongly oxidizing chemicals was a prime objective of our coating program each bisepoxide was considered to be satisfactorily cured when the polymeric coating it produced was unaffected by exposure to concentrated nitric acid at 20°C. for 20 hr. Details of the cure and oxidation tests are given in the Experimental Section.

### Thermal Analyses

The thermal stability of the coatings obtained by polymerizing bisepoxides I, II, and III, was determined in air by thermogravimetric analysis (TGA) and differential thermal analysis (DTA).

The TGA curve for bisepoxide I, suitably polymerized with *p*-toluenesulfonic acid, is shown in Figure 1. Rapid weight loss (ca. 10%) began near 424°C., and 50% of the original weight was lost at 560°C. The data show that polymerized bisepoxide I is stable to approximately 100°C., higher than any previously known epoxy polymer.<sup>6b</sup> The TGA data support the original hypothesis that elimination of the aliphatic ether linkage of the glycidyl group (present in conventional bisphenol-A-epichlorohydrin resins), and methyl substitution of the reactive and oxidation-

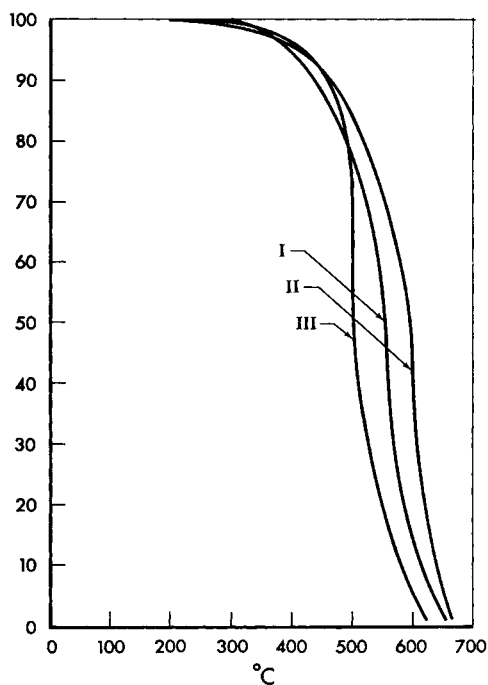


Fig. 1. Thermogravimetric data (4.5°C./min. in air) for bisepoxides I, II, and III.

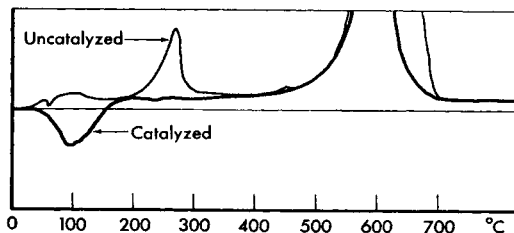
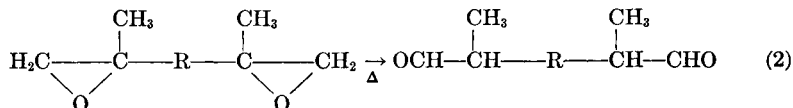


Fig. 2. Differential thermal analysis data for bisepoxide I.

sensitive "benzylic" position of the epoxyethyl group, substantially improve the thermal stability of the epoxy polymer.

Differential thermal analyses (DTA) of bisepoxide I, uncatalyzed and catalyzed, are shown in Figure 2. The uncatalyzed monomer showed a small but distinct endotherm at 45–55°C. (fusion, m.p. 51.5°C.), and an exotherm at about 260°C., which was attributed to rapid thermal rearrangement of the oxirane rings to aldehyde (formyl) groups, as shown in eq. (2).



This rearrangement is well known to occur at 180–280°C.<sup>7</sup> Indeed, the infrared spectrum of small specimens of bisepoxide I heated at 180°C. for 15 min. showed strong absorption for carbonyl at 1675 and 1720 cm.<sup>-1</sup>, but no absorption for the oxirane ring at 910 cm.<sup>-1</sup>, indicating that rearrangement of the oxirane ring to the aldehyde group is very rapid at 180°C.

Catalyzed bisepoxide I showed a strong endothermic doublet at 98 and 110°C. (ascribed to the curing reaction) but no further change until 475–550°C., in which range thermal and oxidative degradation occurred. The fusion endotherm was lost because the monomer did not crystallize in the presence of the *p*-toluenesulfonic acid catalyst. The absence of an exotherm in the 260°C. region strongly suggested that polymerization occurred before the oxirane rings rearranged to aldehyde groups.

The TGA curve for polymerized bisepoxide II ( $n = 2$ ), similarly catalyzed with *p*-toluenesulfonic acid, is shown in Figure 1. A 10% weight loss occurred at 475°C., and a 50% loss at 580°C. These data show an

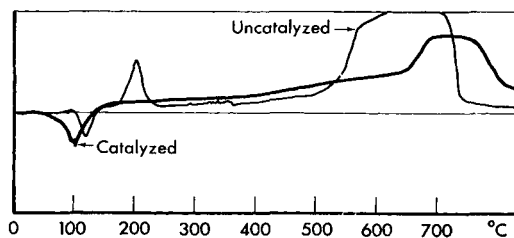


Fig. 3. Differential thermal analysis data for bisepoxide II.

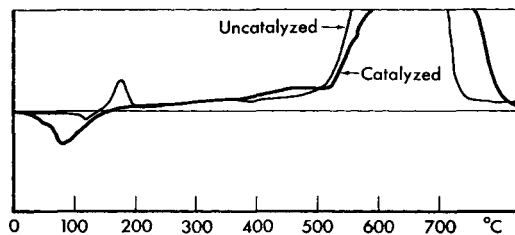


Fig. 4. Differential thermal analysis data for bisepoxide III.

improvement of approximately 50°C. over those for bisepoxide I at the knee of the curve.

The DTA curves for uncatalyzed and catalyzed bisepoxide II are shown in Figure 3. The uncatalyzed epoxy monomer showed a fusion endotherm in the 100–110°C. region (actual m.p. 103.5°C.). The epoxy rearrangement endotherm occurred at 200–220°C. Catalyzed bisepoxide II showed an endothermic doublet in the 100°C. region, attributed to fusion and initiation of polymerization. No further changes were observed until thermal and oxidative degradation occurred in the 550–700°C. region (cf. 475–550°C. for bisepoxide I). From the TGA and DTA data, it is evident that bisepoxide II is thermally and oxidatively superior to bisepoxide I.

The TGA curve for bisepoxide III ( $n = 3$ ) is shown in Figure 1. A 10% weight loss occurred at 460°C. and a 50% loss at 500°C. (cf. 560°C. for bisepoxide I and 580°C. for bisepoxide II). The DTA curve of the uncatalyzed monomer (Fig. 4) showed a fusion endotherm at 115–120°C. (actual m.p. 119°C.), and an exotherm at 165–180°C., attributed to thermal rearrangement of the oxirane rings. The catalyzed resin showed a broad endotherm centered at 85°C., indicating initiation of polymerization. Serious thermal and oxidative degradation began at 530°C. (cf. 550–700°C. for bisepoxide II).

### Formulation Studies

As the epoxy polymers were synthesized to provide superior thermal, oxidative, and hydrolytic stability, the choice of fillers used in formulation was limited to those which would not impair these properties. The catalyst system was that employed in the polymer characterization studies. Plasticizers were not used as modification of the chemical structure of the monomer was relied upon to reduce brittleness in the polymerized coatings.

Only chemically inert pigment fillers were employed. Anatase, strontium chromate, graphite, and mica were used in coating formulations with bisepoxides I and II. Anatase ( $\text{TiO}_2$ ) exhibits good color retention over a wide range of temperatures (–50 to 250°C.). Strontium chromate does not inhibit the polymerization reaction as does, for example, zinc chromate. Graphite is an abrasion-resistant filler in conventional epoxy coatings; and mica is often used in brittle resins to increase flexibility and improve water resistance.<sup>6c</sup> Concentrations of fillers were adjusted to give adequate opacity. In the case of strontium chromate, the concentration was that conventionally employed to give acceptable corrosion inhibition.

### Evaluation of Coatings

All formulations were subjected to standard coating evaluation tests. Coatings were evaluated for adhesion, liquid oxygen impact sensitivity, chemical resistance, weathering characteristics, flexibility, thermal stability, and abrasion resistance (Table I).

By the standard wet-tape adhesion test,<sup>8</sup> all formulations exhibited excellent adhesion to titanium, aluminum, stainless steel, and carbon steel.

TABLE I  
Evaluation Tests on Epoxy Resin Coatings

Test	Bisepoxide I				Bisepoxide II, TiO <sub>2</sub> -filled
	Unfilled	TiO <sub>2</sub> -filled	SrCrO <sub>4</sub> -filled	Graphite-filled	
Adhesion (4 substrates) <sup>a</sup>	Good	Good	Good	Good	Good
Liquid oxygen impact	40/40 passed	40/40 passed	39/40 passed	40/40 passed	20/20 passed
Liquid contact (7 chemicals) <sup>b</sup>	All passed	All passed	All passed	All passed	All passed
Accelerated weathering	80 hr. passed	Severe chalking 100 hr.	Slight chalking 600 hr.	600 hr. passed	Severe chalking 200 hr.
Conical bend	Failed	7/8 in. radius passed	5/8 in. radius passed	5/8 in. radius passed	1/4 in. radius passed
N <sub>2</sub> O <sub>4</sub> immersion	—	Passed	Passed	Passed	Passed
Elevated temperature					
500°F.	96.6% loss, 142 hr.	14.6% loss, 175 hr.	26.5% loss, 142 hr.	32.2% loss, 142 hr.	—
450°F.	45.4% loss, 500 hr.	12.03% loss, 500 hr.	13.83% loss, 500 hr.	17.51% loss, 500 hr.	10.57% loss, 150 hr.
Salt spray	—	Moderate corrosion, 50 hr.	Slight corrosion, 150 hr.	Severe corrosion, 50 hr.	—
Abrasion resistance	Poor	Poor	Poor	Poor	—
Curing conditions	1/2 hr., 350°F.	1 hr., 350°F.	1 hr., 350°F.	1 hr., 350°F.	1 hr., 350°F.
	1 hr., 250°F.	4 hr., 300°F.	4 hr., 300°F.	4 hr., 300°F.	
	48 hr., 120°F.	48 hr., 250°F.	48 hr., 250°F.	48 hr., 250°F.	

<sup>a</sup> Titanium, aluminum, stainless steel, carbon steel.

<sup>b</sup> Concentrated nitric acid, JP-6 jet fuel, ethylene glycol, diester lube oil, Oronite 8200 hydraulic fluid, Oronite 8515 hydraulic fluid, MIL-H-3136 Type III test fuel.

Smooth metal surfaces were cleaned with dichloromethane, and coatings were applied directly.

Tests for liquid oxygen impact sensitivity were conducted by using a standard method.<sup>9</sup> Both anatase-filled coatings and the graphite formulation passed all impact tests (i.e., 40/40). No other epoxy polymeric coatings known will pass this test. The strontium chromate-filled bisepoxide I coating passed 39 of 40 liquid oxygen impact tests. It is believed that the solitary failure was due to sample contamination. The corresponding chromate-filled bisepoxide II coating was completely inert in the liquid oxygen impact test (20/20 passed).

A procedure was developed to determine the resistance of polymerized coatings to splashing with various solvents and strongly corrosive liquids. One drop of liquid was placed on the coated surface and left in a desiccator for 24 hr. at 20°C. The surface was then dried and examined microscopically for etching or deterioration. Liquids employed were: concentrated nitric acid (specific gravity 1.42), JP-6 jet fuel, ethylene glycol, diester lube oil, Oronite 8200 hydraulic fluid, Oronite 8515 hydraulic fluid, and MIL-H-3136 Type III test fuel. No softening, blistering, cracking, corrosion, or other evidence of attack of the polymerized coated surfaces was observed in any of the liquid-contact tests. The outstanding resistance of bisepoxide I and II coatings to concentrated nitric acid was particularly noteworthy.

For aerospace applications, it was necessary to determine the resistance of the coatings to the very strong oxidizers used as rocket fuels. Formulated coatings, cured on stainless steel, were partially immersed for 30 min. in liquid nitrogen tetroxide ( $N_2O_4$ ) at 21°C. After immersion, specimens were air-dried for 30 min. at 20°C., then exposed to 35°C. ( $\pm 2$ ), at a relative humidity of  $95 \pm 3\%$  for 23.5 hr. Anatase-filled and graphite-filled coatings showed no visible effect from the  $N_2O_4$  immersion. The strontium chromate coatings were severely discolored in the area exposed to  $N_2O_4$  vapor. After the humidity exposure, no evidence of blistering, softening, or loss of adhesion was observed. The tests demonstrated that coatings made from these novel bisepoxides are remarkably chemically inert to strongly oxidizing liquids.

Resistance of the unfilled and filled polymerized coatings to splashing with strong solutions of sodium chloride showed that the coatings were not good under these conditions.<sup>10</sup>

Accelerated weathering tests indicated that the anatase-filled coatings did not have sufficient resistance to the combined effects of water and ultraviolet irradiation to be recommended for exterior use. The strontium chromate and graphite filled coatings exhibited far superior accelerated weathering resistance.<sup>11</sup>

Conical bend tests of the coatings on stainless steel showed good film flexibility for all formulations examined.<sup>12</sup> Unfilled coatings and earlier formulations omitting mica exhibited brittleness in this test.

Weight loss on heating unfilled bisepoxide I or II coatings in air was



found to be a function of the area of surface exposed. Coatings were rapidly attacked at about 500°F. with concurrent darkening. Formulated coatings, however, exhibited excellent resistance to thermal aging at 450°F.

All coatings showed poor abrasion resistance in the standard Taber procedure.<sup>13</sup> Where mechanical abrasion is an important factor, therefore, unfilled or filled bisepoxide I or II coatings would require an additional surface coating of an abrasion-resistant polymer.

It became clear early in our study that the conditions for polymerizing the unfilled bisepoxides required modification for formulated coatings. Times for polymerizing the monomers at different temperatures, and other data, are recorded in Table I. No formulation studies were carried out on bisepoxide III, as the TGA and DTA data indicated that this compound was less thermally stable than bisepoxides I and II.

### Laminate Studies

To evaluate bisepoxides I and II as laminating resins for use in structural plastics applications, multilayer resins and glass-cloth laminates were prepared as described in the Experimental Section. These laminates showed excellent aging characteristics at 500°F. in air for periods exceeding 500 hr. Only the exposed surfaces of the laminate showed any evidence of oxidative attack, as indicated by darkening of the polymer. Cross-sectional examination of cut laminate specimens which had been heated continuously in air at 500°F. showed no darkening or other evidence of polymer degradation, demonstrating that the polymers are substantially unaffected when protected from air within the laminate matrix. Glass-cloth laminates ( $1/8$  in. thick) were heated, and  $1 \times 4$  in. flexural specimens were tested at 500°F. Data are presented in Table II.

TABLE II  
Flex and Modulus Data for Laminates

Bisepoxide	Condition	Flex $\times 10^3$ , psi	Flex modulus $\times 10^4$ , psi
I	Unaged	25.96	3.09
	Aged 192 hr., 500°F.	36.0	3.32
	Aged 500 hr., 500°F.	30.6	3.79
II	Unaged	25.35	3.15
	Aged 192 hr., 500°F.	39.05	3.71
	Aged 500 hr., 500°F.	33.73	3.89

## EXPERIMENTAL

### Materials and Equipment

All chemicals used in this study were reagent grade. Thermogravimetric analyses (TGA) and differential thermal analyses (DTA) were determined by using a Stanton thermobalance.

### Polymerization of Bisepoxides I and II

A 25% solution of bisepoxide I in a mixed ketone solvent (40% methyl ethyl ketone, 25% methyl isobutyl ketone, 25% toluene, and 10% xylene) catalyzed with one part per 100 of resin (phr) of *p*-toluenesulfonic acid, added as a 10% solution in isobutyl acetate, gave a concentrated nitric acid-resistant coating on stainless steel in 1 hr. at 116°C. or 48 hr. at 49°C.

Similar systems were employed to catalyze the polymerization of bisepoxides II and III. As the monomers were only sparingly soluble in the mixed ketone solvent, 25% solutions in tetrahydrofuran were used for polymerization studies. Bisepoxide II with 0.3 phr of catalyst was polymerized after 1 hr. at 177°C., 3 hr. at 116°C., or 4 days at 49°C. With the same catalyst concentration, bisepoxide III was polymerized after 2 hr. at 177°C. or 3 hr. at 116°C.

Bisepoxides were considered to be satisfactorily polymerized when the coatings were unaffected by exposure to concentrated nitric acid at 20°C. for 20 hr. Suitable solutions of the bisepoxides (see above) were catalyzed and sprayed onto 1 in. square panels of 321 stainless steel. Solvents were evaporated in a slow current of air, and the coated steel specimens were heated at 49°C. in a circulating air oven for 3 hr. to remove residual solvent. Polymerization was effected by heating as described above. One drop of concentrated nitric acid was placed on the coating and the specimen left for 20 hr. at 20°C. in a desiccator. Acid was removed, and the specimen examined microscopically for etching or corrosion.

### Formulations

In formulations of bisepoxides I and II, the following solvent mixture was used: methyl ethyl ketone, 40%; methyl isobutyl ketone, 25%; toluene, 25%; cyclohexanone, 10%. Formulated mixtures were ground 48 hr. in a ball mill, then diluted with an additional 25% of solvent prior to catalysis.

TABLE III  
Formulations of Bisepoxides I and II

Bisepoxide	Parts					Color (1 hr./350°F.)
	TiO <sub>2</sub>	SrCrO <sub>4</sub>	Graphite	Mica	Solvent	
I, 25	22	—	—	3	25	Pale beige
I, 25	—	22	—	3	25	Bright yellow
I, 25	—	—	10	3	25	Gray-black
II, 25	25	—	—	—	25	Pale beige

The catalyst was 1.0 phr of *p*-toluenesulfonic acid, added as a 10% solution in *n*-butyl acetate. Satisfactorily cured coatings were obtained on heating at 350°F. for 1 hr.

Table III lists the parts by weight of the components in each formulated mixture.

### Laminates

Thirteen-ply glass-cloth laminates (approx.  $9 \times 9$  in.), were suitably impregnated with bisepoxides I or II and press-cured into  $\frac{1}{8}$ -in. thick panels at 25 psi and 350°F. for 2 hr. followed by a post-cure for 10 hr. in a circulating air oven at 500°F. All laminates were "aged" simultaneously at 500°F. in the same circulating air oven. Duplicate specimens were cut from each laminate at required intervals, and the remaining material was returned to the oven for further aging. Flexural specimens ( $1 \times 4$  in.) were tested at 500°F. for all values recorded in Table II.

### CONCLUSIONS

This study shows that the thermal and oxidative stability of conventional epoxy resins are significantly improved by employing phenyl ether or phenylene oxides as the main chain of the monomer. By terminally substituting the main chain with  $\alpha$ -methylepoxyethyl groups, the thermal and oxidative instability inherent in the conventional glycidyl-terminated epoxy resins is improved by at least 100°C.

The four epoxy coating formulations developed perform extremely well when exposed to impact in liquid oxygen and immersions in liquid nitrogen tetroxide. Formulated and unformulated coatings exhibit good resistance to contact with concentrated nitric acid and a variety of organic solvents (hydrocarbons, alcohols, esters); but possess only poor resistance to salt solutions. Coatings exhibit excellent adhesion to titanium, aluminum, and various steels. They display poor resistance to abrasion and weathering and are not suitable for exterior uses where mechanical abrasion may occur. These properties suggest possible use in oxidizer tankage compartments of aerospace vehicles or in ground storage facilities, where there is danger of intermittent exposure to splashing with strong oxidizing agents.

Coatings based on bisepoxide II are significantly more flexible than those with bisepoxide I. A further advantage of bisepoxide II is its higher thermal stability as shown by the TGA and DTA data. Preliminary studies with glass-cloth laminates indicate that these resins may show promise as structural plastics.

The lower solubility of bisepoxide III, its higher melting point and lower thermal stability compared with bisepoxide II, precluded further studies of this resin monomer.

The foregoing data show that bisepoxide II, which contains three benzenoid nuclei, is clearly more oxidatively and thermally stable than bisepoxides I and III, which contain two or four aromatic nuclei, respectively.

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