



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(21) International Application Number: PCT/US82/00666 (22) International Filing Date: 17 May 1982 (17.05.82) (31) Priority Application Number: 270,915 (32) Priority Date: 5 June 1981 (05.06.81) (33) Priority Country: US (71) Applicant: GENERAL ELECTRIC COMPANY [US/ US]; 1 River Road, Schenectady, NY 12305 (US). (72) Inventor: ALLEN, Richard, Brian ; 525 Windsor Road, Dalton, MA 01226 (US). (74) Agents: KING, Arthur, M.; International Patent Opera- tion, General Electric Company, 570 Lexington Ave- nue, New York, NY 10022 (US) et al.		(81) Designated States: AU, DE (European patent), FR (Eu- ropean patent), GB (European patent), JP, NL (Euro- pean patent). Published <i>With international search report.</i>
(54) Title: THERMOSET EPOXY COMPOSITION (57) Abstract These compositions contain the reaction product of a liquid cycloaliphatic epoxy resin having more than one 1,2-epoxy group per molecule with a diphenolic compound. This reaction product, which is solid, may be employed with curing agent, filler and the like in any conventional manner or, as preferred, as molding compound.		

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THERMOSET EPOXY COMPOSITION

Background of the Invention

Epoxy resins are well known for a variety of different uses. The liquid resins are commonly employed for such applications as casting, potting, coating, impregnation and the like. Solid epoxy resins are most popular for injection, compression and transfer molding.

In general, the more rapidly curing cycloaliphatic epoxies do not lend themselves to molding in conventional equipment. However, because they possess generally superior characteristics of weathering resistance, heat distortion temperature and electrical properties the liquid cycloaliphatic epoxy resins would be preferred if articles could be produced by conventional solid resin techniques. Such articles would find particular utility in the field of electric or electronic parts.

It is further known that many epoxy resins may be modified by reaction with an adduct to provide different properties. The results of such an adduct reaction, however, are often unpredictable.

By way of illustration, Japanese Patent application 50,921 of Osaka Soda Co. Ltd., describes the production of thermoset resins from diglycidyl compounds. Those compounds are reacted with divalent phenols in the presence of ammonium bicarbonate or acetate.

British patent 915,767 of Shell Internationale Research Maatshappij N.V. relates to thermoplastic materials. These materials are produced by reaction of a diglycidyl ether with a dihydroxy compound in the presence of a sodium salt.

U.S. Patent No. 3,364,178 of Kreps et al describes production of thermoplastic materials. Equimolar amounts of a dihydric phenol and a diglycidyl ether of a dihydric phenol (at least one of which contains a diarylsulfone group) are reacted in the presence of an alkaline catalyst.



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U.S. Patent No. 3,694,407 of Krikorian describes the reaction of certain polyepoxides having more than one vicepoxide group with special polyhydric phenols. The reaction takes place in the presence of an organic phosphine or hydrocarbon phosphonium halide to produce a thermoset product.

U.S. Patent No. 3,733,305 of Loewrigkeit et al again relates to thermoset products. These are produced without a catalyst by reaction of sulfonyl diphenol (bisphenol-S) with a bis (glycidyloxyphenyl) sulfone (diepoxy bisphenol S) in molten condition.

Not only do the reactions and results for such adduct formation differ, but the prior art lacks particular teaching as to the modification of liquid cycloaliphatic (or alicyclic) epoxy resins. The production of thermoset materials capable of use in solid molding techniques from these resins therefore remains an especially desirable objective.

Introduction of the Invention

The present invention involves improved thermoset compositions. These compositions relate to the reaction product of a liquid cycloaliphatic epoxy resin having more than one 1,2-epoxy group per molecule with a diphenolic compound. The diphenolic compound is utilized in an amount effective to convert the epoxy resin from a normally liquid to solid form.

The present compositions are particularly useful in molding applications. They may additionally contain a curing agent, such as metal acetylacetonate and phenolic accelerator, or a filler. In preferred compositions useful as molding compounds, they contain both these additional components.

Description of the Invention

The epoxy resins employed in this invention can be cycloaliphatic 1,2-epoxy resins having more than 1 epoxy group per molecule. They include resins such as



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3,4-epoxycyclohexylmethyl-(3,4-epoxy) cyclohexane
carboxylate (sold under the trademarks ERL 4221 by
Union Carbide Plastics Company or Araldite CY 179 by
Ciba Products Company); bis(3,4-epoxy-6-methylcyclo-
5 hexylmethyl) adipate (sold under the trademarks ERI 4289
by Union Carbide Plastics Company of Araldite CY 178 by
Ciba Products Company); vinylcyclohexane dioxide (ERL
4206 made by Union Carbide Plastics Company); bis(2,3-
epoxy-cyclopentyl) ether resins (sold under the trademark
10 ERL 4205 by Union Carbide Plastics Company); 2-(3,4-epoxy)
cyclohexyl-5,5-spiro (3,4-epoxy) cyclohexane-m-dioxane
(sold under the trademark ERL 4234 by Union Carbide Corp.
of Araldite CY 175 by Ciba Products Company). These
and other suitable cycloaliphatic epoxy resins are
15 readily available and/or may be made in accordance with
conventional techniques.

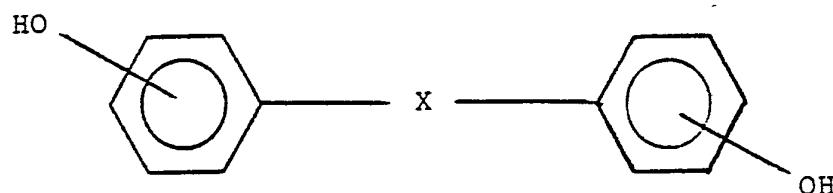
In addition to the cycloaliphatic resins, other epoxy
resins may also be present in the compositions of this
invention. These ancillary epoxy resins need not be
20 cycloaliphatics but desirably are solid, thermoset resins.
In this manner, they are most compatible with the preferred
properties and applications of the present composition.

The other essential reactant for the present composi-
tions is a diphenolic, and preferably a bisphenol,
25 compound. An amount of diphenolic compound effective to
transform the liquid cycloaliphatic epoxy resin into a
solid thermoset reaction product should be utilized.
The equivalent ratio of liquid cycloaliphatic epoxy
resin and the diphenolic compound is desirably between
30 about 40:1 and 2:1, more preferably 20:1 to 3:1,
respectively.

Preferably, the diphenolic compound utilized in
accordance with the present invention has the formula:



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wherein X is selected from the group consisting of lower aliphatic, sulfide and sulfonyl radicals.

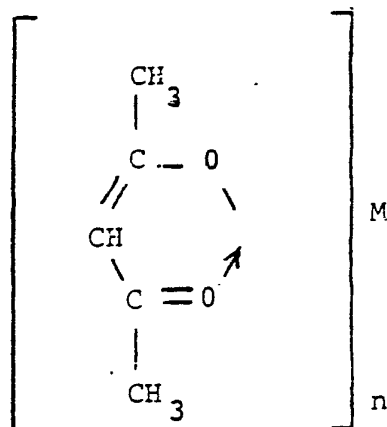
Exemplary of these diphenolic compounds are 4,4'-
 5 sulfonyldiphenol, 4,4'-(3,3',5,5' tetramethyl) sulfonyl
 diphenol, 4,4'-diphenolsulfide and bis(4-hydroxyphenyl)
 methane, ethane, propane or butane.

The reaction between the cycloaliphatic epoxy resin
 and diphenolic compound is readily achieved. They need
 10 only be admixed and, desirably, heated to yield the
 present reaction product. Preferably, the admixture is
 heated to between about 100° and 180°C. This ensures
 rapid and complete reaction. The resultant solid product
 is an improved thermoset epoxy resin.

15 This thermoset resin may be admixed with curing agent
 where it is to be utilized in conventional manner.
 While there is no criticality as to the curing agent
 utilized, it is preferred to employ the combination of
 a metal acetylacetonate and a phenolic accelerator. This
 20 is particularly true where the composition is to be
 employed as a molding compound. In this embodiment of
 the present invention, such a curing agent imparts
 improved properties of storage stability with activatable
 rapid curing.

25 The metal acetylacetonates of the present invention
 desirably have the following structural formula:

- 5 -



wherein M is a metal ion and n is 1 to 4 corresponding to the valence number of the metal ion.

They are characterized by the presence of solely metal to oxygen bonds. Included within the scope of the invention are metal acetylacetonates in which one or more hydrogen atoms of the methyl or methylene groups are substituted by a halogen atom or by a alkyl, aryl, or an alkaryl substituent. An example of a halogen-substituted metal acetylacetonate is a metal hexafluoroacetylacetonate or trifluoroacetylacetonate. An example of an alkyl-substituted acetylacetonate is dipivaloylenthane in which the three hydrogen atoms on each of the methyl groups are substituted with a methyl group.

The acetylacetonate hardeners of the present invention should not be confused with similar compositions containing a labile halogen atom. In the present compositions, the halogens, if present, are attached directly to a carbon atom of the methylene or methyl groups and are therefore extremely stable. Labile halogen atoms in epoxy resin curing agents normally form halogen acids, and the presence of such an ionic constituent in the cured resin would raise many problems, including poor electrical properties.

Metal acetylacetonates in which the metal is aluminum, titanium, zinc or zirconium are a particularly preferred class of metal acetylacetonates within the scope of the



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invention. However, essentially any metallic acetyl-
acetate may be used, including aluminum, barium,
beryllium, cadmium, calcium, cerous, chromic, cobaltic,
cobaltous, cupric, ferric, ferrous, gallium, hafnium,
5 indium, lead, lithium, magnesium, manganic, manganous,
molybdenum, molybdenyl, nickel, palladium, platinum,
potassium, rhodium, tungstyl, uranyl, vandium, vanadyl,
zinc, and zirconium. Acetylacetonates of the rare earth
elements, scandium, cerium, yttrium, lanthanum,
10 praseodymium, neodymium, samarium, europium, gadolinium,
terbium, dysprosium, holmium, erbium, thulium, ytterbium,
and lutetium are known and can reasonably be expected to
be useful in the practice of the present invention.

The metal acetylacetonates are used in small catalytic
15 quantities of from 0.01 to 5.0 percent, based upon the
weight of the epoxy resin. Optimum results have been
achieved with from 0.05 to 3.0 percent. It is important
to note that the acetylacetonates of the invention are
catalytic hardeners, which do not in a significant way
20 become a part of the hardened epoxy molecule as do curing
agents added in much larger or near stoichiometric
amounts.

Among the phenolic accelerators which can be effectively
used in this invention are bisphenol A (i.e., 2,2-bis
25 (4-hydrophenyl) propane), pyrogallol, dihydroxydiphenyls
as well as ortho-, meta- and para- hydroxbenzaldehydes,
(such as salicylaldehyde), catechol, resorcinol, hydro-
quinone, and phenol-formaldehyde and resorcinol-formal-
dehyde condensates. Examples of other phenolic accelera-
30 tors suitably employed in this invention also include
halogenated phenols such as ortho-, meta- and para-
chlorophenols or bromophenols and ortho-, meta-, and
para- nitrophenols. The phenolic accelerator may be
present in concentrations ranging from 0.1 to 30 percent .
35 based on epoxy resin, with optimum cure rates being



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produced with phenolic accelerator concentration between 0.5 and 10 percent by weight of the epoxy resin. As in the case of the acetylacetonate, the phenolics are added in relatively small amounts because they are accelerators or catalysts rather than curing agents of the stoichiometric type which form a significant reaction with the epoxy resin and become a significant part of the epoxy molecule.

Another preferred but optional component of the present compositions is a filler. Filler is again especially important where these compositions are to be utilized as molding compounds.

The fillers of the present invention are generally chemically inert. They ordinarily will not react with any of the epoxy resins, metal acetylacetonate or accelerator. They instead function by stabilizing the physical properties of the molding compound, particularly during and after resin cure.

Any conventional filler may be utilized in the present compositions. Representative fillers include: clays, like kaolin and calcined clays; silica, like novaculities, ground sand and amorphous glass; mica; talc; carbon black; alumina; and wollastonite. Alternatively or in addition, a structural filler may be employed. These fillers include such fibrous materials as glass fiber, mineral wool and the like which may provide enhanced product strength.

The present compositions, particularly where utilized as molding compounds, should contain from about 10 to 95% filler by total weight. More preferably, from about 50 to 95% filler is utilized. This optimizes the advantages of the present invention.

In preparing the present compositions, the various optional ingredients may simply be admixed with the epoxy resin/diphenolic compound reaction product, usually at ambient temperatures. In thereafter utilizing the present



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compositions, conventional techniques may be employed. For example, the solid composition may be compression or transfer molded. While under molding pressure, the composition should be heated, generally to from about
5 50 to 200°C. Under these conditions, curing may occur in minutes. Molded articles having virtually any configuration or size may be formed.

In some preferred embodiments of the present invention, the present compositions may contain one or more further
10 auxiliary components. For example, up to about 10%, desirably from 1 to 5%, by weight of epoxy resin of a flame retardant may be used. Most commercial retardants, including antimony oxide or halogenated hydrocarbon may be utilized. Mold release agent such as wax, ordinarily
15 in an amount of from 0.2 to 4% by weight of epoxy resin are also highly desirable. These and other components may simply be admixed with the composition prior to molding to obtain the benefits for which they are already known.

20 The following examples are given by way of illustration only and are not intended as a limitation on the scope of this invention. Many variations are possible without departing from its spirit and scope. Unless otherwise specified herein, all proportions are provided on a
25 weight basis.

EXAMPLE I

180 grams of liquid cycloaliphatic resin (ERL 4234 having the structure of epoxycyclohexylspiroepoxy-cyclohexane dioxide) is heated to 140°C under mechanical
30 agitation. 20 grams of 4,4'-sulfonyldiphenol is added to the hot resin under rapid stirring. The admixture is maintained at 140°C for about 40 minutes and until its Brookfield viscosity reaches 560 centipoises. The admixture is then cooled to yield a slightly yellow
35 transparent solid having a resin/diphenol equivalent ratio of about 7.8.



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EXAMPLE II

The process of Example 1 is repeated utilizing the epoxy resin and diphenolic compound in an equivalent ratio of 16.25. Under similar conditions a Brookfield viscosity of 460 is reached after 320 minutes. Upon then cooling, the results were similar.

EXAMPLE III

The process of Example 1 is repeated, substituting 4,4'-(3,3',5,5' tetramethyl) sulfonyl diphenol as the diphenolic compound. An equivalent ratio of 2.24 is employed. The reagents are maintained at 150°C for 3 hours. On cooling, the resin separates as a transparent, but dark straw-colored solid.

EXAMPLE IV

The process of Example 3 is repeated substituting bisphenol A as the diphenolic compound. An equivalent ratio of about 2.5 is utilized. The resultant reaction product is a transparent solid.

EXAMPLE V

85 grams of liquid cycloaliphatic resin (ERL 4221) having the structure of epoxy cyclohexyl methylepoxy-cyclohexane carboxylate is heated to 150°C under mechanical agitation. 15 grams of 4,4'-sulfonyl diphenol is added and the admixture is maintained for 15 minutes at 150°C. On cooling, a solid transparent resin having an equivalent ratio of about 5.3 is separated.

EXAMPLE VI

Using the resin reaction product produced in Example IV, a molding compound is produced by dry blending the following composition:

Cycloaliphatic resin adduct	97 grams
Aluminum acetylacetonate	1 gram
Phenolic resin	3 grams
Amorphous silica	300 grams
Ester lubricant	1 gram



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The composition is then roll milled at 65°C for 3 minutes and then sheeted and chopped.

The chopped composition is transfer molded at 175°C for 2 minutes and then tempered for an additional 4 hours.

5 The molded article has the following properties.

Hot rigidity (175°C)	12 mils
Linear Shrinkage	.004 in/in
Glass transition temperature	180°C
Coefficient of linear expansion,	17×10^{-6} in./in./°C.

10 These properties reflect the superiority of the present compositions as molding compounds, especially for encapsulation of electronic parts.

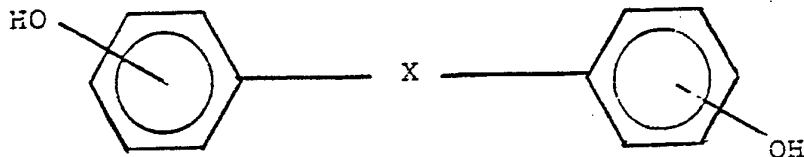
The above mentioned patents and/or publications are incorporated herein by reference. Obviously, other
15 modifications and variations of the present invention are possible in light of the above teachings. It is therefore to be understood that changes may be made in the particular embodiments of the invention described which are within the full intended scope of the invention as defined
20 by the appended claims.



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CLAIMS

1. A thermoset composition comprising the solid reaction product of a liquid cycloaliphatic/epoxy resin having more than one 1,2-epoxy group per molecule with an effective amount of a diphenolic compound.
2. The composition of Claim 1, wherein the diphenolic compound is a bisphenol compound.
3. The composition of Claim 1, wherein the epoxy resin and diphenol compound are reacted in an equivalent ratio of between 40:1 and 2:1.
4. The composition of Claim 1, wherein the diphenol compound has the formula:



5 wherein X is selected from the group consisting of lower aliphatic sulfide and sulfonyl radicals.

5. The composition of Claim 1, which additionally contains an effective amount of curing agent for the reaction product.
6. The composition of Claim 5, wherein the curing agent comprises the combination of metal acetylacetonate and phenolic accelerator.
7. The composition of Claim 5, which additionally contains from 10 to 95% by total weight of filler.



INTERNATIONAL SEARCH REPORT

International Application No PCT/US82/00666

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) ³ According to International Patent Classification (IPC) or to both National Classification and IPC INT. CL. <u>3</u> C08G 59/24, 59/50 U.S. CL. <u>525/523; 528/87, 92, 99, 104</u>																													
II. FIELDS SEARCHED <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black;">Minimum Documentation Searched ⁴</div> <table style="width: 100%; border-collapse: collapse;"> <tr> <th style="width: 20%; text-align: left; border-bottom: 1px solid black;">Classification System</th> <th style="text-align: left; border-bottom: 1px solid black;">Classification Symbols</th> </tr> <tr> <td style="vertical-align: top; padding: 5px;">U.S.</td> <td style="vertical-align: top; padding: 5px;">525/523; 523/400 528/87, 92, 99, 104</td> </tr> </table> <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black;">Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched ⁵</div>			Classification System	Classification Symbols	U.S.	525/523; 523/400 528/87, 92, 99, 104																							
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<div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p>[*] Special categories of cited documents: ¹⁵</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance.</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</p> <p>"Z" document member of the same patent family</p> </div> </div>																													
IV. CERTIFICATION <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%; vertical-align: top; padding: 5px;"> Date of the Actual Completion of the International Search ² 30 JULY 1982 </td> <td style="width: 50%; vertical-align: top; padding: 5px;"> Date of Mailing of this International Search Report ² <div style="font-size: 1.2em; font-weight: bold;">04 AUG 1982</div> </td> </tr> <tr> <td style="vertical-align: top; padding: 5px;"> International Searching Authority ¹ ISA/US </td> <td style="vertical-align: top; padding: 5px;"> Signature of Authorized Officer ¹⁰ E. Nielsen <i>E. Nielsen</i> </td> </tr> </table>			Date of the Actual Completion of the International Search ² 30 JULY 1982	Date of Mailing of this International Search Report ² <div style="font-size: 1.2em; font-weight: bold;">04 AUG 1982</div>	International Searching Authority ¹ ISA/US	Signature of Authorized Officer ¹⁰ E. Nielsen <i>E. Nielsen</i>																							
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