

PATENT SPECIFICATION

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(54) AQUEOUS EPOXY RESIN PAINT COMPOSITION

(71) We, DAI NIPPON TORYO CO., LTD., of No. 38, Nishinoshimonocho, Konohana-ku, Osaka-shi, Japan, a Japanese corporation, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to aqueous epoxy resin paint compositions.

According to the present invention, there is provided an aqueous epoxy resin paint composition comprising an epoxy resin emulsion formed by emulsifying in water containing, as a non-ionic surface active agent, a polyoxyethylene benzylated phenyl ether having an HLB value of at least 15 (A) 98 to 50% by weight of a diglycidyl ether of a bisphenol and (B) 2 to 50% by weight of at least additive resin selected from (1) novolak type epoxy resins, (2) glycidyl esters of dimer or trimer acids and (3) condensates of β -methylepichlorohydrin and a bisphenol.

The diglycidyl ether of a bisphenol (A) used in this invention can be, for example an epoxy resin obtained by condensation between bisphenol A (2,2-bis(4'-hydroxyphenyl)-propane) and epichlorohydrin, and is commercially available. Commercially available diglycidyl ethers of bisphenols are liquid or solid and usually have a molecular weight of 350 to 3750 and an epoxy equivalent of 180 to 3500. Typical instances of these commercially available resins are as follows:

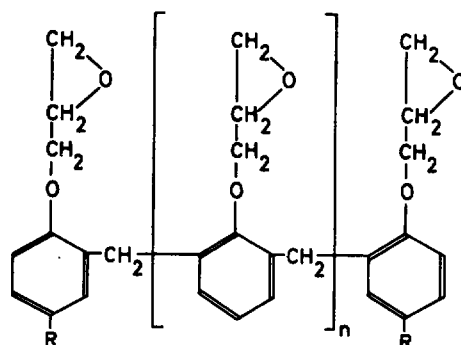
- (1) Epikote 815, Epikote 827, Epikote 834, Epikote 1001, Epikote 1004, Epikote 1007 and Epikote 1009 (trademarks for products manufactured by Shell Chemical Co.)
- (2) DER 330, DER 331, DER 334, DER 337, DER 661, DER 664, DER 667 and DER 669 (trademarks for products manufactured by Dow Chemical Co.)
- (3) Araldite GY250, Araldite GY252, Araldite GY260, Araldite GY280, Araldite GY6071, Araldite GY6084, Araldite GY6097 and Araldite GY6099 (trademarks for products manufactured by Ciba Limited)
- (4) ERL 2774 and ERL 2795 (trademarks for products manufactured by Union Carbide Corporation)
- (5) Epiclon 850 (trademark for product manufactured by Dainippon Ink and Chemicals Incorp.)

These commercially available resins can be used singly, or mixtures of two or more of them can also be employed.

The diglycidyl ether of a bisphenol is used as a vehicle. Use of a liquid vehicle epoxy resin is preferred because it is readily emulsified, but when the vehicle epoxy resin is solid, it can be emulsified or dispersed in water by mixing it with another liquid epoxy resin to dissolve it or by melting it under heating.

In this invention, in combination with the above vehicle epoxy resin (A) is used as an additive resin (1), (2) or (3).

As the additive resin (1), there can be used, for example, those having the following structure:

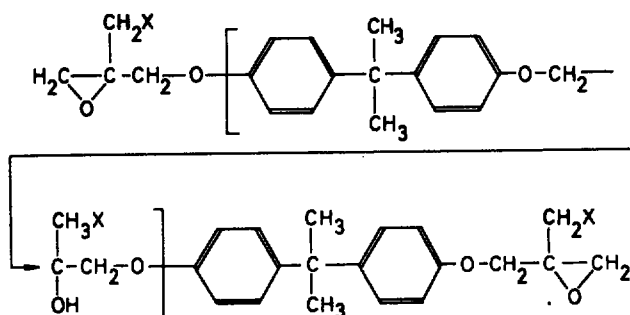


wherein n is a number of from 0 to 2 and R stands for a hydrogen atom or a methyl, ethyl or propyl group.

As commercially available additive resins (1), there can be mentioned Epikote 152 (product of Shell Chemical Co. having an epoxy equivalent of 172—179), Epikote 154 (product of Shell Chemical Co. having an epoxy equivalent of 176—181), DEN 431 (product of Dow Chemical Co. having an epoxy equivalent of 172—179), DEN 438 (product of Dow Chemical Co. having an epoxy equivalent of 175—182), EPN 1138 (product of Ciba Limited having an epoxy equivalent of 172—179), etc. These resins are generally prepared by reacting a novolak type phenolic acid-formaldehyde resin with epichlorohydrin.

The additive resin (2) is usually prepared by reacting a dimer or trimer acid with epichlorohydrin (the preparation process is disclosed in, for example, Japanese Patent Publication No. 7454/62 published on July 7, 1962 by Shell International Research Maatschappij N.V., Netherlands), and as commercially available epoxy resins of this type, there can be mentioned, for example, Epikote 871 (product of Shell Chemical Co. having an epoxy equivalent of 390—470) and Epikote 872 (product of Shell Chemical Co. having an epoxy equivalent of 600—700).

The additive resin (3) used in this invention can be, for example, a resin having the following structure:



wherein n is 0 to 3 and X is hydrogen or chlorine. The epoxy resin (3) can be prepared by, for example, condensation of β -methyl epichlorohydrin or β -chloromethyl epichlorohydrin with bisphenol-A. The preparation process is disclosed in, for example, U.S. No. 3,533,165 (Patented on January 5, 1971 by Chiyoda Kako Kensetsu Kabushiki Kaisha).

As a commercially available resin of this type, there can be mentioned, for example, Epiclon 800 (product of Dianippon Ink and Chemicals Incorp. having an epoxy equivalent of 195—225).

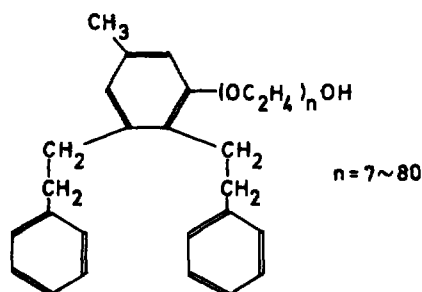
In this invention, in order to prevent crystallization, there is adopted the combined use of the above mentioned vehicle resin with at least one member selected from the foregoing resins (1) to (3), each of which has a compatibility with the vehicle resin and a structure similar to that of the vehicle resin but is different therefrom in a part of the structure.

The additive epoxy resin (1), (2) or (3) is incorporated in the vehicle resin in such an amount that the additive resin accounts for 2 to 50% by weight of the sum of the vehicle resin and the additive epoxy resin. When the amount of the additive epoxy resin (1), (2) or (3) is less than 2% by weight, no substantial effect of

preventing crystallization can be obtained, and when the amount of the additive resin exceeds 50% by weight, the saline resistance, water resistance and moisture resistance of the resulting film are reduced.

As pointed out above, in this invention, by incorporating in the vehicle epoxy resin a suitable amount of at least one of the additive epoxy resins (1) to (3), each of which has a compatability with the vehicle epoxy resin and a structure similar to that of the vehicle epoxy resin but is quite different therefrom in a part of the structure, it is possible to inhibit arrangement of the molecules of the vehicle epoxy resin, which results in crystallization, and thus prevent crystallization even at low temperatures.

The mixture of the above-mentioned vehicle epoxy resin with the above-mentioned additive epoxy resin (1), (2) or (3) is emulsified in water in the presence of a polyoxyethylene benzylated phenyl ether having an HLB value of at least 15 as a non-ionic surface active agent, according to customary procedures. The use of this non-ionic surface active agent aids in preventing crystallization. There can be used, for example, a polyoxyethylene benzylated phenyl ether having the structure



and having an HLB value of at least 15.

As the polyoxyethylene benzylated phenyl ether, there can be used, for example, such commercially available products as Newcol 714 (HLB = 15) and Newcol 723 (HLB = 16.6) (trademarks for products manufactured by Nippon Nyukazai Co., Ltd.), and Emulgen A-500 (HLB = 18) (trademark for the product manufactured by Kao-Atlas Co., Ltd.).

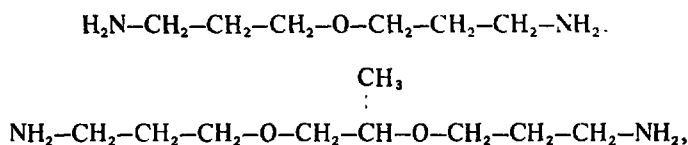
For emulsification of the epoxy resin, the polyoxyethylene benzylated phenyl ether having an HLB value of at least 15 is preferably used in an amount of 0.1 to 20% by weight based on the epoxy resin.

As pointed above, undesired phenomenon of crystallization can be effectively prevented in this invention by adding to the vehicle epoxy resin at least one member selected from the above additive resins (1) to (3) having a compatibility with the vehicle epoxy resin and a structure similar to that of the vehicle epoxy resin but being quite different therefrom in a part of the structure, and by emulsifying the epoxy resin mixture in water with a polyoxyethylene benzylated phenyl ether having an HLB value of at least 15. The above two means are adopted in combination, and the crystallization-preventing effect is greatly enhanced.

In the composition of this invention, a curing agent should be added in order to cure the above-mentioned epoxy resin. As the curing agent, there can be employed known compounds such as water-soluble and water-dispersible polyamide resins, amine adducts, polyamines and the like.

When it is intended to improve the film-forming property especially under high-humidity conditions, good results are obtained when a polyamide resin prepared from a dimer acid and an ether diamine, preferably such polyamide resin having an amine value of 250 to 500, is employed as the curing agent.

The dimer acid to be used for forming the above polyamide resin is a product prepared by employing an unsaturated fatty acid having 18 carbon atoms as the starting material, and the product is composed mainly of a dimer acid and includes small amounts of a trimer acid and a monomer acid. As the ether diamine to be used for forming the above polyamide resin, there can be mentioned, for example,

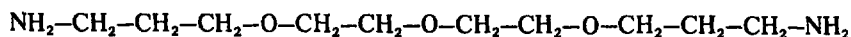
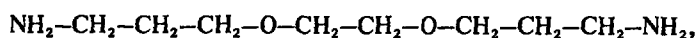


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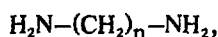


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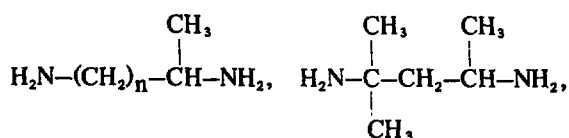
The intended polyamide resin can be obtained by reacting such ether diamine with the dimer acid at 150 to 250°C. for 1 to 4 hours according to customary procedures.

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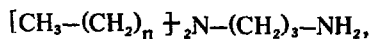
In this invention, the intended polyamide resin, preferably having an amine value of 250 to 500, can also be prepared by substituting a part of the ether diamine by one or more of aliphatic amines such as



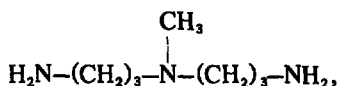
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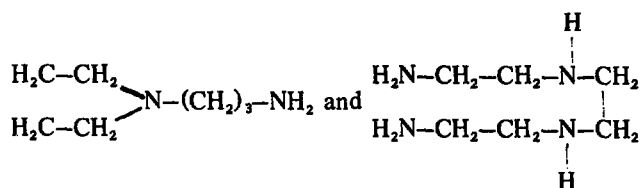
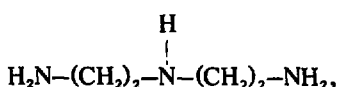
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It is preferred that the aliphatic amine be used in an amount of 0.1 to 4 moles per mole of the ether diamine.

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In order to increase the molecular weight of the polyamide resin curing agent, it is possible to add and react a chain extender at the reaction between the dimer acid and ether diamine or the reaction among the dimer acid, ether diamine and aliphatic amine. As the chain extender, there can be used, for example, acrylonitrile, methyl methacrylate, formaldehyde, a polyglycol type epoxy resin or a glycerin type epoxy resin. When the so obtained high-molecular-weight polyamide resin, preferably having an amine value of 250 to 500, is used as the curing agent, the resulting composition is excellent in film-forming property under high-humidity conditions and gives a film excellent in anti-cracking property.

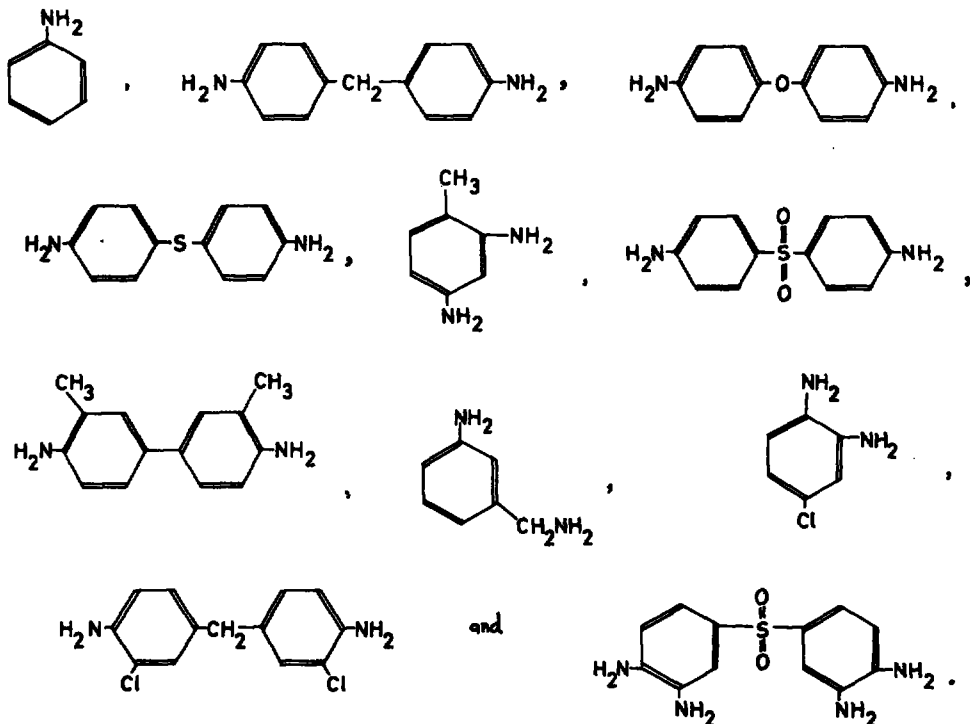
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Further, an aromatic amine compound can be used in combination with the above polyamide resin. As such aromatic amine compound, the following compounds can be mentioned:

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- (1) Aromatic amines having a structure such as shown below (when these amines are solid at room temperature, they are used in the state melt-mixed with the above polyamide resin or other curing agent mentioned below):



- (2) Reaction products between an aromatic amine and an epoxy resin obtained according to customary procedures (aromatic amine adducts).

- (3) Polyamide resins prepared by employing an aromatic amine.

When such aromatic amine compound is used in combination with the above-mentioned polyamide resin curing agent, the moisture resistance, saline dipping resistance and salt spraying resistance are highly improved in the resulting film.

It is preferred that the ration of the above aromatic amine to the polyamide resin derived from a dimer acid and an ether diamine be with a range of from 5/95 to 50/50 as expressed in the non-volatile component weight ratio.

The so obtained curing agent or curing agent composition is mixed with an epoxy resin emulsion according to customary procedures to form an aqueous epoxy resin paint composition of this invention.

In this invention, it is preferred that the mixing ratio of the epoxy resin and curing agent be such that the ratio of the epoxy groups to the active hydrogen group is within a range of form 0.5 to 1.5.

The aqueous epoxy resin paint composition of this invention may further comprise additives customarily used in this field, such as organic pigments, inorganic pigments, surface active agents and thickeners.

The use of a conventional anti-corrosive pigment of the chromate type is restricted because of the toxicity of chromium ions dissolved out of the film; it is preferred that at least one member selected from the group consisting of molybdcic acid salts of strontium, calcium, potassium, iron (divalent), barium and ammonium be used instead of the chromate type anti-corrosive pigment.

The molybdate type anti-corrosive pigment is non-toxic and its corrosion resistance is superior or comparable to that of the conventional chromate type anti-corrosive pigment, and it is further characterized in that it promotes the film-forming property of the resulting composition under high-humidity conditions and even when the composition is applied in a large thickness, the curing is accomplished uniformly throughout the film so that the degree of curing does not differ between the surface portion and the interior of the film (this property will hereinafter be referred to as "curing uniformity").

It is preferred that this molybdate type anti-corrosive pigment be incorporated in the paint composition in an amount of 0.1 to 20% by weight. If the amount incorporated of the pigment is smaller than 0.1% by weight, no substantial anti-corrosive effect is obtained and the film-forming property under high-humidity conditions is not promoted. In contrast, if the amount incorporated of the pigment is larger than 20% by weight, the pigment concentration increases and tendency of reduction of physical properties of the film is observed.

In view of the rust-preventive effect and from the economical viewpoint, it is especially preferred that the molybdate type anti-corrosive pigment be incorporated in an amount of 5 to 10% by weight.

It has been found that especially good results are obtained when among the foregoing molybdate type anti-corrosive pigments, strontium molybdate or calcium molybdate obtained by calcining a mixture of molybdenum oxide and calcium carbonate or strontium carbonate at 600 to 800°C. is chosen and used.

In the aqueous epoxy resin paint composition of this invention, at the time of application the main components (the epoxy resin emulsion, the pigment, etc.) are mixed with the curing agent, the mixture is diluted appropriately with water and the resulting composition can be applied to a substrate to be coated according to customary coating methods, for example, brush-coating, spray-coating and air spray-coating methods. Since the paint composition of this invention has a very high thixotropic property, a thick coating having a thickness of more than 100 μ can be obtained by one coating.

This invention will now be illustrated by the following Examples, in which all "parts" and "%" are by weight.

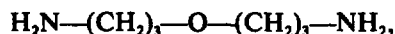
Example 1.

A condensate of β -methylepichlorohydrin and a bisphenol (Epilcon 800 having an epoxy equivalent of 195 to 225), a novolak type epoxy resin (Epikote 152 having an epoxy equivalent of 172 to 179) or a glycidyl ester of a dimer or trimer acid (Epikote 871 having an epoxy equivalent of 390 to 470) was incorporated into a diglycidyl ether of a bisphenol (Epikote 828) in amounts indicated in Table 1. 20 parts of the so prepared resin mixtures were emulsified in 5 parts of water with a polyoxyethylene benzylated phenyl ether having an HLB value of 16.6. The so obtained emulsions were mixed with a composition comprising 10 parts of red iron oxide, 35 parts of talc and 30 parts of water to form main components (samples 1 to 9).

The state of formation and growth of crystals was examined with respect to each of the so obtained components. The results are shown in Table 1.

From the results obtained, it will readily be understood that crystallization can be effectively prevented by using a specific surface active agent and this effect can be further enhanced by using an epoxy resin of a specific structure in combination with the diglycidyl ether of a bisphenol.

1 mole of a dimer acid was mixed with 12 moles of an ether diamine having the structural formula



and the mixture was subjected to condensation reaction at 160°C. When the reaction had consumed all water present, the temperature was further elevated to 200 to 250°C and the condensation product was maintained at this temperature for 3 hours, to obtain a polyamide resin of an amine value of 350 to be used as a curing agent.

The so obtained curing agent was incorporated into the main component of the above samples Nos. 1, 5 and 8 so that the ratio of the epoxy groups to the active hydrogen groups was 1.0. Each paint exhibited excellent film-forming properties even when applied under high-humidity conditions and gave a film excellent in both physical and chemical properties.

Example 2.

In the main component of Example 11 of Application No. 48224/76, the diglycidyl ether of a bisphenol was replaced by the mixture used in sample No. 3 of Example 1 above, 5 parts of calcium molybdate was used as the anti-corrosive pigment and the amount of talc was changed to 15 parts. Crystallization at low temperatures did not occur in the resulting aqueous epoxy resin emulsion.

A mixture of the polyamide resin obtained in Example 11 of Application No.

48224/76 (Serial No. 1,472,199) with an aromatic amine adduct was used as the curing agent.

The main component was mixed with the curing agent so that the epoxy groups in the emulsion and the active hydrogen atom groups in the curing agent were equivalent to each other. The resulting paint composition was tested and the results obtained are shown in Table 2.

Example 3.

In the main component of Example 11 of Application No. 48224/76, the diglycidyl ether of a bisphenol was replaced by the mixture used in sample No. 5 of Example 1 above, and it was mixed with 9 parts of red iron oxide, 15 parts of talc and 2 parts of ammonium molybdate.

20% by weight of the polyamide resin prepared in Example 13 of Application No. 48224/76 (Serial No. 1,472,199) was replaced by an amine adduct obtained by reacting a diglycidyl ether of a bisphenol (Epikote 828) with metaphenylene diamine at a ratio of 1:2, and the resulting mixture was used as a curing agent.

The above main component was mixed with the curing agent so that the epoxy groups in the emulsion and the active hydrogen atom groups in the curing agent were equivalent to each other to obtain an aqueous epoxy resin paint composition. The paint composition was tested, and the results obtained are shown in Table 2.

Example 4.

An aqueous epoxy resin paint composition was prepared in the same manner as in Example 2 except that the epoxy resin mixture was replaced by that used in sample No. 8 of Example 1, the amount of talc was changed to 15 parts and 5 parts of strontium molybdate was used as an anti-corrosive pigment. The paint composition was tested, and the results obtained are shown in Table 2.

TABLE 1

Sample	Additive Resin		Amount of Epikote 828	State of Formation and Growth of Crystals*	Remarks
	Kind	Amount			
Sample No. 1	Epiclon 800	2%	98%	not observed	this invention
Sample No. 2	ditto	5%	95%	not observed	this invention
Sample No. 3	ditto	10%	90%	not observed	this invention
Sample No. 4	Epikote 152	5%	95%	not observed	this invention
Sample No. 5	ditto	10%	90%	not observed	this invention
Sample No. 6	ditto	15%	85%	not observed	this invention
Sample No. 7	Epikote 871	10%	90%	not observed	this invention
Sample No. 8	ditto	15%	85%	not observed	this invention
Sample No. 9	ditto	20%	80%	not observed	this invention

Note:

* The state of formation and growth of crystals was examined after samples were allowed to stand still for 12 months under the same conditions as described in the note to Table 1 of Application No. 48224/76. (Serial No. 1472199.)

TABLE 2

Results of Tests

Sample	Film-Forming Property under High-Humidity 1)	Curing Uniformity of Film 2)	Salt Spraying Test 3)	Moisture Resistance 4)	Saline Dipping Test 5)	Water Resistance 6)
Example 2	◎	◎	◎	◎	◎	◎
Example 3	◎	◎	◎	○	◎	◎
Example 4	◎	◎	◎	◎	◎	◎

◎ : very good, ○ : good.

- Notes) 1) After the main component was mixed with the curing agent, the composition was applied on the inner face of a certain vessel and the film was allowed to stand still in the sealed state for 7 days. After passage of 7 days the vessel was opened and the state of the film was examined with the fingers. Namely, it was checked whether the film was sticky or the film adhered to the fingers.
- 2) The paint was coated in a thickness of about 1 cm on a substrate, and after passage of 7 days, the film was cut with a knife to examine the curing uniformity.
- 3) A 3% aqueous solution of NaCl was sprayed at 35°C for 500 hours.
- 4) The film was allowed to stand still at a temperature of 50°C and a relative humidity of 100% for 500 hours.
- 5) The film was immersed in a 5% aqueous solution of NaCl for 30 days.
- 6) The film was immersed in water for 30 days.

(18)

Attention is drawn to our copending Application No. 48224/76 (Serial No. 1,472,199), which describes and claims an aqueous epoxy resin paint composition comprising an epoxy resin emulsion formed by emulsifying, in water containing a non-ionic surface active agent, a diglycidyl ether of a bisphenol, there being incorporated into the epoxy resin emulsion a curing agent comprising (i) a polyamide resin derived from a dimer acid and an ether diamine, (ii) a polyamide resin derived from a dimer acid, an ether diamine and an aliphatic amine, (iii) a polyamide resin derived from a dimer acid, an ether diamine and a chain extender, or (iv) a polyamide resin derived from a dimer acid, an ether diamine, an aliphatic amine and a chain extender.

WHAT WE CLAIM IS:—

1. An aqueous epoxy resin paint composition comprising an epoxy resin emulsion formed by emulsifying in water containing, as a non-ionic surface active agent, a polyoxyethylene benzylated phenyl ether having an HLB value of at least 15 (A) 98 to 50% by weight of a diglycidyl ether of a bisphenol and (B) 2 to 50% by weight of at least additive resin selected from (1) novolak type epoxy resins, (2) glycidyl esters of dimer or trimer acids and (3) condensates of β -methylepichlorohydrin and a bisphenol.

2. A composition as claimed in claim 1, wherein the epoxy resin composition contains a curing agent.

3. A composition as claimed in claim 2, wherein the curing agent comprises (i) a polyamide resin derived from a dimer acid and an ether diamine, (ii) a polyamide resin derived from a dimer acid, an ether diamine and an aliphatic diamine, (iii) a polyamide derived from a dimer acid, an ether diamine and a chain extender, or (iv) a polyamide derived from a dimer acid, an ether diamine, an aliphatic diamine and a chain extender.

4. A composition as claimed in claim 3, wherein the curing agent further comprises (i) an aromatic amine, (ii) an aromatic amine/epoxy resin adduct, or (iii) a polyamide derived from an aromatic amine.

5. A composition as claimed in claim 4, wherein the weight ratio of the further component of the curing agent to the polyamide resin of the curing agent is from 5:95 to 50:50.

6. A composition as claimed in any of claims 1 to 5, further comprising from 0.1 to 20% by weight, based on the total weight of the composition, of at least one member selected from molybdcic acid salts of strontium, calcium, potassium, iron (divalent), barium and ammonium.

7. An aqueous epoxy resin paint composition in accordance with claim 1 or 2, substantially as described in any of the foregoing Examples.

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Attention is drawn to our copending Application No. 48224/76 (Serial No. 1,472,199), which describes and claims an aqueous epoxy resin paint composition comprising an epoxy resin emulsion formed by emulsifying, in water containing a non-ionic surface active agent, a diglycidyl ether of a bisphenol, there being incorporated into the epoxy resin emulsion a curing agent comprising (i) a polyamide resin derived from a dimer acid and an ether diamine, (ii) a polyamide resin derived from a dimer acid, an ether diamine and an aliphatic amine, (iii) a polyamide resin derived from a dimer acid, an ether diamine and a chain extender, or (iv) a polyamide resin derived from a dimer acid, an ether diamine, an aliphatic amine and a chain extender.

WHAT WE CLAIM IS:—

1. An aqueous epoxy resin paint composition comprising an epoxy resin emulsion formed by emulsifying in water containing, as a non-ionic surface active agent, a polyoxyethylene benzylated phenyl ether having an HLB value of at least 15 (A) 98 to 50% by weight of a diglycidyl ether of a bisphenol and (B) 2 to 50% by weight of at least additive resin selected from (1) novolak type epoxy resins, (2) glycidyl esters of dimer or trimer acids and (3) condensates of β -methylpichlorohydrin and a bisphenol.

2. A composition as claimed in claim 1, wherein the epoxy resin composition contains a curing agent.

3. A composition as claimed in claim 2, wherein the curing agent comprises (i) a polyamide resin derived from a dimer acid and an ether diamine, (ii) a polyamide resin derived from a dimer acid, an ether diamine and an aliphatic diamine, (iii) a polyamide derived from a dimer acid, an ether diamine and a chain extender, or (iv) a polyamide derived from a dimer acid, an ether diamine, an aliphatic diamine and a chain extender.

4. A composition as claimed in claim 3, wherein the curing agent further comprises (i) an aromatic amine, (ii) an aromatic amine/epoxy resin adduct, or (iii) a polyamide derived from an aromatic amine.

5. A composition as claimed in claim 4, wherein the weight ratio of the further component of the curing agent to the polyamide resin of the curing agent is from 5:95 to 50:50.

6. A composition as claimed in any of claims 1 to 5, further comprising from 0.1 to 20% by weight, based on the total weight of the composition, of at least one member selected from molybdic acid salts of strontium, calcium, potassium, iron (divalent), barium and ammonium.

7. An aqueous epoxy resin paint composition in accordance with claim 1 or 2, substantially as described in any of the foregoing Examples.

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