

Curable coating composition

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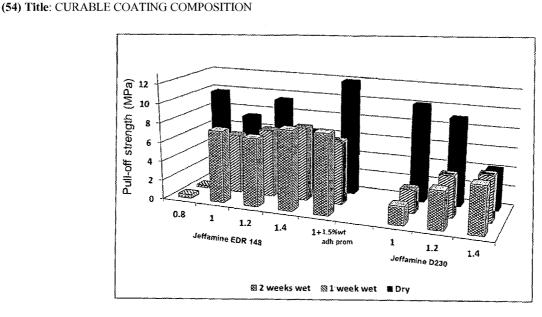
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(57) Abstract: The invention relates to a curable coating composition comprising an aliphatic epoxy resin comprising two or more epoxide groups and a compound having two or more amine functionalities, wherein two of the two or more amine functionalities are connected to each other by a chain comprising a segment consisting of an alkyl ether or a poly(alkyl ether), the alkyl ether or poly(al-kyl ether) having alkylene units of 2 to 5 carbon atoms or a mixture of such alkylene units, the poly(alkyl ether) having from 2 to 20 ether oxygen atoms in the chain.

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CURABLE COATING COMPOSITION

The invention relates to a curable coating composition, a coating obtainable by curing such curable coating composition and an object comprising such coating.

Aluminum and aluminum alloys have characteristic properties that make them useful for application in aircrafts, in particular their high strength stiffness to weight ratio and their good formability. However, aluminum and aluminum alloys are susceptible to corrosion. The aircraft industry is therefore challenged to provide protective coatings capable of protecting aluminum and aluminum alloys used in aircrafts.

Many protective coatings for aluminum alloys have been developed to meet the needs in the aircraft industry. Frequently used materials for such coatings are amine-cured epoxy resins. These resins have been

- 15 commercially available for more than 50 years. Their good mechanical properties, strong adhesion and excellent chemical resistance have made them attractive as an adhesive or protective coating for many major industrial applications, among which the aircraft industry. However, a disadvantage of these resins - when applied as a coating on aluminum or aluminum alloys -
- 20 is their vulnerability to water. When exposed to aqueous conditions, water can accumulate at the aluminum-coating interface. This then leads to corrosion, blistering and delamination of the coating from the substrate as described by A. Foyet et al. in ECS Transactions, 25 (29) 31-39 (2010).

Given the relevance of the performance of a coating in a certain 25 application under dry conditions on the one hand and aqueous conditions on the other hand, the adhesion under these conditions is in the art usually referred to as "dry adhesion" and " wet adhesion", respectively. Other materials that are used as protective coatings are e.g. based on chromates. These coatings have a good anti-corrosive performance, but their toxicity is a 30 main disadvantage from an environmental point of view as is generally known.

A relatively new technique is plasma deposition of coatings on a substrate. Although promising, a major disadvantage is the vast amount of energy required for deposition.

It is therefore an object of the invention to provide a coating having a strong dry and wet adhesion to aluminum and aluminum alloys.

It is in particular an object of the invention to provide a coating that is less vulnerable to water than coatings known in the art. Herewith is understood that, when exposed to aqueous conditions, such coating exhibits a stronger wet adhesion to its aluminum/aluminum alloy substrate than coatings known in the art, or is at least less susceptible to blistering and delamination than coatings known in the art.

It is also an object of the invention to provide a coating that provides better protection of aluminum/aluminum alloys against water than coatings known in the art, and/or that provides better protection against corrosion of aluminum alloys than coatings known in the art.

It is in particular an object of the invention to provide a coating that is suitable for application on aircrafts.

It is also an object of the invention to provide a coating that can be applied to a substrate in an energy-efficient manner. A further object of the 20 invention is to provide a curable coating composition and/or a coating that comprises less toxic constituents than coating compositions and/or coatings known in the art, respectively.

It has now been found that at least one of these objects can, at least in part, be reached when a curable coating composition is used comprising a particular combination of an epoxide resin and a curing agent.

Therefore, the invention relates to a curable coating composition comprising

- an aliphatic epoxy resin comprising two or more epoxide groups;

a compound having two or more amine functionalities, wherein two of the two or more amine functionalities are connected to each other by a chain comprising a segment consisting of an alkyl ether or a poly(alkyl ether), the alkyl ether or poly(alkyl ether) having alkylene units of 2 to 5 carbon atoms or

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a mixture of such alkylene units, the poly(alkyl ether) having from 2 to 20 ether oxygen atoms in the chain.

The chain that connects two amine functionalities may in particular consist of the segment as defined in claim 1.

The aliphatic epoxy resin usually has an Epoxide Equivalent Weight of 100-1000, preferably of 150-500, more preferably of 180-240 g/eg. The Epoxide Equivalent Weight (EEW) of a resin is defined as the weight of the resin in grams that contains one gram equivalent of epoxy.

In a preferred embodiment, the aliphatic epoxy resin of a curable coating composition of the invention is a cycloaliphatic epoxy resin. In particular, such cycloaliphatic epoxy resin comprises one or more cyclohexyl moieties.

In another preferred embodiment, the aliphatic epoxy resin is the diglycidyl ether of a diol. Preferably, such diol is selected from the group of cyclohexanediols, cyclohexanedimethanols and bis(hydroxycyclohexyl)alkanes. More preferably, the diol is 2,2-bis(4-hydroxycyclohexyl)propane (also known as "hydrogenated Bisphenol A"), *i.e.* the aliphatic epoxy resin in a curable coating composition of the invention is then 2,2-bis(4-hydroxycyclohexyl)propane diglycidylether.

20 This compound is commercially available under the name "Eponex 1510".

A coating composition of the invention may comprise - next to the aliphatic epoxy resin - one or more further epoxy resins. Such one or more further epoxy resins may comprise a further aliphatic epoxy resin selected from the group of 1,4-butanediol diglycidyl ether (BD-DGE), 1,6-hexanediol diglycidyl ether (HD-DGE) and poly(ethylene glycol) diglycidyl ether (PEG-DGE).

A curable coating composition of the invention comprises a compound having two or more amine functionalities, wherein two of the two or more amine functionalities are connected to each other by a chain as defined in claim 1. The two amine functionalities that are connected by the chain usually are primary amine groups (*i.e.* $-NH_2$) or secondary amine groups (*i.e.* -RNH, R representing an alkyl group).

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In the art of amine-cured epoxy resins, the amine is usually regarded as having a certain amount of "active NH's". The hydrogen atom in an active NH is "active" since it is capable of being substituted by a carbon atom of a reacting epoxide group. A primary amine group is considered to contain two active NH's and a secondary amine group is considered to contain one active NH. For example, when the compound with two or more amine functionalities in a composition of the invention has two secondary amine groups, it has two active NH's. When it has two primary amine groups, it has four active NH's. When it has one primary and one secondary amine group, it has three active NH's.

The alkylene units in the alkyl ether or poly(alkyl ether) are selected from the group of alkylene units having 2–10 carbon atoms. Preferably, they are selected from the group of alkylene units having 2–5 carbon atoms, more preferably from the group of alkylene units having 2–4 carbon atoms.

The alkylene units may be linear or branched. Linear alkylene units having 2–4 carbon atoms are ethylene (- $(CH_2)_2$ -), propylene (- $(CH_2)_3$ -) and butylene (- $(CH_2)_4$ -). Preferably, linear alkylene units having 2–4 carbon atoms are ethylene units.

Branched alkylene units having 2–4 carbon atoms are 1,2-propylene (-CH(CH₃)-CH₂-), 1,2-butylene (-CH(C₂H₅)-CH₂-), 1,3-butylene (-CH₂-CH(CH₃)-CH₂-) and 2,3 butylene (-CH(CH₃)-CH(CH₃)-).

The alkylene units in the alkyl ether or poly(alkyl ether) may be a plurality of identical alkylene units or a mixture of different alkylene units. Thus, the alkyl ether or poly(alkyl ether) may comprise two or more different types of alkylene units. For example, a chain may contain one or more ethylene units as well as one or more propylene units in the same chain. The different alkylene units in a chain may be present in an alternating fashion, but they may also be present in a different order such as a random order.

An alkyl ether chain segment having two identical units is for example: $-(CH_2)_x$ -O- $(CH_2)_x$ -. Particular examples of such segments are chains wherein x=2 or wherein x=3

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A poly(alkyl ether) chain segment having a plurality of identical units is for example: $-(CH_2)_x$ -O- $(CH_2)_x$ -O- $(CH_2)_x$ -. Particular examples of such segments are chains wherein x=2 or wherein x=3.

An alkyl ether chain segment having a mixture of different alkylene units is for example: $-(CH_2)_x$ -O- $(CH_2)_y$ -. In this example, x and y may independently be chosen from the range of 2 to 5 (with x and y being different). Particular examples of such segments are chains wherein x=2 and y=3, or wherein x=2 and y=4.

A poly(alkyl ether) chain segment having a mixture of different 10 alkylene units is for example: $-(CH_2)_x$ -O- $(CH_2)_y$ -O- $(CH_2)_x$ -. In this example, x and y may independently be chosen from the range of 2 to 5 (with x and y being different). Particular examples of such segments are chains wherein x=2 and y=3, or wherein x=3 and y=2, or wherein x=4 and y=2, or wherein x=4 and y=3.

The poly(alkyl ether) chain segment contains 2–20 ether oxygen atoms in the chain. Preferably, the number of ether oxygen atoms in the chain is 2–5, more preferably it is 2.

In a preferred curable coating composition of the invention, the compound having two or more amine functionalities is $H_2N-(CH(CH_3)-CH_2-O)_x-(CH_2)-CH(CH_3)-NH_2$. These compounds are commercially available under the name "Jeffamine D-Series". In this series, x may for example be 2 or 3.

In a preferred embodiment, a curable coating composition according to the invention comprises

- 2,2-bis(4-hydroxycyclohexyl)propane diglycidylether and

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 H₂N-(CH(CH₃)-CH₂-O)_x-(CH₂)-CH(CH₃)-NH₂, wherein x=2 or 3. The two amine functionalities may be connected to each other by a linear chain. For the purpose of the invention, the term "linear chain" has the meaning and scope it normally has in the art: a chain that has no side-groups or branches. A linear chain may contain monovalent atoms on the chain.

In another preferred curable coating composition of the invention, the compound having two or more amine functionalities is $H_2N-(CH_2)_2-O-($

compounds are commercially available under the names "Jeffamine EDR-148" and "Jeffamine EDR-176", respectively.

In another preferred embodiment, a curable coating composition according to the invention comprises

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- 2,2-bis(4-hydroxycyclohexyl)propane diglycidylether and

- $H_2N-(CH(CH_3)-CH_2-O)_x-(CH_2)-CH(CH_3)-NH_2$.

In yet another preferred embodiment, a curable coating composition according to the invention comprises

2,2-bis(4-hydroxycyclohexyl)propane diglycidylether and

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- $H_2N-(CH_2)_2-O-(CH_2)_2-O-(CH_2)_2-NH_2$ or $H_2N-(CH_2)_3-O-(CH_2)_2-O-(CH_2)_3-NH_2$

A coating composition of the invention may further comprise – next to the compound having two or more amine functionalities as defined in the invention – one or more compounds having one or more amine functionalities.

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A coating composition of the invention may in particular comprise one or more further compounds having two or more amine functionalities. Such coating composition may for example comprise a mixture of two or more different compounds of the type $H_2N-(CH(CH_3)-CH_2-O)_x-(CH_2)-CH(CH_3)-NH_2$. Such mixture may then comprise two of such compounds, *i.e.* one compound wherein x=2 and one compound wherein x=3, in a ratio of *e.g.* 1/1. It may also comprise three of such compounds, *i.e.* one compound wherein x=2, one compound wherein x=3 and one compound wherein x=4.

In case of a mixture of compounds with different values for certain repeating units, a weighted average value of x may be assigned to that 25 mixture. For example, x may be approximately 2.5 or approximately 6.1. These compounds are commercially available under the names "Jeffamine D-230" and "Jeffamine D-400", respectively.

The epoxy/active NH ratio (*i.e.* the ratio of the total molar amount of epoxy groups to the total molar amount of active NH's in the mixture) may vary in curable coating compositions according to the invention. The epoxy/active NH ratio is usually from 0.5/1.0 to 2.0/1.0, preferably it is from 0.8/1.0 to 1.8/1.0. Preferred ratios in this range are for example 0.85/1.0,

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0.9/1.0, 0.95/1.0, 1.0/1.0, 1.05/1.0, 1.1/1.0, 1.15/1.0, 1.20/1.0, 1.25/1.0, 1.30/1.0, 1.35/1.0, 1.40/1.0, 1.50/1.0, 1.60/1.0 and 1.70/1.0.

A curable coating composition of the invention may comprise an adhesion promoter. An adhesion promoter is a substance that is applied to a substrate to improve the adhesion of a coating to the substrate. Typical adhesion promoters that are suitable for the coating of aluminum and aluminum alloys are based on silanes and silicones with hydrolyzable groups on one end of their molecules. These groups can react with moisture to yield silanol groups, which in turn react with or adsorb to inorganic surfaces

10 enabling the formation of strong bonds. At the other ends of the molecules, reactive, and non-hydrolyzable groups are present that are capable of reacting with one or more component(s) of the curable coating composition. An adhesion promoter may be selected from the group of (3glycidoxypropyl)trimethoxysilane, (3-aminopropyl)triethoxysilane and (3-15 aminopropyl)trimethoxysilane.

When an adhesion promoter is used in a curable coating composition of the invention, it is usually present in an amount of 0.01-10 wt%, based on the total weight of the curable coating composition.

It has in particular been found that when an adhesion promoter is 20 present in a coating of the invention, the coating has a higher dry adherence than when the adhesion promoter is absent in the coating. Further, the wet adherence of a coating comprising an adhesion promoter is comparable to that of a coating that lacks an adhesion promoter.

The invention further relates to a coating obtainable by curing a 25 curable coating composition of the invention.

The curing process comprises exposing the curable coating composition to a temperature of more than 100 °C during at least 4 hours. Usually, the temperature is increased stepwise from room temperature (20 °C) to 150 °C. If applied on aluminum the maximum temperature should not be higher than 110 °C. The duration of the curing may last up to 10 days. The person skilled in the art is able to find the appropriate conditions for the curing of a certain curable composition by routine experimentation.

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The invention further relates to an object comprising a coating obtainable by curing a curable coating composition of the invention. An object that comprises a coating according to the invention is selected from the group of airplanes, helicopters, boats, yachts, cars and bikes. A coating of the invention may in particular be present on constructive aluminum parts such as window frames or on external window blinds such as those with horizontal or vertical slats.

When an object comprises a coating of the invention, the coating is preferably present on a surface area (of that object) that comprises aluminum or an aluminum alloy. This means that the coating adheres to aluminum or an aluminum alloy. The aluminum alloy may be selected from the group of Al 2024 and Al 7075.

It has been found that coatings of the invention are capable to adhere strongly to a surface comprising aluminum or an aluminum alloy. The strength of this adherence is comparable to that of coatings known in the art.

Surprisingly, it has also been found that coatings of the invention are substantially less vulnerable to water than coatings known in the art. Accordingly, coatings of the invention have a substantially higher wet adhesion.

The adherence strength of coatings known in the art to aluminum/aluminum alloy substrates usually decreases significantly during exposure to aqueous conditions. The adherence strength of coatings of the invention to aluminum/aluminum alloy substrates, however, hardly changes during exposure to aqueous conditions. This means that when coatings of the invention are exposed to aqueous conditions, these coatings exhibit a stronger wet adhesion to the aluminum/aluminum alloy substrate than coatings known in the art that are exposed to the same aqueous conditions and during the same period of time. As a result, the coatings are also much less susceptible to blistering and delamination than coatings known in the art.

The high water-resistance of coatings of the invention is very advantageous for application of the coating on aircrafts. The conditions under which aircrafts operate are often aqueous conditions. Under such conditions, unprotected aluminum/aluminum alloys are prone to corrosion. When

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aluminum/aluminum alloys are provided with a coating of the invention, a better protection against corrosion is obtained than when they are provided with coatings known in the art.

5 EXAMPLES

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Coating preparation

All chemicals were used as received and without any further
purification. The standard commercial Aluminum 2024-T3 grade is used. Four different types of epoxy-amine coatings were prepared using
two different epoxy resins and two different curing agents (amines). The two different epoxy resins used are: 1) a bisphenol A based
epoxy (Epikote 828, Resolution Nederland BV) with an epoxide equivalent
weight (eew) of 187 g/mol; and 2) a medium viscosity hydrogenated DDP
epoxy resin (Eponex 1510, Brenntag Nederland B.V, eew 210 g/mol). These
epoxy resins are further denoted as E828 and E1510 (Figure 1a). The two different amines used are 1) Jeffamine EDR-148 (NH-eqw
37 g/mol) and 2) Jeffamine D-230 (NH-eqw 60 g/mol). These amines are

further denoted as JEDR148 and JD230 (Figure 1b). Further, (3-glycidoxypropyl)trimethoxysilane was used as adhesion promoter for the coating prepared from E1510 and JEDR148 in a 1/1 ratio.

The adhesion promoter was applied in 1.5 wt% based on the total weight of the curable coating composition.

Curable coating compositions were prepared by mixing the appropriate epoxy resin with the appropriate amine at initially different epoxy/active NH (E/NH) ratios: 1.0/1.0, 1.2/1.0 and 1.4/1.0. Additionally, for samples E828-JEDR148 and E1510-JEDR148, a composition with an 0.8/1.0 ratio was also prepared. Aluminum alloy 2024-T3 (10x15 cm) was pretreated by using sand paper (Scotch-Brite, 3M) and acetone to remove oil and dust. The four curable coating compositions, each having the different E/NH ratios as mentioned above, were applied on aluminum alloy. The panels were blown dry using clean N₂. Thereafter, a quadruple film applicator (Erichsen GmbH &

Co. KG) was employed, providing controlled film thicknesses of 60 µm and an applying rate of 10 mm.s-1. The coated aluminum alloy substrates were cured at 100 °C for 4 hours and allowed to reach room temperature in an oven. The final coating thickness was measured by a TQC thickness gauge after curing to confirm that the obtained values were in the required range of 40-60 µm.

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Pull-off test

Adhesion tests (pull-off tests) were used to measure the adhesion
strength of the different coatings to the substrate (Figure 2). From each ratio of each of the four coatings, three sets of six identical samples were made. The samples were panels of 2x3 cm. The first set was stored in dry conditions, the second set was immersed in distilled water during one week, and the third set was immersed in distilled water during two weeks. The
samples immersed in distilled water were sealed by water-proof tape before immersion. After the one or two weeks of immersion, the samples were thoroughly dried with tissue paper and dry N₂. The coating surface of the three different sets of samples was ground by sand paper (#1200, 3M) to enhance the mechanical anchoring of the glue (3M scotch-weld DP460)

which attaches the stud perpendicularly to the coating surface. After application, the glue was cured at RT for two days before pull-off test. An electrical drill was used to remove the coating around the stud. The pull-off test was performed using an Easy TEST (EZ 20) tensile equipment, with a velocity of 1 mm/min. The force (stress) needed to detach the coating at an angle of 90° from the substrate was monitored as a function of the stud displacement. Only the loading of adhesive failure was recorded. Further details are provided by "Absorption of Water and Corrosion Performance of a Clear and Pigmented Epoxy Coating on Al-2024 Alloy in ECS Trans., Adolphe Foyet, Te Hui Wu, A. Kodentsov, Leo G. Van der Ven, Gijsbertus De With, and Rolf A. Van Benthem, 25, 31 (2010).

Results of the pull-off tests

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The four different clear coatings, each of which is present with the different epoxy/amine (E/NH) ratios, were subjected to pull-off tensile tests. Six samples of each coating were measured and the average value was taken. The outcome of each of the measurements shown in Figure 3 is an average of the six samples. Figure 3a shows the results from the pull-off test of the coatings obtained with E828 and each of the JEDR148 and JD230 amines. Figure 3b shows the results of the coatings obtained with E1510 and each of the JEDR148 and JD230 amines. The values shown refer to storage under dry conditions (most distant row), to storage under wet conditions during one week (middle row), and to storage under wet conditions during two weeks (most nearby row). Under dry conditions, all prepared samples demonstrated good dry adhesion to the aluminum substrate. Further, the addition of the adhesion promoter to the E1510-JEDR148 coating (with a 1/1 ratio) demonstrated an improved dry adhesion compared to the same coating lacking the adhesion promoter. The average value increased from ~7 MPa to ~12 MPa.

After one week under wet conditions, all E1510 coatings appeared to perform much better than the E828 coatings. The latter showed a strong decrease in adhesion, while the decrease was much less pronounced in the former. Moreover, the wet adherence of the E1510-JEDR148 coatings remained comparable to the dry adherence of these coatings. On the other hand, the effect of the adhesion promoter that was seen under dry conditions, had largely disappeared after exposure to wet conditions.

After two weeks exposure to wet conditions, the overall values of almost all E828 coatings were below 2 Mpa (the only coating with a value still above 2 Mpa is E828/JEDR148 1.4/1). Some coatings showed delamination or studs from samples broke during drilling. The wet adherence of most E1510 coatings, however, was still comparable to that obtained after one week under wet conditions. The coating of E1510-JEDR148 again demonstrated an outstanding wet adherence for the ratios 1/1, 1/1.2 and 1/1.4. Only when an excess of amine was present (ratio of 0.8/1), the wet adhesion was substantially weaker.

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<u>Claims</u>

- 1. Curable coating composition comprising
 - an aliphatic epoxy resin comprising two or more epoxide groups;
- a compound having two or more amine functionalities, wherein two of the two or more amine functionalities are connected to each other by a chain comprising a segment consisting of an alkyl ether or a poly(alkyl ether), the alkyl ether or poly(alkyl ether) having alkylene units of 2 to 5 carbon atoms or a mixture of such alkylene units, the poly(alkyl ether) having from 2 to 20 ether oxygen atoms in the chain.
 - 2. Curable coating composition of claim 1, wherein the aliphatic epoxy resin has an Epoxide Equivalent Weight of 100–1000, preferably of 150–500, more preferably of 180-240 g/eq.
 - Curable coating composition of any of claims 1–2, wherein the aliphatic epoxy resin is a cycloaliphatic epoxy resin.
 - 4. Curable coating composition of any of claims 1–3, wherein the aliphatic epoxy resin comprises one or more cyclohexyl moieties.
 - 5. Curable coating composition of claim 1 or 2, wherein the aliphatic epoxy resin is a diglycidyl ether of a diol.
- Curable coating composition of claim 5, wherein the diol is selected from the group of cyclohexanediols, cyclohexanedimethanols and bis(hydroxycyclohexyl)alkanes.
 - Curable coating composition of claim 5, wherein the diol is
 2,2-bis(4-hydroxycyclohexyl)propane.
- Curable coating composition of any of claims 1–7, wherein the compound having two amine functionalities is a compound of the formula H₂N-(CH(CH₃)-CH₂-O)_x-(CH₂)-CH(CH₃)-NH₂, wherein x=2 or 3.
 - 9. Curable coating composition of any of claims 1–8, wherein the two amine functionalities are connected to each other by a linear chain.
- 30 10. Curable coating composition of claim 9, wherein the compound having two amine functionalities is a compound of the formula H₂N-(CH₂)_x-O-(CH₂)₂-O-(CH₂)_x-NH₂, wherein x=2 or 3.

- 11. Curable coating composition of any of claims 1–9, wherein the ratio of the total molar amount of epoxy groups to the total molar amount of active NH's is from 0.8/1.0 to 1.8/1.0, preferably from 1.0/1.0 to 1.4/1.0.
- 12. Coating obtainable by curing a curable coating composition as claimed in any

- one of claims 1-11.
 - 13 Coating according to claim 12, wherein the curing comprises exposing the curable coating composition to a temperature of more than 100 °C during at least 4 hours.
 - 14. Object comprising the coating of claim 12 or 13, in particular an airplane.

comprising aluminum or an aluminum alloy.

10 15. Object according to claim 14, wherein the coating is present on a surface

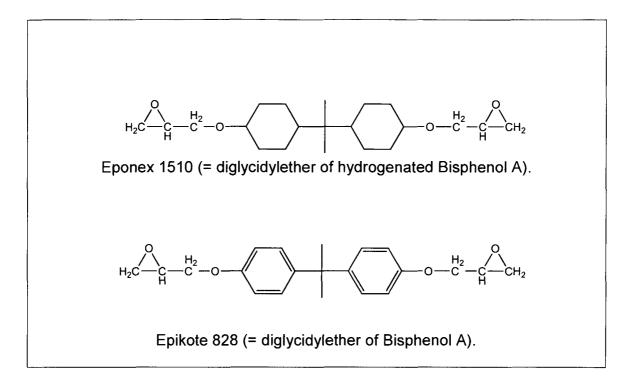


Figure 1A

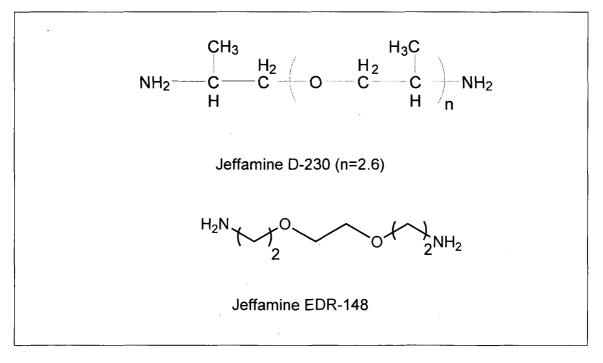
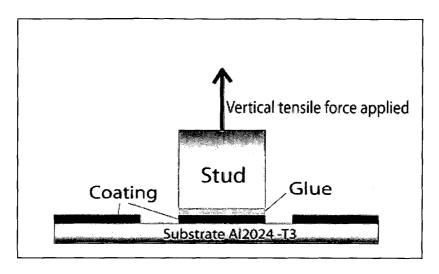


Figure 1B.

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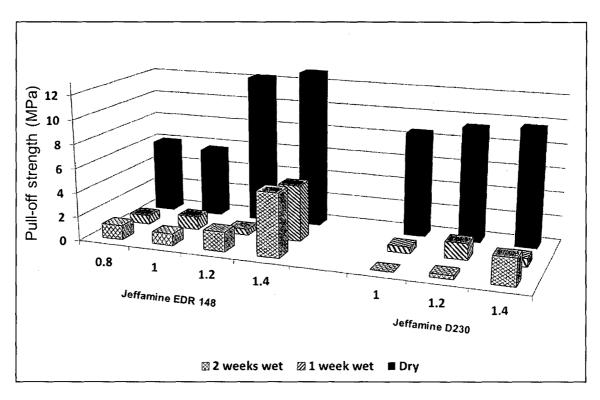


Figure 3A.

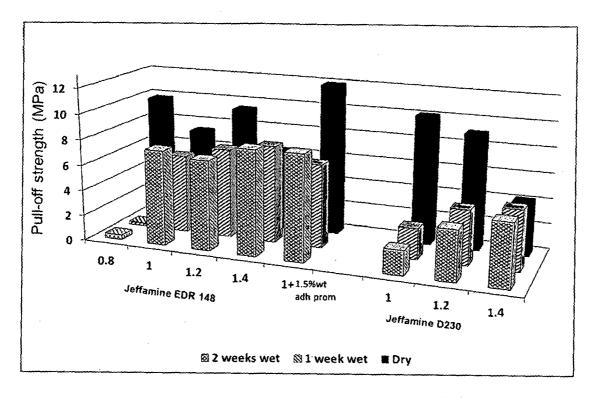


Figure 3B