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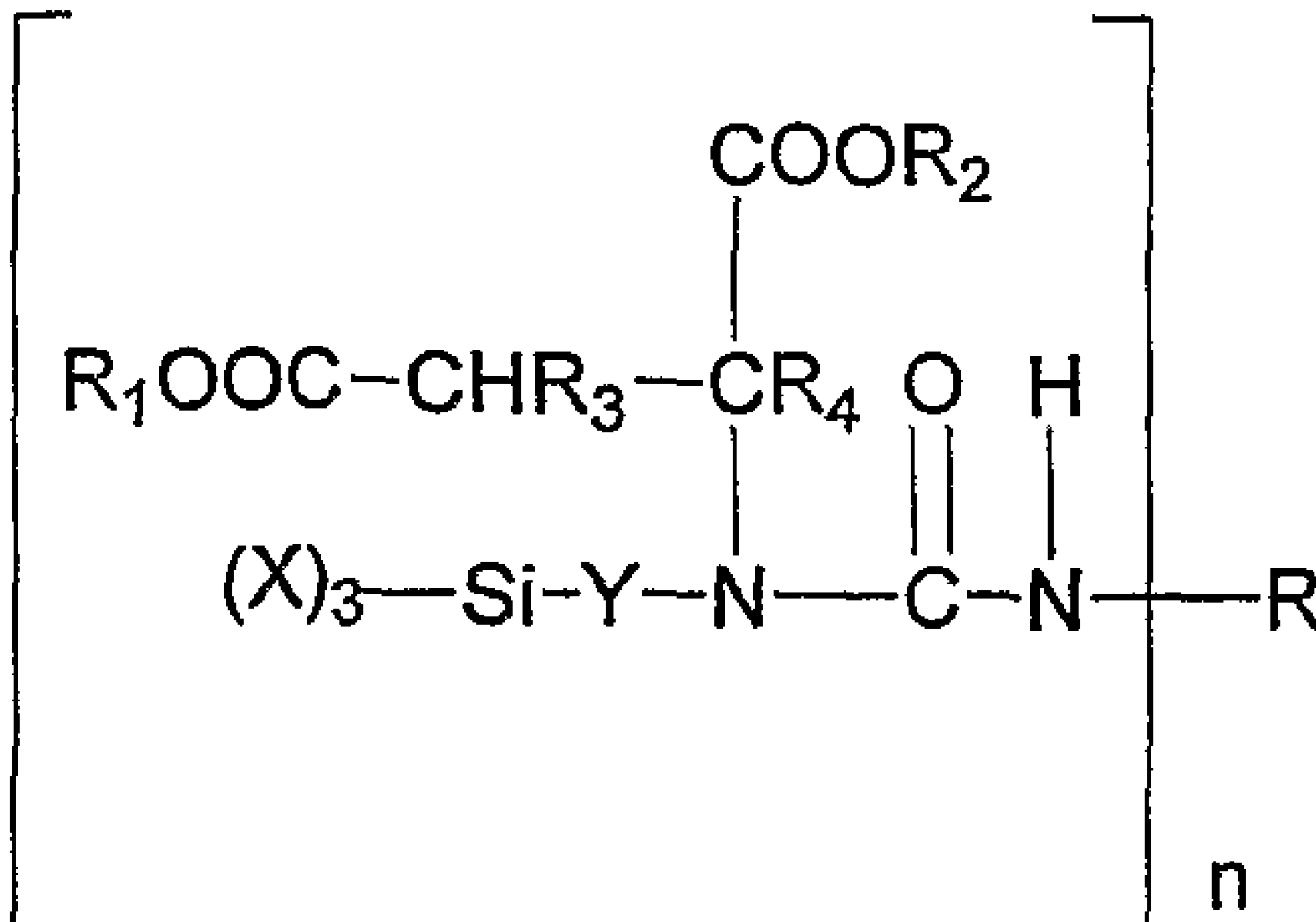
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(54) Titre : COMPOSITIONS DE REVETEMENT A DEUX COMPOSANTS RENFERMANT DES PROMOTEURS  
D'ADHESION A BASE DE SILANE

(54) Title: TWO-COMPONENT COATING COMPOSITIONS CONTAINING SILANE ADHESION PROMOTERS



(57) Abrégé/Abstract:

The present invention relates to two-component coating compositions containing a) a polyisocyanate component, b) an isocyanate-reactive component and c) 0.1 to 20 wt.%, based on the weight of components a) and b), of a compound containing alkoxysilane

## (57) Abrégé(suite)/Abstract(continued):

groups and urea groups corresponding to the formula (I), wherein X represents identical or different organic groups which are inert to isocyanate groups below 100°C, provided that at least one of these groups is an alkoxy group, Y represents a linear or branched alkylene group containing 1 to 8 carbon atoms, R represents the residue obtained by removing the isocyanate groups from a monoisocyanate or a polyisocyanate, R<sub>1</sub> and R<sub>2</sub> are identical or different and represent organic groups which are inert to isocyanate groups at a temperature of 100°C or less, R<sub>3</sub> and R<sub>4</sub> are identical or different and represent hydrogen or organic groups which are inert to isocyanate groups at a temperature of 100°C or less, and n has a value of 1 to 6. The present invention also relates to substrates coated with these coating compositions, in particular metal substrates.

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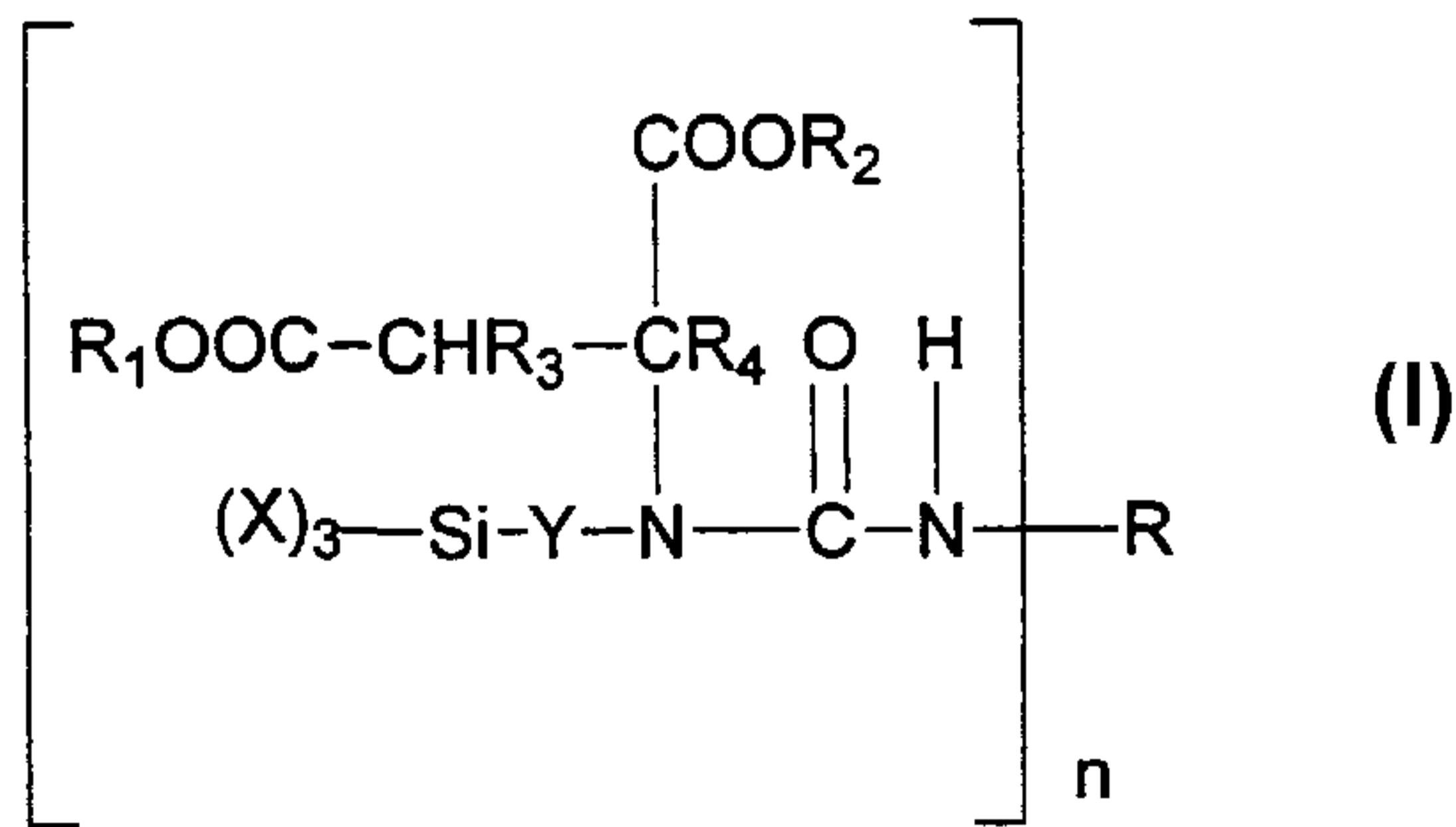
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(54) Title: TWO-COMPONENT COATING COMPOSITIONS CONTAINING SILANE ADHESION PROMOTERS



(57) **Abstract:** The present invention relates to two-component coating compositions containing a) a polyisocyanate component, b) an isocyanate-reactive component and c) 0.1 to 20 wt.%, based on the weight of components a) and b), of a compound containing alkoxysilane groups and urea groups corresponding to the formula (I), wherein X represents identical or different organic groups which are inert to isocyanate groups below 100°C, provided that at least one of these groups is an alkoxy group, Y represents a linear or branched alkylene group containing 1 to 8 carbon atoms, R represents the residue obtained by removing the isocyanate groups from a monoisocyanate or a polyisocyanate, R<sub>1</sub> and R<sub>2</sub> are identical or different and represent organic groups which are inert to isocyanate groups at a temperature of 100°C or less, R<sub>3</sub> and R<sub>4</sub>

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are identical or different and represent hydrogen or organic groups which are inert to isocyanate groups at a temperature of 100°C or less, and n has a value of 1 to 6. The present invention also relates to substrates coated with these coating compositions, in particular metal substrates.

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## TWO-COMPONENT COATING COMPOSITIONS CONTAINING SILANE ADHESION PROMOTERS

### BACKGROUND OF THE INVENTION

#### Field of the Invention

10 The present invention relates to two-component coating compositions containing adhesion promoters based on aspartate silanes and to their use for preparing coatings having improved adhesion, especially wet adhesion.

#### Description of the Prior Art

15 Two-component coating compositions containing a polyisocyanate component and an isocyanate-reactive component, preferably a polyhydroxyl compound, are known and can be used to coat a variety of substrates. One of the deficiencies of these coatings is that they do not possess adequate wet adhesion, which is a critical requirement for  
20 coatings used underground, e.g., as coatings for oil and gas transmission pipelines.

Coatings previously used for this application have suffered from other problems. Asphaltic coatings are biodegradable and not suitable for long term (20 to 50 years) use. Coal tar coatings are not used because of  
25 toxicity. Liquid epoxy coatings require application temperatures of >55°C and cure too slowly for production use in the field. Polyolefin coatings do not maintain adhesion under cathodic protection.

High performance coatings are needed. Known polyurethane coatings have the ability to be rapidly cured, but do not have the wet  
30 adhesion necessary for this application.

Accordingly, it is an object of the present invention to provide coating compositions that are suitable for preparing coatings having improved adhesion, especially wet adhesion.

This object may be achieved with the two-component coating  
35 compositions according to the present invention which contain certain

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silane adhesion promoters. The silane compounds are prepared by the reaction of compounds containing one or more isocyanate groups with aspartate silanes as described hereinafter.

The adhesion promoters used in the compositions of the present  
5 invention are described in U.S. Patent 5,908,948. However, the  
compounds disclosed in the patent are used as coating compositions; their  
use as adhesion promoters to improve the wet adhesion of two-component  
coating compositions is not disclosed.

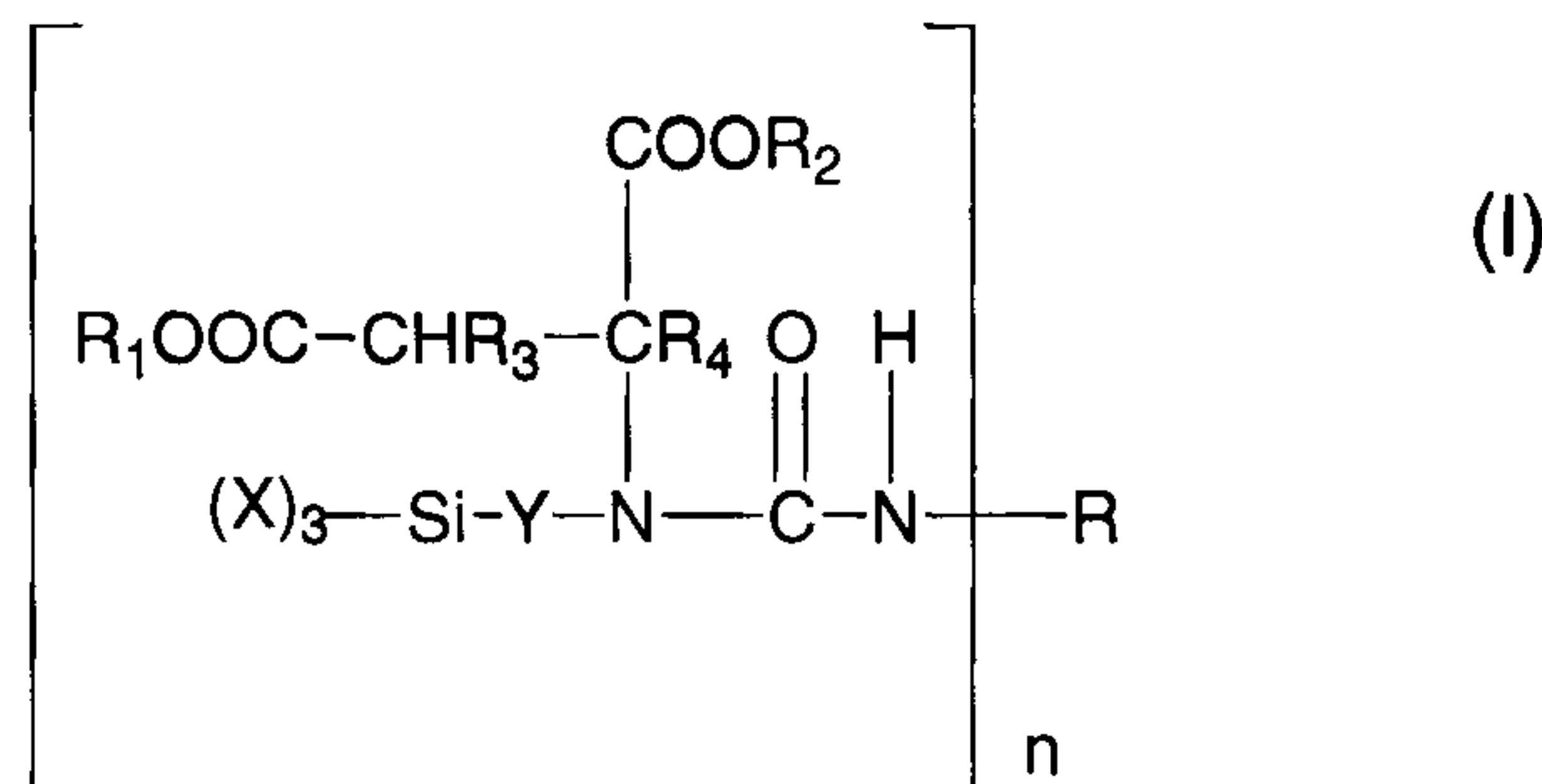
The use of silanes as adhesion promoters has been suggested. A  
10 product information bulletin from OSi Specialties discloses the use of  
several silane compounds as adhesion promoters for various coating  
resins, including polyurethanes. However, as shown by the comparison  
examples of the present application, most of the disclosed silane  
compounds were not very effective for improving the adhesion, especially  
15 the wet adhesion, of two-component polyurethane coating compositions.  
Accordingly, it is apparent that the reference to polyurethanes was a  
reference to one-component, fully reacted polyurethanes.

It must be regarded as surprising and unexpected that the adhesion  
promoters according to the present invention provide good adhesion,  
20 especially wet adhesion, since they contain less silane per unit weight than  
the silane compounds disclosed in the OSi bulletin when added in similar  
amounts. In addition, because they contain less silane the adhesion  
promoters according to the invention are less costly, thereby reducing the  
cost of the resulting coating compositions.

25 SUMMARY OF THE INVENTION

The present invention relates to two-component coating  
compositions containing

- a) a polyisocyanate component,
- b) an isocyanate-reactive component and
- 30 c) 0.1 to 20 wt.%, based on the weight of components a) and b), of a  
compound containing alkoxy silane groups and urea groups  
corresponding to the formula



wherein

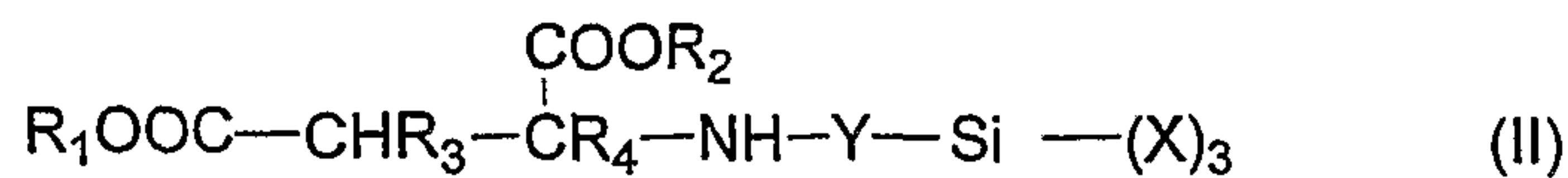
- X represents identical or different organic groups which are inert to isocyanate groups below 100°C, provided that at least one of these groups is an alkoxy group,
- 5 Y represents a linear or branched alkylene group containing 1 to 8 carbon atoms,
- R represents the residue obtained by removing the isocyanate groups from a monoisocyanate or a polyisocyanate,
- 10 R<sub>1</sub> and R<sub>2</sub> are identical or different and represent organic groups which are inert to isocyanate groups at a temperature of 100°C or less,
- R<sub>3</sub> and R<sub>4</sub> are identical or different and represent hydrogen or organic groups which are inert to isocyanate groups at a
- 15 temperature of 100°C or less, and
- n has a value of 1 to 6.

The present invention also relates to substrates coated with these coating compositions, in particular metal substrates.

#### DETAILED DESCRIPTION OF THE INVENTION

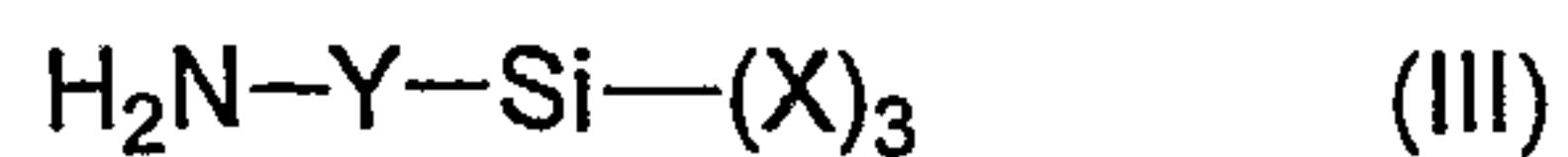
- 20 The compounds containing alkoxy silane groups that are used as component c) in the compositions according to the present invention are based on the reaction products of compounds containing one or more isocyanate groups and compounds containing alkoxy silane groups and aspartate groups (secondary amino groups) corresponding to formula II)

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The compounds of formula II are prepared by reacting aminoalkyl alkoxysilanes corresponding to formula III)

5



with maleic or fumaric acid esters corresponding to formula IV)



10

In formulas I to IV

- X represents identical or different organic groups which are inert to isocyanate groups below 100°C, provided that at least one of these groups is an alkoxy group, preferably alkyl or alkoxy groups having 1 to 4 carbon atoms and more preferably alkoxy groups;
- 15 Y represents a linear or branched alkylene group containing 1 to 8 carbon atoms, preferably a linear group containing 2 to 4 carbon atoms or a branched group containing 5 to 6 carbon atoms, more preferably a linear group containing 3 carbon atoms,
- 20 R represents the residue obtained by removing the isocyanate groups from a monomeric polyisocyanate, a polyisocyanate adduct or an NCO prepolymer, preferably a monomeric polyisocyanate or a polyisocyanate adduct,
- 25 R<sub>1</sub> and R<sub>2</sub> are identical or different and represent organic groups which are inert to isocyanate groups at a temperature of 100°C or less, preferably alkyl groups having 1 to 9 carbon atoms, more preferably methyl, ethyl or butyl groups,

$R_3$  and  $R_4$  are identical or different and represent hydrogen or organic groups which are inert towards isocyanate groups at a temperature of 100°C or less, preferably hydrogen, and

n has a value of 1 to 6, preferably 1 to 4.

5 With regard to the preceding definitions  $R_1$  and  $R_2$  may be different when the compounds of formula II) are prepared from mixed maleates, such as methylethyl maleate. In addition, one  $R_1$  may be different from another  $R_1$ . For example, when a mixture of compounds containing alkoxysilane groups (e.g. those prepared from dimethyl and diethyl 10 maleate) is used to prepare the compounds of formula I), one pair of  $R_1$  and  $R_2$  groups will be methyl and the other will be ethyl.

Especially preferred are compounds in which X represents methoxy, ethoxy groups or propoxy groups, more preferably methoxy or ethoxy groups and most preferably methoxy groups, and Y is a propylene 15 group.

Examples of suitable aminoalkyl alkoxysilanes of formula III include 2-aminoethyl-dimethylmethoxysilane; 6-aminoethyl-tributoxysilane; 3-aminopropyl-trimethoxysilane; 3-aminopropyl-triethoxysilane; 3-aminopropyl-methyldiethoxysilane; 5-aminopentyl-trimethoxysilane; 5-20 aminopentyl-triethoxysilane and 3-aminopropyl-triisopropoxysilane. 3-aminopropyl-trimethoxysilane and 3-aminopropyl-triethoxysilane are particularly preferred.

Examples of optionally substituted maleic or fumaric acid esters suitable for use in the preparation of polyaspartates II) include dimethyl, 25 diethyl, dibutyl (e.g., di-n-butyl), diamyl, di-2-ethylhexyl esters, the previously discussed mixed maleates and fumarates, and the corresponding maleic or fumaric acid esters substituted by methyl in the 2- and/or 3-position. The dimethyl, diethyl and dibutyl esters of maleic acid are preferred and the diethyl and dibutyl esters are especially preferred.

30 The reaction of primary amines with maleic or fumaric acid esters to form aspartates is disclosed in U.S. Patents 5,126,170, 5,236,741 and 5,364,955.

The compounds of formula II are colorless to pale yellow. They may be reacted with polyisocyanates to form the compounds of formula I without further purification.

Suitable isocyanates for preparing the compounds of formula I are 5 selected from monoisocyanates, monomeric polyisocyanates, polyisocyanate adducts and NCO prepolymers, preferably monomeric polyisocyanates and polyisocyanate adducts. The polyisocyanates have an average functionality of 1.8 to 6, preferably 2 to 6 and more preferably 2 to 4.

10 Suitable monomeric diisocyanates may be represented by the formula



in which R represents an organic group obtained by removing the isocyanate groups from an organic diisocyanate having a molecular weight 15 of about 112 to 1,000, preferably about 140 to 400. Diisocyanates preferred for the process according to the invention are those in which R represents a divalent aliphatic hydrocarbon group having 4 to 40, preferably 4 to 18 carbon atoms, a divalent cycloaliphatic hydrocarbon group having 5 to 15 carbon atoms, a divalent araliphatic hydrocarbon 20 group having 7 to 15 carbon atoms or a divalent aromatic hydrocarbon group having 6 to 15 carbon atoms.

Examples of the suitable organic diisocyanates include 1,4-tetramethylene diisocyanate, 1,6-hexamethylene diisocyanate, 2,2,4-trimethyl-1,6-hexamethylene diisocyanate, 1,12-dodecamethylene 25 diisocyanate, cyclohexane-1,3- and -1,4-diisocyanate, 1-isocyanato-2-isocyanatomethyl cyclopentane, 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethyl-cyclohexane (isophorone diisocyanate or IPDI), bis-(4-iso-cyanatocyclohexyl)-methane, 2,4'-dicyclohexyl-methane diisocyanate, 1,3- and 1,4-bis-(isocyanatomethyl)-cyclohexane, bis-(4-isocyanato-3-30 methyl-cyclohexyl)-methane,  $\alpha,\alpha,\alpha',\alpha'$ -tetramethyl-1,3- and/or -1,4-xylylene diisocyanate, 1-isocyanato-1-methyl-4(3)-isocyanatomethyl cyclohexane, 2,4- and/or 2,6-hexahydrotoluylene diisocyanate, 1,3- and/or

1,4-phenylene diisocyanate, 2,4- and/or 2,6-toluylene diisocyanate, 2,4- and/or 4,4'-diphenyl-methane diisocyanate, 1,5-diisocyanato naphthalene and mixtures thereof.

Suitable monoisocyanates include compounds corresponding to the 5 formula R-NCO, wherein R is defined as above.

Polyisocyanates containing 3 or more isocyanate groups such as 4-isocyanantomethyl-1,8-octamethylene diisocyanate and aromatic polyisocyanates such as 4,4',4"-triphenylmethane triisocyanate and polyphenyl polymethylene polyisocyanates obtained by phosgenating 10 aniline/formaldehyde condensates may also be used.

Preferred organic diisocyanates include 1,6-hexamethylene diisocyanate, 1-isocyanato-3-isocyanatomethyl-3,5,5-trimethylcyclohexane (isophorone diisocyanate or IPDI), bis-(4-isocyanato-cyclohexyl)-methane, 1-isocyanato-1-methyl-4(3)-isocyanatomethyl cyclohexane, 2,4- and/or 2,6-toluylene diisocyanate, and 2,4- and/or 4,4'-diphenylmethane diisocyanate.

Suitable polyisocyanate adducts for preparing the compounds of formula I are those prepared from the preceding monomeric polyisocyanates and containing isocyanurate, uretdione, biuret, urethane, 20 allophanate, iminoxadiazine dione, carbodiimide and/or oxadiazinetrione groups. The polyisocyanates adducts, which preferably have an NCO content of 5 to 30% by weight, include:

1) Isocyanurate group-containing polyisocyanates which may be prepared as set forth in DE-PS 2,616,416, EP-OS 3,765, 25 EP-OS 10,589, EP-OS 47,452, US-PS 4,288,586 and US-PS 4,324,879. The isocyanato-isocyanurates generally have an average NCO functionality of 3 to 3.5 and an NCO content of 5 to 30%, preferably 10 to 25% and most preferably 15 to 25% by weight.

2) Uretidine diisocyanates which may be prepared by 30 oligomerizing a portion of the isocyanate groups of a diisocyanate in the presence of a suitable catalyst, e.g., a trialkyl phosphine catalyst, and which may be used in admixture with other aliphatic and/or cycloaliphatic

polyisocyanates, particularly the isocyanurate group-containing polyisocyanates set forth under (1) above.

- 3) Biuret group-containing polyisocyanates which may be prepared according to the processes disclosed in U.S. Patent Nos. 5 3,124,605; 3,358,010; 3,644,490; 3,862,973; 3,906,126; 3,903,127; 4,051,165; 4,147,714; or 4,220,749 by using co-reactants such as water, tertiary alcohols, primary and secondary monoamines, and primary and/or secondary diamines. These polyisocyanates preferably have an NCO content of 18 to 22% by weight and an average NCO functionality of 3 to 10 3.5.
- 4) Urethane group-containing polyisocyanates which may be prepared in accordance with the process disclosed in U.S. Patent No. 3,183,112 by reacting excess quantities of polyisocyanates, preferably diisocyanates, with low molecular weight glycols and polyols having 15 molecular weights of less than 400, such as trimethylol propane, glycerine, 1,2-dihydroxy propane and mixtures thereof. The urethane group-containing polyisocyanates have a most preferred NCO content of 12 to 20% by weight and an (average) NCO functionality of 2.5 to 3.
- 5) Allophanate group-containing polyisocyanates which may be 20 prepared according to the processes disclosed in U.S. Patent Nos. 3,769,318, 4,160,080 and 4,177,342. The allophanate group-containing polyisocyanates have a most preferred NCO content of 12 to 21% by weight and an (average) NCO functionality of 2 to 4.5.
- 6) Isocyanurate and allophanate group-containing 25 polyisocyanates which may be prepared in accordance with the processes set forth in U.S. Patents 5,124,427, 5,208,334 and 5,235,018, preferably polyisocyanates containing these groups in a ratio of monoisocyanurate groups to mono-allophanate groups of about 10:1 to 1:10, preferably about 5:1 to 1:7.
- 7) Iminooxadiazine dione and optionally isocyanurate group-containing polyisocyanates which may be prepared in the presence of 30

special fluorine-containing catalysts as described in DE-A 19611849. These polyisocyanates generally have an average NCO functionality of 3 to 3.5 and an NCO content of 5 to 30%, preferably 10 to 25% and most preferably 15 to 25% by weight.

5 8) Carbodiimide group-containing polyisocyanates which may be prepared by oligomerizing di- or polyisocyanates in the presence of known carbodiimidization catalysts as described in DE-PS 1,092,007, US-PS 3,152,162 and DE-OS 2,504,400, 2,537,685 and 2,552,350.

10 9) Polyisocyanates containing oxadiazinetrione groups and containing the reaction product of two moles of a diisocyanate and one mole of carbon dioxide.

Preferred polyisocyanate adducts are the polyisocyanates containing isocyanurate, uretdione, biuret, iminoxadiazine dione and/or allophanate groups.

15 15 The NCO prepolymers, which may also be used to prepare the compounds of formula I, are prepared from the previously described monomeric polyisocyanates or polyisocyanate adducts, preferably monomeric diisocyanates, and polyhydroxyl compounds containing at least two hydroxyl groups. These polyhydroxyl compounds include high 20 molecular weight compounds having molecular weights of 500 to about 10,000, preferably 800 to about 8,000, and more preferably 1800 to 8,000, and optionally low molecular weight compounds having molecular weights below 500. The molecular weights are number average molecular weights ( $M_n$ ) and are determined by end group analysis (OH number). Products 25 obtained by reacting polyisocyanates exclusively with low molecular weight compounds are polyisocyanates adducts containing urethane groups and are not considered to be NCO prepolymers.

30 Examples of the high molecular weight compounds are polyester polyols, polyether polyols, polyhydroxy polycarbonates, polyhydroxy polyacetals, polyhydroxy polyacrylates, polyhydroxy polyester amides and polyhydroxy polythioethers. The polyether polyols, polyester polyols and

polycarbonate polyols are preferred. Especially preferred are the polyether polyols.

Examples of suitable high molecular weight polyhydroxyl compounds include polyether polyols, which may be obtained in known manner by the alkoxylation of suitable starter molecules. Examples of suitable starter molecules include polyols, water, organic polyamines having at least two N-H bonds and mixtures thereof. Suitable alkylene oxides for the alkoxylation reaction are preferably ethylene oxide and/or propylene oxide, which may be used in sequence or in admixture.

Also suitable for preparing the prepolymers are polyester polyols prepared from low molecular weight alcohols and polybasic carboxylic acids such as adipic acid, sebacic acid, phthalic acid, isophthalic acid, tetrahydronaphthalic acid, hexahydronaphthalic acid, maleic acid, the anhydrides of these acids and mixtures of these acids and/or acid anhydrides. Polylactones having hydroxyl groups, particularly poly-,-caprolactone, are also suitable for producing the prepolymers.

Other suitable polyols include polycarbonates having hydroxyl groups, which may be produced by the reaction of diols with phosgene or diaryl carbonates such as diphenyl carbonate.

Further details concerning the low molecular weight compounds and the starting materials and methods for preparing the high molecular weight polyhydroxy compounds are disclosed in U.S. Patent 4,701,480.

These NCO prepolymers preferably have an isocyanate content of 0.3 to 35% by weight, more preferably 0.6 to 25% by weight and most preferably 1.2 to 20% by weight. The NCO prepolymers are produced by reacting the diisocyanates with the polyol component at a temperature of 40 to 120°C, preferably 50 to 100°C, at an NCO/OH equivalent ratio of 1.3:1 to 20:1, preferably 1.4:1 to 10:1. If chain extension via urethane groups is desired during the preparation of the isocyanate prepolymers, an NCO/OH equivalent ratio of 1.3:1 to 2:1 is selected. If chain extension is not desired, an excess of diisocyanate is preferably used, corresponding

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to an NCO/OH equivalent ratio of 4:1 to 20:1, preferably 5:1 to 10:1. The excess diisocyanate may optionally be removed by thin layer distillation when the reaction is completed. In accordance with the present invention NCO prepolymers also include NCO semi-prepolymers which contain 5 unreacted starting polyisocyanates in addition to the urethane group-containing prepolymers.

The compounds of formula I containing alkoxy silane groups and urea groups are prepared by reacting the polyisocyanate monomers and/or adducts with the compounds of formula II at an equivalent ratio of 10 aspartate groups (i.e., secondary amino groups) to isocyanate groups of approximately 1:1. The reaction is preferably carried out by incrementally adding the aspartate to the polyisocyanate. The reaction to form the urea groups is conducted at a temperature of 10 to 100°C, preferably 20 to 80°C and more preferably 20 to 50°C.

15 In accordance with the present invention the urea groups initially formed by the reaction of the compounds containing alkoxy silane groups and aspartate groups of formula II with the polyisocyanate may be converted to hydantoin groups in known manner, e.g., by heating the compounds at elevated temperatures, optionally in the presence of a 20 catalyst. Therefore, the term "urea groups" is also intended to include other compounds containing the group, N-CO-N, such as hydantoin groups.

25 Suitable polyisocyanates for use as component a) in the compositions of the present invention are selected from the polyisocyanates previously described for use in the preparation of the compounds of formula I), preferably polyisocyanate adducts and NCO prepolymers and more preferably NCO prepolymers.

30 Suitable isocyanate-reactive components for use as component b) in the compositions of the present invention are the high and low molecular weight, polyols previously disclosed for preparing the NCO prepolymers. Also suitable are the known high molecular weight amine-functional compounds, which may be prepared by converting the terminal

hydroxy groups of the polyols previously described to amino groups, and the high molecular weight polyaldimines disclosed in U.S. Patent 5,466,771. The high molecular weight polyols are preferred.

The two-component coating compositions of the present invention 5 may be prepared by mixing the individual components. Components a) and b) are present in an amount of sufficient to provide an equivalent ratio of isocyanate groups to isocyanate-reactive, preferably hydroxyl groups of 0.8:1 to 1.2:1, preferably 0.9:1 to 1.1:1 and more preferably 1:1 to 1.1:1. Component c) is present in an amount of 0.1 to 20%, preferably 0.5 to 10 10%, more preferably 0.5 to 5% and most preferably 1 to 3%, based on the weight of components a) and b).

The two-component compositions generally may be either solvent-free or contain up to 70%, preferably up to 60% organic solvents, based on the weight of components a) and b). Suitable organic solvents include 15 those which are known from polyurethane chemistry.

The compositions may also contain known additives, such as catalysts, leveling agents, wetting agents, flow control agents, antiskinning agents, antifoaming agents, fillers (such as silica, aluminum silicates and high-boiling waxes), viscosity regulators, plasticizers, pigments, dyes, UV 20 absorbers and stabilizers against thermal and oxidative degradation.

The two-component compositions may be applied to any desired substrates, such as wood, plastics, leather, paper, textiles, glass, ceramics, plaster, masonry, metals and concrete. They may be applied by standard methods, such as spray coating, spread coating, flood coating, 25 casting, dip coating, roll coating. The coating compositions may be clear or pigmented.

The two-component compositions are especially suitable for coating substrates, preferably metal substrates such as pipes, which will be exposed to a wet environment.

The two-component compositions may be cured at ambient temperature or at elevated temperatures, preferably at ambient temperatures.

The invention is further illustrated but is not intended to be limited  
5 by the following examples in which all parts and percentages are by weight  
unless otherwise specified.

### EXAMPLES

#### Polyisocyanate 1

An aniline/formaldehyde condensation product containing 44.4%  
10 4,4'-diphenylmethane diisocyanate, 19.0% 2,4'-diphenylmethane  
diisocyanate, 2.6% 2,2'-diphenylmethane diisocyanate and the remainder  
higher functional homologs, and having an isocyanate content of 32.6%  
and a viscosity at 25EC of about 41 mPa.s.

#### Polyisocyanate 2

15 An isocyanurate group-containing polyisocyanate prepared from  
1,6-hexamethylene diisocyanate and having an isocyanate content of  
21.6%, a content of monomeric diisocyanate of <0.2% and a viscosity at  
25°C of 3000 mPa.s (available from Bayer Corporation as Desmodur\* N  
3300).

#### Polyisocyanate 3

556 g of a distilled diphenylmethanediisocyanate (MDI) composition  
containing 65% 2,4'-MDI and 35% 4,4'-MDI were stirred at 60°C. Then a  
mixture of 222 g of a polyether diol made by propoxylation of propylene  
glycol (OH number 112) and 222 g of a tetrafunctional polyether made by  
25 propoxylation of ethylene diamine (OH number 60) was added dropwise,  
so that a maximum temperature of 65°C was maintained. When an NCO  
content of 16% was reached, the prepolymer was diluted with 1000 g of a  
crude MDI containing 40.5% 2,4-MDI, 49.5% 4,4'-MDI and 10% polymeric  
MDI. The final semi-prepolymer had an NCO content of 24% and a  
30 viscosity of 220 mPa.s at 23°C.

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Polyether polyol 1

A polypropylene oxide polyol having a functionality of 3, an OH number of 370 and an equivalent weight of 152 and initiated with trimethylolpropane.

5 Polyether polyol 2

A polypropylene oxide polyol initiated with glycerine, tipped with ethylene oxide (PO:EO equivalent ratio 83:17), having a functionality of 3, an OH number of 35 and an equivalent weight of 1600 and containing 20% of the equimolar reaction product toluene diisocyanate and hydrazine

10 (overall OH number 28).

Silane Aspartate 1

1482 parts (8.27 equiv.) of 3-aminopropyltrimethoxysilane were added to a 5 liter flask fitted with agitator, thermocouple, nitrogen inlet and addition funnel with condenser. 1423.2 parts (8.27 equiv.) of diethyl

15 maleate were added dropwise through the addition funnel over a period of 2 hours. The temperature of the reactor was maintained at 25°C during the addition. The reactor was maintained at 25°C for an additional 5 hours at which time the product was poured into glass containers and sealed under a blanket of nitrogen. After one week the unsaturation number was

20 0.6 indicating the reaction was ~99% complete. The product, N-(3-trimethoxysilylpropyl) aspartic acid diethyl ester, had a viscosity of 11 mPa.s at 25°C.

Compound 1 containing alkoxy silane groups of formula I

367 parts (1 equiv.) of silane aspartate 1 and 129 parts (1 equiv.) of 25 polyisocyanate 1 were added to a three neck, 1 liter, round bottom flask fitted with agitator, thermocouple, nitrogen inlet and condenser. The reaction to form the urea groups was accompanied by an exotherm. A water bath was used to maintain the temperature at 80°C for 3 hours until an IR spectrum showed no residual isocyanate in the urea product.

30 Compound 2 containing alkoxy silane groups of formula I

669 parts (1.7 equiv.) of silane aspartate 1 and 331 parts (1.7 equiv.) of polyisocyanate 2 were added to a three neck, 5 liter, round

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bottom flask fitted with agitator, thermocouple, nitrogen inlet and condenser. The reaction to form the urea groups was accompanied by an exotherm which took the reaction mixture to 80°C. The reaction was held at 80°C for 14 hours until an IR spectrum showed no residual isocyanate  
5 in the urea product.

Compound 3 containing alkoxy silane groups of formula I

100 parts (0.254 equiv.) of silane aspartate 1 and 21.4 parts (0.254 equiv.) of 1,6-hexamethylene diisocyanate were added to a three neck, 500 ml flask fitted with agitator, thermocouple, nitrogen inlet and  
10 condenser. The reaction to form the urea groups was accompanied by an exotherm which took the reaction mixture to 80°C. The reaction was held at 80°C for 3 hours until an IR spectrum showed no residual isocyanate in the urea product.

Two-component coating compositions

15 Two-component coating compositions were prepared from the following ingredients:

	<u>Component I</u>	<u>Parts</u>
	Polyether polyol 1	120
	Polyether polyol 2	60
20	1,4-butane diol	130
	3,5-diethyl-toluene diamine	5
	Talc	125
	Lamilar silicate <sup>1</sup>	70
	Titanium dioxide	30
25	1:1 Blend of molecular sieve/caster oil <sup>2</sup>	50
	Dibutyltin dilaurate	0.4
	Silicone flow aid	0.2

Component II

Polyisocyanate 3	490
Silane additive	See Table
Black paste	5

5

- 1 - Novacite\* 207 (available from Malvern)
- 2 - Baylith\* L paste (available from Bayer)

Two-component coating compositions were prepared from the preceding ingredients and the amounts and types of the silane additives set forth in the following tables. The compositions were applied at 20-30 mils on hot-rolled, sand-blasted steel panels. The panels were cured at room temperature for two weeks.

To determine dry and wet adhesion the coated panels were placed on end in a container or water such that about two thirds of the coating was submerged. After 30 days the panel was removed and immediately tested. The test was conducted by scribing an "X" on both the dry and wet areas of the panel. A knife was used to pick at the corners of the "X." A rating scale of 1 to 10 was used as follows:

20	1	-	no adhesion, coating falls off of panel
	5	-	moderate adhesion, but coating can be peeled off of panel
	7	-	strong adhesion, but coating can be peeled off of panel
	8	-	coating tears as it is peeled off of panel
	10	-	coating can only be cut off of panel

25 A rating of 7 is acceptable, a rating of 8 is a good result and a rating of 10 is an excellent result. A rating of 6 or less is unacceptable for use as a pipeline coating.

**Table 1 - According to the Invention**

Adhesion Promoter	Amount (wt. %)	Adhesion	
		Dry	Wet
Compound 1 containing alkoxy silane groups	1	10	10
	2	-	10
	3	10	10
	5	10	10
	10	8	9
	15	10	10
Compound 2 containing alkoxy silane groups	1	-	8
	2	-	9
	3	8	9
	5	9.5	9
	10	8	9
	15	10	10
Compound 3 containing alkoxy silane groups	5	7	8
	10	8	8
	20	8	7

**Table 2 - Comparison Examples**

Adhesion Promoter	Amount (wt. %)	Adhesion	
		Dry	Wet
None	0	2	2
3-isocyanatopropyltriethoxysilane <sup>3</sup>	1	7	7
	1	8	8
	2	-	10
Tris-[3-(trimethoxysilyl)propyl]isocyanurate <sup>4</sup>	3	9	8.5
	5	8	9
	10	7	8
	15	7	7.5
3-glycidoxypropyl-trimethoxysilane <sup>5</sup>	1	10	10
	2	10	10
3-mercaptopropyl-trimethoxysilane <sup>6</sup>	1	3	3
	2	3	7
3-aminopropyl-trimethoxysilane <sup>7</sup>	1	3	6
	2	10	10
N-(2-aminoethyl)-3-aminopropyl-trimethoxysilane <sup>8</sup>	1	2	2
	2	3	5
Triaminofunctionalsilane <sup>9</sup>	1	3	3
	2	incom	incom <sup>10</sup>
3-ureidopropyl-trialkoxysilane <sup>11</sup>	1	3	3
	2	4	3
Bis-(3-trimethoxysilylpropyl)amine <sup>12</sup>	1	3	3
	2	8	6
Silquest* Wet Link 78 additive	1	4	5
	2	8	9

\*trade-mark

- 3 - Silquest A-1310
- 4 - Silquest Y-11597
- 5 - Silquest A-187
- 5 6 - Silquest A-189
- 7 - Silquest A-1110
- 8 - Silquest A-1120
- 9 - Silquest A-1130
- 10 - Incompatible
- 10 11 - Silquest A-1160
- 12 - Silquest A-1170

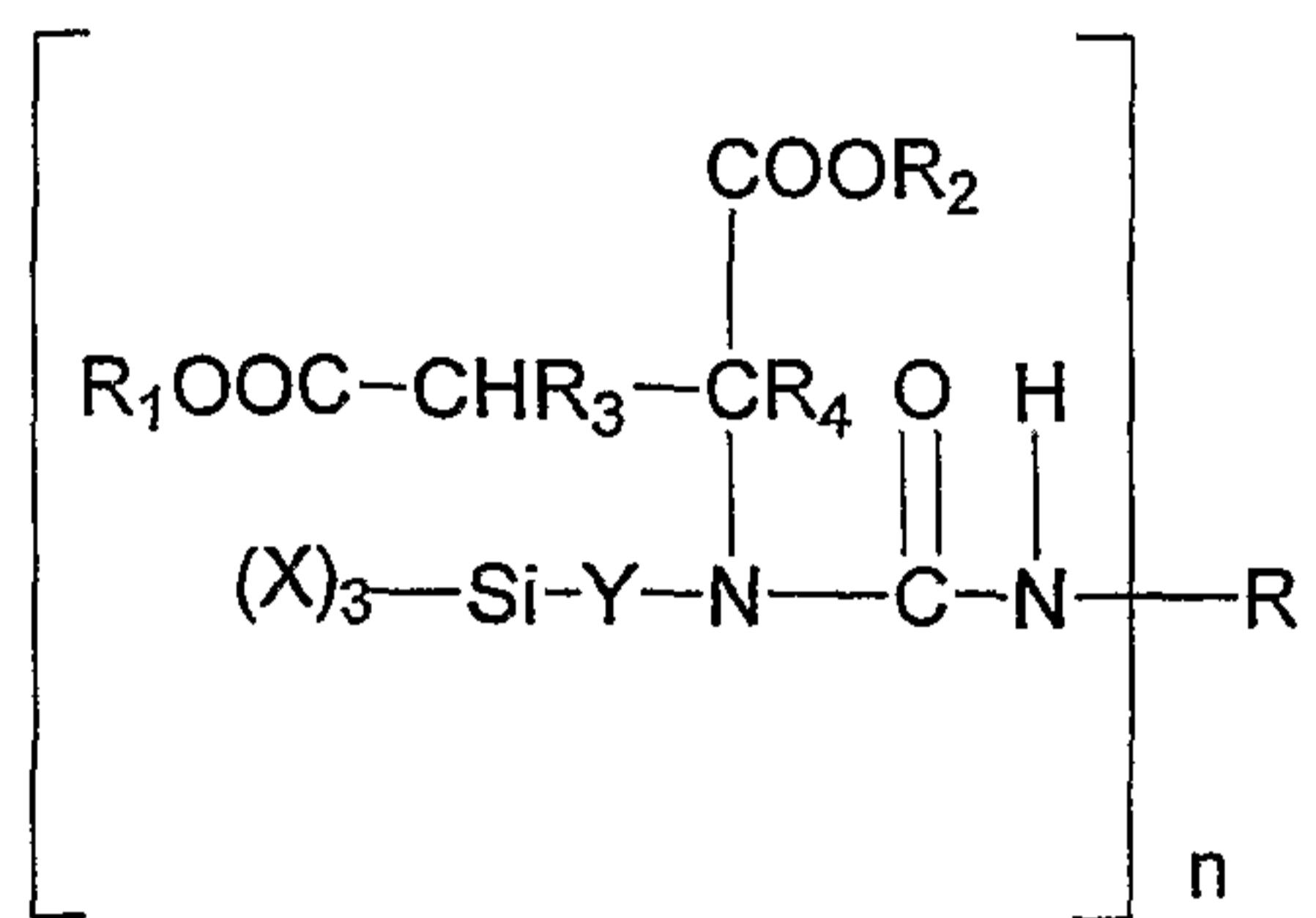
The examples demonstrate that it is possible using the adhesion promoters according to the present invention to match and even exceed 15 the adhesion provided by the known silane adhesion promoters, while at the same time lowering the cost of the additives.

Although the invention has been described in detail in the foregoing for the purpose of illustration, it is to be understood that such detail is solely for that purpose and that variations can be made therein by those 20 skilled in the art without departing from the spirit and scope of the invention except as it may be limited by the claims.

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WHAT IS CLAIMED IS:

1. A two-component coating composition comprising
  - a) a polyisocyanate component,
  - b) an isocyanate-reactive component and
  - 5 c) 0.1 to 20 wt.%, based on the weight of components a) and b), of a compound containing alkoxy silane groups and succinyl urea groups corresponding to the formula



wherein

- 10 X represents identical or different organic groups which are inert to isocyanate groups below 100°C, provided that at least one of these groups is an alkoxy group,
- Y represents a linear or branched alkylene group containing 1 to 8 carbon atoms,
- 15 R represents the residue obtained by removing the isocyanate groups from a monoisocyanate or a polyisocyanate,
- R<sub>1</sub> and R<sub>2</sub> are identical or different and represent organic groups which are inert to isocyanate groups at a temperature of 100°C or less,
- R<sub>3</sub> and R<sub>4</sub> are identical or different and represent hydrogen or
- 20 organic groups which are inert to isocyanate groups at a temperature of 100°C or less and
- n has a value of 1 to 6.

2. The composition of Claim 1 wherein
- R<sub>1</sub> and R<sub>2</sub> are identical or different and represent alkyl groups having
- 25 1 to 9 carbon atoms,

R<sub>3</sub> and R<sub>4</sub> represent hydrogen and

n has a value of 1 to 4.

3. The composition of Claim 1 wherein component c) is present in an amount of 0.5 to 10 wt.%, based on the weight of components a) and 5 b), and

R<sub>1</sub> and R<sub>2</sub> are identical or different and represent methyl, ethyl or butyl,

R<sub>3</sub> and R<sub>4</sub> represent hydrogen and

n has a value of 1 to 4.

4. The composition of Claim 1 wherein 10 R represents the residue obtained by removing the isocyanate groups from a monomeric polyisocyanate or a polyisocyanate adduct.

5. The composition of Claim 2 wherein

R represents the residue obtained by removing the isocyanate groups from a monomeric polyisocyanate or a polyisocyanate adduct.

15 6. The composition of Claim 3 wherein

R represents the residue obtained by removing the isocyanate groups from a monomeric polyisocyanate or a polyisocyanate adduct.

7. The composition of Claim 1 wherein polyisocyanate 20 component a) comprises an NCO prepolymer prepared from a polyisocyanate and a polyether polyol and component b) comprises a polyhydroxyl compound.

8. The composition of Claim 2 wherein polyisocyanate 25 component a) comprises an NCO prepolymer prepared from a polyisocyanate and a polyether polyol and component b) comprises a polyhydroxyl compound.

9. The composition of Claim 3 wherein polyisocyanate component a) comprises an NCO prepolymer prepared from a polyisocyanate and a polyether polyol and component b) comprises a polyhydroxyl compound.

30 10. The composition of Claim 4 wherein polyisocyanate component a) comprises an NCO prepolymer prepared from a

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polyisocyanate and a polyether polyol and component b) comprises a polyhydroxyl compound.

11. The composition of Claim 5 wherein polyisocyanate component a) comprises an NCO prepolymer prepared from a 5 polyisocyanate and a polyether polyol and component b) comprises a polyhydroxyl compound.

12. The composition of Claim 6 wherein polyisocyanate component a) comprises an NCO prepolymer prepared from a polyisocyanate and a polyether polyol and component b) comprises a 10 polyhydroxyl compound.

13. The composition of Claim 7 wherein component b) comprises a polyether polyol.

14. The composition of Claim 8 wherein component b) comprises a polyether polyol.

15. The composition of Claim 9 wherein component b) comprises a polyether polyol.

16. The composition of Claim 10 wherein component b) comprises a polyether polyol.

17. The composition of Claim 11 wherein component b) 20 comprises a polyether polyol.

18. The composition of Claim 12 wherein component b) comprises a polyether polyol.

19. A substrate coated with coating composition of Claim 1.

20. A metal substrate coated with the coating composition of 25 Claim 1.

