

HYDROPHILICIZING SEALER TREATMENT FOR METAL OBJECTS

BACKGROUND OF THE INVENTION

Field of the Invention

This invention relates to a hydrophilicizing treatment for a surface with underlying metal, normally one already bearing a corrosion protective conversion coating such as a chromate or phosphate conversion coating. After treatment according to this invention, the surface will have a tendency for water to spread spontaneously over the surface. The invention is particularly advantageously applicable to provide aluminum evaporators, heat exchangers, and condensers with hydrophilic coatings that have good corrosion resistance, especially to sea water, and a durably hydrophilic character with little or no tendency to develop undesirable odors during use.

Statement of Related Art

Although any of the common structural metals can be used in constructing practical heat exchanging surfaces, aluminum and its alloys are among those most often

used, because of their high heat conductivity. In heat exchanger surfaces, metals are normally used without any relatively thick protective coating such as a paint or lacquer that would normally be used to improve the resistance of the metals to corrosion in most other types of metal structures, but is avoided in heat exchangers because such
5 a coating would also reduce the efficiency of heat exchange.

During the cooling of hot air, a common use of these heat exchangers, moisture contained as vapor in the hot air condenses and initially forms water drops or beads on the fins of the heat exchanger. If the surface of the heat exchanger fins is not sufficiently hydrophilic, the water beads accumulate on the fin surface and impede the
10 air flow between fins and also reduce the heat transfer efficiency. The condensed water beads also tend to absorb dust and contaminants in the air, such as carbon dioxide, nitrogen oxides, and sulfur oxides, which often promote aluminum corrosion. Therefore, the formation of water beads on the fins of aluminum heat exchanger not only decreases heat transfer efficiency but also can cause the corrosion of aluminum.

15 In order to achieve a desirable combination of a hydrophilic nature and corrosion resistance on metal, particularly aluminum, surfaces, various coatings and treatments have been tried, but no perfectly satisfactory result has yet been achieved. This is particularly true for uses in which the hydrophilicized surface comes into contact with sea water, which is especially corrosive, so that currently available hydrophilicizing
20 treatments leave aluminum and its alloys with often inadequate corrosion protection.

DESCRIPTION OF THE INVENTION

Except in the claims and the operating examples, or where otherwise expressly indicated, all numerical quantities in this description indicating amounts of material
25 or conditions of reaction and/or use are to be understood as modified by the word "about" in describing the broadest scope of the invention. Practice within the numerical limits stated is generally preferred. Furthermore, unless expressly stated to contrary below, a description of a material as selected or preferably selected from a group of specified chemical materials shall be considered to include selection from a mixture
30 of materials each of which is a member of the stated group. Also, the terms "solution", "soluble", and the like are to be understood as including not only true equilibrium solutions but also dispersions that show no visually detectable tendency toward

phase separation over a period of observation of at least 1000 hours. Furthermore, the description of materials in ionic form shall be understood as implying the presence of counterions as needed for electrical neutrality of the total composition. To the extent possible, such counterions are preferably chosen from other specified necessary or optional components of the compositions concerned. Otherwise, such counterions generally may be freely chosen, except for avoiding any that are detrimental to the objects of the invention. Also, unless otherwise stated, percentages are to be understood as percentages by mass or weight.

Object of the Invention

The principal object of the invention is to achieve a combination of adequate hydrophilicity and improved corrosion resistance in sea water, compared with the prior art, while avoiding the use of polluting constituents, and of highly volatile constituents, particularly organic solvents, with potential toxicity or unpleasant odors for workers, in the treatment compositions and achieving durability of the hydrophilicity under thermal aging. Another object is to reduce the energy input needed for the hydrophilicizing process. Other objects will be apparent from the description below.

Summary of the Invention

It has been found that a desirable combination of hydrophilicity and corrosion resistance can be achieved on the surface of metals, particularly aluminum and its alloys containing at least 75 % by weight aluminum, by contacting the surface, preferably after the surface has already been supplied with a corrosion protective conventional conversion coating, preferably a chromium oxide or chromium phosphate conversion coating, with an aqueous liquid composition comprising, preferably consisting essentially of, or more preferably consisting of, water and:

- (A) from 0.05 to 20 %, preferably from 0.1 to 10.0 %, more preferably from 0.15 to 3.0 %, still more preferably from 0.35 to 1.50 %, measured as dissolved solids, of a component selected from the group of water soluble homopolymers and copolymers, preferably homopolymers, of alkylene imines, preferably of alkylene imines containing no more than 4 carbon atoms per molecule, most preferably of ethylene imine, and salts of such alkylene imine polymers;
- (B) from 0.1 to 60 %, preferably from 0.5 to 19 %, more preferably from 0.7 to 10.0 %, still more preferably from 0.9 to 4.8 %, measured as dissolved solids,

of a component of water soluble alkali metal silicate; and, optionally,

- (C) up to 20 %, preferably from 0.2 to 20 %, more preferably from 0.45 to 6.0 %, still more preferably from 1.4 to 3.0 %, measured as dissolved active ingredient, of a component selected from the group of water soluble, hydroxyl terminated polymers of ethylene oxide and, preferably, of mixtures of ethylene oxide and propylene oxide (these components also being known as "polyether polyols"); and, optionally,
- (D) an anti-microbial effective amount of a anti-microbial agent; and, optionally,
- (E) a component of water soluble non-silicate alkalizing agents to adjust the pH.

One embodiment of the invention is a composition as described above suitable for direct use in imparting hydrophilicity to a metal surface; such a composition may be described hereinafter as a "working composition". Another embodiment of the invention is a concentrate composition, which can be diluted with water to produce a working composition as specified above. A concentrate composition according to this invention preferably comprises from 1.5 to 10, more preferably from 1.5 to 5, or still more preferably from 1.6 to 2.4, times the concentrations of each of the components, except for water, of a working composition.

Still another embodiment of the invention is a process of contacting a metal surface with a working composition according to the invention as described above.

Description of Preferred Embodiments

With increasing preference in the order stated, component (A) as defined above should have a solubility in water of at least 0.01, 0.04, 0.11, 0.27, 0.50, 1.2, 4, 10, 28, 55, or 100 g/L, measured as dissolved solids. The number average molecular weight of component (A) preferably is in the range from 5×10^2 to 10^6 , more preferably from 5×10^3 to 5.0×10^5 , or still more preferably from 9.9×10^3 to 2.0×10^5 .

With increasing preference in the order stated, component (B) as defined above should have a solubility in water of at least 0.35, 0.56, 1.7, 4, 11, 28, 55, or 100 g/L, measured as dissolved solids. With increasing preference in the order given, at least 35, 52, 70, 84, 90, 96, or 99 % by weight of component (B) consists of silicates of sodium and/or potassium, in which the atomic ratio between alkali metal and silicon atoms is in the range from 0.35:1.0 to 5.0:1.0, more preferably from 0.40:1.0 to 4.0:1.0, still more preferably from 0.4:1.0 to 2.0:1.0. (In calculating these ratios, the

alkali metal ions present should be allocated to provide counterions for any stoichiometrically well defined anions, such as hydroxide and phosphates, also found to be present, with only the remaining alkali metal ions being considered as part of the alkali metal silicates.)

5 With increasing preference in the order stated, component (C) as defined above should have a solubility in water of at least 0.02, 0.05, 0.18, 0.35, 0.56, 1.7, 4, 11, 28, 55, or 100 g/L, measured as dissolved solids. The number average molecular weight of component (C), with increasing preference in the order given, is in the range from 10^3 to 5×10^6 , 4×10^3 to 5×10^5 , 7×10^3 to 1.0×10^5 , or 1.0×10^4 to 5.0×10^4 .

10 A working composition according to the invention preferably has a pH in the range from 7 to 14, more preferably from 9.0 to 14, still more preferably from 10.0 to 13.0. If an alkalinizing agent is needed to bring the pH into these ranges when the other necessary and/or preferred ingredients are used alone, alkali metal hydroxides and/or phosphates, including condensed phosphates, are preferably used for this purpose, with a combination of potassium hydroxide and tetrapotassium pyrophosphate
15 particularly preferred.

 Under certain conditions, compositions containing only components (A) - (C) as described above have been found to develop undesired growth of microbes such as fungi during storage. A wide variety of antimicrobial agents are known in the art and
20 are suitable for use in compositions according to this invention, in order to prevent such growth. Examples include zinc 2-pyridinethiol-1-oxide, poly(hexamethylenebiguanide) hydrochloride, 1,2-benzisothiazoline-3-one, 4-chloro-3,5-xyleneol, amine 4,4-dimethyloxazolidines, abietic acid, tetrachloroisophthalonitrile, diiodomethyl-p-tolylsulfone, 3-iodo-2-propylbutyl carbonate, 2[(hydroxymethyl)amino]-2-methyl-1-propanol,
25 2[(hydroxymethyl)amino]ethanol, 1-(4-thiazolyl)benzimidazole, 1-methyl-3,5,7-triazol-1-azoniatricylcodecane chloride, and hexahydro-1,3,5-tris(2-hydroxyethyl)-S-triazine. Generally, total amounts of from 0.03 to 0.20 % of antimicrobial agent, preferably 1-(4-thiazolyl)benzimidazole in a powder or paste form, are preferred in a concentrate composition according to this invention.

30 The hydrophilicizing process according to the invention is preferably accomplished by contacting a metal surface already bearing a corrosion protective conversion coating with a composition according to the invention as described above at tempera-

ture within the range from 10 to 90, more preferably from 12 to 80, still more preferably from 15 to 70, ° C for a time within the range from 5 to 600, more preferably from 10 to 300, still more preferably from 10 to 200, seconds (hereinafter often abbreviated "sec"). Contact may be accomplished by any convenient means, such as immersion, spray, brush, squeegee roll, and the like; many such means of contact are known *per se* in the art. After the hydrophilicizing treatment, the treated surface is normally dried, preferably without any intermediate rinsing. While drying in the natural atmosphere without any use of heat can be used, normally faster drying by any convenient method is preferred; drying methods also are well known in the art. It is preferred that the temperature of the treated metal surface be maintained within the range from 10 to 300, more preferably from 15 to 250, still more preferably from 30 to 200, ° C during drying. The mass added to the surface treated by the hydrophilicizing treatment according to the invention is preferably within the range from 0.05 to 3.0, more preferably from 0.1 to 2.0, still more preferably from 0.1 to 1.5, grams per square meter (hereinafter often abbreviated as "g/m²") of surface treated.

For various reasons it is often preferred that compositions according to the invention as defined above should be substantially free from many ingredients used in compositions for similar purposes in the prior art. Specifically, it may be increasingly preferred in the order given, independently for each preferably minimized component listed below, that these compositions, when directly contacted with metal in a process according to this invention, contain no more than 1.0, 0.35, 0.10, 0.08, 0.04, 0.02, 0.01, or 0.001 % of each of the following constituents: hexavalent chromium, ferricyanide, ferrocyanide, anions containing sulfur, molybdenum, or tungsten, any metallic elements (other than silicon) in an oxidation state of +2 or higher, nitrate and other oxidizing agents (the others being measured as their oxidizing stoichiometric equivalent as nitrate), organic liquids with a boiling point below 120° C, organic compounds containing amino and/or amido groups, and undissolved dispersed silica exclusive of that chemically combined with an alkali metal as part of component (C) as described above.

For aluminum substrates, it is preferred that the surfaces to be treated according to this invention first be provided with a conventional chromium containing conversion coating treatment selected from the types of treatments generally known in the

art as "chromium phosphate" and "chromium oxide" treatments. The former are illustrated, without limitation, by U. S. Patents 4,668,305 and 4,131, 489 and the latter is illustrated, without limitation, by U. S. Patent 4,146,410. The add-on mass from the conversion coating treatment is preferably within the range from 0.1 to 10, more preferably from 0.5 to 5.0, or still more preferably from 1.0 to 4.0, g/m² of surface treated when either a chromium phosphate treatment or a chromium oxide treatment is used. Preferably there is at least one water rinse after application of the conversion coating treatment before contact of the surface with a hydrophilicizing treatment according to this invention. Before the conversion coating treatment, or before the hydrophilicizing treatment if no conversion coating is used, the surfaces to be treated are preferably cleaned of any foreign matter and then rinsed, in one of the many ways known in the art to be effective.

The practice and benefits of the invention may be further appreciated by consideration of the following non-limiting working and comparison examples.

General Conditions for the Examples and Comparison Examples

The surfaces treated were those on sample sheets (panels) of Type 3003 aluminum alloy. The processing sequence for all experiments included the following steps:

1. Acid cleaning by immersion of the samples sheets in a solution of 1.5 % by volume of a concentrated acid cleaner formulation in water at 38° C for 2.0 minutes.
2. Rinse with cold tap water for 30 sec.
3. Chromate conversion coating by immersion in either a 7.7 % by weight solution in water of ALODINE® 407-47 at 38° C for 120 sec to produce a coating add-on mass of 2.2 - 2.7 g/m² or a 7.2 % by weight solution in water of BOND-
DERITE® 713 at 43° C for 90 sec to produce an add-on mass of 1.5 - 1.9 g/m².
4. Rinse with cold tap water for 30 sec.
5. Hydrophilicizing treatment at 29° C for 60 sec by immersion of the samples, producing a wet coating mass of about 0.75 g/m².
6. Air blowoff by impinging pressurized air against sample surface until no more liquid is blown off by the air stream.
7. Oven dry at 138° C for 20 minutes.

ALODINE® 407-47 (keyed in the table below as "A") and BONDERITE® 713 (keyed in the table below as "B") are both commercially available from the Parker+Amchem Div. of Henkel Corp., Madison Heights, Michigan, USA; the former is an example of a concentrate for a chromium phosphate type conversion coating treatment and the latter is an example of a concentrate for a chromium oxide type conversion coating treatment. Prior art conventional hydrophilicizing treatments are exemplified by PARCOLENE® HX-2020 and PALENE® 210, also both available from the Parker+Amchem Div. of Henkel Corp., Madison Heights, Michigan, USA.

The hydrophilicizing treatment compositions used and their keys in the table below were:

H-1 (not according to the invention): PARCOLENE HX-2020

H-2 (not according to the invention): PALENE 210

H-3 (according to the invention):

0.5 % (as dry solids) of POLYMIN™ p, a commercially available aqueous solution of poly(ethylene imine) with an average molecular weight of about 70,000 and about 50 % dissolved solids from BASF Corp;

2.0 % (as active ingredient) of MACOL™ 27, a block copolymer of ethylene and propylene oxides from PPG Corp;

1.57 % by weight (dry solids) of potassium silicate with an alkali metal to silicon atomic ratio of 0.606: 1.0;

0.09 % of METASOL™ TK-100, an antimicrobial agent commercially available from Calgon Corp. and reported to contain 1-(4-thiazolyl)benzimidazole as its principal active ingredient;

and the balance water. The pH was 11.8 ± 0.5 .

H-4 (according to the invention): This was the same as H-3, except that the antimicrobial agent was omitted.

H-5 (according to the invention): This was the same as H-3, except that the MACOL™ 27 was omitted, and the amount of POLYMIN™ p was increased to give a concentration of 1.0 %.

The samples prepared as described above were subjected to the following evaluation tests:

1. Sea water acidified accelerated test (abbreviated "SWAAT") according to

American Society for Testing Materials (hereinafter often abbreviated "ASTM") standard method G 85-85.

2. Water wettability, with and without hot aging. The water wettability was tested as follows: A syringe was placed at a distance between the test panel and the delivery point of the syringe of about 5 millimeters (hereinafter often abbreviated as "mm"). Then, an amount of 0.5 milliliter of deionized water was dropped onto the panel over a time of 2 sec. About 25 seconds after the delivery of the water onto the panel had been completed, the diameter of the water wetted area on the sample was measured and recorded. Hot aging was performed by placing the panels in a 90° C oven for 168 hours, then removing the panels were set at room temperature for 3 hours, before measuring the wettability.

The conversion coating and hydrophilicizing treatment compositions used and the test results obtained are shown in Table 1 below.

TABLE I

Example No.	Conversion Coating	Hydrophilicizing Composition	pH	Wetted Area Diameter, mm		Days To Failure in SWATT Test	Salt Spray Rating, 1000 Hours
				Un-aged	Aged		
1	A	None	-	19	13	-	pits and pin holes
2	B	None	-	23	13	-	N
3	A	H-1	11.45	65	26	6-8	N
4	B	H-2	4.57	20	17	2-3	N
5	A	H-3	11.80	51	56	-	N
6	A	H-4	11.51	42	50	10-11	N
7	A	H-5	11.90	50	65	-	N

The results in Table 1 show that the aluminum treated according to the invention has adequate hydrophilicity, which is well retained or even improved by heating, and superior resistance to sea water corrosion.

The invention claimed is:

CLAIMS

1. A concentrate composition for preparing a hydrophilicizing treatment by dilution with water, said concentrate composition consisting essentially of:

5 (A) from about 0.3 to about 60 % by weight of a component selected from the group consisting of water soluble homopolymers and copolymers of alkylene imines and salts of such alkylene imine polymers;

(B) from about 0.5 to about 40 % by weight of a component of water soluble alkali metal silicates; and, optionally,

10 (C) not more than about 60 % by weight of a component of water soluble hydroxyl terminated polymers of (i) ethylene oxide or (ii) mixtures of ethylene oxide and propylene oxide; and, optionally,

(D) an anti-microbial effective amount of a anti-microbial agent; and, optionally,

(E) up to about 20 % by weight of a component of water soluble non-silicate alkalizing agents; and, optionally.

15 (F) water.

2. A composition according to claim 1, wherein the concentration of component (A) is in the range from about 0.7 to about 40 % by weight, the concentration of component (B) is in the range from about 1.0 to about 30 % by weight, and the concentration of component (C) is not more than about 40 % by weight.

20 3. A composition according to claim 2, wherein the concentration of component (A) is in the range from about 0.7 to about 20 % by weight, the concentration of component (B) is in the range from about 1.0 to about 20 % by weight, the concentration of component (C) is not more than about 20 % by weight, and the concentration of component (D) is in the range from about 0.03 to about 0.20 % by weight.



4. A composition according to claim 3, wherein at least 85 % of component (A) consists of polymers, or salts of polymers, of ethylene imine and component (A) has an average molecular weight in the range from about 5×10^2 to 10^6 ; at least 84 % of component (B) consists of silicates of sodium, potassium, or a mixture thereof in which the atomic ratio between alkali metal and silicon atoms is in the range from about 0.40:1.0 to about 2.0:1.0; and at least 95 % of component (C) if present consists of molecules containing both oxyethylene and oxypropylene oligomer blocks and the average molecular weight of component (C) if present is in the range from about 7×10^3 to 1.0×10^5 .

5. A composition according to claim 2, wherein at least 73 % of component (A) consists of polymers of alkylene imines containing no more than 4 carbon atoms per molecule and component (A) has an average molecular weight in the range from about 5.0×10^3 to 5.0×10^5 ; at least 52 % of component (B) consists of silicates of sodium, potassium, or a mixture thereof in which the atomic ratio between alkali metal and silicon atoms is in the range from about 0.35:1.0 to about 5.0:1.0; and at least 75 % of component (C) if present consists of molecules containing both oxyethylene and oxypropylene oligomer blocks and the average molecular weight of component (B) is in the range from about 4×10^3 to 5×10^5 .

6. A composition according to claim 1, wherein component (A) has an average molecular weight in the range from about 10^3 to 10^6 ; at least 52 % of component (B) consists of silicates of sodium, potassium, or a mixture thereof in which the atomic ratio between alkali metal and silicon atoms is in the range from about 0.35:1.0 to about 5.0:1.0; and the average molecular weight of component (C) if present is in the range from about 10^3 to 5×10^6 .



7. A process of hydrophilicizing a metal surface, said surface optionally bearing a pre-existing conversion coating, said process comprising contacting the metal surface for a time within the range from about 5 to about 600 seconds at a temperature within the range from about 10 to about 90 ° C with a hydrophilicizing composition consisting essentially of water and:

- (A) from about 0.05 to about 20 % by weight of a component selected from the group consisting of water soluble homopolymers and copolymers of alkylene imines and salts of such alkylene imine polymers;
- (B) from about 0.1 to about 60 % by weight of a component of water soluble alkali metal silicates; and, optionally,
- (C) up to about 20 % by weight of a component of water soluble hydroxyl terminated polymers of (i) ethylene oxide or (ii) mixtures of ethylene oxide and propylene oxide; and, optionally,
- (D) an anti-microbial effective amount of an anti-microbial agent; and, optionally,
- (E) a component of water soluble non-silicate alkalizing agents to adjust the pH.

8. A process according to claim 7, wherein, in the hydrophilicizing composition, the concentration of component (A) is in the range from about 0.1 to about 10.0 %, the concentration of component (B) is in the range from about 0.5 to about 19 %, and the concentration of component (C) is not more than about 6.0 %.

9. A process according to claim 8, wherein, in the hydrophilicizing composition, the concentration of component (A) is in the range from about 0.15 to about 3.0 %, the concentration of component (B) is in the range from about 0.7 to about 10.0 %, and the concentration of component (C) is in the range from about 0.45 to about 6.0 %.

10. A process according to claim 9, wherein, in the hydrophilicizing composition, the concentration of component (A) is in the range from about 0.35 to about 1.50 %, the concentration of component (B) is in the range from about 0.9 to about 4.8 %, and the concentration of component (C) is in the range from about 1.4 to 3.0 %.



11. A process according to claim 10, wherein at least 73 % of component (A) has an average molecular weight in the range from about 5×10^2 to 10^6 ; at least 52 % of component (B) consists of silicates of sodium, potassium, or a mixture thereof in which the atomic ratio between alkali metal and silicon atoms is in the range from about 0.35:1.0 to about 5.0:1.0; and the average molecular weight of component (C) is in the range from about 10^3 to 5×10^6 .

12. A process according to claim 11, wherein at least 85 % of component (A) consists of polymers of alkylene imines containing no more than 4 carbon atoms per molecule and component (A) has an average molecular weight in the range from about 5.0×10^3 to 5.0×10^5 ; at least 52 % of component (B) consists of silicates of sodium, potassium, or a mixture thereof in which the atomic ratio between alkali metal and silicon atoms is in the range from about 0.35:1.0 to about 5.0:1.0; and at least 75 % of component (C) consists of molecules containing both oxyethylene and oxypropylene oligomer blocks and the average molecular weight of component (C) is in the range from about 4×10^3 to 5×10^5 ; and the pH is in the range from about 9.0 to about 14.0.

13. A process according to claim 12, wherein the surface treated is an aluminum or aluminum alloy surface precoated with a chromium oxide or chromium phosphate conversion coating; the mass added to the surface treated by the hydrophilicizing treatment is within the range from about 0.1 to 1.5 g/m^2 of surface treated; the temperature during contact between the hydrophilicizing composition and the metal is within the range from about 15 to about 70°C ; and the time of contact is within the range from about 10 to about 200 seconds.

14. A process according to claim 11, wherein the surface treated is an aluminum or aluminum alloy surface precoated with a chromium oxide or chromium phosphate conversion coating; the mass added to the surface treated by the hydrophilicizing treatment is within the range from about 0.1 to about 2.0 g/m^2 of surface treated; the temperature during contact between the hydrophilicizing composition and the metal is within the range from about 12 to about 80°C ; and the time of contact is within the range from about 10 to about 200 seconds.



15. A process according to claim 10, wherein the surface treated is an aluminum or aluminum alloy surface precoated with a chromium oxide or chromium phosphate conversion coating; the mass added to the surface treated by the hydrophilicizing treatment is within the range from about 0.05 to about 3.0 g/m² of surface treated; and the time of contact is within the range from about 10 to about 300 seconds.

16. A process according to claim 9, wherein the surface treated is an aluminum or aluminum alloy surface precoated with a chromium oxide or chromium phosphate conversion coating; the mass added to the surface treated by the hydrophilicizing treatment is within the range from about 0.05 to about 3.0 g/m² of surface treated; and the time of contact is within the range from about 10 to about 300 seconds.

17. A process according to claim 8, wherein the surface treated is an aluminum or aluminum alloy surface precoated with a chromium oxide or chromium phosphate conversion coating; the mass added to the surface treated by the hydrophilicizing treatment is within the range from about 0.1 to about 2.0 g/m² of surface treated; the temperature during contact between the hydrophilicizing composition and the metal is within the range from about 12 to about 75 ° C; and the time of contact is within the range from about 10 to 200 seconds.

18. A process according to claim 7, wherein the surface treated is an aluminum or aluminum alloy surface precoated with a chromium oxide or chromium phosphate conversion coating; the mass added to the surface treated by the hydrophilicizing treatment is within the range from about 0.05 to about 3.0 g/m² of surface treated; the temperature during contact between the hydrophilicizing composition and the metal is within the range from about 12 to about 75 ° C; and the time of contact is within the range from about 10 to about 300 seconds.

19. A process according to claim 18, wherein the mass added to the surface treated by the conversion coating treatment is within the range from about 1.0 to about 4.0 g/m² of surface treated.

20. A process according to claim 13, wherein the mass added to the surface treated by the conversion coating treatment is within the range from about 0.1 to about 10 g/m² of surface treated.

