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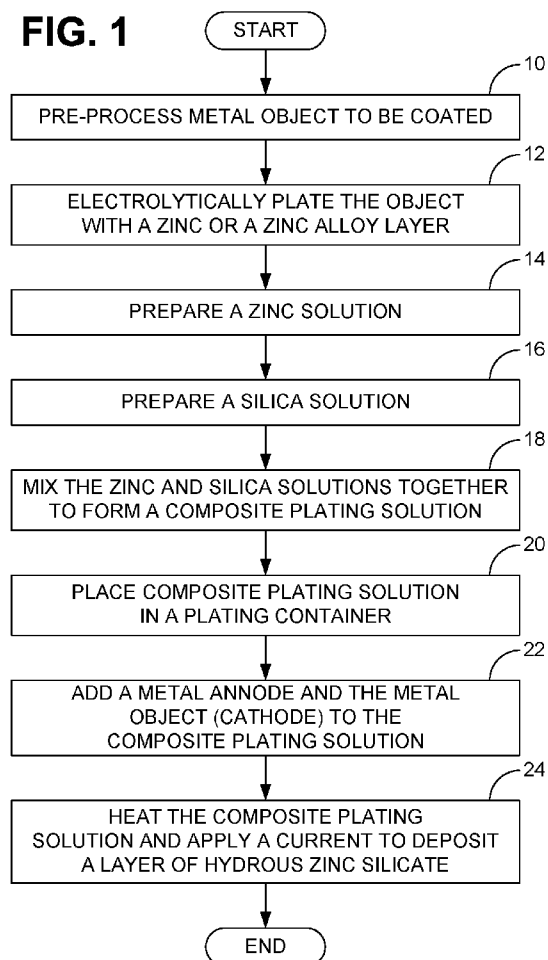
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[Continued on next page]

(54) Title: COMPOSITE CERAMIC COATINGS FOR ANTI-CORROSION PROTECTION

FIG. 1



(57) Abstract: In some embodiments, an anti-corrosive composite ceramic coating includes an inner metal layer formed on the surface of a metal object to be protected and an outer composite ceramic layer formed on the inner metal layer.



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COMPOSITE CERAMIC COATINGS FOR ANTI-CORROSION PROTECTION

Background

Hot-dip galvanizing is one of the most commonly used methods for protecting steel surfaces against corrosion. Another common corrosion protection method is to apply anti-corrosive paint to steel surfaces. Although hot-dip galvanized steel resists corrosion well in numerous environments, there has recently been concern of health damage to the human body caused by zinc exposure. Regarding anti-corrosive paint, the hexavalent chromium used in the production of such paint presents an environmental and health hazard. There is a need for an anti-corrosion technology that does not involve the use of hazardous chemicals or processes.

Brief Description of the Drawings

The present disclosure may be better understood with reference to the following Figures. Matching reference numerals designate corresponding parts throughout the Figures, which are not necessarily drawn to scale.

Fig. 1 is a flow diagram of an example method for forming and applying a

composite ceramic coating.

Fig. 2 is a schematic drawing of an embodiment of a metal object having a composite ceramic coating.

Fig. 3 is a light-optical micrograph of a cross-section of an example composite ceramic coating formed on a steel plate.

Fig. 4 is a scanning electron microscopy (SEM)/energy-dispersive X-ray spectroscopy (EDS) analysis of the interface between carbon steel and the zinc layer.

Fig. 5 is an X-ray diffraction (XRD) pattern of the composite ceramic coating.

Fig. 6 is a model of the crystal structure of $\text{Zn}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$.

Fig. 7 includes images of specimens in Q-FOG racks prior to and after salt spray testing.

Fig. 8 includes images of multiple samples (Samples 1-9) tested in the salt spray testing and evaluation of the degree of rusting in accordance with ASTM D 610-01.

Fig. 9 includes optical micrographs of the coated samples (specimens 1-6) after the salt spray testing.

Detailed Description

As described above, there is a need for an anti-corrosion technology that does not involve the use of hazardous chemicals or processes. Disclosed herein are composite ceramic coatings that can provide anti-corrosion protection to metal without such hazards. In some embodiments, the composite ceramic coating comprises a hydrous zinc silicate layer that is applied to a metal object, such as a steel object, using an electrolytic process. The composite ceramic coating is

extremely resistant to corrosion and has a crystalline structure that is extremely stable.

In the following disclosure, various specific embodiments are described. It is to be understood that those embodiments are example implementations of the disclosed inventions and that alternative embodiments are possible. All such embodiments are intended to fall within the scope of this disclosure.

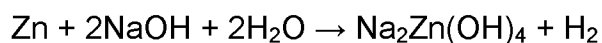
As expressed above, disclosed herein are composite ceramic coatings that can be formed on metal using an electrolytic process. In some embodiments, the composite ceramic coating includes a composite ceramic layer that comprises both metal, such as zinc (Zn), and silicon (Si).

Fig. 1 is a flow diagram of an example method for preparing and applying a composite ceramic coating. Beginning with block 10 of Fig. 1, a metal object to be coated is pre-processed. The object can comprise any object that would benefit from anti-corrosion protection. For example, the object can comprise a low-carbon steel object. In some embodiments, pre-processing comprises cleaning and/or polishing the surface of the object to improve adhesion of materials that are to be applied to the object. In other embodiments, the roughness of the surface of the object can be increased to improve mechanical adhesion.

Turning next to block 12, the object is electrolytically plated with Zn or a Zn alloy, such as Zn-Ni, Zn-Fe, and Zn-Sn. This results in a layer of Zn or a Zn alloy being deposited on the surface of the object in similar manner to conventional galvanization. In some embodiments, the Zn or Zn alloy layer is approximately 4 to 25 μm thick. In other embodiments, the Zn or Zn alloy layer is approximately 200 to 300 μm thick.

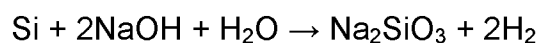
Once the Zn or Zn alloy layer has been deposited, the object can be coated

with a composite ceramic material. A composite plating solution can be prepared by separately preparing a Zn solution (block 14) and an Si solution (block 16). In some embodiments, the Zn solution can be prepared by placing Zn flakes in a container with water (H₂O) and adding pellets of sodium hydroxide (NaOH) over a period of time (e.g., 1 hour) at an elevated temperature (e.g., 80°C) to cause a reaction. The reaction is continued for an extended period of time (e.g., 48 hours) and produces a solution of sodium zincate:



In some embodiments, the resulting sodium zincate solution has molar ratio of approximately Zn:NaOH:H₂O = 1:1:6. Although a Zn solution have been specifically identified, it is noted that, in other embodiments, a plating solution can comprise another metal material, such as aluminum (Al), nickel (Ni), tin (Sn), titanium (Ti), beryllium (Be), or copper (Cu).

In some embodiments, the Si solution can be prepared by placing chunks of a high purity Si mineral, which is commercially available as "Si metal," in a container with H₂O and adding pellets of NaOH over a period of time (e.g., 1 hour) to cause a reaction. The reaction is continued for an extended period of time (e.g., 6 hours) and produces a clear and colorless solution of sodium silicate:



In some embodiments, the sodium silicate solution has a molar ratio of approximately Si:NaOH:H₂O = 6:1:10 and a specific gravity (SG) of approximately 1.8

to 2.0. The sodium silicate solution can be diluted with water to a SG of approximately 1.3.

With reference back to Fig. 1, the Zn solution (i.e., sodium zincate solution) can be mixed with the Si solution (i.e., sodium silicate solution) to form a composite plating solution (block 18). In some embodiments, one part of the Zn solution is mixed with two parts of the Si solution to form the composite plating solution having an Si/Zn ratio of approximately 2 to 5. This ratio enables efficient plating.

The composite plating solution can then be placed in a plating container in which the metal object is to be plated with the solution (block 20). A metal (e.g., Zn) anode and the metal object (cathode) can be added to the composite solution (block 22). Next the composite plating solution can be heated and a current can be applied to the solution for several minutes to form a composite ceramic layer (block 24). In some embodiments, a current of approximately 1 to 4 amps (e.g., 1 amps) is applied to the composite plating solution for approximately 1 to 30 minutes (e.g., 5 minutes) at a temperature of approximately 25 to 90°C (e.g., 60°C). In other embodiments, a current of approximately 0.5 to 1 amp is applied to the composite plating solution for approximately 15 to 30 minutes at a temperature of approximately 30 to 40°C. During this time, a composite ceramic layer comprising hydrous zinc silicate is electrolytically deposited on the surface of the object over the Zn/Zn alloy layer. In some embodiments, the hydrous zinc silicate layer has a thickness of approximately 40 to 300 μm . In other embodiments, the hydrous zinc silicate layer has a thickness of approximately 260 to 300 μm .

Fig. 2 schematically illustrates a coated metal object 30 of the type that can be produced using the method of Fig. 1. As shown in Fig. 2, a composite ceramic coating 32 has been applied to a metal object 34 having a steel body. As indicated in

this figure, the coating 32 includes a first or inner metal layer 36 formed directly on the metal object and a second or outer composite ceramic layer 38 formed directly on the inner metal layer. In some embodiments, the inner metal layer 36 is a layer of Zn or a Zn alloy and the outer composite ceramic layer 38 is a layer of hydrous zinc silicate and the coating 32 comprises no other materials.

Fig. 3 shows an actual example of a steel object coated in the manner described in relation to Fig. 1. As can be seen in Fig. 3, the composite ceramic coating includes a first or inner metal layer comprising Zn and a second or outer composite ceramic layer comprising hydrous zinc silicate. In this example, the Zn layer is approximately 49 μm thick and the hydrous zinc silicate layer is approximately 140 μm .

Scanning electron microscopy (SEM) analysis was performed using a Hitachi SU1510 variable pressure electron microscope at an accelerating voltage of 30 kV. The results of the study are presented in the Fig. 4. The image in Fig. 4A shows the SEM micrograph of the interface between the carbon steel and zinc layer. The images in Figs. 4B-4D depict the element distribution maps: Zn (Fig. 4B), Fe (Fig. 4C) and O (Fig. 4D). The SEM micrographs indicate that there is an inter-diffusion of Zn atoms onto the steel structure. Due to this inter-diffusion, an interface having a composition of Fe_xZn_y is produced. The values of x and y depend on the exact deposition conditions and can be controlled with the temperature of the deposition. The Fe_xZn_y layer composition changes with the thickness. The images in Figs. 4B-4D reveal the presence of a clear boarder between iron of the steel and Zn of the inner layer of the coating.

The X-ray diffraction (XRD) pattern of the composite ceramic coating was collected using a Rigaku Ultima IV diffractometer with Cu $\text{K}\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$)

between 2θ of 2 to 75° at a power settings of 40 kV and 44 mA. Fig. 5 shows the XRD pattern of the composite ceramic coating. The peak positions and intensities in the pattern indicate that the sample is composed of hydrous zinc silicate, $\text{Zn}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$. The structure of the silicate is composed of Si_2O_5 sheets with zinc sandwiched between sheets in octahedral sites. Fig. 6 shows a model of the crystal structure of $\text{Zn}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$.

The salt spray (fog) test is a standardized test method used to evaluate corrosion resistance of coated samples. Five hundred hours of salt spray testing was conducted in accordance with the ASTM B117-11 standard in a Q-FOG cyclic corrosion tester. During an ASTM B117 test, specimens are exposed to a continuous fog of a 5 wt. % solution of sodium chloride (greater concentration than sea water, which is only 1.8% to maximum 3%) at an elevated temperature of 35°C . The justification for these extreme conditions is that a coating system that will resist these test conditions should also perform well in aggressive service environments. Five hundred hours of salt fog testing is roughly equivalent to 2.5 years in a coastal environment. Nine test specimens were used and no previous treatment was performed prior to the testing. The specimens included six steel plates coated with the composite ceramic coating of this disclosure (specimens 1-6), two Zn-coated samples (specimens 7-8), and one uncoated carbon steel plate (specimen 9). Fig. 7 shows images of the specimens in their Q-FOG racks prior to and after salt spray testing. The images labeled "A" contained (from bottom to top) samples 1, 2, 3, 4, and 5 and the images labeled "B" contained (from bottom to top) samples 6, 7, 8, and 9. The thicknesses of the composite ceramic coatings were measured on polished cross-sections by using an Olympus DSX500 opto-digital microscope. The results of the thickness measurements are presented in the Table 1.

Table 1: Thickness of the Ceramic-Zinc Coatings

Sample number	Coating thickness (μm)	Standard deviation
1	44.0	9.9
2	73.0	4.1
3	80.6	8.4
4	122.6	9.1
5	161.4	13.1
6	298.0	8.8

The results of the salt spray testing for specimens 1-9 are illustrated in Fig. 8. Fig. 9 includes optical micrographs of the coated samples (specimens 1-6) after the salt spray testing (R_a is the mean surface roughness). As can be appreciated from these figures, the specimens having the composite ceramic coatings remained free of rust after 500 hours of exposure. This significantly exceeds the Society of Automotive Engineers (SAE) requirement of 96 hours of salt spray without red rust. The obtained results demonstrate that the composite ceramic coated plates have excellent corrosion protection compared to the uncoated and Zn-coated plates, which suggests that the coating will be able to withstand harsh operating environments, particularly those that promote the rapid onset of red rust. The best anticorrosion performance was obtained for the specimens 5 and 6, which had the highest coating thickness. Notably, however, thinner layers can still be used for less demanding applications, such as those in which the rate of corrosion is lower.

Micro-hardness measurements were performed on the outer layer of the composite ceramic coating and it was determined that its average hardness was approximately 58 HV, which is within range of hardness reported for electrodeposited Zn. Pull-off strength testing was also performed and showed very good adhesion of the hydrous zinc silicate to the Zn layer on the steel surface with

an average value of stress at break of approximately 3.40 MPa. These hardness and adhesion results indicate that the material is resistant to mechanical scratches.

Cyclic voltammetry tests were also performed and indicated that the composite ceramic coating effectively protects the coated steel from the electrochemical attack by aggressive ions from solution, thereby providing an effective anticorrosion performance.

Tribological testing was additionally performed and indicated that the composite ceramic coating is effective in promoting the lubrication performance of engine oil by reducing the coefficient of friction by up to 10%.

CLAIMS

Claimed are:

1. An anti-corrosive composite ceramic coating comprising:
an inner metal layer formed on the surface of a metal object to be protected;
and
an outer composite ceramic layer formed on the inner metal layer.
2. The coating of claim 1, wherein the inner metal layer is a zinc or zinc alloy layer.
3. The coating of claim 1, wherein the inner metal layer is approximately 4 to 300 μm thick.
4. The coating of claim 1, wherein the composite ceramic layer contains zinc and silicon.
5. The coating of claim 1, wherein the composite ceramic layer is a hydrous zinc silicate layer.
6. The coating of claim 5, wherein the hydrous zinc silicate layer is made of $\text{Zn}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$.
7. The coating of claim 1, wherein the composite ceramic layer is approximately 40 to 300 μm thick.

8. A coated metal object protected by an anti-corrosive composite ceramic coating, the coated metal object comprising:

a steel body having a surface; and

an anti-corrosive composite ceramic coating formed on body, the coating including an inner metal layer formed on the surface and an outer composite ceramic layer formed on the inner metal layer.

9. The object of claim 8, wherein the inner metal layer is a zinc or zinc alloy layer.

10. The object of claim 8, wherein the composite ceramic layer is a hydrous zinc silicate layer.

11. A method for forming an anti-corrosive coating on a metal object, the method comprising:

electrolytically plating the surface of the object with a metal to form an inner layer; and

electrolytically depositing a composite ceramic layer on top of the inner layer.

12. The method of claim 11, wherein electrolytically plating the surface of the object comprises electrolytically plating the surface with zinc or a zinc alloy.

13. The method of claim 11, wherein electrolytically depositing a composite ceramic layer comprises electrolytically depositing a hydrous zinc silicate layer on top of the inner layer.

14. The method of claim 13, wherein electrolytically depositing a composite ceramic layer further comprises mixing a zinc solution with a silicon solution to form a composite plating solution.

15. The method of claim 14, wherein the composite plating solution has an Si:Zn ratio of approximately 2:5.

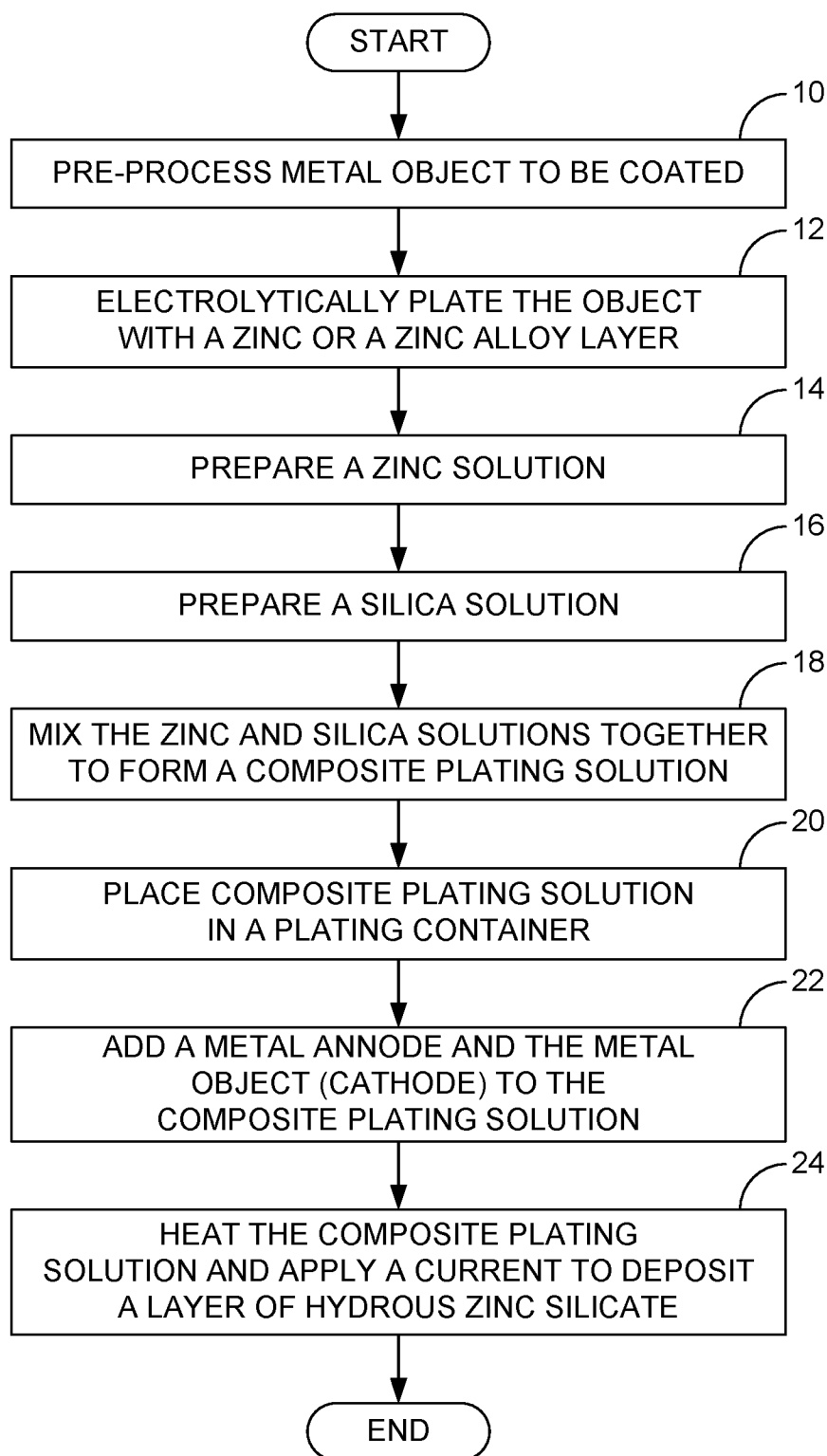
16. The method of claim 14, wherein electrolytically depositing a composite ceramic layer further comprises applying a current of approximately 0.5 to 4 amps to the composite plating solution.

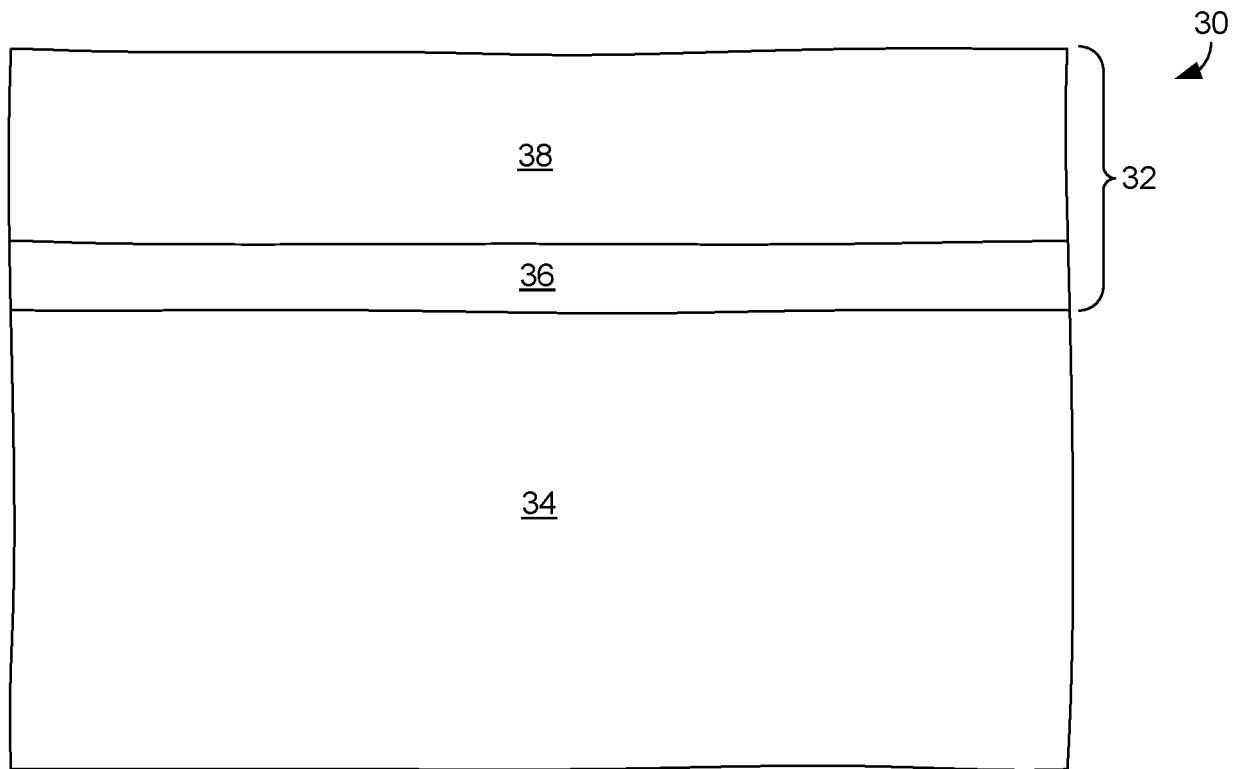
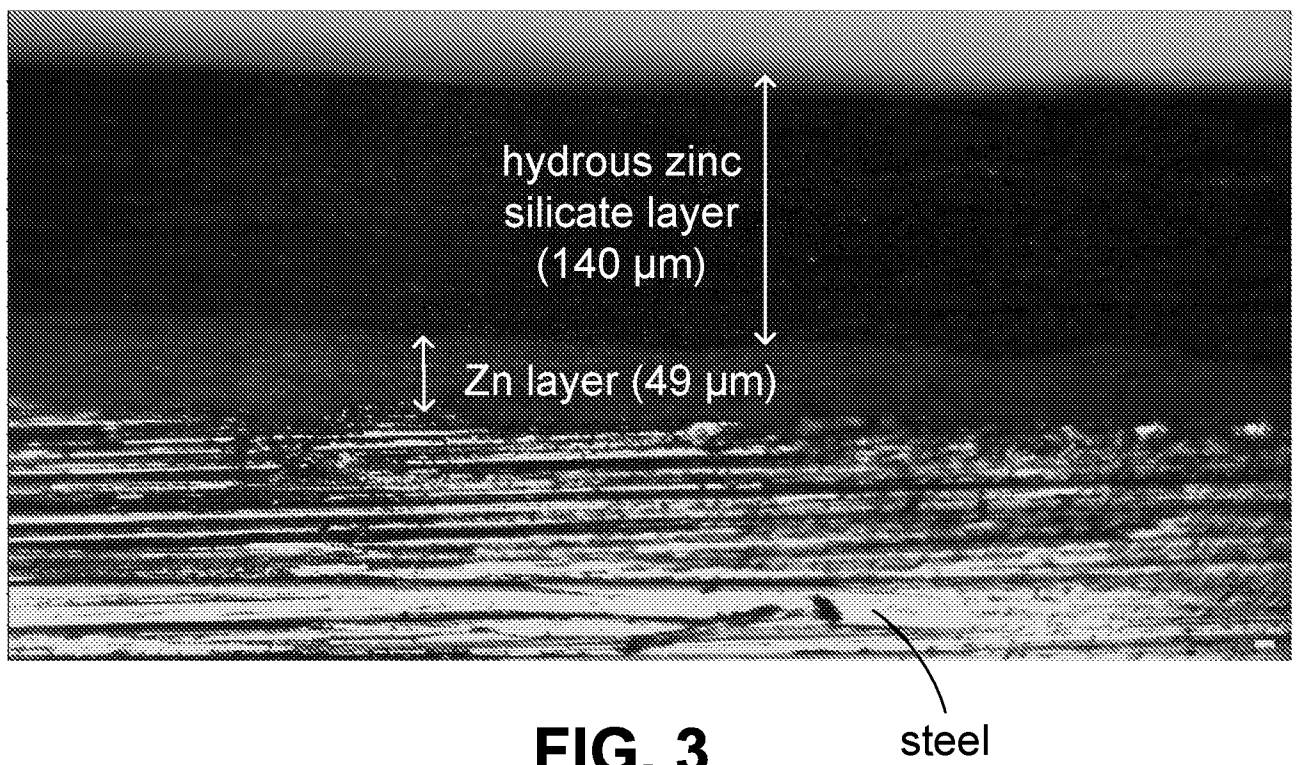
17. The method of claim 16, wherein electrolytically depositing a composite ceramic layer further comprises heating the composite plating solution to a temperature of approximately 25 to 90°C.

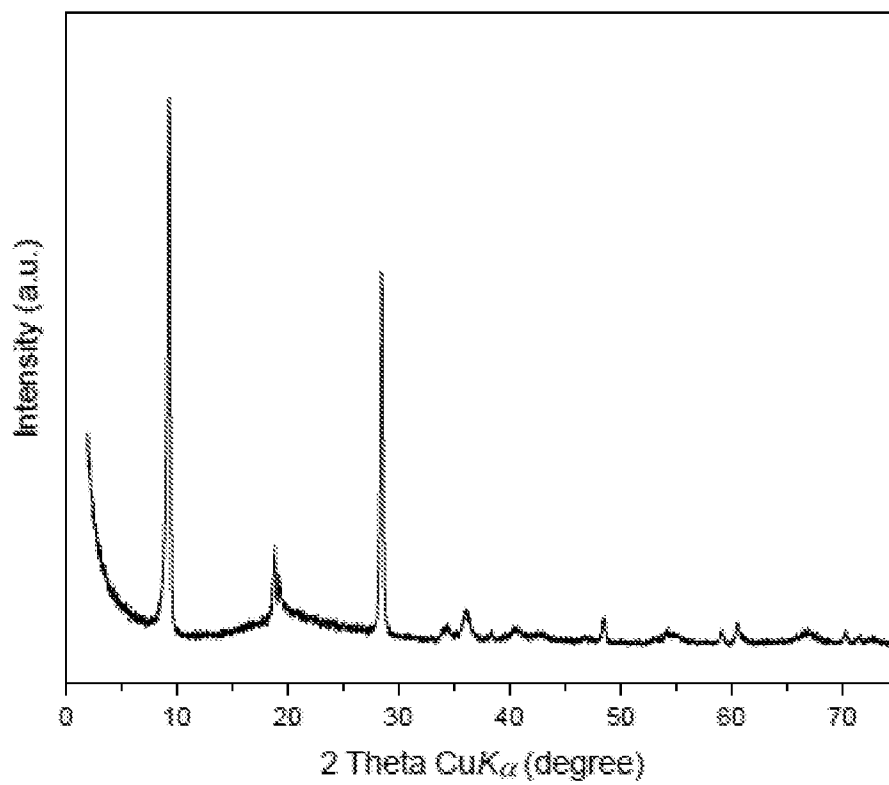
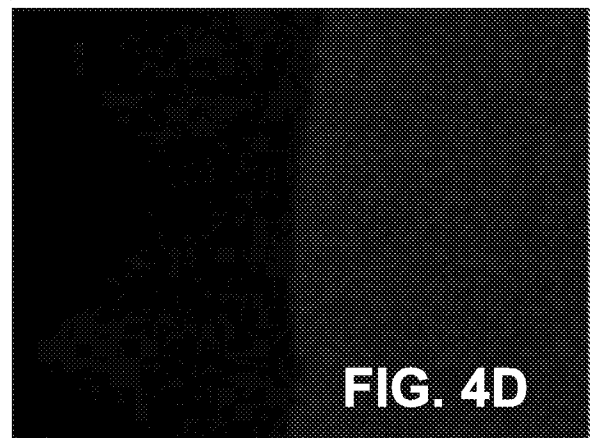
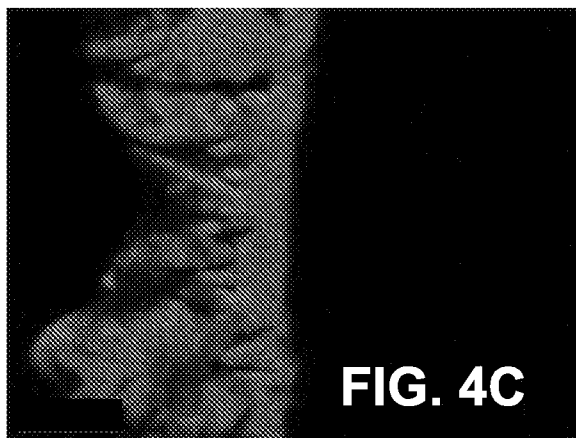
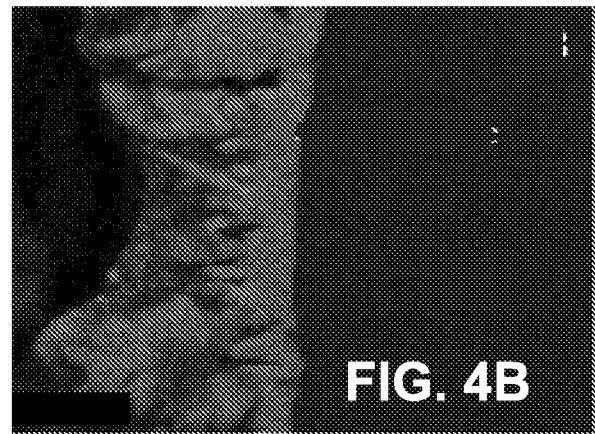
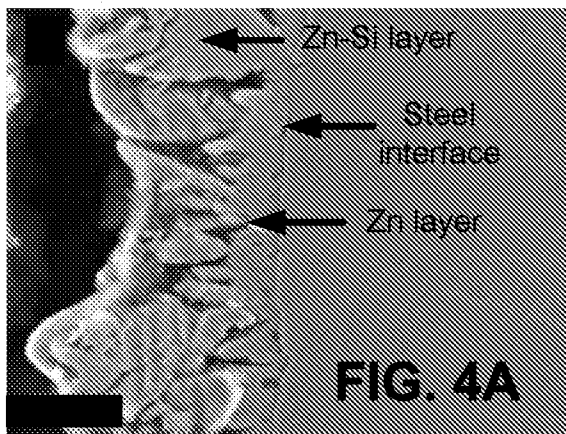
18. The method of claim 11, further comprising pre-processing the metal object before electrolytically plating the surface of the object.

19. The method of claim 19, wherein pre-processing comprises cleaning and polishing the surface of the object

20. The method of claim 18, wherein pre-processing comprises increasing the roughness of the surface of the metal object.

**FIG. 1**

**FIG. 2****FIG. 3**

**FIG. 5**

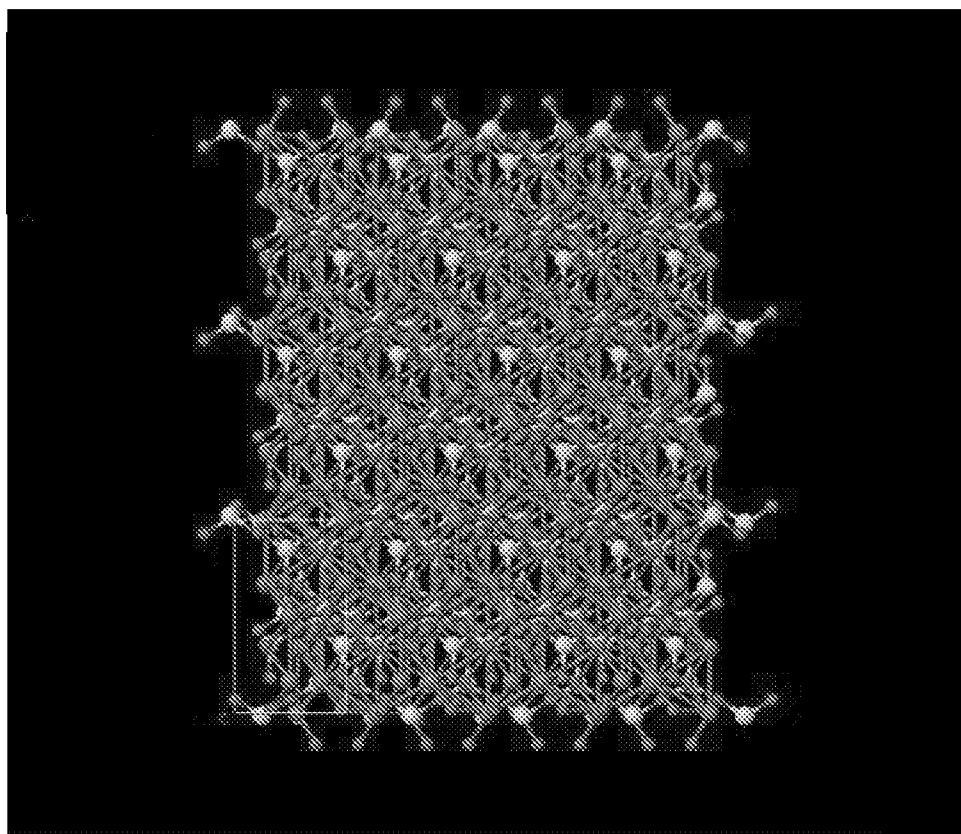
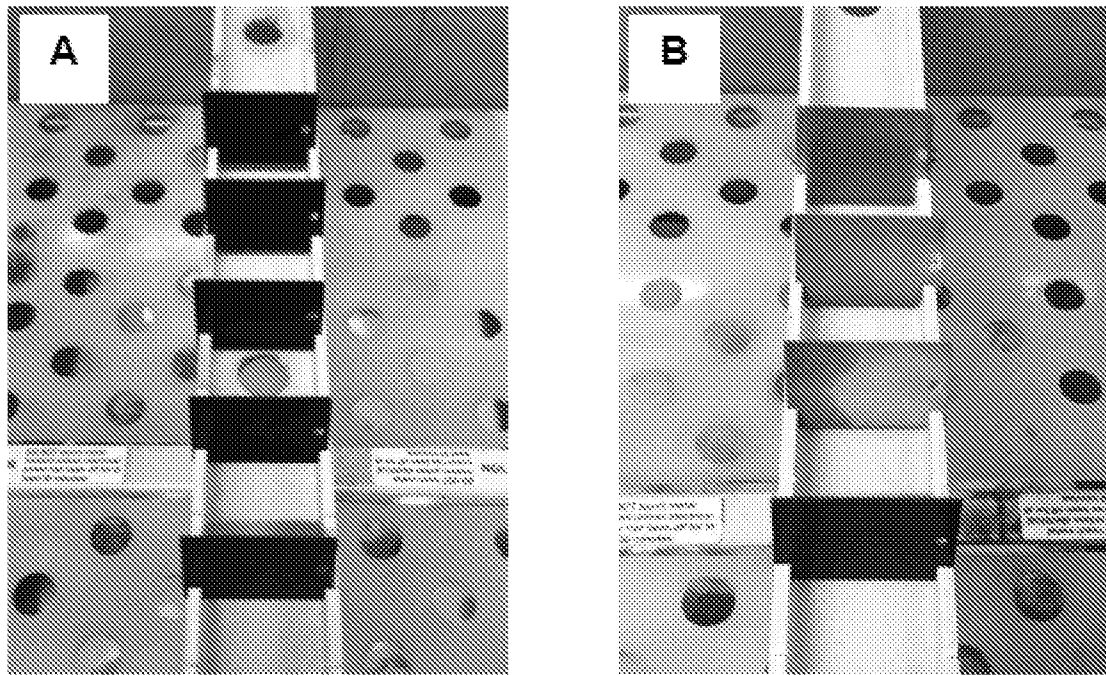


FIG. 6

Before salt spray testing



After 504 hours of salt spray testing

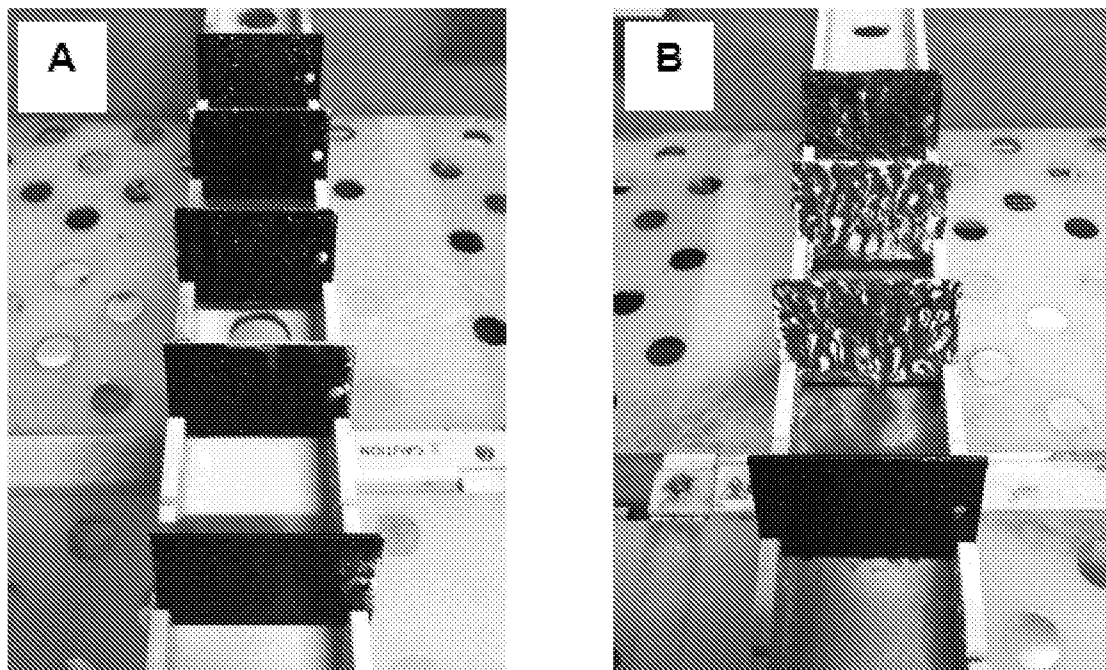


FIG. 7

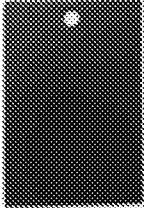
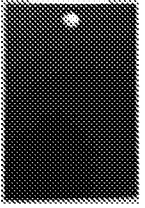
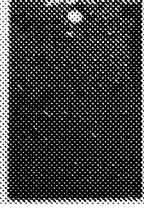
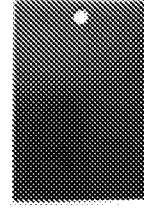
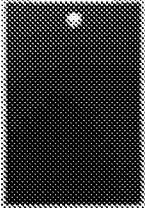
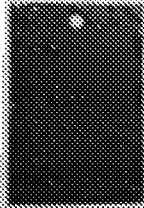
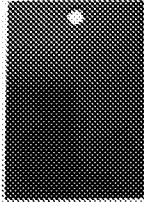
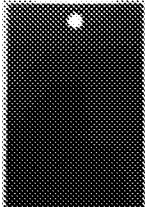
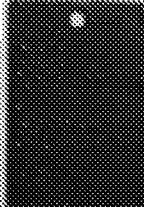
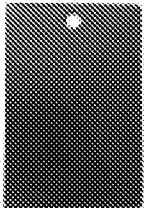
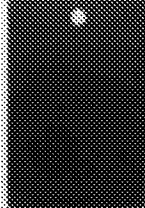
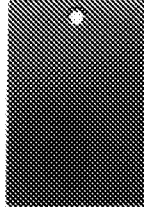
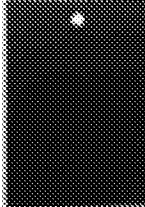
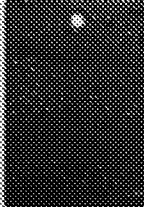
Sample	0 hours	48 hours	192 hours
1		 No changes	 White rust, Grade 6-S
2		 No changes	 White rust, Grade 6-S
3		 No changes	 No change
4		 No changes	 White rust, Grade 5-S
5		 No changes	 No changes

FIG. 8

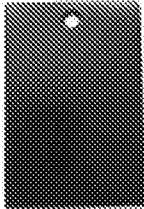
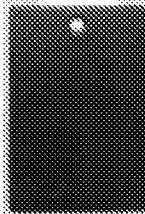
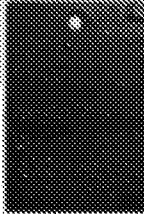
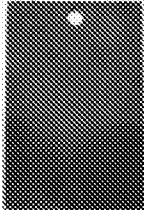
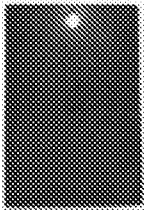
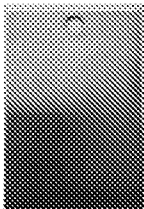
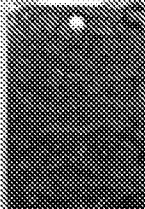
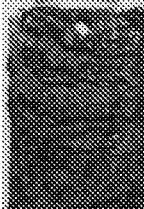
Sample	0 hours	48 hours	192 hours
6		 No changes	 No changes
7		 White and yellow rust, Grade 0-G	 Yellow and red rust, Grade 0-G
8		 White and yellow rust, Grade 0-G	 Yellow and red rust, Grade 0-G
9		 Red rust, Grade 0-G	 Red rust, Grade 0-G

FIG. 8 (con't)

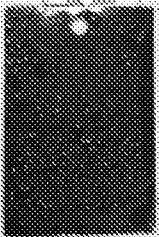
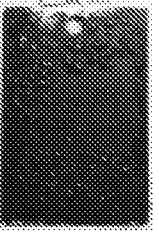
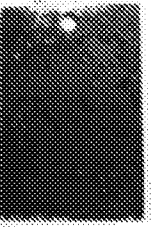
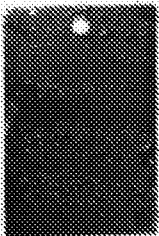
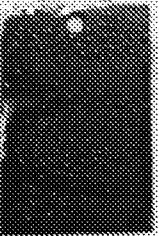
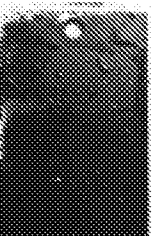
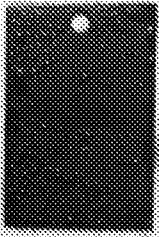
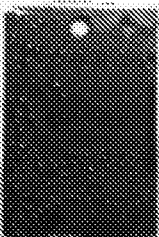
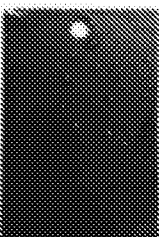
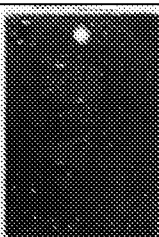
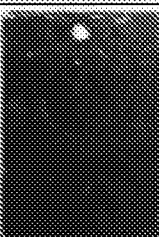
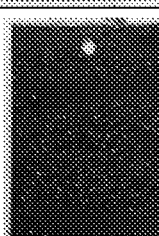
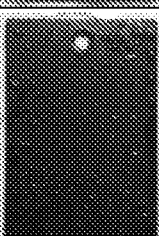
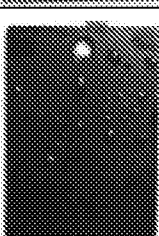
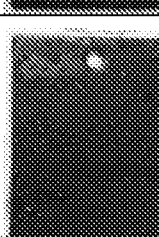
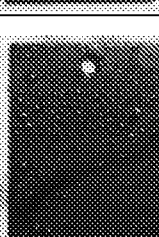
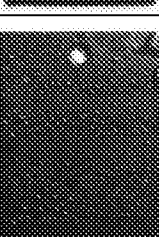
Sample	264 hours		408 hours		504 hours	
1		White rust, Grade 6-S		White rust, Grade 3-S		White rust, Grade 3-S
2		White rust, Grade 6-S		White rust, Grade 3-S		White rust, Grade 3-S
3		No changes		White rust, Grade 6-S		White rust, Grade 6-S
4		White rust, Grade 6-S		White rust, Grade 3-G		White rust, Grade 3-G
5		No changes		No changes		No changes
6		No changes		No changes		No changes

FIG. 8 (con't)

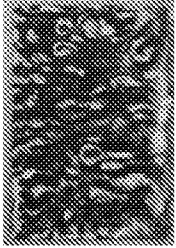
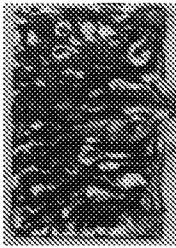
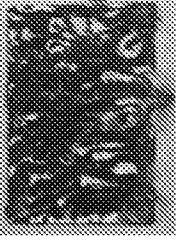
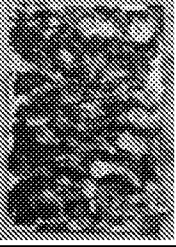
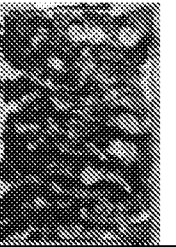
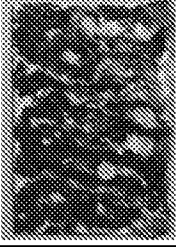
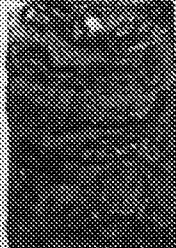
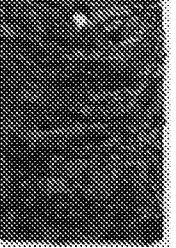
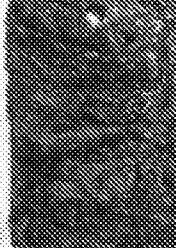
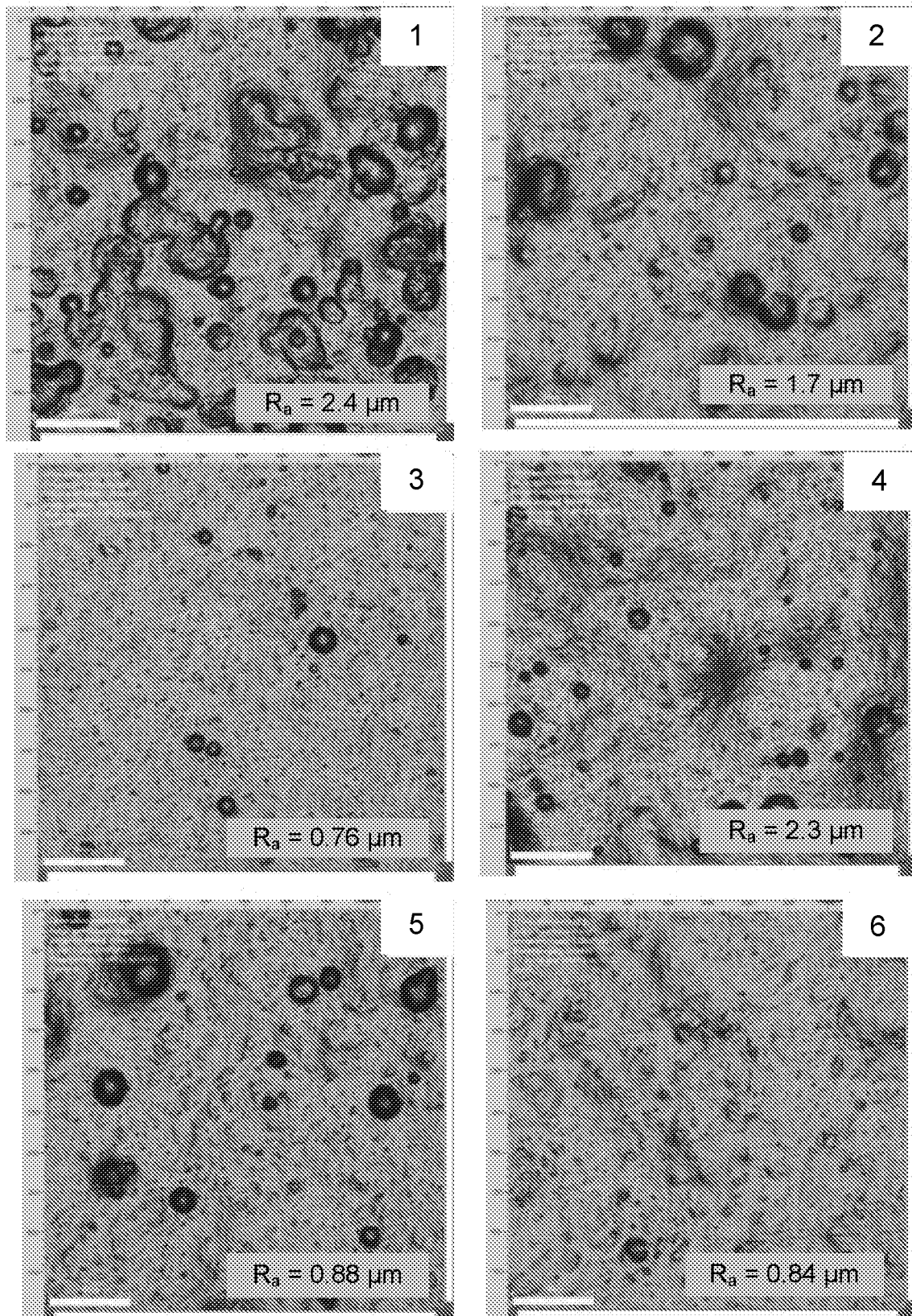
Sample	264 hours		408 hours		504 hours	
7		Yellow and red rust, Grade 0-G		Red with some yellow rust, Grade 0-G		Red with some yellow rust, Grade 0-G
8		Yellow and red rust, Grade 0-G		Yellow and red rust, Grade 0-G		Red with some yellow rust, Grade 0-G
9		Red rust, Grade 0-G		Red rust, Grade 0-G		Red rust, Grade 0-G

FIG. 8 (con't)

**FIG. 9**

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US16/21066

A. CLASSIFICATION OF SUBJECT MATTER

IPC(8) - C25D 3/02, 3/22; C23C 22/05, 22/60 (2016.01)

CPC - C25D 3/02, 3/22; C23C 22/05, 22/60

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC(8) Classifications: C25D 3/02, 3/22; C23C 22/05, 22/60 (2016.01)

CPC Classifications: C25D 3/02, 3/22; C23C 22/05, 22/60

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

PatSeer (US, EP, WO, JP, DE, GB, CN, FR, KR, ES, AU, IN, CA, INPADOC Data); Google Scholar; Google; EBSCO

Anti-corrosive coating, zinc, inner layer, outer layer, sodium zincate, sodium silicate, Electrolytic deposition

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X --- Y	US 2002/0012804 A1 (HEIMANN, RL et al.) 31 January 2002; paragraphs [0016], [0019]-[0021], [0026], [0029], [0032], [0074], [0087], [0099], [0108], [0114]; claim 1	1-2, 4-5, 8-14, 16-20 ----- 3, 6-7, 15
X	US 5,068,134 A (COLE, JA et al.) 26 November 1991; claim 1	1
Y	US 4,013,488 A (RAMQVIST, LH et al.) 22 March 1977; column 5, lines 8-9; column 6, lines 6-11	3, 7
Y	(YURIEVA, IM et al.) Non-hydrothermal synthesis of copper-, zinc and copper-zinc hydrosilicates. Materials Research Innovations. 2001. Vol. 5. Pages 3-11; page 5, right column, last paragraph; page 4, left column, 4th paragraph; page 7, left column, 2nd paragraph; table 1	6, 15



Further documents are listed in the continuation of Box C.



See patent family annex.

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"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

28 April 2016 (28.04.2016)

Date of mailing of the international search report

23 MAY 2016

Name and mailing address of the ISA/

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