

PATENT SPECIFICATION

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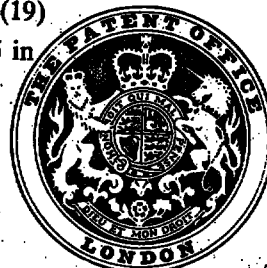
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(54) PROCESS FOR ELECTROLYTICALLY COLORING ALUMINUM AND ALUMINUM ALLOYS

(71) We, YOSHIDA KOGYO K.K., a corporation duly organized under the laws of Japan and existing at No. 1, Kanda Izumicho, Chiyoda-ku, Tokyo, Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:-

This invention relates to a process for electrolytically coloring aluminum or any of its various alloys.

For electrolytic coloring of aluminum or aluminum alloy, there has been suggested and practiced extensively a process wherein the basis metal is first anodized to form an oxide film thereon and then electrolyzed in an electrolytic solution containing a metallic salt such as a nickel salt by use of alternating or direct current. The basis metal is colored by electrodeposition of the metal or metal oxide in the pores of the oxide film thereon.

Such a prior art process is generally poor in throwing power, so that particularly in the case of an irregularly shaped workpiece, its protuberant and recessed portions tend to be colored in noticeably different shades. Moreover, since a darker shade is usually produced on the end faces of workpieces, the counter electrode requires intricate masking.

It has also been proposed to make suitably combined use, in the electrolysis of the anodized basis metal in accordance with the above described prior art process, of alternating and direct currents or of other currents similar to or quite dissimilar from such currents in waveform. This practice also has drawbacks such as the complexity of equipment required and the difficulty of control. Another disadvantage arises from the fact that according to such known methods, the degree of depth of coloring of successive workpieces must be controlled by the period of electrolysis. It has been highly difficult to

eliminate differences in the coloring degree of the workpieces because of the inevitable personal differences of personnel engaged in color matching operation.

The present invention seeks to provide an improved process for uniformly coloring aluminum and aluminum alloys by electrolysis.

The invention also seeks to provide a process of the character described such that aluminum or aluminum alloy can be colored to a desired constant degree.

The invention further seeks to provide a process of the character described such that colored coatings formed on aluminum or aluminum alloy have excellent weather resistance and other properties.

According to the invention, there is provided a process for electrolytically coloring aluminum or aluminum alloy which comprises anodizing a desired basis metal, and electrolyzing the anodized basis metal in an electrolytic coloring solution by use of alternating current, the voltage at which the anodized basis metal is electrolyzed being lowered at least once in the course of the electrolysis.

The above and other objects, features and advantages of this invention will become more apparent and understandable from the following detailed description, Examples and claims.

It is understood that aluminum and aluminum alloys to be colored by the process of this invention comprise pure aluminum and the alloys of pure aluminum and one or more of such elements as silicon, magnesium, copper, zinc, chromium, lead, bismuth, iron, titanium, and manganese.

For anodising aluminium or any of such aluminium alloys, the basis metal may first be degreased, rinsed and otherwise suitably pretreated in the conventional manner. The pretreated basis metal is made anodic in the usual acid electrolytic solution containing

sulfuric acid, oxalic acid, sulfamic acid or the like, and electric current is passed through the solution between the anodic basis metal and a cathode also immersed therein as the counter electrode.

5 In accordance with this invention, the anodized basis metal is then subjected to electrolysis by use of alternating current at a preselected starting voltage, with the voltage
10 being lowered at least once during the progress of the electrolysis. The electrolysis is preferably effected in an electrolytic solution which contains either at least three
15 metallic salts or at least two metallic salts and a strongly reducing compound, selected from dithionites, thiosulfates, hydrogen sulfites, sulfurous acid, sulfites, thioglycolic acid and thioglycolates. An electrolytic coloring process involving the latter kind of
20 bath is described and claimed in our copending Application No. 7990/76 (Serial No. 1519702).

The metallic salts for use in such an electrolytic solution can be selected, for
25 example, from such inorganic acid salts as nitrates, sulfates, phosphates, chlorides and chromates, and such organic acid salts as oxalates, acetates, and tartrates, of various metals typically comprising nickel, cobalt,
30 chromium, copper, magnesium, iron, cadmium, titanium, manganese, molybdenum, calcium, vanadium, tin, lead, and zinc. The electrolytic solution preferably contains at least three of such metallic salts, or at least
35 two of such metallic salts and a strongly reducing compound, selected from the compounds listed above. The concentration of the total amount of any two or more selected metallic salts in the electrolytic solution is preferably in the range from 5 to
40 500 g/l and, for the best results, from about 10 to 250 grams per litre.

Examples of strongly reducing compound to be added required to the electrolytic solution
45 used in the invention include for example, such dithionites as sodium dithionite and zinc dithionite; such thiosulfates as ammonium thiosulfate, sodium thiosulfate, potassium thiosulfate and iron thiosulfate;
50 such hydrogen sulfites as sodium hydrogen sulfite and potassium hydrogen sulfite; sulfurous acid; such sulfites as ammonium sulfite, sodium sulfite and potassium sulfite; thioglycolic acid; and such thioglycolates as
55 ammonium thioglycolate, sodium thioglycolate, potassium thioglycolate and lithium thioglycolate. The concentration of any selected reducing compound in the electrolytic solution is preferably in the range of
60 from 0.05 to 10 grams per liter and, for the best results, from about 0.5 to 3 grams per liter.

The preferred use of an aqueous solution of at least three selected metallic salts, or of
65 at least two selected metallic salts and a

selected strongly reducing compound, as the electrolytic solution in the process of this invention is effective not only to produce coatings of desired colors but also to speed the progress of the coloring operation and to improve the uniformity of the colored coatings produced.

Usually, there is further added to the electrolytic solution for use in the process of this invention at least one of such inorganic acids
75 as sulfuric acid, nitric acid, hydrochloric acid, phosphoric acid, boric acid, thiocyanic acid and chromic acid; such organic acids as oxalic acid, acetic acid, propionic acid, formic acid, tartaric acid and malic acid; and
80 their ammonium salts, amino salts and imino salts. The concentration of any selected one of these substances in the electrolytic solution should be in the range of from about 5 to 250 grams per liter.

Still more improved results will be realized by further adding to the electrolytic solution an organic substance e.g. toluenesulfonic acid, sulfosalicylic acid, hydrazine sulfate, and maleic acid anhydride.

For electrolytically coloring aluminum or aluminum alloy in accordance with this invention, the anodized basis metal together with a counter electrode is immersed in the electrolytic solution which has been prepared as above described, and alternating current is passed through the solution. It is an essential feature of this invention that the voltage impressed across the electrodes is lowered at least once during the progress of the electrolysis.

The uniformity of coloring on various surfaces of aluminum or aluminum alloy workpieces can be achieved by suitably selecting the instant at which the voltage is decreased, the voltage values at which the electrolysis is conducted, and the difference between the starting and the subsequently lowered voltages. It is also possible, through suitable control of the voltages, to substantially limit the degree or depth to which the workpieces can be colored, so that the finished workpieces will have little or no difference in the shade of their color. Furthermore, the throwing power can be so improved that, as has been confirmed experimentally, three extruded aluminum or aluminum alloy specimens each sized 150 millimeters by 70 by 1.3 can be colored uniformly on all their surfaces when anodized and subsequently subjected to simultaneous electrolysis with constant spacings of 10 millimeters therebetween and with the use of a single counter electrode, in accordance with the process of this invention.

The effects of lowering the voltage in the course of electrolysis according to the invention are explained in the following, on the assumption that the electrolytic solution in use has the composition set forth in Inven-

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tive Example I given hereinbelow. The greater is the difference between the starting and the final voltages, the better is the uniformity of color produced, but the lower is the progress of coloring. Colors produced are poor in uniformity if the voltage is lowered too early or too late following the start of electrolysis, so that the optimum instant for the voltage change should be ascertained previously. Also, if the electrolysis, so that the optimum instant for the voltage change should be ascertained previously. Also, if the electrolysis is carried out at voltages at or near the lower limit of the voltage range in which coloring is possible, the basis metal will be colored only to a substantially limited degree.

In consideration of the foregoing, the starting voltage of electrolysis according to the invention is preferably in the range of from 6 to 50 volts and, for the best results, from about 10 to 30 volts. In the course of electrolysis the voltage may be lowered by 1 to 10 volts. Generally, the higher is the starting voltage, the greater should be the difference between the starting and the final voltages. The voltage is preferably lowered within 2 minutes and, for the best results, in 5 to 60 seconds, following the start of electrolysis.

It is to be noted that only a decrease in voltage is effective to attain the objects of this invention. The degree and speed of coloring depend on the specific voltages in use, the difference therebetween, and the instant at which the voltage is decreased. These conditions, when suitably determined in relation to each other, provide improved results, particularly if the electrolytic solution has a suitably elevated temperature. In practice, however, the bath temperature should be determined in consideration of such factors as the composition of the bath, its stability, the progress of sealing due to its temperature rise, and the drying of the workpieces at the time of the subsequent color matching operation. Usually, the bath temperature may range from room temperature to about 50°C.

The pores in the oxide films on workpieces which have been colored by the process of the invention as above described may be sealed by boiling water, by steam, or by superheated steam, as has been known heretofore. After, or without, the sealing treatment, the colored surfaces may be coated with a suitable resin paint as by the dipping or electrodeposition method for protection purposes.

The inventive process is hereinafter described more specifically in terms of several Inventive Examples, which, however, are meant purely to illustrate or explain and not to impose limitations upon the invention. Also given hereinbelow are some

Comparative Examples which are intended to make clear the advantages of the inventive process.

Inventive Example I

Two specimens each consisting of an aluminum extrusion sized 150 millimeters by 70 by 1.3 were degreased, etched and desmutted in the usual manner. The thus pretreated specimens were each made anodic in an aqueous solution of 17.5 W/V % sulfuric acid, and a DC voltage of 15 volts was impressed for 35 minutes across the anodic specimen and an aluminum cathode connected as the counter electrode in the bath. The current density was 1.2 amperes per square decimeter. An anodic oxide film with a thickness of about 12 microns was thus formed on each specimen, which was then rinsed.

For electrolytically coloring the above anodized specimens, there was used a vessel with a length of 300 millimeters, a width of 100 millimeters and a height of 150 millimeters. This vessel was filled with an electrolytic solution of the following composition:

Nickel sulfate (hexahydrate)	30 g/l
Magnesium sulfate (heptahydrate)	15 g/l
Boric acid	20 g/l
Ammonium sulfate	30 g/l
Sodium dithionite	0.5 g/l

The pH of the above electrolytic solution was 5.6, and its temperature was 20°C.

The two anodized specimens which had been prepared as above were immersed in the solution with a spacing of 10 millimeters therebetween, and a single counter electrode was positioned at a distance of 250 millimeters from the specimens. The specimens were then subjected to electrolysis for 20 seconds by use of alternating current at 24 volts, and then for three minutes by use of alternating current at 18 volts. All the four surfaces of the two specimens were colored uniformly in bronze.

The above obtained colored films on the specimens were then subjected to sealing treatment for 30 minutes by live steam, under pressure of five kilograms per square centimeter. A 3000-hour accelerated weathering test of the finished specimens by means of a weatherometer developed no change in their colored surfaces. Also, no change in color took place when the specimens were heated to 200°C for two hours, and the specimens remained intact when subjected to a 16-hour CASS (copper-accelerated acetic acid salt spray) test. It has thus been confirmed that aluminum or aluminum alloy colored by the process of this invention lends itself to use, for example, as structural members which will sufficiently withstand exterior use.

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Inventive Example II

Two aluminum extrusions each sized 150 millimeters by 70 by 1.3 were anodized through the same procedure as in Inventive Example I to form thereon an oxide film with a thickness of about 12 microns. The anodized specimens were rinsed and were subsequently electrolyzed, first for 20 seconds by use of alternating current at 21 volts and then for three minutes by use of alternating current at 16 volts, in an electrolytic solution of the following composition:

Nickel sulfate (hexahydrate)	25 g/l
Magnesium sulfate (heptahydrate)	15 g/l
Boric acid	20 g/l
Ammonium sulfate	30 g/l
Cobalt sulfate (heptahydrate)	25 g/l

The pH of this electrolytic solution was 5.6, and its temperature was 20°C. The specimens were colored uniformly in deep bronze as in Inventive Example I.

The pores in the thus obtained colored films on the specimens were sealed in the manner set forth in Inventive Example I. The finished specimens exhibited the same favorable results as those of the preceding Example when subjected to a 3000-hour accelerated weathering test by means of a weatherometer, a 2-hour heating test at 200°C, and a 16-hour CASS test.

Inventive Example III

Two aluminum extrusions each sized 150 millimeters by 70 by 1.3 were anodized through the procedure of Inventive Example I to form thereon an oxide film with a thickness of about 12 microns. After having been rinsed, the anodized specimens were electrolyzed, first for 15 seconds by use of alternating current at 19 volts and then for 10 minutes by use of alternating current at 16 volts, in the same electrolytic solution as in Inventive Example II. The specimens were colored uniformly in black on all their surfaces.

The pores in the thus produced colored films on the specimens were sealed by the same means as in Inventive Example I. The finished specimens exhibited the same favorable results as those of Inventive Example I when subjected to a 3000-hour accelerated weathering test by means of a weatherometer, a 2-hour heating test at 200°C, and a 16-hour CASS test.

Inventive Example IV

Through the procedure of Inventive Example I, two aluminum extrusions of the same size as above were anodized to form thereon an oxide film with a thickness of about 12 microns. After having been rinsed, the anodized specimens were electrolyzed, first for 30 seconds by use of alternating current at 21 volts and then for three minutes by use of alternating current at 16 volts, in an electrolytic solution of the following

composition:

Nickel sulfate (hexahydrate)	30 g/l	
Magnesium sulfate (heptahydrate)	30 g/l	
Boric acid	25 g/l	
Ammonium sulfate	30 g/l	70
Ferrous sulfate (heptahydrate)	20 g/l	

The pH of this electrolytic solution was 5.6, and its temperature was 20°C. The specimens were colored uniformly in greenish bronze on all their surfaces.

The pores in the thus produced colored films on the specimens were sealed by the same means as in Inventive Example I. The finished specimens, when subjected to the same tests as in Inventive Example I, exhibited the same favorable results as above.

Inventive Example V

Through the procedure of Inventive Example I, two aluminum extrusions of the same size as above were anodized to form thereon an oxide film with a thickness of about 12 microns. After having been rinsed, the anodized specimens were electrolyzed, first for 20 seconds by use of alternating current at 15 volts and then for three minutes by use of alternating current at 12 volts, in an electrolytic solution of the following composition:

Nickel sulfate (hexahydrate)	30 g/l	
Magnesium sulfate (heptahydrate)	10 g/l	
Boric acid	10 g/l	
Ammonium sulfate	30 g/l	
Sodium dithionite	0.5 g/l	100

The pH of this electrolytic solution was 5.6, and its temperature was 40°C. The specimens were colored uniformly in bronze on all their surfaces.

The pores in the thus produced colored films on the specimens were sealed by the same means as in Inventive Example I. The finished specimens, when subjected to the same tests as in Inventive Example I, exhibited the same favorable results as above.

Inventive Example VI

Through the procedure of Inventive Example I, two aluminum extrusions of the same size as above were anodized to form thereon an oxide film with a thickness of about 12 microns. After having been rinsed, the anodized specimens were electrolyzed, first for 30 seconds by use of alternating current at 24 volts and then for 4 minutes by use of alternating current at 18 volts, in an electrolytic solution of the following composition:

Nickel sulfate (hexahydrate)	25 g/l	125
Magnesium sulfate (heptahydrate)	30 g/l	
Boric acid	25 g/l	
Ammonium sulfate	30 g/l	
Ammonium thiosulfate	0.5 g/l	
p-toluenesulfonic acid	1.0 g/l	130

The pH of this electrolytic solution was 5.6, and its temperature was 25°C. The specimens were colored uniformly in deep bronze on all their surfaces.

The pores in the thus produced colored films on the specimens were sealed by the same means as in Inventive Example I. The finished specimens, when subjected to the same tests as in Inventive Example I, exhibited the same favorable results as above.

Inventive Example VII

Through the procedure of Inventive Example I, two aluminum extrusions of the same size as above were anodized to form thereon an oxide film with a thickness of about 12 microns. After having been rinsed, the anodized specimens were electrolyzed, first for 20 seconds by use of alternating current at 17 volts and then for four minutes by use of alternating current at 13 volts, in an electrolytic solution of the following composition:

Copper sulfate (pentahydrate)	7 g/l
Magnesium sulfate (heptahydrate)	7 g/l
Ammonium sulfate	30 g/l
Cobalt sulfate (heptahydrate)	7 g/l

The pH of this electrolytic solution was 4.0, and its temperature was 20°C. The specimens were colored uniformly in reddish brown on all their surfaces.

The pores in the thus produced colored films on the specimens were then sealed by the same means as in Inventive Example I. The finished specimens, when subjected to the same tests as in Inventive Example I, exhibited the same favorable results as above.

Comparative Example I

Through the procedure of Inventive Example I, two aluminum extrusions of the same size as above were anodized to form thereon an oxide film with a thickness of about 12 microns. The specimens were rinsed and then electrolyzed for four minutes by use of alternating current at 16 volts in an electrolytic solution of the following composition:

Nickel sulfate (hexahydrate)	25 g/l
Magnesium sulfate (heptahydrate)	20 g/l
Boric acid	25 g/l
Ammonium sulfate	30 g/l

The pH of the above electrolytic solution was 5.6, and its temperature was 20°C.

The two specimens were colored in bronze. One of their surfaces which has been located closest to the counter electrode, however, was colored in a significantly darker shade than the other three surfaces. Moreover, the color of the said other three surfaces became generally lighter with the increase in distance from the counter electrode, and the color of each of these three surfaces became still lighter

toward its center.

Inventive Example VIII

Two aluminum extrusions of the same size as above were anodized and rinsed through the same procedure as in Inventive Example I. The anodized specimens were then electrolyzed in the electrolytic solution of Comparative Example I, first for 20 seconds by use of alternating current at 24 volts and then for four minutes by use of alternating current at 16 volts. The specimens were colored in extremely light beige on all their surfaces. The shade of the color on these surfaces remained substantially unchanged when the specimens were further electrolyzed in the solution for five minutes at 16 volts.

Inventive Example IX

To the electrolytic solution of Comparative Example I was added, 1.0 gram per liter of ammonium thiosulfate, and the pH of the overall solution was regulated to 5.6. Two aluminum extrusions of the same size as above, which had been anodized and rinsed through the same procedure as in Inventive Example I, were electrolyzed in this electrolytic solution through the procedure of Comparative Example II, that is, first for 20 seconds by use of alternating current at 24 volts and then for four minutes by use of alternating current at 16 volts. The two specimens were colored uniformly in bronze on all their surfaces.

Comparative Example II

Through the procedure of Inventive Example I, two aluminum extrusions of the same size as above were anodized to form thereon an oxide film with a thickness of about 12 microns. The anodized specimens were rinsed and then electrolyzed for four minutes by use of alternating current at 16 volts in an electrolytic solution of the following composition:

Nickel sulfate (hexahydrate)	25 g/l
Magnesium sulfate (heptahydrate)	20 g/l
Boric acid	25 g/l
Ammonium sulfate	30 g/l
Cobalt sulfate (heptahydrate)	25 g/l

The pH of this electrolytic solution was 5.6, and its temperature was 20°C.

The two specimens were colored in bronze. One of their surfaces which had been located closest to the counter electrode, however, was colored in a significantly darker shade than the other three surfaces. Moreover, the color of the said other three surfaces became lighter with the increase in distance from the counter electrode, and the color of each of these three surfaces became still lighter toward its center.

Inventive Example X

Two aluminum extrusions of the same size as above were anodized and rinsed through

the same procedure as in Inventive Example I. The anodized specimens were then electrolyzed in the electrolytic solution of Comparative Example III in accordance with the teaching of this invention, that is, first for 20 seconds by use of alternating current at 24 volts and then for four minutes by use of alternating current at 16 volts. The aluminum extrusions were colored

uniformly in bronze on all their four surfaces.

Inventive Examples XI-XIV

In the electrolytic solution of Inventive Example I, the following strongly reducing compounds were used in lieu of sodium dithionite to prepare four different solutions:

Example	Strongly Reducing Compound	Concentration (g/l)	pH
X	Thioglycolic acid	1.5	4.5
XI	Ammonium thioglycolate	1.5	5.6
XII	Ammonium sulfite	2.0	5.6
XIII	Ammonium hydrogen sulfite	1.0	5.6

Two aluminum extrusions of the same size as above were anodized, rinsed, and electrolyzed in each of the above electrolytic solutions, through the procedure of Inventive Example I. The results were as favorable as those set forth in Inventive Example I.

WHAT WE CLAIM IS:-

1. A process for electrically colouring aluminium or aluminium alloy which comprises anodising a desired basis metal, and electrolyzing the anodised basis metal in an electrolytic colouring solution by use of alternating current, the voltage at which the anodised basis metal is electrolysed being lowered at least once in the course of the electrolysis.

2. The process as claimed in Claim 1, wherein the anodised basis metal is electrolysed at a starting voltage in the range of from 6 to 50 volts.

3. The process as claimed in Claim 2, wherein the voltage is lowered by 1 to 10 volts in the course of the electrolysis.

4. The process as claimed in any one of Claims 1 to 3, wherein the voltage is lowered within 2 minutes following the start of the electrolysis.

5. The process as claimed in any one of the preceding Claims, wherein the electrolytic solution contains at least three metallic salts.

6. The process as claimed in any one of Claims 1 to 4, wherein the electrolytic solution contains at least two metallic salts and a strongly reducing compound, selected from dithionites, thiosulfates, hydrogen sulfites, sulfurous acid, sulfites, thioglycolic acid and thioglycolates.

7. The process as claimed in Claim 5 or Claim 6, wherein the metallic salts are selected from inorganic and organic salts of nickel, cobalt, chromium, copper, magnesium, iron, cadmium, titanium, manganese, molybdenum, calcium, vanadium, tin, lead, and zinc.

8. The process according to Claim 1 substantially as hereinbefore described in the Examples.

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the same procedure as in Inventive Example I. The anodized specimens were then electrolyzed in the electrolytic solution of Comparative Example III in accordance with the teaching of this invention, that is, first for 20 seconds by use of alternating current at 24 volts and then for four minutes by use of alternating current at 16 volts. The aluminum extrusions were colored

uniformly in bronze on all their four surfaces.

Inventive Examples XI-XIV

In the electrolytic solution of Inventive Example I, the following strongly reducing compounds were used in lieu of sodium dithionite to prepare four different solutions:

Example	Strongly Reducing Compound	Concentration (g/l)	pH
X	Thioglycolic acid	1.5	4.5
XI	Ammonium thioglycolate	1.5	5.6
XII	Ammonium sulfite	2.0	5.6
XIII	Ammonium hydrogen sulfite	1.0	5.6

Two aluminum extrusions of the same size as above were anodized, rinsed, and electrolyzed in each of the above electrolytic solutions, through the procedure of Inventive Example I. The results were as favorable as those set forth in Inventive Example I.

WHAT WE CLAIM IS:-

1. A process for electrically colouring aluminium or aluminium alloy which comprises anodising a desired basis metal, and electrolysing the anodised basis metal in an electrolytic colouring solution by use of alternating current, the voltage at which the anodised basis metal is electrolysed being lowered at least once in the course of the electrolysis.
2. The process as claimed in Claim 1, wherein the anodised basis metal is electrolysed at a starting voltage in the range of from 6 to 50 volts.
3. The process as claimed in Claim 2, wherein the voltage is lowered by 1 to 10 volts in the course of the electrolysis.
4. The process as claimed in any one of Claims 1 to 3, wherein the voltage is lowered within 2 minutes following the start of the electrolysis.

5. The process as claimed in any one of the preceding Claims, wherein the electrolytic solution contains at least three metallic salts.

6. The process as claimed in any one of Claims 1 to 4, wherein the electrolytic solution contains at least two metallic salts and a strongly reducing compound, selected from dithionites, thiosulfates, hydrogen sulfites, sulfurous acid, sulfites, thioglycolic acid and thioglycolates.

7. The process as claimed in Claim 5 or Claim 6, wherein the metallic salts are selected from inorganic and organic salts of nickel, cobalt, chromium, copper, magnesium, iron, cadmium, titanium, manganese, molybdenum, calcium, vanadium, tin, lead, and zinc.

8. The process according to Claim 1 substantially as hereinbefore described in the Examples.

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