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(54) COATING FILM FORMATION METHOD

(57) An object of the present invention is to provide a method for forming a coating film, the method capable of producing a coating color having a soft texture, the coating color showing a large lightness change from the highlight (in the vicinity of specularly reflected light) to the shade (in an oblique direction), and having high chroma from the highlight to the shade, the method being applicable to industrial products, such as two-wheeled automobile exteriors and automobile interior parts. The invention relates to a method for forming a coating film, the method comprising forming a metallic base coating film containing an aluminum flake pigment on a substrate, and forming a color clear coating film containing a color pigment and a flatting agent on the metallic base coating film, wherein the color pigment in the color clear coating film is a transparent pigment, and wherein when the transparent pigment is contained in a coating composition as a sole coating material in an amount of 1 part by mass, per 100 parts by mass of the resin solids content, which is a vehicle formation component, and when this coating composition is used to form a coating film having a thickness of 100 μm, this coating film achieves a haze value of 0.1 to 10.0.

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Description

Technical Field

Cross Reference of Related Application

[0001] This application claims priority to JP2012-092156A, filed April 13, 2012, the disclosure of which is incorporated herein by reference in its entirety. The present invention relates to a method for forming a coating film, the method being capable of producing a coating color having a soft texture, the coating color showing a large lightness change from the highlight (in the vicinity of specularly reflected light) to the shade (in an oblique direction) and having high chroma from the highlight to the shade.

Background Art

[0002] Matte design, which is visually soft and has a relaxing texture, is gaining more popularity in industrial products, such as two-wheeled automobile exterior materials and automobile interior parts. In particular, a matte and metallic coating color, which shows a lightness change according to the observation angle, provides a luxurious appearance, and is thus one of the most attention-grabbing coating colors among matte designs.

[0003] In relation to a coating color that shows a lightness change according to the observation angle, Patent Literature (PTL) 1 discloses a method for obtaining a gloss-adjusted design. In relation to this method, PTL 1 discloses a matte coating film obtained by forming a matte clear coating layer on a metallic coating layer containing an effect metal powder pigment. Although this method can form a matte coating film having a metallic texture, this film suffers from insufficient chroma.

Citation List

Patent Literature

[0004] PTL 1: JPH04-317774A

Summary of Invention

Technical Problem

[0005] An object of the present invention is to provide a method for forming a coating film, the method being capable of producing a coating color having a soft texture, the coating color showing a large lightness change from the highlight (in the vicinity of specularly reflected light) to the shade (in an oblique direction) and having high chroma from the highlight to the shade.

Solution to Problem

[0006] The present invention provides the following Items.

1. A method for forming a coating film, the method comprising:

   forming a metallic base coating film comprising an aluminum flake pigment on a substrate; and
   forming a color clear coating film comprising a color pigment and a flatting agent on the metallic base coating film, wherein the color pigment in the color clear coating film is a transparent pigment, and wherein when the transparent pigment is contained in a coating composition as a sole coating material in an amount of 1 part by mass, per 100 parts by mass of the resin solids content, which is a vehicle formation component, and when this coating composition is used to form a coating film having a thickness of 100 μm, this coating film achieves a haze value of 0.1 to 10.0.

2. The method for forming a coating film according to Item 1, wherein the flatting agent in the color clear coating film is at least one member selected from the group consisting of silica fine powder (hydrous silicon dioxide), polyethylene powder, resin beads, and ceramic beads.
3. The method for forming a coating film according to Item 1, wherein the color clear coating film is formed as a two-layer film in which a second clear coating film comprising a flatting agent is laminated on a first clear coating film

comprising a color pigment.

4. The method for forming a coating film according to Item 3, wherein each of the metallic base coating film and the first clear coating film comprises at least one type of resin as a vehicle formation component, and wherein at least a portion of the resin as the vehicle formation component in the metallic base coating film is the same as the resin as the vehicle formation component in the first clear coating film.

Advantageous Effects of Invention

[0007] The present invention enables provision of a method for forming a coating film, the method being capable of producing a coating color having a soft texture, the coating color showing a large lightness change from the highlight (in the vicinity of specularly reflected light) to the shade (in an oblique direction) and having high chroma from the highlight to the shade.

Description of Embodiments

[0008] In the method for forming a coating film of the present invention, the metallic base coating film and color clear coating film described below are laminated on a substrate.

[0009] Examples of the substrate include iron, zinc, aluminum, magnesium, and like metals; alloys comprising these metals; shaped articles plated or vapor-deposited with these metals; shaped articles of, for example, glass, plastic, and a foamed body; and the like. These materials may be suitably subjected to a degreasing treatment, a surface treatment, or the like, to obtain substrates. Further, these materials may be coated with an undercoating film, an intermediate coating film, and the like, to obtain substrates. Substrates comprising an undercoating film, an intermediate coating film, and the like, are particularly preferable.

[0010] The undercoating film is formed to conceal the material surface and to impart corrosion resistance, rust resistance, and the like, to the material. The undercoating film can be formed by application of an undercoating composition, followed by drying and curing. Examples of the undercoating composition include, but are not particularly limited to, an electrodeposition coating composition, a solvent-based primer, and the like.

[0011] The intermediate coating film is formed to conceal the material surface, the undercoating film surface, or the like, and to impart an adhesion property, chipping resistance, and the like. The intermediate coating film can be formed by applying an intermediate coating composition to the material surface, the undercoating film surface, or the like, followed by drying and curing. There is no particular limitation on the types of the intermediate coating composition, and known intermediate coating compositions may be used. It is preferable to use, for example, organic solvent based- or aqueous based-intermediate coating compositions comprising a thermosetting resin composition and a color pigment, as essential components.

[0012] In particular, when the undercoating film or intermediate coating film is formed on a substrate, the undercoating film or intermediate coating film is heated and cured by crosslinking, followed by application of the metallic base coating composition described below. It is also possible to perform the application of the metallic base coating composition when the undercoating film and/or intermediate coating film is in an uncured state.

[0013] The metallic base coating film used in the method of the present invention is obtained by application of a metallic base coating composition. The metallic base coating composition comprises an aluminum flake pigment to conceal the substrate surface, to obtain a lightness change from the highlight (in the vicinity of specularly reflected light) to the shade (in an oblique direction), and to increase the lightness of the multilayer coating film.

[0014] Aluminum flake pigments are generally produced by grinding or milling aluminum in a ball mill or an attritor mill in the presence of a grinding liquid medium using a grinding lubricant. As the grinding lubricant, higher fatty acids, such as oleic acid, stearic acid, isostearic acid, lauric acid, palmitic acid, and myristic acid, as well as aliphatic amine, aliphatic amide, and aliphatic alcohol, are used. As the grinding liquid medium, aliphatic-based hydrocarbons, such as mineral spirit, are used.

[0015] Aluminum flake pigments are roughly categorized into leafing type and non-leafing type aluminum flake pigments, according to the type of the grinding lubricant. When a leafing-type aluminum flake pigment is incorporated in a coating composition, the aluminum particles align themselves (leafing) on the surface of a coating film obtained by application of this coating composition, providing a finished appearance with strong metallic texture. This coating film has a heat reflection effect and rust resistance, and is thus often used for plant equipment, such as tanks, ducts, pipes, and roofing materials; and various building materials. It is possible to use a leafing-type aluminum flake pigment in the metallic base coating composition used in the method of the present invention. When a leafing-type aluminum flake pigment is used, however, care must be taken because the aluminum particles align themselves on the metallic base coating film surface due to the surface tension effect of the grinding lubricant during the coating film formation process, possibly resulting in overly increased lightness at the highlight of the multilayer coating film; however, this depends on the amount of the aluminum flake pigment. Considering this point, it is preferable to use a non-leafing type aluminum

flake pigment.

[0016] The aluminum flake pigment has an average particle diameter of preferably 5 to 30 μm, from the viewpoint of the finished appearance, the lightness at the highlight, and the lightness change from the highlight to the shade of the resulting coating film. The average particle diameter is more preferably 7 to 18 μm. The thickness is preferably 0.01 to 1.0 μm, and particularly preferably 0.02 to 0.5 μm. The average particle diameter as used herein refers to a median diameter of volume-based particle size distribution measured by a laser diffraction scattering method using a Microtrack particle size distribution analyzer MT3300 (product name, produced by Nikkiso Co., Ltd.). The thickness is defined as an average value of 100 or more measured values obtained by observing the cross-section of a coating film comprising the aluminum flake pigment by using a microscope, and measuring the thickness by using image-processing software.

[0017] If the average particle diameter exceeds the upper limit of the above range, it is possible that the obtained multilayer coating film will undesirably have a particulate texture due to the aluminum flake pigment. If the average particle diameter is smaller than the lower limit, it is possible for the lightness change from the highlight to the shade to be too small.

[0018] To increase the lightness at the highlight in the multilayer coating film, the aluminum flake pigment in the metallic base coating composition used in the method of the present invention is preferably such that when it is contained in a coating composition as a sole coloring material in the vehicle formation component, and when this coating composition is used to form a coating film, this coating film achieves an IV value of 100 to 550, and more preferably 160 to 350. In this specification, the IV value in relation to the aluminum flake pigment is defined as a value measured by the following method. The aluminum flake pigment is used in an amount of 15 parts by mass on a solids basis per 100 parts by mass of the resin solids content of acric No. 1000 clear (product name, produced by Kansai Paint Co., Ltd., a cellulose nitrate-modified acrylic clear coating composition), and the mixture is diluted with a dedicated thinner to obtain a viscosity appropriate for coating application, followed by stirring and mixing. The obtained coating composition is applied to a plate coated with a gray (N-5) coating film to form a cured coating film having a film thickness of 15 μm using a doctor blade. After the resulting film is left to stand at room temperature for 3 minutes, a clear coating composition (LUGA BAKE clear, produced by Kansai Paint Co., Ltd.; product name, acrylic resin/amino resin base, organic solvent type) is applied thereto by air spray to form a cured coating film having a thickness of 30 μm. After the application, the film is left to stand at a room temperature for 15 minutes and heated in a hot-air circulating oven at 140°C for 30 minutes to dry and cure the multilayer coating film simultaneously. In terms of the resulting coating film, a value Y in the XYZ color space based on the spectral reflectance of light received at an angle of 15 degrees with respect to the specular reflection light of light illuminated at an angle of 45 degrees is calculated using an MA-68II (product name, produced by X-Rite; a multi-angle spectrophotometer), and the calculated value Y is used as an IV value. The XYZ color space is the basis of the CIE color space, and three values Yxy are used in relation to the chromaticity diagram. Y represents reflectance, which corresponds to lightness, while xy represents chromaticity.

[0019] The aluminum flake pigment is preferably incorporated in a total amount of preferably 1 to 25 parts by mass, more preferably 3 to 20 parts by mass, and particularly preferably 5 to 18 parts by mass, per 100 parts by mass of the resin solids content of the coating composition, from the viewpoint of the finished appearance, the lightness at the highlight, and the lightness change from the highlight to the shade of the coating film obtained by application of the coating composition.

[0020] The metallic base coating composition may further comprise a color pigment to make minor adjustment in the hue of the multilayer coating film. Specific examples of the color pigment include, but are not particularly limited to, organic pigments, such as azo pigments, quinacridone pigments, diketopyrrolopyrrole pigments, perylene pigments, perinone pigments, benzimidazolone pigments, isoindoline pigments, isoindolinone pigments, metal chelate azo pigments, phthalocyanine pigments, indanthrone pigments, dioxazine pigments, threne pigments, and indigo pigments; carbon black pigments; and the like. These may be used singly, or in a combination of two or more.

[0021] In the present invention, when the metallic base coating composition comprises a color pigment, the amount is generally preferably 0.01 to 15 parts by mass, and particularly 0.05 to 10 parts by mass, on a solids basis, per 100 parts by mass of the resin solids content of the coating composition, from the viewpoint of the lightness, and the like, of the multilayer coating film.

[0022] In the metallic base coating composition and the below-described color clear coating composition used in the method of the present invention, the color pigment may be incorporated as a powder. It is also possible to mix and disperse the color pigment with a portion of the resin composition to preliminarily prepare a pigment dispersion, and mix the pigment dispersion with the rest of the resin component and other components to obtain the coating compositions. In the preparation of the pigment dispersion, commonly used coating composition additives, such as antifoaming agent, dispersion agent, and surface control agent, may optionally be used.

[0023] The metallic base coating composition used in the method of the present invention may generally contain a resin component as a vehicle. Specific examples of resin components include a combination of a base resin with a crosslinking agent. Examples of the base resin include acrylic resin, polyester resin, alkyd resin, and urethane resin, which have a crosslinkable functional group (e.g., hydroxyl). Examples of the crosslinking agent include melamine resin,

urea resin, and a polyisocyanate compound (including a blocked polyisocyanate compound). Such resin components are used after being dissolved or dispersed in an organic solvent and/or a solvent, such as water.

[0024] The metallic base coating composition used in the method of the present invention may optionally further comprise a solvent, such as water or an organic solvent; various additives, such as a dispersion agent, an antisettling agent, a curing catalyst, an antifoaming agent, an antioxidant, a UV absorber, a surface control agent, and a rheology control agent; an extender pigment; and the like.

[0025] The metallic base coating composition used in the method of the present invention is prepared by mixing and dispersing the above-described components. It is preferable to adjust the solids content at the time of application to 12 to 60 mass%, more preferably 15 to 50 mass%, based on the coating composition, and the viscosity at 20°C to 17 to 23 seconds (Ford cup No. 3).

[0026] The application of the metallic base coating composition used in the method of the present invention may be performed by electrostatic coating, air spray, airless spray, or the like. It is preferable that the thickness of the coating film is 5 to 30 μm when cured, from the viewpoint of the smoothness of the resulting coating film. In general, the metallic base coating composition used in the method of the present invention is applied to obtain a predetermined film thickness, and the coating film is heated, dried, and cured, followed by application of the color clear coating composition described below. It is also possible to perform the application of the color clear coating composition when the coating film is in an uncured state. When the metallic base coating composition is of a baking-drying type, the metallic base coating film of the metallic base coating composition used in the method of the present invention can generally be cured by crosslinking at a temperature of about 50°C to 150°C. When the metallic base coating composition is of a normal temperature drying type or a forced drying type, the film can be cured by crosslinking at a temperature in the range of an ordinary temperature to about 80°C.

[0027] In the method for forming a coating film of the present invention, a color clear coating film is laminated on the coating film obtained by application of the metallic base coating composition.

[0028] In the method for forming a coating film of the present invention, the color clear coating film may be formed as a one-layer coating film that is obtained by application of a color clear coating composition comprising a color pigment and a flatting agent. It is also possible to form the color clear coating film as a two-layer coating film in which a second clear coating film comprising a flatting agent is laminated on a first clear coating film comprising a color pigment. The two-layer coating film is preferable to increase the chroma of the multilayer coating film.

[0029] First, a method for forming a one-layer coating film is described below. When a one-layer coating film is formed, a color clear coating composition is applied to the metallic base coating film described above.

[0030] The color clear coating composition comprises a color pigment to adjust the color tone of the multilayer coating film and increase the chroma. Specific examples of the color pigment include organic pigments, such as azo pigments, quinacridone pigments, diketopyrrolopyrrole pigments, perylene pigments, perinone pigments, benzimidazolone pigments, isoindoline pigments, isoindolinone pigments, metal chelate azo pigments, phthalocyanine pigments, indanthrone pigments, dioxazine pigments, threne pigments, and indigo pigments; carbon black pigments; and the like. These may be used singly, or in a combination of two or more.

[0031] As the color pigment in the color clear coating composition, a transparent pigment having high transparency and a small primary particle diameter is used to increase the chroma of the multilayer coating film. It is preferable for the color pigment to have a primary particle diameter of 10 to 250 nm, from the viewpoint of transparency and coloring power. The primary particle diameter is particularly preferably 20 to 200 nm. The transparent pigment used in the present invention preferably satisfies the following feature from the viewpoint of the chroma of the multilayer coating film: when a coating composition comprising a coloring material and a resin as a vehicle formation component is prepared, and when the transparent pigment used in the present invention is contained in this coating composition as the sole coloring material in an amount of 1 part by mass, per 100 parts by mass of the resin solids content, a coating film formed of this coating composition to have a film thickness of 100 μm achieves a haze value of 0.1 to 10.0. The haze value is more preferably 0.1 to 7.5, particularly preferably 0.1 to 5, and further preferably 0.1 to 3.

[0032] In the present invention, the haze value is defined as a value calculated using the following equation based on the diffuse light transmittance (DF) and parallel light transmittance (PT) of a coating film formed and cured on a smooth PTFE plate, and peeled off from the plate. The DF and PT of the coating film are measured using a Turbidimeter COH-300A (product name, produced by Nippon Denshoku Industries Co., Ltd.).

$$\text{Haze value} = 100 \times DF/(DF + PT)$$

[0033] The color clear coating composition used in the method of the present invention comprises the color pigment in an amount of preferably 0.01 to 5 parts by mass, more preferably 0.02 to 4 parts by mass, and particular preferably 0.03 to 3 parts by mass, per 100 parts by mass of the resin solids content of the coating composition, from the viewpoint

of the chroma and the lightness difference between the highlight and shade of the multilayer coating film.

[0034] The color clear coating composition further comprises a flatting agent. The presence of the flatting agent in a coating film as a particle component having a refractive index different from the vehicle component of the coating film causes adequate diffuse reflection of incident light. In this manner, the flatting agent has an effect of adjusting the gloss of the coating film. In general, silica fine powder (hydrous silicon dioxide), polyethylene powder, resin beads, ceramic beads, dispersion liquids in which these materials are pre-dispersed, or the like, are used. Among these, silica fine powder, a dispersion liquid in which silica fine powder is pre-dispersed, and ceramic beads are preferable, with silica fine powder being particularly preferable. However, there is no limitation thereon, and a single flatting agent or a combination of two or more flatting agents may be used, depending on the desired texture.

[0035] The flatting agent is incorporated preferably in an amount of 1 to 50 parts by mass, per 100 parts by mass of the resin solids content of the color clear coating composition. If the amount is less than 1 part by mass, it is possible that the resulting coating film will be transparent, and that a desired texture cannot be obtained when a multilayer coating film is formed. If the amount exceeds 50 parts by mass, it is possible that the appearance of the resulting coating film will be deteriorated. The amount is more preferably 5 to 30 parts by mass, and particular preferably 5 to 30 parts by mass.

[0036] The color clear coating composition used in the method of the present invention may generally contain a resin component as a vehicle. Specific examples of the resin component include a combination of a base resin with a crosslinking agent. Examples of the base resin include acrylic resin, polyester resin, alkyd resin, and urethane resin, which have a crosslinkable functional group (e.g., hydroxyl). Examples of the crosslinking agent include amino resin (melamine resin and urea resin), and a polyisocyanate compound (including a blocked polyisocyanate compound). Such resin components are used after being dissolved or dispersed in an organic solvent and/or a solvent, such as water.

[0037] The color clear coating composition used in the method of the present invention may optionally further comprise a solvent, such as water or an organic solvent; various additives, such as a dispersion agent, an antisettling agent, a curing catalyst, an antifoaming agent, an antioxidant, a UV absorber, a surface control agent, and a rheology control agent; an extender pigment; and the like. As stated above, the color clear coating composition used in the method of the present invention comprises, as a color pigment, a transparent pigment that achieves a haze value of the above range. As long as the effect of the present invention is obtained, the color clear coating composition may further comprise a small amount of non-transparent pigment in addition to the above color pigment.

[0038] The color clear coating composition used in the method of the present invention is prepared by mixing and dispersing the above-described components. It is preferable to adjust the solids content at the time of application to 12 to 60 mass%, more preferably 15 to 50 mass%, based on the coating composition, and the viscosity at 20°C to 17 to 23 seconds (Ford cup No. 3).

[0039] The application of the color clear coating composition used in the method of the present invention may be performed by electrostatic coating, air spray, airless spray, or the like. It is preferable that the thickness of the coating film is 5 to 30 μm when cured, from the viewpoint of the smoothness of the resulting coating film. When the color clear coating composition is of a baking-drying type, the color clear coating film can generally be cured by crosslinking at a temperature of about 50°C to 150°C. When the color clear coating composition is of a normal temperature drying type or a forced drying type, the film can be cured by crosslinking at a temperature in the range of an ordinary temperature to about 80°C.

[0040] Next, a method for forming a two-layer color clear coating film is described below. When a two-layer coating film is formed, a first clear coating composition comprising a color pigment is applied to the metallic base coating film described above, and a second clear coating composition is applied to the obtained coating film to form a coating film in which the first clear coating film and the second clear coating film are laminated.

[0041] The first clear coating composition comprises a color pigment mentioned above as a color pigment included in the color clear coating composition, and a resin component as a vehicle. As the resin component, resins that can be used in the color clear coating composition may also be used.

[0042] The resin component in the second clear coating composition of the present invention may comprise a resin component that is partially or entirely the same as the resin component used in the metallic base coating composition, from the viewpoint of the dispersion stability of the color pigment in the first clear coating composition.

[0043] The first clear coating composition comprises the color pigment in an amount of preferably 0.01 to 5 parts by mass, more preferably 0.02 to 4 parts by mass, and particular preferably 0.03 to 3 parts by mass, per 100 parts by mass of the resin solids content of the coating composition, from the viewpoint of the chroma and the lightness difference between the highlight and shade of the multilayer coating film.

[0044] The first clear coating composition may further optionally comprise a solvent, such as water or an organic solvent; various additives, such as a dispersion agent, an antisettling agent, a curing catalyst, an antifoaming agent, an antioxidant, a UV absorber, a surface control agent, and a rheology control agent; an extender pigment; and the like.

[0045] The first clear coating composition may be prepared by mixing and dispersing the above-described components. It is preferable to adjust the solids content at the time of application to 12 to 60 mass%, more preferably 15 to 50 mass%, based on the coating composition, and the viscosity at 20°C to 17 to 23 seconds (Ford cup No. 3).

[0046] The application of the first clear coating composition of the present invention may be performed by electrostatic coating, air spray, airless spray, or the like. It is preferable that the thickness of the coating film is 5 to 40 μm, more preferably 5 to 35 μm, when cured, from the viewpoint of the smoothness of the resulting coating film. When the first clear coating composition is of a baking-drying type, the first clear coating film can generally be cured by crosslinking at a temperature of about 50°C to 150°C. When the first clear coating composition is of a normal temperature drying type or a forced drying type, the film can be cured by crosslinking at a temperature in the range of an ordinary temperature to about 80°C. The first clear coating composition of the present invention is generally applied to a predetermined film thickness, heated, dried, and cured, followed by application of the second clear coating composition described below. It is also possible to apply the second clear coating composition when the coating film is in an uncured state.

[0047] The second clear coating composition used in the method of the present invention comprises a flatting agent as an essential component, and a resin component as a vehicle. As the flatting agent and the resin component, those that can be used in the color clear coating composition may also be used.

[0048] The second clear coating composition comprises a flatting agent in an amount of preferably 1 to 50 parts by mass, per 100 parts by mass of the resin solids content of the second clear coating composition. If the amount is less than 1 part by mass, it is possible that the resulting coating film will be transparent, and that a desired texture cannot be obtained when a multilayer coating film is formed. If the amount exceeds 50 parts by mass, it is possible that the appearance of the resulting coating film will be deteriorated. The amount is more preferably 5 to 30 parts by mass, and particular preferably 5 to 30 parts by mass.

[0049] The second clear coating composition may further optionally comprise a solvent, such as water or an organic solvent; various additives, such as a pigment dispersant, an antisettling agent, a rheology control agent, a curing catalyst, an antifoaming agent, an antioxidant, and a UV absorber; an extender pigment; and the like.

[0050] The application of the second clear coating composition of the present invention may be performed by electrostatic coating, air spray, airless spray, or the like. It is preferable that the thickness of the coating film is 15 to 70 μm when cured. When the second clear coating composition is of a baking-drying type, the film can generally be cured by crosslinking at a temperature of about 50°C to 150°C. When the second clear coating composition is of a normal temperature drying type or a forced drying type, the film can be cured by crosslinking at a temperature in the range of an ordinary temperature to about 80°C.

Examples

[0051] Hereinafter, the present invention is described in more detail with reference to Examples and Comparative Examples. However, the present invention is not limited to these Examples. Note that "parts" and "%" are expressed on a mass basis. In the Examples, the "PHR" represents a proportion of a predetermined component (parts by mass) relative to 100 parts by mass of a resin solids content.

Production Example 1

Production of hydroxyl-containing acrylic resin

[0052] 50 parts of ethylene glycol monoethyl ether acetate was placed in a reaction vessel equipped with a thermometer, a thermostat, a stirrer, a reflux condenser, and a dropping funnel, and stirred and mixed. The mixture was heated to 135°C. Subsequently, a mixture of monomer/polymerization initiator described below was added dropwise over 3 hours to the reaction vessel maintained at the same temperature. After the completion of the dropwise addition, the mixture was aged for 1 hour. Thereafter, a mixture comprising 10 parts of ethylene glycol monoethyl ether acetate and 0.6 part of 2,2'-azobis(2-methylpropionitrile) was added dropwise to the reaction product over 1 hour and 30 minutes while maintaining the same temperature, and the resulting product was aged for another 2 hours. Then, the ethylene glycol monoethyl ether acetate was distilled off under a reduced pressure, thereby obtaining a hydroxyl-containing acrylic resin having a hydroxyl value of 54 mg KOH/g, a number average molecular weight of 20,000, and a resin solids content of 65 mass%. The term "number average molecular weight" used herein represents a value determined by gel permeation chromatography (GPC) using the calibration curve of standard polystyrene.

Mixture of monomer/polymerization initiator:

[0053] a mixture comprising 38 parts of methyl methacrylate, 17 parts of ethyl acrylate, 17 parts of *n*-butyl acrylate, 7 parts of hydroxyethyl methacrylate, 20 parts of lauryl methacrylate, and 1 part of acrylic acid; and 2 parts of 2,2'-azobis(2-methylpropionitrile).

1) Substrate to be coated

[0054] A cationic electrodeposition coating composition "Electron 9400HB" (product name; produced by Kansai Paint Co., Ltd.; an epoxy resin and polyamine cationic resin comprising a blocked polyisocyanate compound as a curing agent) was applied to a degreased and zinc phosphate-treated steel plate (JIS G3141, size: 400 x 300 x 0.8 mm) by electrodeposition to a film thickness of 20 μm when cured. The resulting film was heated at 170°C for 20 minutes to be cured by crosslinking, thereby obtaining an electrodeposition coating film.

[0055] "LUGA BAKE Intermediate Gray" (product name, produced by Kansai Paint Co., Ltd.; polyester resin/melamine resin base, organic solvent type), an intermediate coating composition, was applied to the surface of the obtained electrodeposition coating film by air spray coating to a film thickness of 30 μm when cured. The resulting film was heated at 140°C for 30 minutes to be cured by crosslinking. The thus-obtained coated plate comprising an intermediate coating film was regarded as a substrate to be coated.

2) Application

(Examples 1 to 3 and Comparative Examples 1 and 2)

[0056] A coloring material was added at a ratio shown in Table 1 per 100 parts (solids content) of a resin component comprising 75 parts of the hydroxyl-containing acrylic resin obtained in Production Example 1 and 25 parts of U-Van 28-60 (product name, butyl-etherified melamine resin; produced by Mitsui Chemicals, Inc.), and the mixture was stirred and mixed to prepare a base coating composition containing the coloring material. The base coating composition was diluted to a viscosity appropriate for coating to form an organic solvent-based coating composition having a solids content of about 25%. The coating composition was applied to the substrate to be coated by air spray coating to a film thickness of 15 μm when cured. After the application, the resulting product was allowed to stand in a laboratory at a room temperature of about 20°C for about 15 minutes. Thereafter, a color clear coating composition containing a coloring material at a ratio shown in Table 1 per 100 parts by mass of a resin component of a clear coating composition (LUGA BAKE Clear, produced by Kansai Paint Co., Ltd., product name, acrylic resin/amino resin base, organic solvent type) was applied by air spray coating to a film thickness of 30 μm when cured. After the application, the resulting product was allowed to stand at a room temperature for 15 minutes, and heated in a hot-air circulating oven at 140°C for 30 minutes to simultaneously dry and cure the multilayer coating film, thereby obtaining a test plate.

Examples 4-11 and Comparative Examples 3 and 4

[0057] A coloring material was added at a ratio shown in Table 2 per 100 parts (solids content) of a resin component comprising 75 parts of the hydroxyl-containing acrylic resin obtained in Production Example 1 and 25 parts of U-Van 28-60 (product name, butyl-etherified melamine resin, produced by Mitsui Chemicals, Inc.), and the mixture was stirred and mixed to prepare a base coating composition containing the coloring material. The base coating composition was diluted to a viscosity appropriate for coating to form an organic solvent-based coating composition having a solids content of about 25%. The coating composition was applied to the substrate to be coated by air spray coating to a film thickness of 15 μm when cured. After the application, the resulting product was allowed to stand in a laboratory at a room temperature of about 20°C for about 15 minutes. Thereafter, a first clear coating composition containing a coloring material at a ratio shown in Table 2 per 100 parts by mass of a resin component of a clear coating composition (LUGA BAKE Clear, produced by Kansai Paint Co., Ltd.; product name, acrylic resin/amino resin base, organic solvent type) was applied by air spray coating to a film thickness of 10 μm when cured. After the application, the resulting product was allowed to stand at a room temperature for 15 minutes. A second clear coating composition containing a coloring material at a ratio shown in Table 2 per 100 parts by mass of a resin component of a clear coating composition (LUGA BAKE Clear, produced by Kansai Paint Co., Ltd.; product name, acrylic resin/amino resin base, organic solvent type) was further applied by air spray coating to a film thickness of 30 μm when cured. After the application, the resulting product was allowed to stand at a room temperature for 15 minutes, and heated in a hot-air circulating oven at 140°C for 30 minutes to simultaneously dry and cure the multilayer coating film, thereby obtaining a test plate.

Examples 12 and 13

[0058] Test plates were obtained in the same manner as in Examples 4 to 11 except that the first clear coating composition containing a coloring material at a ratio shown in Table 2 per 100 parts (solids content) of a resin component comprising 75 parts of the hydroxyl-containing acrylic resin obtained in Production Example 1 and 25 parts of U-Van 28-60 (product name, butyl-etherified melamine resin, produced by Mitsui Chemicals, Inc.) was used as the first clear coating composition.

Table 1

	Base coating composition		Color clear coating composition		Aesthetic		
	Type of coloring material	Amount (PHR)	Type of coloring material (flatting agent)	Amount (PHR)	A	B	C
Example 1	A	8.0	a/c	1.0/13.0	3	4	3
Example 2	B	10.0	a/c	1.0/13.0	3	4	3
Example 3	C	10.0	a/c	1.0/13.0	3	4	3
Comp. Example 1	g/h	80.0/0.5	a/c	1.0/13.0	1	3	1
Comp. Example 2	D/E/d/f	5.0/6.0/5.0/5.0	a	13.0	2	4	2

*In the table, the amounts of the coloring material and flatting agent were represented by parts by mass (PHR) per 100 parts by mass of the resin component in the coating composition.
Coloring material and flatting agent
A: Metasheen 71-0010 (product name, a vacuum-metalized aluminum flake; produced by Toyo Aluminium K.K., IV=168)
B: Aluminum paste GX-40A (product name, an aluminum flake pigment, average particle diameter: 17 μm; produced by Asahi Kasei Chemicals, Corp., IV=517)
C: Aluminum paste 91-4056 (product name, aluminum flake pigment, average particle diameter: 8.2 μm; produced by Toyo Aluminium K.K., IV=126)
D: Paliocrom Orange L 2800 (product name, iron oxide-coated aluminium pigment; produced by BASF, IV=30.5)
E: Iriodin Ultra 7235 WNT (product name, titanium oxide-coated mica pigment; produced by Merck & Co., Inc.)
a: Acematt 3300 (product name, dry silica; produced by Evonik Degussa Co., Ltd.)
b: Sylicia 435 (product name, wet silica; produced by Fuji Sylicia Chemical, Ltd.)
c: Perrindo Maroon 179-229 6436 (product name, Pigment Red 179; produced by DIC Sun Chemical, haze=0.9)
d: Perrindo Maroon 179-229 6440 (product name, Pigment Red 179; produced by DIC Sun Chemical, haze=2.0)
e: Blue G-314 (product name, Pigment Blue 15:1; produced by Sanyo Color Works, Ltd., haze=0.8)
f: Cinquasia Magenta RT-355D (product name, Pigment Red 202; produced by Chiba Specialty Chemicals, haze=2.0)
g: Tipaque CR-95 (product name, white titanium oxide pigment; produced by Ishihara Sangyo Kaisha, Ltd., haze=86)
h: Monarch 1300 (product name, carbon black pigment; produced by Cabot Corporation, haze: 15)

Table 2

	Base coating composition		First clear coating composition		Second clear coating composition		Aesthetic		
	Type of coloring material	Amount (PHR)	Type of coloring material	Amount (PHR)	Type of flatting agent	Amount (PHR)	A	B	C
Example 4	A	8.0	c	1.0	a	13.0	4	4	4
Example 5	B	10.0	c	1.0	a	13.0	4	4	3
Example 6	C	10.0	c	1.0	a	13.0	3	4	3
Example 7	A	8.0	c	1.0	b	20.0	4	4	4
Example 8	A	8.0	d	1.0	a	13.0	3	3	3

(continued)

	Base coating composition		First clear coating composition		Second clear coating composition		Aesthetic		
	Type of coloring material	Amount (PHR)	Type of coloring material	Amount (PHR)	Type of flatting agent	Amount (PHR)	A	B	C
Example 9	A	8.0	e	1.0	a	13.0	4	4	4
Example 10	B	10.0	e	1.0	a	13.0	4	4	3
Example 11	C	10.0	e	1.0	a	13.0	3	4	3
Example 12	A	8.0	c	1.0	a	13.0	4	4	4
Example 13	A	8.0	d	1.0	a	13.0	3	4	4
Comp. Ex. 3	g/h	80.0/0.5	c	1.0	a	13.0	1	3	1
Comp. Ex. 4	g/h	80.0/0.5	e	1.0	a	13:0	1	3	1

*In the table, the amounts of the coloring material and flatting agent were represented by parts by mass (PHR) per 100 parts by mass of the resin component in the coating composition.
Coloring material and flatting agent
A: Metasheen 71-0010 (product name, vacuum-metalized aluminum flake; produced by Toyo Aluminium K.K., IV=168)
B: Aluminum paste GX-40 (product name, aluminum flake pigment, average particle diameter: 17 μm; produced by Asahi Kasei Chemicals, Corp., IV=517)
C: Aluminum paste 91-4056 (product name, aluminum flake pigment, average particle diameter: 8.2 μm; produced by Toyo Aluminium K.K., IV=126)
D: Paliocrom Orange L 2800 (product name, iron oxide-coated aluminium pigment; produced by BASF, IV=30.5)
E: Iriodin Ultra 7235 WNT (product name, titanium oxide-coated mica pigment; produced by Merck & Co., Inc.)
a: Acematt 3300 (product name, dry silica; produced by Evonik Degussa Co., Ltd.)
b: Sylicia 435 (product name, wet silica; produced by Fuji Sylicia Chemical, Ltd.)
c: Perrindo Maroon 179-229 6436 (product name, Pigment Red 179; produced by DIC Sun Chemical, haze=0.9)
d: Perrindo Maroon 179-229 6440 (product name, Pigment Red 179; produced by DIC Sun Chemical, haze=2.0)
e: Blue G-314 (product name, Pigment Blue 15:1; produced by Sanyo Color Works, Ltd., haze=0.8)
f: Cinquasia Magenta RT-355D (product name, Pigment Red 202; produced by Chiba Specialty Chemicals, haze=2.0)
g: Tipaque CR-95 (product name, white titanium oxide pigment; produced by Ishihara Sangyo Kaisha, Ltd., haze=86)
h: Monarch 1300 (product name, carbon black pigment; produced by Cabot Corporation, haze: 15)

Aesthetic Evaluation

[0059] The resulting test plates were evaluated in the following manner. Tables 1 and 2 show the results.

[0060] Each obtained test plate was illuminated with an artificial sunlight lamp (produced by Seric Ltd., color temperature: 6500 K), and the angle of the test plate relative to the lamp was changed to observe and evaluate the following A, B, and C. Tables 1 and 2 shows the results. The evaluation was performed by five people (two designers and three engineers) engaged in color development for three years or more, and the average values were employed:

A: brightness at the highlight;
B: chroma at the highlight; and
C: sense of depth (lightness change from the highlight to the shade; high chroma at the shade)

[0061] A to C were rated on a one-to-four scale. The larger the value, the higher the brightness at the highlight, the

higher the chroma at the highlight, and the greater the sense of depth.

Production Example 2

Production of acrylic resin emulsion

[0062] 130 parts by mass of deionized water and 0.52 part by mass of Aqualon KH-10 (product name, surfactant; produced by Dai-Ichi Kogyo Seiyaku Co., Ltd.) were placed in a reaction vessel equipped with a thermometer, a thermostat, a stirrer, a reflux condenser, and a dropping funnel, then stirred and mixed under a nitrogen stream, and heated to 80°C. Subsequently, 1% of the total amount of monomer emulsion (1) described below and 5.3 parts by mass of a 6% aqueous ammonium persulfate solution were introduced into the reaction vessel, and maintained at 80°C for 15 minutes. Thereafter, the remaining monomer emulsion (1) was added dropwise over 3 hours to the reaction vessel maintained at the same temperature. After the completion of the dropwise addition, the mixture was aged for 1 hour. Subsequently, monomer emulsion (2) described below was added dropwise over 1 hour, followed by aging for 1 hour. Thereafter, the mixture was cooled to 30°C while 40 parts by mass of a 5% aqueous dimethylethanolamine solution was gradually added thereto, and filtered through a 100-mesh nylon cloth, thereby obtaining an acrylic resin emulsion having a average particle diameter of 100 nm (after diluted with deionized water, the average particle diameter was measured at 20°C using a "Coulter N4" submicron particle size distribution analyzer (produced by Beckman Coulter)) and a solids content of 30%. The obtained acrylic resin had an acid value of 33 mg KOH/g and a hydroxyl value of 25 mg KOH/g.

[0063] Monomer emulsion (1): 42 parts by mass of deionized water, 0.72 part by mass of Aqualon KH-10, 2.1 parts by mass of methylene bis acrylamide, 2.8 parts by mass of styrene, 16.1 parts by mass of methyl methacrylate, 28 parts by mass of ethyl acrylate, and 21 parts by mass of *n*-butyl acrylate were mixed and stirred, thereby obtaining monomer emulsion (1).

[0064] Monomer emulsion (2): 18 parts by mass of deionized water, 0.31 part by mass of Aqualon KH-10, 0.03 part by mass of ammonium persulfate, 5.1 parts by mass of methacrylic acid, 5.1 parts by mass of 2-hydroxyethyl acrylate, 3 parts by mass of styrene, 6 parts by mass of methyl methacrylate, 1.8 parts by mass of ethyl acrylate, and 9 parts by mass of *n*-butyl acrylate were mixed and stirred, thereby obtaining monomer emulsion (2).

Production Example 3

Production of polyester resin

[0065] 109 parts by mass of trimethylolpropane, 141 parts by mass of 1,6-hexanediol, 126 parts by mass of hexahydrophthalic anhydride, and 120 parts by mass of adipic acid were placed in a reaction vessel equipped with a thermometer, a thermostat, a stirrer, a reflux condenser, and a water separator. The mixture was heated from 160°C to 230°C over a period of 3 hours, followed by a condensation reaction at 230°C for 4 hours. Subsequently, in order to add carboxyl groups to the obtained condensation reaction product, 38.3 parts by mass of trimellitic anhydride was further added thereto, and reacted at 170°C for 30 minutes. The reaction product was diluted with 2-ethyl-1-hexanol, thereby obtaining a polyester resin solution having a solids content of 70%. The obtained polyester resin had an acid value of 46 mg KOH/g, a hydroxyl value of 150 mg KOH/g, and a weight average molecular weight of 6,400. The term "weight average molecular weight" used herein represents a value determined by gel permeation chromatography (GPC) using the calibration curve of standard polystyrene.

Production Example 4

Production of phosphoric acid-containing resin solution

[0066] A mixed solvent comprising 27.5 parts of methoxypropanol and 27.5 parts of isobutanol was placed into a reaction vessel equipped with a thermometer, a thermostat, a stirrer, a reflux condenser, and a dropping funnel, and heated to 110°C. Subsequently, while maintaining the temperature at 110°C, 121.5 parts of a mixture comprising 25 parts of styrene, 27.5 parts of *n*-butyl methacrylate, 20 parts of branched higher alkyl acrylate (product name: Isostearyl Acrylate, produced by Osaka Organic Chemical Industry, Ltd.), 7.5 parts of 4-hydroxybutyl acrylate, 15 parts of a phosphoric acid-containing polymerizable monomer described below, 12.5 parts of 2-methacryloyloxy ethyl acid phosphate, 10 parts of isobutanol, and 4 parts of *tert*-butyl peroxyoctanoate were added to the above mixed solvent over 4 hours. Further, a mixture comprising 0.5 part of *tert*-butyl peroxyoctanoate and 20 parts of isopropanol was added dropwise thereto over 1 hour, followed by aging with stirring for 1 hour, thereby obtaining a phosphoric acid-containing resin solution having a solids content of 50%. The phosphoric acid-containing resin had an acid value of 83 mg KOH/g, a hydroxyl value of 29 mg KOH/g, and a weight average molecular weight of 10,000.

Phosphoric acid-containing polymerizable monomer:

[0067] 57.5 parts of monobutyl phosphate and 41 parts of isobutanol were placed into a reaction vessel equipped with a thermometer, a thermostat, a stirrer, a reflux condenser, and a dropping funnel, and heated to 90°C. Subsequently, 42.5 parts of glycidyl methacrylate was added dropwise over 2 hours. After aging for 1 hour with stirring, 59 parts of isopropanol was added thereto, thereby obtaining a phosphoric acid-containing polymerizable monomer solution with a solids content of 50%. The resulting monomer had an acid value of 285 mg KOH/g.

Production Example 5

Preparation of color pigment dispersion 1

[0068] 25.7 parts (solids content: 18 parts) of polyester resin solution, 18 parts of "Perrindo Maroon 179-229 6436" (product name, Pigment Red 179; produced by DIC Sun Chemical, 1.4 parts of "Solsperse 12000" (product name, phthalocyanine pigment derivative; produced by Lubrizol), and 61.3 parts of deionized water were placed in a stirring and mixing container, and uniformly mixed. Further, 2-(dimethylamino)ethanol was added thereto, and the pH of the mixture was adjusted to 7.5. The obtained mixture was then placed into a resin-made bottle having a capacity of 225 ml. Zirconia beads (130 parts) having a diameter of about 1.5 mm were added to the bottle, which was then hermetically sealed, and the mixture was dispersed for 120 minutes by a shaker-type paint conditioner. After dispersion, the mixture was filtered through a 100-mesh wire screen to remove zirconia beads, thereby obtaining color pigment dispersion 1.

Production Example 6

Preparation of concentrated aluminum pigment solution

[0069] In a stainless steel beaker, 13.5 parts (solids content: 10 parts) of aluminum paste GX-30A (product name, aluminum flake pigment, average particle diameter: 17 μm, aluminum content: 74%), 8 parts (solids content: 4 parts) of the phosphoric acid-containing resin solution obtained in Production Example 4, 38.5 parts of 2-ethyl-1-hexanol (mass dissolved in 100 g of water at 20°C), and 0.5 part of 2-(dimethylamino)ethanol were uniformly mixed, thereby obtaining a concentrated aluminum pigment solution.

Production Example 7

Aqueous base coating composition

[0070] 100 parts of the acrylic resin emulsion obtained in Production Example 2, 57.1 parts of the polyester resin solution obtained in Production Example 3, 60.5 parts of the concentrated aluminum pigment solution obtained in Production Example 6, and 37.5 parts of Cymel 325 (product name, produced by Nihon Cytec Industries Inc., melamine resin, solids content: 80%) were uniformly mixed. Further, Primal ASE-60 (product name, polyacrylic acid thickener; produced by Rohm & Haas Co., Ltd.), 2-(dimethylamino)ethanol, and deionized water were added thereto, thereby preparing an aqueous coating composition having a pH of 8.0, a coating composition solids content of 25%, and a viscosity of 40 seconds as measured by Ford cup No. 4 at 20°C. In the coating composition prepared in Example 14, the ratio of the coloring material is the same as that in the base composition of Example 2.

Production Example 8

Aqueous first clear coating composition

[0071] 100 parts of the acrylic resin emulsion obtained in Production Example 2, 56.1 parts of the polyester resin solution obtained in Production Example 3, 5.9 parts of the pigment dispersion obtained in Production Example 5, and 37.5 parts of Cymel 325 (product name, melamine resin; produced by Nihon Cytec Industries Inc., solids content: 80%) were uniformly mixed. Further, Primal ASE-60 (product name, polyacrylic acid thickener; produced by Rohm & Haas Co., Ltd.), 2-(dimethylamino)ethanol, and deionized water were added thereto, thereby preparing an aqueous coating composition having a pH of 8.0, a coating composition solids content of 25%, and a viscosity of 40 seconds as measured by Ford cup No. 4 at 20°C.

Example 14

[0072] The aqueous base coating composition obtained in Production Example 7 was applied to the substrate to be coated by air spray coating to a film thickness of 15 μm when cured. After the application, the resulting product was allowed to stand at a room temperature of 20°C for about 5 minutes in a lavatory, and heated in a hot-air circulating oven at 80°C for 10 minutes. Thereafter, a color clear coating composition comprising a coloring material at a ratio shown in Table 3 per 100 parts by mass of a resin component of a clear coating composition ("LUGA BAKE Clear" (product name, produced by Kansai Paint Co., Ltd.; acryl resin/amino resin base, organic solvent type)) was applied by air spray coating to a film thickness of 30 μm when cured. After the application, the resulting product was allowed to stand at room temperature for 15 minutes, and heated in a hot-air circulating oven at 140°C for 30 minutes to simultaneously dry and cure the multilayer coating film, thereby obtaining a test plate. The test plate was evaluated in accordance with the above-described "Aesthetic Evaluation."

Table 3

	Base coating composition		Color clear coating composition		Aesthetic		
	Type of coloring material	Amount (PHR)	Type of coloring material and flatting agent	Amount (PHR)	A	B	C
Example 14	B	10.0	a/c	1.0/13.0	3	4	3

*In the table, the amounts of the coloring material and flatting agent were represented by parts by mass (PHR) per 100 parts by mass of the resin component in the coating composition.
Coloring material and flatting agent
B: Aluminum paste GX-40 (product name, an aluminum flake pigment, average particle diameter: 17 μm; produced by Asahi Kasei Chemicals, Corp., IV=517)
a: Acematt 3300 (product name, dry silica; produced by Evonik Degussa Co., Ltd.)
c: Perrindo Maroon 179-229 6436 (product name, Pigment Red 179; produced by DIC Sun Chemical, haze=0.9)

Example 15

[0073] The aqueous base coating composition obtained in Production Example 7 was applied to the substrate to be coated by air spray coating to a film thickness of 15 μm when cured. After the application, the resulting product was allowed to stand at a room temperature of about 20°C for about 5 minutes in a laboratory, and heated in a hot-air circulating oven at 80°C for 10 minutes. Thereafter, a first clear coating composition comprising a coloring material at a ratio shown in Table 4 per 100 parts by mass of a resin component of a clear coating composition (product name: LUGA BAKE Clear, acryl resin/amino resin base, organic solvent type; produced by Kansai Paint Co., Ltd.) was applied by air spray coating to a film thickness of 10 μm when cured. After the application, the resulting product was allowed to stand at room temperature for 15 minutes, and a second clear coating composition comprising a coloring material at a ratio shown in Table 4 per 100 parts by mass of a resin component of a clear coating composition (product name: LUGA BAKE Clear, acryl resin/amino resin base, organic solvent type; produced by Kansai Paint Co., Ltd.) was applied by air spray coating to a film thickness of 30 μm when cured. After the application, the resulting product was allowed to stand at room temperature for 15 minutes, and heated in a hot-air circulating oven at 140°C for 30 minutes to simultaneously dry and cure the multilayer coating film, thereby obtaining a test plate.

Example 16

[0074] The aqueous base coating composition obtained in Production Example 7 was applied to the substrate to be coated by air spray coating to a film thickness of 15 μm when cured. After the application, the resulting product was allowed to stand at a room temperature of about 20°C for about 5 minutes in a laboratory, and heated in a hot-air circulating oven at 80°C for 10 minutes. Thereafter, the aqueous first clear coating composition produced in Production Example 8 was applied by air spray coating to a film thickness of 10 μm when cured. After the application, the resulting product was allowed to stand at room temperature for 5 minutes, and a second clear coating composition comprising a coloring material at a ratio shown in Table 4 per 100 parts by mass of a resin component of a clear coating composition (product name: LUGA BAKE Clear, acryl resin/amino resin base, organic solvent type; produced by Kansai Paint Co., Ltd.) was applied by air spray coating to a film thickness of 30 μm when cured. After the application, the resulting product was allowed to stand at room temperature for 15 minutes, and heated in a hot-air circulating oven at 140°C for 30 minutes

to simultaneously dry and cure the multilayer coating film, thereby obtaining a test plate. The test plate was evaluated in accordance with the above-described "Aesthetic Evaluation."

Table 4

	Base coating composition		First clear coating composition		Second clear coating composition		Aesthetic		
	Type of coloring material	Amount (PHR)	Type of coloring material	Amount (PHR)	Type of flatting agent	Amount (PHR)	A	B	C
Ex. 15	B	10.0	c	1.0	a	13.0	4	4	3
Ex. 16	B	10.0	c	1.0	a	13.0	4	4	3

*In the table, the amounts of the coloring material and flatting agent were represented by parts by mass (PHR) per 100 parts by mass of the resin component in the coating composition.
Coloring material and flatting agent
B: Aluminum paste GX-40 (product name, aluminum flake pigment, average particle diameter: 17 μm; produced by Asahi Kasei Chemicals, Corp., IV=517)
a: Acematt 3300 (product name, dry silica; produced by Evonik Degussa Co., Ltd.)
c: Perrindo Maroon 179-229 6436 (product name, Pigment Red 179; produced by DIC Sun Chemical, haze=0.9)

Industrial Applicability

[0075] The method for forming a coating film of the present invention is applicable to various industrial products, in particular, an automobile exterior panel.

Claims

1. A method for forming a coating film, the method comprising:

forming a metallic base coating film comprising an aluminum flake pigment on a substrate; and
forming a color clear coating film comprising a color pigment and a flatting agent on the metallic base coating film, wherein the color pigment in the color clear coating film is a transparent pigment, and wherein when the transparent pigment is contained in a coating composition as a sole coating material in an amount of 1 part by mass, per 100 parts by mass of the resin solids content, which is a vehicle formation component, and when this coating composition is used to form a coating film having a thickness of 100 μm, this coating film achieves a haze value of 0.1 to 10.0.

2. The method for forming a coating film according to claim 1, wherein the flatting agent in the color clear coating film is at least one member selected from the group consisting of silica fine powder, polyethylene powder, resin beads, and ceramic beads.

3. The method for forming a coating film according to claim 1 or 2, wherein the color clear coating film is formed as a two-layer film in which a second clear coating film comprising a flatting agent is laminated on a first clear coating film comprising a color pigment.

4. The method for forming a coating film according to claim 3, wherein each of the metallic base coating film and the first clear coating film comprises at least one type of resin as a vehicle formation component, and wherein at least a portion of the resin as the vehicle formation component in the metallic base coating film is the same as the resin as the vehicle formation component in the first clear coating film.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2013/060976

A. CLASSIFICATION OF SUBJECT MATTER
B05D1/36(2006.01)i, *B05D5/06*(2006.01)i

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)
B05D1/36, B05D5/06

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
Jitsuyo Shinan Koho 1922-1996 Jitsuyo Shinan Toroku Koho 1996-2013
Kokai Jitsuyo Shinan Koho 1971-2013 Toroku Jitsuyo Shinan Koho 1994-2013

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 2006-87978 A (Kansai Paint Co., Ltd.), 06 April 2006 (06.04.2006), claims; paragraphs [0011], [0015], [0024] to [0033], [0043] to [0050]; examples (Family: none)	1-4
X	JP 2011-20021 A (Kansai Paint Co., Ltd.), 03 February 2011 (03.02.2011), claims; paragraphs [0001], [0010], [0023] to [0024], [0034] to [0043]; examples (Family: none)	1
A		2-4
A	JP 2003-245603 A (Nippon Paint Co., Ltd.), 02 September 2003 (02.09.2003), claims; paragraphs [0015], [0023] to [0024], [0044] to [0053]; examples (Family: none)	1-4

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:
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"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
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Date of the actual completion of the international search	Date of mailing of the international search report
01 July, 2013 (01.07.13)	09 July, 2013 (09.07.13)
Name and mailing address of the ISA/ Japanese Patent Office Facsimile No.	Authorized officer Telephone No.

Form PCT/ISA/210 (second sheet) (July 2009)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2013/060976

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 2010-194429 A (Nippon Paint Co., Ltd.), 09 September 2010 (09.09.2010), claims; paragraphs [0048] to [0053] (Family: none)	1-4
A	JP 2004-313983 A (Kansai Paint Co., Ltd.), 11 November 2004 (11.11.2004), claims; paragraph [0031] (Family: none)	1-4
P,X	JP 2012-232236 A (Kansai Paint Co., Ltd.), 29 November 2012 (29.11.2012), claims; paragraphs [0006], [0020], [0044] to [0052], [0056] to [0061]; examples (Family: none)	1-4

Form PCT/ISA/210 (continuation of second sheet) (July 2009)

REFERENCES CITED IN THE DESCRIPTION

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Patent documents cited in the description

- JP 2012092156 A **[0001]**