



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<p>(51) International Patent Classification <sup>6</sup> : C09C 1/62, C04B 14/20</p>	<p>A1</p>	<p>(11) International Publication Number: <b>WO 99/35194</b></p> <p>(43) International Publication Date: 15 July 1999 (15.07.99)</p>
<p>(21) International Application Number: PCT/US98/23761</p> <p>(22) International Filing Date: 9 November 1998 (09.11.98)</p> <p>(30) Priority Data: 09/005,064                      9 January 1998 (09.01.98)                      US</p> <p>(71) Applicant: FLEX PRODUCTS, INC. [US/US]; 1402 Mariner Way, Santa Rosa, CA 95407-7370 (US).</p> <p>(72) Inventors: COULTER, Kent; 2128 Contra Costa Avenue, Santa Rosa, CA 95405 (US). MAYER, Thomas; 2490 Mark West Springs Road, Santa Rosa, CA 95404 (US). PHILLIPS, Roger, W.; 466 Jacqueline Drive, Santa Rosa, CA 95405 (US).</p> <p>(74) Agents: SEELEY, David, O. et al.; Workman, Nydegger &amp; Seeley, 1000 Eagle Gate Tower, 60 East South Temple, Salt Lake City, UT 84111 (US).</p>		<p>(81) Designated States: AU, CA, CN, JP, KR, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).</p> <p><b>Published</b> <i>With international search report.</i></p>
<p>(54) Title: BRIGHT METAL FLAKE</p> <p>(57) Abstract</p> <p>A rigid and brittle bright metal flake is formed of a central layer of a reflective material supported on both sides by dielectric layers. In a preferred embodiment, the metal layer is aluminum having a thickness of about 100 nm and the dielectrics are either silicon dioxide or magnesium fluoride, each having a thickness of about 100 nm. The result is a very thin three-layered metal flake about 300 nm thick that exhibits a uniaxial compressive strength of about 8 times a corresponding uniaxial tensile strength. As a result, the metal flake is then afforded the benefits of rigidity and brittle fracture during the manufacturing and applicational processes which ultimately provides favorable planar and specular reflectance characteristics in the visible wavelength range.</p>		

**FOR THE PURPOSES OF INFORMATION ONLY**

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece			TR	Turkey
BG	Bulgaria	HU	Hungary	ML	Mali	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MN	Mongolia	UA	Ukraine
BR	Brazil	IL	Israel	MR	Mauritania	UG	Uganda
BY	Belarus	IS	Iceland	MW	Malawi	US	United States of America
CA	Canada	IT	Italy	MX	Mexico	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NE	Niger	VN	Viet Nam
CG	Congo	KE	Kenya	NL	Netherlands	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NO	Norway	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	NZ	New Zealand		
CM	Cameroon		Republic of Korea	PL	Poland		
CN	China	KR	Republic of Korea	PT	Portugal		
CU	Cuba	KZ	Kazakstan	RO	Romania		
CZ	Czech Republic	LC	Saint Lucia	RU	Russian Federation		
DE	Germany	LI	Liechtenstein	SD	Sudan		
DK	Denmark	LK	Sri Lanka	SE	Sweden		
EE	Estonia	LR	Liberia	SG	Singapore		

**BRIGHT METAL FLAKE****BACKGROUND OF THE INVENTION****1. The Field of the Invention**

5           The present invention relates generally to metal flakes. More specifically the present invention relates to thin metal flakes useful as pigments.

**2. The Relevant Technology**

10           The prior art is replete with various applications utilizing metallic flakes or platelets (hereinafter collectively referred to as flakes) to improve the lustre, sparkle, shine, and/ or reflective properties of the application. Such applications are well known and include coating compositions, inks, extrusions, paints, electrostatic coatings, infrared interference pigments, glass, ceramics and cosmetics. In general, it is known that for the application to achieve the greatest specular reflectance across visible wavelengths (300 -  
15           800 nm), the metallic flakes should individually lay as flat as possible. As a collection of numerous flakes, the greatest reflectance, and hence greatest brightness, occurs when the flakes are collectively planar oriented to expose the greatest amount of surface area of the metallic flakes to the incident light and reflect as much of that light as possible.

20           A major factor, however, affecting those reflectance characteristics is the size or dimensions of the flake as the flake is used in a particular application. For example, if the flakes are thick, a plurality of thick flakes combined together in an application are prevented from lying together in a generally flat or horizontal singular plane because adjacent flakes cannot easily overlap each other due to their thickness. As a result, many flakes are adversely caused to be oriented in a substantially vertical manner and the plurality of flakes are formed into a radically non-planar layer. Incident light then  
25           exposed upon the non-planar layer is subject to extreme scatter and diffraction. Thus, the favorable reflective properties of the application are diminished by thick flakes. To a lesser extent, thick flakes frequently cause other difficulties such as the clogging of automatic-spray paint guns during painting applications.

30           However, it is also well known that as the thicknesses of the flakes is reduced, the point is reached where the flakes become so flimsy (non-rigid, flaccid) that they begin to curl and/ or wrinkle. This decreases favorable planarity and reflective properties because incident light exposed upon the flakes is subject to scatter and diffraction. Additionally, if the flakes are too thin when applied onto a surface during applicational use, the flakes will assume any microscopic defects in the contour of that surface. For example, if that  
35           contour is rough, the flakes will correspondingly be rough or non-planar. Again,

disfavored planarity and reflective properties result because incident light exposed on the surface is subject to scatter and diffraction.

Some manufacturing processes form flakes from a singular, larger sheet or film of metal which is "fractured" by well known means into smaller, flake-sized particles. Two types of fracture may result, "ductile" or "brittle." Ductile fractures cause the metal to undergo substantial plastic deformation near the vicinity of fracture before fracture occurs. This deformation causes numerous malformed regions having unfavorable planar characteristics to appear. As before, these malformed regions, such as regions having curled or wrinkled metal, disadvantageously tend to scatter and diffuse incident light exposed thereupon. Brittle fractures, on the other hand, tend to cause little or no plastic deformation of the metal before the fracture occurs which enables the produced metal flake to maintain, as much as possible, the original planarity of the larger metal sheet. Consequently, brittle fracture is the desired manufacturing fracture means.

However, brittle fracture does not occur with most metals having high reflectivity. In fact, brittle fracture is only likely to occur with materials having a large compressive strength as compared to its corresponding tensile strength. This is because the internal bond strength distributed throughout a material is composed of tensile and compressive components. The tensile strength compensates for forces out of the plane of the material and the compressive strength is related to forces in the plane. Thus, similar compressive and tensile strengths will allow ductile deformations since the relative strength into and out of the plane is equivalent. In contrast, brittle deformation occurs when the compressive strength is greater than the tensile strength and the material strength is directed into the plane, not out of the plane. Consequently, a high compressive strength relative to tensile strength results in bond rupture and material cracking when a force is applied. Thus, aluminum, for example, which has a tensile strength of about 13-24 lb/in<sup>2</sup> and a compressive strength of about 13-24 lb/in<sup>2</sup>, would most likely undergo a ductile fracture under a uniaxial stress which would cause the aluminum to exhibit disfavored reflective characteristics. Moreover, once the aluminum is bent or deformed, as would occur with ductile fracture, the aluminum remains deformed and the disfavored reflective characteristics would persist. Consequently, it is difficult to manufacture metal flakes, such as aluminum, without malformations that reduce reflectance.

As is well known, fracture mechanics are not only important for metal flakes during the manufacturing process, but are as equally important during use. For example, applicational processes, such as the drying of a paint or ink solvent, also induce stresses on the flake. These stresses, caused by surface tension, again cause the flake to undergo

fracture or malformation. Since brittle fracture of the flake during the applicational process also tends to produce smaller flakes that maintain much of the original planarity of the larger flake, instead of curled or deformed flakes, flake planarity and reflective properties are improved. Thus, flake brittleness is a characteristic not only preferred during the manufacture process but also preferred during the applicational use. Accordingly, the prior art has attempted to produce thin, rigid and brittle flakes facilitating both the manufacturing thereof and the reflective properties of the application.

Yet all prior solutions have involved compromises. For example, in U.S. Patent No. 5,198,042, entitled "Aluminum Alloy Powders for Coating Materials and Materials Containing the Alloy Powders," it is taught to alloy the metal flake with other materials and metals to reduce the adverse curling, wrinkling and malleability of thin flakes. Alloying, however, dilutes the reflectance properties of the flake.

In U.S. Patent No. 4,213,886, entitled "Treatment of Aluminum Flake to Improve Appearance of coating Compositions," a surface bound species that pulls the flake flat in a coating resin. This method, however, requires chemical tailoring of the flake and the resin in order achieve chemical compatibility with the species. Such compatibility is difficult and has not proved to be practical.

In U.S. Patent No. 4,629,512, entitled "Leafing Aluminum Pigments of Improved Quality," flakes are floated on a resin coating. Adversely, this method submits the flake to durability attacks because the flake is unprotected. Such attacks primarily include corrosion which not only corrodes the flake but tends to give the application a mottled or discolored appearance. Additionally, if this method were used in conjunction with another resinous application, such as a clear overcoat paint, the overcoat itself would tend to disfavorably disrupt the planar orientation of the flake because of solvent penetration. Again, reflectance properties are decreased.

In U.S. Patent No. 5,593,773, entitled "Metal Powder Pigment," pre-cracked flakes are disclosed having such a small aspect ratio that malformation of the flake is essentially impossible. A shrinking aspect ratio, however, also correspondingly shrinks the inherent reflectance capability of the flake. This is because, as the aspect ratio becomes smaller, any non-planar flake orientation during applicational use exposes such a small surface area of the flake to the incident light that reflection of that light is minimal. Other prior art proposals, such as encapsulating a metal flake in a metallic coating, also decrease the flake aspect ratio which adversely eliminates the ability of the flake to reflect incident light.

In U.S. Patent No. 3,622,473, entitled "Method of Providing Aluminum Surfaces with Coatings," flake rigidity is increased by oxidizing the reflector of the flake to form a rigid, outer oxide layer. Whenever an oxide is used, however, the inherent reflectance properties of the flake are decreased. Additionally, oxides are typically formed at defect sites on the flakes which then tends to prevent a uniform application across the surface of the flake. This non-uniformity introduces a reduction in reflectance and can also cause a mottled applicational appearance.

In U.S. Patents Nos. 3, 988,494 and 4,978,394 attempts have been made to improve flake rigidity by applying singular or multiple layer coatings about the surfaces thereof. Thus far, the singular layer coatings have been so thick that reflective properties are detrimentally damaged because the coatings have greatly contributed to the scatter of light. The multiple layer coatings have induced even more scatter and adversely caused light to diffuse at the boundaries between various layers. In addition, all coatings thus far have essentially been organic and inherent within the crystalline structure of these organic coatings is a natural limitation as to how thinly applied the coatings can be manufactured and still provide structural rigidity to a flimsily thin metal flake. Disadvantageously, the natural thickness limitation is still so large that other applicational processes remain burdened by this thickness. Such processes include spraying the flakes through an automatic-spray paint gun. Moreover, organic coatings when applicationally used in a solvent are eventually caused to lose structural rigidity because of dissolution related effects.

Although some reflective coatings exist that are rigid and facilitate brittle fracture, the coatings are unlike most of the other prior art because they do not even use a metal flake. In U.S. Patent No. 4,309,075, entitled "Multilayer Mirror with Maximum Reflectance," for example, multiple layer coatings are taught that merely simulate a metal flake and its reflective properties. The coatings, known as "high-low" coatings after their alternatingly layers of high-low indices of refraction, are used to create a reflector that simulates the reflective properties of a metal flake. Another example is described in U.S. Patent 3,123,490 issued to Balomey wherein a layer of ZnS coated on a top and bottom thereof with MgF<sub>2</sub>. Although rigid and subject to brittle fracture, this structure is typically very thick (about 215 microns) and cannot be used in many applications requiring thin flakes. Moreover, it is often necessary to have numerous layers of alternating high-low coatings to achieve simulation of the metal flake. But as thicknesses and layers increase, manufacturing complexities and economic burdens correspondingly increase.

Accordingly, it is desirous to find alternatives for inexpensively producing a thin, rigid and brittle metal flake to provide favorable reflective characteristics to various other applications.

### SUMMARY

5 In accordance with the invention as embodied and broadly described herein, the foregoing is achieved by preparing a sheet comprising a thin metal layer having dielectric coatings disposed on the surfaces thereof. This structure has the favorable properties of being both brittle and rigid so that it is easily fractured into small bright metal flakes during the manufacturing processes without the flakes becoming curled or wrinkled. The  
10 flakes so-formed have been observed as having a large aspect ratio capable of favorably reflecting substantial amounts of incident light during applicational use.

In a preferred embodiment, the metal layer is aluminum having a thickness of about 100 nm. Each side of the aluminum is preferably coated with an inorganic dielectric, such as silicon dioxide or magnesium fluoride, each having a thickness of  
15 about 100 nm. The result is a very thin three-layered metal flake about 300 nm thick that exhibits a uniaxial compressive strength of about 8 times its uniaxial tensile strength. As a result, the metal flake is then afforded the benefits of brittle fracture and rigidity during the manufacturing and applicational processes which ultimately provides favorable planar and reflective characteristics for both the flake and the applicational use.

### BRIEF DESCRIPTION OF THE DRAWINGS

In order to more fully understand the manner in which the above-recited and other advantages and objects of the invention are obtained, a more particular description of the invention will be rendered by reference to specific embodiments thereof which are illustrated in the appended drawings. Understanding that these drawings depict only  
25 typical embodiments of the invention and are not therefore to be considered to be limiting of its scope, the invention in its presently understood best mode for making and using the same will be described and explained with additional specificity and detail through the use of the accompanying drawings in which:

Figure 1 is a cross section view of a bright metal flake in accordance with the  
30 present invention; and

Figure 2 is a graphic representation of the total reflectance of an aluminum pigment prepared with a plurality of bright metal flakes of the present invention.

### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

The present invention is directed to thin flakes having improved specular  
35 reflectance characteristics in the wavelength range of about 300 to about 800 nm suitable

for use as pigments in various applications. It is a feature of the present invention to support a thin reflector layer, typically metal, with dielectric coatings disposed on the two opposing planar surfaces thereof. The dielectric layers provide rigid support and brittle fracture characteristics for the metal layer. The uses are well known and include, but are not limited to, coating compositions, inks, extrusions, paints, electrostatic coatings, infrared interference pigments, glass, ceramics and cosmetics.

A preferred embodiment of a flake in accordance with the present invention is depicted schematically in Figure 1. Flake 20 is shown as being a three-layered structure having an inner reflector layer 22 and two outer dielectric layers 24 and 26. Although not illustrated, it should be understood that other layers could be included as long as the functionality of the structure of Figure 1 remains.

The materials of reflector layer 22 is selected so as to have reflective characteristics suitable for the intended use of the flake. The presently preferred reflector material is aluminum because aluminum has good reflectance characteristics while remaining inexpensive and easy to form into a thin layer. It will be appreciated in view of the teachings herein, however, that other reflective materials may be used in place of aluminum. For example, silver, gold, platinum and palladium, or alloys of these or other metals might be used in place of aluminum. Other useful materials include, but are not limited to, metal carbides, oxides, nitrides and sulfides, or mixtures of metals and one or more of these materials. For purposes of convenience and brevity, the flake of the present invention shall sometimes be referred as a "bright metal flake." This term is not meant to exclude reflector layers formed of a material other than a pure metal.

Situated on the opposing surfaces of reflector layer 22 are two dielectric layers 24 and 26. The presently preferred dielectrics are inorganic, because inorganic dielectric materials have been found to have good characteristics of brittleness and rigidity. Most preferred are dielectric layers of either silicon dioxide ( $\text{SiO}_2$ ) or magnesium fluoride ( $\text{MgF}_2$ ) because these materials are readily available and easily applied. It will be appreciated from the teachings herein, however, that any dielectric providing the functionality described would be an acceptable substitute for one of these materials. Such acceptable substitutes include, but are not limited to,  $\text{Al}_2\text{O}_3$ ,  $\text{TiO}_2$ ,  $\text{ZnS}$  or any dielectrics having an index of refraction below about 1.65. Advantageously, these materials will also protect against the adverse affects of solvents during applicational use thereof.

The thickness of an aluminum reflector layer is in a preferred range of about 40 - 150 nm with the limits of 40 and 150 nm being merely representative and not restrictive. For example, the lower limit of 40 nm is selected so that the aluminum layer is other than

a transparent layer. Other reflector materials may justify higher or lower minimum thicknesses in order to obtain a non-transparent thickness. The upper limit of 150 nm, in this embodiment, is selected primarily to obtain a high aspect ratio in the final flake. Even greater thicknesses could be sustained for some purposes.

5           In a more preferred range, the thickness of an aluminum reflector layer is from about 80 to about 150 nm with a most preferred thickness of about 100 nm. With respect to the lower limit, 80 nm is selected to obtain a substantially opaque thickness of aluminum for the purposes of facilitating reflectance. Other reflector materials may justify higher or lower minimum thicknesses in order to obtain opaque thicknesses. The upper limit of 150 nm in is still selected to maintain a high aspect ratio in the final flake. It is contemplated that the reflector thickness selected, however, will be flimsy if unsupported, having adverse properties such as curling, wrinkling and malleability of the layer.

10           The preferred thickness of each of the dielectric layers is between about 50 - 200 nm, with a most preferred thickness of about 100 nm. Similar to the thickness of the metal layer, the preferred range of coating thicknesses is subject to variation based upon the actual metal layer and the dielectric coating chosen. With an aluminum layer and either silicon dioxide or magnesium fluoride, the lower thickness limit of about 50 nm is selected based upon the strength of the coating beyond which, when smaller, will not be strong enough to maintain structural integrity, hence flake rigidity, under the stress and strain imposed by a flexing aluminum layer. The upper limit of about 200 nm is selected based upon the observation that color interference between dielectric layers commences at thicknesses beyond 200 nm. In situations where color interference is useful, thicker dielectric layers may be used.

25           From the foregoing, it will be appreciated that the presently preferred flake in accordance with the present invention is only about 300 nm thick: 100 nm for each of the two dielectric layers and another 100 nm for the reflector layer. Despite this exceedingly small flake thickness, it has been surprisingly discovered that a flake having this three-layered structure has sufficient rigidity for use as a highly reflective pigment, primarily because of the inherent uniaxial strengths of the dielectrics. For example, a dielectric coating of silicon dioxide, which has a uniaxial compressive strength of about 30           160,000 (lb/in<sup>2</sup>) and a uniaxial tensile strength of about 7000 (lb/in<sup>2</sup>), prevents the internal reflector layer from flexing, bowing or otherwise deforming.

Aluminum has a tensile strength approximately equal to its compressive strength. Yet when the aluminum layer of the present invention is layered with the dielectrics of the present invention, the uniaxial compressive strength (lb/in<sup>2</sup>) of the flake is surprisingly about 8 times greater than the uniaxial tensile strength (lb/in<sup>2</sup>). This surprising result is empirically supported under the well known theory of brittle fracture known as *Griffith's* theory. For a further discussion of *Griffith's* theory, see e.g., William D. Callister, Jr., *Materials Science and Engineering* (John Wiley & Sons, Inc., 2d ed., 1991). This substantial difference in strengths beneficially transforms an aluminum layer tending to fracture by ductility into a layer tending to fracture by brittleness. Brittle fracture characteristics then advantageously serve to facilitate the separation of a plurality of metal flakes from a larger metal film during the manufacturing processes and to facilitate the fracture of individual metal flakes as stresses are imposed during applicational use.

In a preferred brittleness range, the metal layer is satisfactorily strengthened by the dielectric coatings when the uniaxial compressive strength of the metal flake is about 6 times or higher than the uniaxial tensile strength. Although the metal flake can be strengthened in amounts less than about 6, the metal flake then tends to exhibit adverse fracture characteristics similar to those of ductile fractures. It should be appreciated that although the foregoing rigidity and brittleness was achieved in a two-sided coating, it is even contemplated that the layer can also be coated upon a singular side of the reflector and still achieve favorable properties during manufacturing and use. The singular layer coating, however, must be stress balanced by means well known to those skilled in the art to prevent curling of the flake. The two sided coating, however, is the preferred embodiment to satisfactorily improve flake rigidity and to facilitate protection against detrimental attacks, such as corrosion, upon the metal layer. Corrosion is even contemplated within the scope of this invention as being more substantially fended off by further providing an encapsulating material about any exposed regions of the metal flake or the entire three-layered structure itself. Such encapsulating materials are well known in the art and are not described herein in detail.

The flakes of the present invention are not of a uniform shape. Nevertheless, for purposes of brevity, the flakes will be referred to as having a "diameter." It is presently preferred that the diameter of the flakes be in a preferred range of about 1 - 50 microns with a more preferred range of about 5 - 25 microns. Thus, the aspect ratio of the flakes of the present invention is in a preferred range of about 6.5 - 625 with a more preferred range of about 50 - 250. As is well known, the greater the aspect ratio, the flatter the

flakes will lie, hence increasing reflectance. Thus, since many prior art flakes have an optimal aspect ratio of only about 15, it should be apparent that the aspect ratio of the present invention will inherently yield substantially improved reflectance properties.

5 In general, the presently preferred process for manufacture of the metal flakes involves the providing of a flexible web onto which one of the outer dielectric layers is deposited, followed by deposition of the reflector layer and the remaining dielectric layer.

10 Since the deposition of the dielectric layers is preferably accomplished by vapor deposition means, it is observed that the dielectric layer will crack under the stresses imposed as the dielectric transitions from the vapor into the solid phase. Next, the reflector layer is deposited, taking on the characteristics of the underlying cracked dielectric layer. Finally, the other outer dielectric layer is deposited on the metal film. In this manner, the two dielectric layers bolster an extremely flimsy and thin metal film into a rigid metal film possessing characteristics tending to fracture the metal along the cracks of the dielectric layer in a brittle, instead of ductile manner. Thereafter, as the flexible web is removed, either by dissolution in a preselected liquid or by means of release agents, both well known in the art, a plurality of metal flakes are fractured out according to the cracks of the dielectric layer. This type of manufacturing technique is similar to that more fully described in U.S. Patent No. 5,135,812, issued on August 4, 1992 to Phillips et al., entitled "Optically Variable Thin Film Flake and Collection of the Same," expressly incorporated herein by reference.

20 Once manufactured, however, each individual metal flake possesses exceptional planar characteristics which allow each flake to be extremely flat even in the vicinity of the fracture. In applicational use with plurality of similar metal flakes, not only can the flakes advantageously overlap and lie in a substantially planar orientation but each individual metal flake still possesses characteristics tending to fracture the flake by brittleness. Thus, when an application in which the flakes are used imposes additional stresses upon the flake, such as paint drying, the flake will fracture due to brittle fracture characteristics rather than bend due to ductile fracture characteristics. Such rigidity and fracture characteristics of a metal flake are heretofore unknown.

30

### Example 1

Several samples of applicational pigments were prepared, each sample containing a plurality of bright metal flakes in accordance with the present invention. The metal layers each had a thickness of about 100 nm of aluminum and ranged in diameter from about 1 to about 50 microns. Layered upon the opposing planar surfaces of the metal layer was an inorganic dielectric coating having a thickness in a range of about 100 to

35

about 200 nm. The inorganic dielectric coatings were selected as either silicon dioxide or magnesium fluoride. With reference to Figure 2, the actual reflectance results obtained as a function of various wavelengths were observed and recorded as reflectance curve 40. The reflectance curve has also been empirically supported by the results obtained from various other measurement devices (not shown) such as ink drawdowns, paint sprayouts and microscopy.

Super-imposed with the reflectance curve 40 on the reflectance-wavelength graph in Figure 2 is the actual reflectance curve for a pure aluminum flake 42 and the predicted reflectance curves of an aluminum flake coated with either magnesium fluoride 44 or silicon dioxide 46. The predicted curves are modeled upon the reflectance of aluminum as a function of various wavelengths for a singular aluminum flake, vice a plurality of flakes. What should be appreciated is that although the actual observed results, reflectance curve 40, are below the predicted reflectance values below wavelengths of about 680 nm, the actual observed reflectance is above the predicted reflectance response for silicon dioxide above about 680 nm. While it might appear that the theoretical or predicted responses have been incorrectly modeled, it has been surprisingly discovered that the better-than-modeled reflectance response is a twofold advantageous result of the bright metal flakes of the present invention. Not only are the bright metal flakes of the present invention so rigid to exhibit favorable planar characteristics individually, but the bright metal flakes, as a plurality of flakes, are so thin that the flakes are believed capable of overlapping each other and still lie collectively in a substantially singular plane. This planar orientation is believed to provide for an enlarged surface area being exposed to incident light which enables a larger reflectance to be exhibited therefrom. The plurality of metal flakes are surprisingly allowed to overlap without radically changing the planar structure of the individual flakes which is quite unlike the flakes of the prior art.

It should also be appreciated from Figure 2 that the measured reflectance results for all known prior art aluminum based pigments fall below 75%. Thus, the bright metal flakes of the present invention achieve, at a minimum, a 5% improvement in total reflectance and a 10% improvement in specular reflectance over the prior art. Not only are these results significantly better than the prior art but these rigid metal flakes have achieved results better than what has heretofore ever been predicted.

The present invention may be embodied in still other specific forms without departing from its spirit or essential characteristics. The described embodiments are to be considered in all respects only as illustrative and not restrictive. The scope of the invention is, therefore, indicated by the appended claims rather than by the foregoing

description. All changes which come within the meaning and range of equivalency of the claims are to be embraced within their scope.

What is claimed is:

1. A flake having substantial rigidity so as to provide high reflectance during use as a pigment, said flake comprising:

a reflector layer having a thickness in a range of about 40 to about 150 nm; and  
a dielectric layer on each side of said reflector layer, each said dielectric layer  
5 having a thickness in a range of about 50 to about 200 nm.

2. A flake as defined in claim 1, wherein said dielectric layers are formed of an inorganic material and the reflector layer includes a metal.

3. A flake as defined in claim 1, wherein said reflector layer has an aspect ratio greater than 6.5.

10 4. A flake as defined in claim 1, wherein said reflector layer is formed of aluminum, silver, gold, platinum and palladium, or mixtures thereof.

5. A flake as defined in claim 1, wherein said dielectric layers are formed of a dielectric having an index of refraction below about 1.65.

15 6. A flake as defined in claim 1, wherein said dielectric layers are formed of magnesium fluoride, silicon dioxide, alumina or titanium dioxide.

7. A flake as defined in claim 4, wherein the thickness of said reflector layer is about 100 nm.

8. A flake as defined in claim 1, wherein said thickness of each said dielectric layer is about 100 nm.

20 9. A flake as defined in claim 4, further comprising an encapsulant substantially formed over any exposed regions of said reflector layer.

10. A flake as defined in claim 1, wherein a uniaxial compressive strength is at least about 6 times a uniaxial tensile strength of said flake.

25 11. A thin metal flake having improved planar and reflectance properties, comprising:

an aluminum layer having a thickness of about 100 nm; and  
two inorganic dielectric layers, each said inorganic dielectric layer being disposed on an opposing surface of said aluminum layer and having a thickness of about 100 nm;  
a compressive strength of said flake being about 8 times greater than a tensile strength thereof in order to facilitate brittle fracture thereof.

30 12. A thin metal flake according to claim 11, wherein said aspect ratio is in a range of about 6.25 to about 625.

13. A thin metal flake according to claim 11, wherein said aspect ratio is in a preferred range of about 50 to about 250.

14. A thin metal flake according to claim 11, wherein said two inorganic dielectric layers are selected from dielectrics having an index of refraction below about 1.65.

5 15. A thin metal flake according to claim 11, wherein said two inorganic dielectric layers are selected from the group consisting of magnesium fluoride, silicon dioxide alumina and titanium dioxide.

16. A thin metal flake according to claim 11, further comprising an encapsulant substantially about any exposed regions of said aluminum layer for facilitating protection of said aluminum layer from corrosion.

10 17. A thin film having a uniaxial compressive strength and a uniaxial tensile strength for manufacturing a plurality of thin metal flakes having improved planar and reflective properties therefrom, comprising:

a reflector film having a thickness in a range of about 40 to about 150 nm; and

15 a dielectric layer on each side of said reflector film to cause said uniaxial compressive strength to be at least about 6 times greater than said tensile strength, each said dielectric layer having a thickness in a range of about 50 to about 200 nm.

18. A thin film according to claim 17, wherein said reflector film is one of aluminum, silver, gold, platinum and palladium.

20 19. A thin film according to claim 17, wherein said reflector film is one of a metal carbide, a metal oxide, a metal nitride and a metal sulfide.

20. A thin film according to claim 17, wherein said dielectric layers are dielectrics having an index of refraction below about 1.65.

21. A thin film according to claim 17, wherein said dielectric layers one of magnesium fluoride, silicon dioxide, alumina and titanium dioxide.

25 22. A thin film according to claim 17, wherein said thickness of said reflector is about 100 nm.

23. A thin film according to claim 17, wherein said thickness of each said dielectric layer is about 100 nm.

30 24. A thin film according to claim 15, wherein said uniaxial compressive strength is about 8 times said uniaxial tensile strength.

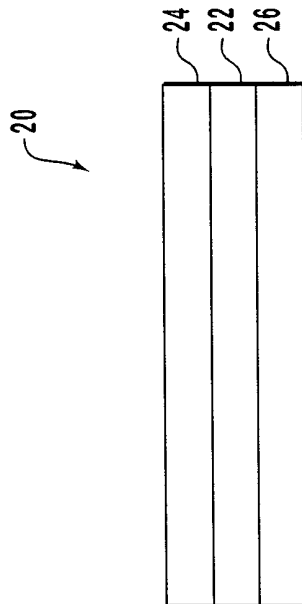


FIG. 1

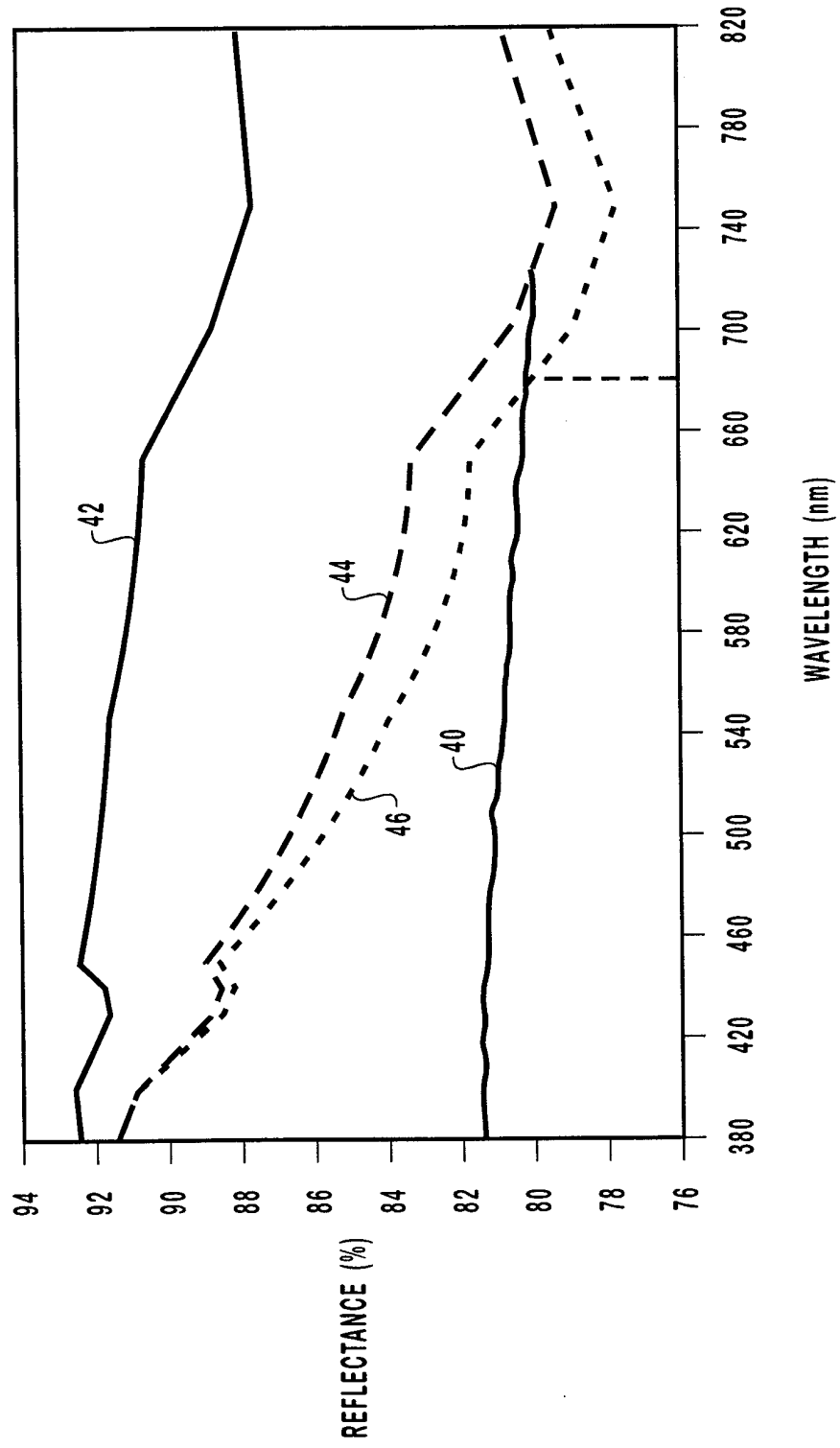


FIG. 2

INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US98/23761

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) : C09C 1/62; C04B 14/20  
US CL : Please See Extra Sheet.

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)

U.S. : Please See Extra Sheet.

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

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 5,135,812 A (PHILLIPS et al.) 04 August 1992, col. 7, line 65- col. 8, line 7; col. 9, lines 25-60; col. 16, lines 8-14; col. 20, line 10.	1-24
Y,P	US 5,763,086 A (SCHMID et al.) 09 June 1998, col. 3, lines 57, 66, 67; col. 4, lines 8-13, 40-42;	1-24
Y,P	US 5,766,334 A (HASHIZUME et al.) 16 June 1998, col. 3, lines 31, 45, 46; col. 4, lines 45-48; col. 5, lines 32, 33; col. 6, lines 6, 18, 57-63; col. 7, lines 10-11.	1-24
Y,P	US 5,766,335 A (BUJARD et al.) 16 June 1998, col. 3, lines 6, 7; col. 4, lines 54-56; col. 5, lines 23-26; col. 7, lines 62, 63; col. 11, examples 5-11;	1-24

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

* Special categories of cited documents:	*T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
*A* document defining the general state of the art which is not considered to be of particular relevance	*X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
*E* earlier document published on or after the international filing date	*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
*L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	*A* document member of the same patent family
*O* document referring to an oral disclosure, use, exhibition or other means	
*P* document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search

09 FEBRUARY 1999

Date of mailing of the international search report

17 FEB 1999

Name and mailing address of the ISA/US  
Commissioner of Patents and Trademarks  
Box PCT  
Washington, D.C. 20231

Facsimile No. (703) 305-3230

Authorized officer

SCOTT L. HERTZOG

Telephone No. (703) 308-3456

INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US98/23761

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1.  Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
  
2.  Claims Nos.:  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
  
3.  Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

Please See Extra Sheet.

1.  As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2.  As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3.  As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
  
4.  No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- The additional search fees were accompanied by the applicant's protest.  
 No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US98/23761

A. CLASSIFICATION OF SUBJECT MATTER:

US CL :

106/403, 404, 415, 437, 461, 482, 489, 286.5; 428/328, 329, 331, 332, 403, 404, 426, 432, 433, 434, 446, 448, 450, 457, 469, 470, 471, 472

B. FIELDS SEARCHED

Minimum documentation searched

Classification System: U.S.

106/403, 404, 415, 437, 461, 482, 489, 286.5; 428/328, 329, 331, 332, 403, 404, 426, 432, 433, 434, 446, 448, 450, 457, 469, 470, 471, 472

BOX II. OBSERVATIONS WHERE UNITY OF INVENTION WAS LACKING

This ISA found multiple inventions as follows:

This application contains the following inventions or groups of inventions which are not so linked as to form a single inventive concept under PCT Rule 13.1. In order for all inventions to be searched, the appropriate additional search fees must be paid.

Group I, claim(s) 1-16, drawn to a thin metal flake.

Group II, claim(s) 17-24, drawn to a thin film.

The inventions listed as Groups I and II do not relate to a single inventive concept under PCT Rule 13.1 because, under PCT Rule 13.2, they lack the same or corresponding special technical features for the following reasons: the only similar feature is the thickness of the reflective and dielectric films, which feature is not special to the claims, but is suggested by the prior art, and therefore not a special technical feature.