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(54) Title: PEARLESCENT COLOR PIGMENTS

(57) Abstract: Described are a series of pearlescent color pigments and methods of making such pearlescent color pigments that derive color from both light interference and light absorption. In one embodiment, the pearlescent color pigment includes an inorganic substrate having a metal oxide coating, an organic polymer coated on the metal oxide coating, and a colorant, wherein the colorant is dispersed or dissolved in the organic polymer. The pearlescent color pigments may be formed by first providing a suspension of metal oxide coated inorganic substrate in water; adding a thermosetting organic polymer to the suspension; adding a colorant to the suspension, wherein the colorant dissolves or disperses in the thermosetting organic polymer; and sufficiently heating the suspension to cause the thermosetting organic polymer to coat the metal oxide coated inorganic substrate to provide the pearlescent color pigment, hi accordance with this method, the colorant remains dissolved or dispersed in the thermosetting organic polymer when the metal oxide coated inorganic substrate is coated with the thermosetting organic polymer.

PEARLESCENT COLOR PIGMENTS

CROSS-REFERENCE TO RELATED APPLICATION

[0001] This application claims priority to and any other benefits of U.S. Provisional Application No. 60/952,410 (Attorney Docket No 06821/08146) filed July 27, 2007, entitled PEARL COLOR PIGMENTS, which is expressly incorporated by reference herein.

TECHNICAL FIELD

[0002] The present invention relates generally to colored lustrous pigments and more particularly to a new series of pearlescent color pigments that derive color from both light interference and light absorption and to methods of making and using the same.

BACKGROUND ART

[0003] Colored lustrous pigments are available in which the lustrous pigment part is a natural pearlescent material or synthetic pearlescent substance (also called a nacreous pigment) and the colored part or dyestuff includes a wide variety of inorganic and organic coloring agents. The processes that have been used to make colored lustrous pigments, however, have many difficulties, among which the following are noteworthy: severe bleeding out of the color upon filtration of the coated product from suspension; poor adherence of dyestuff to the surface of the pigment such that the color could be washed off with water; and difficulty of retaining luster with increased color intensity. Furthermore, these processes may be restricted to the use of dyes that are typically water soluble materials, which severely limit the coloration selection.

[0004] In one attempt to overcome these problems, colored lustrous pigments may be prepared by chemically binding the dyestuff on the surface of the lustrous pigment with the help of a laking reagent, such as aluminum chloride. On hydrolysis, this produces layers of aluminum hydroxide with which the dye reacts to form the insoluble color lake, thereby permitting the dye to deposit on the surface of the lustrous pigment. Although this approach particularly addresses the problem of poor adherence of the organic dye on the surface of the

lustrous pigment, the problem of heavy bleeding of any uncoated dye remains severe. The failure to control the heavy bleeding has been a serious obstacle to developing a suitable process of preparing colored lustrous pigments of superior quality.

[0005] Another attempt at producing colored lustrous pigments employed a two-step process in which aluminum hydroxide, for example, is first deposited on the plate-like surface of the lustrous pigment and, thereafter, the aluminum hydroxide coated lustrous pigment, with or without isolation from the suspension in which it is formed and re-suspended, is reacted with a dye to form the insoluble color lake. This process depends on how well the dye binds to the surface of the lustrous pigment that has been modified by previous precipitation of aluminum hydroxide. The binding of the dye to form the color lake by this method may become increasingly inefficient because active binding sites on the lustrous pigment are lost as a result of the firmly adhering aluminum hydroxide layers already bound to the surface of the lustrous pigment.

[0006] By following the reverse procedure, it is possible to improve not only the efficiency of the laking process but also the quality of the product. When an aqueous solution of the dye is combined with the suspension of lustrous pigment particles and the lustrous pigment particles are allowed to settle out of the suspension, it has been found that the lustrous pigment particles carry the dye molecules with them by physical adsorption so that much of the dye originally in solution is weakly adsorbed onto the lustrous pigment particles. At this point in time the dye can be easily washed off the lustrous pigment surface with water. The laking reagent (*e.g.* a hydrolyzable aluminum compound) is added and the aluminum hydroxide, which precipitates on hydrolysis, reacts with the already adsorbed dye to form a firmly adhering color lake on the surface of the lustrous pigment.

[0007] Even with such advances, however, these laked, colored lustrous pigments are inherently unstable. The laking process, in which a dye in water solution is chemically precipitated onto a plate-like surface of the lustrous pigment, produces colored lustrous pigments with several key defects. One of the most serious defects of the laked, colored lustrous pigments is their tendency to bleed or become soluble in the dispersing media. This is particularly the case with aqueous solutions. Because these processes require water soluble dyes, which as a class are inherently unstable to light or heat, the laked, colored lustrous pigments suffer the same fate. Thus, there remains a need for improved colored lustrous pigments and methods of making such pigments.

DISCLOSURE

[0008] Pearlescent color pigments and methods of making such pigments are provided. In one embodiment, the pearlescent color pigment includes an inorganic substrate having a metal oxide coating, an organic polymer coated over the metal oxide coating, and a colorant, wherein the colorant is dispersed or dissolved in the organic polymer.

[0009] The pearlescent color pigments may be formed by providing a suspension of metal oxide-coated inorganic substrate in water; adding an organic polymer to the suspension; adding a colorant to the suspension, wherein the colorant dissolves or disperses in the organic polymer; and coating the organic polymer on the metal-oxide coated inorganic substrate to provide the pearlescent pigment. In accordance with this method, the colorant remains dissolved, in the case of a dye, or dispersed, in the case of a pigment, in the organic polymer when the metal oxide-coated inorganic substrate is coated with the organic polymer.

[0010] Provided herein is a new series of pearlescent color pigments and methods of making such pigments. As used herein "pearlescent color pigment" means a compound that contains both a pearlescent component and a colorant, in contrast to the typical and more commonly known organic and inorganic pigments. The pearlescent component is a plate-like particle containing a metal oxide coating that produces the interference color and this is seen as a colored highlight. The added colorant is any variety of organic dye or dyes and/or organic and inorganic pigment or pigments that impart its own unique second color by light absorption and this provides the background color. The pearlescent color pigments derive color from both light interference and light absorption mechanisms. The combination of these two-color-producing mechanisms results in a unique double color.

[0011] The pearlescent color pigments described herein are colored, lustrous pigments of superior quality in that they possess strong color intensity, high luster, soft powder texture and superior light stability, bleed resistance and thermal stability. These products can be produced with a variety of dyes and pigments and are not limited to water soluble dyes that can be laked, a fact that allows the usage of high quality organic pigments that possess superior physical properties. Usage of high quality organic pigments extends the field of usage of the colored, lustrous pigments into the fields of plastics and decorative paints and coatings.

[0012] The new pearlescent color pigments described herein are formed by bonding a colorant (an organic or inorganic coloring agent) to metal oxide coated inorganic substrate using an organic polymer. The colorant is dispersed in or dissolved in the organic polymer, and the polymer is then coated on the metal oxide coated inorganic substrate. In some embodiments, the methods comprise the steps of providing a suspension of metal oxide coated inorganic substrate in water; adding a thermosetting organic polymer to the suspension; adding a colorant to the suspension, wherein the colorant dissolves or disperses in the thermosetting organic polymer; and sufficiently heating the suspension to cause the thermosetting organic polymer to coat the metal oxide-coated inorganic substrate to thereby provide the pearlescent color pigment. The colorant remains dissolved or dispersed in the thermosetting organic polymer when the metal oxide-coated inorganic substrate is coated with the thermosetting organic polymer.

[0013] For example, this process may be accomplished *via* an aqueous process that is environmentally advantageous. In accordance with methods described herein, the metal oxide-coated inorganic substrates are suspended in water. If an optional surface enhancing agent is being used, the pH of this suspension is preferably adjusted to 3-4 by the use of acetic acid before the optional surface enhancement agent is added. The mixture is warmed to 60-70° C and maintained at that temperature for a period of 30-120 minutes. The pH is then adjusted to neutral by means of a 20% NaOH solution. To the pH neutral suspension the thermosetting organic polymer, which may be in the form of a dispersion, is added followed by the pigment concentrate or the organic dye. Finally, a hardener, or cross-linking agent, is added and the temperature of the entire mixture is raised to 95° C and maintained at that temperature for 30 minutes. During the progression of the organic polymer reaction, the polymer dispersion becomes unstable and precipitates onto the surface of the metal oxide-coated inorganic substrate. The crosslinking reaction is then completed on the surface of the substrate. The pearlescent color pigment is filtered, dried and cured to give the final bleed-free highly colored product.

[0014] The inorganic substrates used to manufacture the pearlescent color pigments described herein are metal oxide-coated inorganic substrates. Such substrates may include natural micas; *i.e.*, micas mined from the earth and washed, screened, etc., or synthetically produced aluminum oxide or silicon dioxide, or similar. In an exemplary embodiment, the metal oxide coating comprises titanium dioxide. In other exemplary embodiments, metal

oxides other than titanium dioxide may also be used to prepare similar types of pearlescent color pigments, by coating such other metal oxides on the inorganic substrate. Some examples of such other metal oxides include ferric oxide, zirconium dioxide, tin dioxide, and the like.

[0015] The metal oxide coating on the inorganic substrate has a high refractive index and provides the optical effects, including high luster or reflectivity, coverage, interference reflection color when the metal oxide coating is sufficiently thick, and absorption color when the metal oxide contains color material. The inorganic substrate, on the other hand, has a low refractive index and functions as a carrier or substrate, in addition to contributing to some extent towards the optical effects of the pearlescent color pigment.

[0016] In an exemplary embodiment, the pearlescent color pigments are made with titanium dioxide coated mica, which is composed of a mica platelet having a crystalline titanium dioxide coating adhered thereon. The interference color exhibited is a function of the thickness of the titanium dioxide coating. The exemplary pearlescent color pigment has good reflectivity characteristics, high stability to both heat and chemical agents, and it is non-toxic; making it suitable for cosmetic application. The titanium dioxide may be in the anatase form as described, *e.g.*, in U.S. Pat. Nos. 3,087,827; 3,087,828; 3,418,146; and, 3,437,515, or in the rutile crystalline form as described in U.S. Pat. No. 4,038,099, each of which is incorporated herein by reference.

[0017] In some embodiments, commercially available lustrous (*i.e.*: nacreous) pigments may be used. Such commercially available nacreous pigments include, but are not limited to Lumina Pigments from BASF, the Iriodin series of products available from EMD Chemicals, the Colorstream series of products available from EMD Chemicals, the 200 Rutile series of pigments available from Zhejiang Huake Fine Chemical Co., Ltd., and the like.

[0018] An organic polymer, in which the colorant is dissolved or dispersed, is used to coat the metal oxide-coated inorganic substrate to form the pearlescent color pigments. In an exemplary embodiment, the organic polymer is a thermosetting organic polymer. Although the examples and specific embodiments described herein employ a thermosetting polymer, it should be pointed out that any suitable organic polymer may be used. Some exemplary thermosetting organic polymers include epoxy resins, melamine, urea formaldehyde, and so forth. In an exemplary embodiment, the thermosetting organic polymer is an epoxy resin.

Epoxy resins suitable for the invention compositions have molecular weights generally within the range of about 200 to about 6000.

[0019] In an exemplary embodiment, the molecular weight of the epoxy resin is in the range from about 350 to about 2000. One indication of the specific grade of epoxy resin utilized is the epoxy equivalent weight. In accordance with the methods described herein, many different grades of epoxy resins may be used. In some embodiments, the epoxy resin has an epoxy equivalent weight in the range from about 175 to about 3800. In other embodiments, the epoxy resin has an epoxy equivalent weight in the range from about 175 to about 525. In still other embodiments, the epoxy resin has an epoxy equivalent weight in the range from about 175 to about 197.

[0020] In some embodiments, a commercially available epoxy resin may be used. In one embodiment, EPON 828 from Hexion Specialty Chemicals, a reaction product of epichlorohydrin and bisphenol A having a molecular weight of about 828, and an epoxide equivalent (ASTM D-1652) of about 185 to 192 is used. In this embodiment, EPON 828 is utilized in the form of a water dispersion that is prepared by emulsifying the epoxy resin in water with the aid of typical nonionic surfactants. In an alternate embodiment, EPI-REZ Resin 3510-W-60 available from Hexion Specialty Chemicals, which is a nonionic, aqueous dispersion of a bisphenol A epoxy resin similar to EPON 828, with an epoxide equivalent weight of 187, is used. This product is supplied at 60% resin solids and contains no organic solvents.

[0021] The epoxide group on the ends of these molecules serves as the reactive site for crosslinking in these thermoset polymers. The chemical chosen to react with these epoxides is referred to as the curing agent, and it typically has active hydrogen attached to nitrogen, oxygen, or sulfur. Amine curing agents are the most common and can be primary or secondary, aliphatic, aromatic, or cycloaliphatic. The amines typically have greater than three reactive sites per molecule that facilitate the formation of a three-dimensional polymer network when mixed with the epoxy resin. In some embodiments, the curing agents are aliphatic and cycloaliphatic amines. In one specific embodiment, the aliphatic amine diethylenetriamine is used. In another specific embodiment, amidoamine is used as the curing agent.

[0022] In some embodiments described herein, it is advantageous to improve the surface receptivity of the inorganic substrate with the use of a surface enhancement agent. In these embodiments, the surface treatment effectively allows for a greater amount of the epoxy polymer to be deposited on the surface of the inorganic substrate, which concurrently deposits a greater depth of color. In an exemplary embodiment, the surface enhancement agent is an organosilane. The basic structure of an organosilane is $R_nSi(OR)_{4-n}$, with "R" being an alkyl, aryl, or organofunctional group and "OR" being a methoxy, ethoxy, or acetoxy group. In one embodiment, the organosilane used is an epoxy functional material sold as Z-6040 available from Dow Corning.

[0023] In other embodiments, however, no surface enhancement agent is used. It has been discovered that, in some embodiments, no surface enhancement agent is needed to provide sufficient epoxy coverage, and thus sufficient colorant coverage. The ability to produce high quality pearlescent color pigments without the use of a surface enhancement agent represents a significant cost savings in the production of these pearlescent color pigments.

[0024] Colorants useful in the processes described herein include a wide variety of organic dyes and organic and inorganic pigments, and it will be understood that any suitable colorants may be used. Organic and inorganic pigments may be utilized in the methods described herein in a pre-dispersed format commonly referred to as a colorant dispersion or concentrate. In embodiments using colorant concentrates, the concentrate should be free of additives or other polymers that will interfere with the formation of, or degrade the physical properties of the final thermosetting epoxy polymer.

[0025] There is no limitation of the types of colorants that may be used, with materials as diverse as carbon black, azo salts, and phthalocyanines having been used successfully in various embodiments. In other embodiments, a wide variety of organic dyes may be used in accordance with the processes described herein. In embodiments utilizing organic dyes, the dye used should have adequate solubility in the thermosetting organic polymer to impart desirable coloration.

[0026] Unlike other methods for preparing colored, luminous pigments, when the processes described herein are used, the dyes need not be water-soluble and are not required to form complex salts with laking reagents. Additionally, the dyes used are not limited to reactive

dyes, but are merely dyes that are soluble in the thermosetting organic polymer. A non-exclusive list of pigments and dyes useful in the methods described herein is listed in Table 1.

Table 1 Non-exclusive list of pigments and dyes useful in the methods described herein

<u>Pigment Trade Name</u>	<u>Color Index No.</u>	<u>Vendor Name</u>
PERMANENT ORANGE G	PO 13	JECO
12-1007 PERMANENT ORANGE G	PO 13	CLARIANT
13-3042 COLANYL RED FGRG-100	PR112	CLARIANT
16-2034 HOSTAPERM GREEN GNX	PG 7	CLARIANT
15-1042 HOSTAPERM BLUE BT-617-D	PB 15:4	CLARIANT
13-7012 HOSTAPERM RED E5B-02	PV19	CLARIANT
13-7016 HOST. RED EG TRANSPARENT	PR 209	CLARIANT
QUINDO RED R-6700	PV 19	SUN
SUNBRITE RED 3 216-3300	PR3	SUN
11-3012 NOVOPERM YEL	PY 97	CLARIANT
HOSTAPERM PINK E 13-7000	PR 122	CLARIANT
14-4007 PV FAST VIOL	PV 23	CLARIANT
15-1023 HOSTAPERM BLUE BT-627-D	PB 15:2	CLARIANT
11-2210 COLANYL YELLOW 5GX-100 (82%)	PY74	CLARIANT
12-5005 HANSA RED GG-A	PO 5	CLARIANT
13-3061 NOVOPERM RED	PR 170	CLARIANT
TRICOLITH YELLOW IYP	PY 1	TRICHROMATIC
13-7011 HOST. RED VIOLET ER-02	PV19	CLARIANT
HANSA YELLOW G	PY 1	JECO
HELIOGEN BLUE L 6875	PB 15:2	DOWD & GUILD
BRILLIANT RED 2R 30AC0186	N/A	LIPSCOMB
11-3015 F2G-A YELLOW	PY 194	CLARIANT
MONOLITE GREEN GLX	PG 7	HEUCOTECH
RED B NRT-742-D	PV 19	CIBA
11-1418 NOVOPERM YELLOW HR-70	PY 83	CLARIANT
248-0156 FASTOGEN BL	PB15:6	SUN
ULTRAMARINE BLUE E-37	PB29	TAVCO
13-2100 HANSA SCARLE	PR 3	CLARIANT
234-6485 PERM RED 2B	PR 48:4	SUN
SICOTAN YELLOW L 1012	PY 53	DOWD & GUILD
MANOX EASISPERSE 154	PB27	D & J
13-3610 NOVOPERM RED	PR 188	CLARIANT
VCD-4272	PV23	SUN
13-3050 PERMANENT RED FGR	PR112	CLARIANT
11-2527 HANSA BRIL Y	PY 74	CLARIANT
11-2520 HANSA BRIL Y	PY 74	CLARIANT
QHD-6040	PR 22	SUN
11-3030 HOSTAPERM YELLOW H6G	PY 175	CLARIANT
11-1101 PERMANENT YE	PY 14	CLARIANT
2915 DIANISIDINE ORANGE 16	PO 16	ENGELHARD
FAST SCARLET RED BBC	PR 48:1	JECO

YT-805-D DALAMAR M.A	PY 65	CLARIANT
TRICOLITH GREEN GN-1	PG 7	TRICHROMATIC
DALAMAR YELLOW R YT-820-D	PY 65	CLARIANT
13-3070 PERM RED 2R	PR266	CLARIANT
TRICOLITH YELLOW SFY	PY 74	TRICHROMATIC
FAST SCARLET RED BBN	PR 48:1	JECO
11-1400 NOVOPERM YEL	PY 14	CLARIANT
12-3004 NOVOPERM HL-70	PO 36	CLARIANT
ULTRAMARINE BLUE EP-37	PB29	TAVCO
11-2310 HANSA BRILL. YELLOW 4GX	PY73	CLARIANT
UNIPURE REDU LC 3079	PR 57	SENSIENT
UNIPURE BLACK LC902	PB 7	SENSIENT
Dye Trade Name		
FD&C YELLOW 5	ACID YELLOW 23	SENSIENT
D&C RED 28	ACID RED 92	SPECTRA COLORS
D&C RED 22	ACID RED 87	SPECTRA COLORS
MACROLEX VIOLET B	SOLVENT VIOLET 13	LANXESS
MACROLEX YELLOW 3G	SOLVENT YELLOW 93	LANXESS
SAVANNA YELLOW	DISPERSE YELLOW 82	DAYGLO COLOR

EXAMPLES

[0027] **Example 1:** 200 g of HK-625, a red interference pigment available from Wenzhou Huake Pearl Luster Pigment Co., Ltd., was suspended in 50 ml of water by means of mechanical stirring. To this mixture were added 25 g of epoxy dispersion and 60 g of AIT-644, an aqueous Pigment Blue 15:1 dispersion available from DayGlo Color Corp. The mixture was heated to a temperature of 65°C at which time 2.0 g of diethylenetriamine was added to the slurry. The temperature was then raised to 95°C and maintained at that temperature for 45 minutes. The solids were then removed from the suspension *via* vacuum filtration and the wet presscake was transferred to a 130°C oven to completely dry the product and complete the crosslinking reaction. The dried and cured product was then ready for usage.

[0028] **Example 2:** 200 g of Lumina Gold 9Y30D available from BASF was suspended in 500 ml of water by the means of a mechanical stirrer. To this mixture 1.0 g of glacial acetic acid was added followed by 2.5 g of an epoxy functional silane. The temperature of this suspension was raised to 65° C and maintained at 65-70° C for 60 minutes. The pH of this mixture was then adjusted to pH 7.0 by using a 20% aqueous solution of NaOH. 25 g of epoxy dispersion was then added and mixed thoroughly, followed by 60 g of Hostafine Red HF3S an aqueous dispersion of Pigment Red 188 sold by Clariant Pigments and Additives.

After an additional 5 minutes of mixing, 2.0 g of diethylenetriamine was added and the slurry temperature was raised to 95°C and maintained at that temperature for 30 minutes. The solids were then removed from the suspension *via* vacuum filtration and the wet presscake was transferred to a 130°C oven to completely dry the product and complete the crosslinking reaction. The dried and cured product was then ready for usage.

[0029] The product possesses both red and gold hues depending on the viewing angles.

[0030] **Example 3:** Following the same procedure as Example 2, the following materials were utilized:

- 200 g Lumina Blue Red 9B30D from BASF
- 1.0 g Glacial acetic acid
- 2.5 g epoxy functional silane
- 25 g epoxy dispersion
- 0.5 g D&C Red 22 (primarily the disodium salt of 2',4',5',7'-tetrabromofluorescein)
- 2.2 g diethylenetriamine

[0031] The product has both fluorescent red and blue tones that vary with viewing angle.

[0032] **Example 4:** Following the same procedure as Example 2, the following materials were utilized:

- 200 g Iriodin7225 Blue pearl from EMD Chemicals
- 1.0 g Glacial acetic acid
- 2.5 g Epoxy functional silane
- 20 g Epoxy dispersion
- 75 g Pigment Red 264 Dispersion
- 1.85 g diethylenetriamine

[0033] However, the colorant dispersion utilized was prepared as follows. 20 g of E-Sperse321 a polyoxyethylene styrenated phenyl ether available from Ethox Chemicals, LLC

was added to 80 g of water and 2.0 g of DEE-FO 97-3, an emulsifiable mineral oil that was used as a defoamer, available from Ultra Additives. To this mixture 100 g of Casacolor Red 264030, Pigment Red 264 available from Thomas Swan & Co. was added with stirring. The thick paste was then homogenized with a Janke & Kunkel Ultra Turrax T-50 homogenizer for 30 minutes.

[0034] The product has both blue and red hues that are viewing angle dependant.

[0035] **Example 5:** Following the same procedure as Example 2, the following materials were utilized:

100 g T10-02 Colorstream, Artic Fire from EMD Chemicals

0.5 g Glacial acetic acid

1.25 g epoxy functional silane

14.5 g epoxy dispersion

10 g AIT-644 Green Shade Phthalo Blue dispersion from DayGlo Color Corp.

1.0 g norbornanediamine

[0036] The product has multiple shades of violet, blue and green that are viewing angle dependant.

[0037] **Example 6:** Following the same procedure as Example 2, the following materials were utilized:

200 g HK-223 Violet Satin Pearl from Zhejiang Huake Fine Chemical Co., Ltd.

1.0 g Glacial acetic acid

2.5 g epoxy functional silane

25 g epoxy dispersion

100 g AIT-734 Carbon Black Dispersion from DayGlo Color Corp.

1.0 g diethylenetriamine

[0038] The product has violet and black satin hues that are viewing angle dependant.

[0039] **Example 7:** This example was performed the same as Example 4, the following materials were utilized:

200 g Iriodin 7219 Lilac
1.0 g Glacial acetic acid
2.5 g epoxy functional silane
25 g epoxy dispersion
2.2 g diethylenetriamine

[0040] The following materials were utilized for the colorant dispersion:

20 g E-Sperse 321
1.0 g DEE-FO 97-3
50 g Unipure LC-304, D&C Red 6
43 g Water

[0041] The product has violet and red hues that are viewing angle dependant.

[0042] The examples described herein are for illustrative purposes only, and do not limit the scope of the invention, as defined by the claims.

CLAIMS

The invention claimed is:

1. A pearlescent color pigment comprising an inorganic substrate having a metal oxide coating, an organic polymer coated on the metal oxide coating, and a colorant, wherein the colorant is dispersed or dissolved in the organic polymer.
2. The pearlescent color pigment of claim 1 wherein the inorganic substrate is selected from the group consisting of natural micas, synthetic micas, aluminum oxide, silicon dioxide, and combinations thereof.
3. The pearlescent color pigment of claim 1 wherein the metal oxide coating is selected from the group consisting of titanium dioxide, ferric oxide, zirconium dioxide, tin dioxide, and combinations thereof.
4. The pearlescent color pigment of claim 3 wherein the metal oxide coating is titanium dioxide.
5. The pearlescent color pigment of claim 1 wherein the organic polymer comprises a thermosetting organic polymer.
6. The pearlescent color pigment of claim 5 wherein the thermosetting organic polymer is selected from the group consisting of epoxy resins, melamine, urea formaldehyde polymers, and combinations thereof.
7. The pearlescent color pigment of claim 6 wherein the thermosetting organic polymer is an epoxy resin.
8. The pearlescent color pigment of claim 7 wherein the epoxy resin has an average molecular weight in the range from about 200 to about 6000.
9. The pearlescent color pigment of claim 8 wherein there epoxy resin has an average molecular weight in the range from about 350 to about 2000.
10. The pearlescent color pigment of claim 7 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 3800.

11. The pearlescent color pigment of claim 10 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 525.
12. The pearlescent color pigment of claim 11 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 197.
13. The pearlescent color pigment of claim 12 wherein the epoxy resin has an epoxide equivalent in the range from about 185 to about 192.
14. The pearlescent color pigment of claim 1 wherein the colorant comprises a pigment.
15. The pearlescent color pigment of claim 14 wherein the colorant is selected from the group consisting of Permanent Orange G, 12-1007 Permanent Orange G, 13-3042 Colanyl Red FGRG-100, 16-2034 Hostaperm Green GNX, 15-1042 Hostaperm Blue BT-617-D, 13-7012 Hostaperm Red E5B-02, 13-7016 Host. Red EG Transparent, Quindo Red R-6700, Sunbrite Red 3 216-3300, 11-3012 Novoperm Yel, Hostaperm Pink E 13-7000, 14-4007 PV Fast Viol, 15-1023 Hostaperm Blue BT-627-D, 11-2210 Colanyl Yellow 5GX-100 (82%), 12-5005 Hansa Red GG-A, 13-3061 Novoperm Red, Tricolith Yellow IYP, 13-7011 Host. Red Violet ER-02, Hansa Yellow G, Heliogen Blue L 6875, Brilliant Red 2R 30AC0186, 11-3015 F2G-A Yellow, Monolite Green GLX, Red B NRT-742-D, 11-1418 Novoperm Yellow HR-70, 248-0156 Fastogen BL, Ultramarine Blue E-37, 13-2100 Hansa Scarle, 234-6485 Perm Red 2B, Sicotan Yellow L 1012, Manox Easisperse 154, 13-3610 Novoperm Red, VCD-4272, 13-3050 Permanent Red FGR, 11-2527 Hansa Bril Y, 11-2520 Hansa Bril Y, QHD-6040, 11-3030 Hostaperm Yellow H6G, 11-1101 Permanent YE, 2915 Dianisidine Orange 16, Fast Scarlet Red BBC, YT-805-D Dalamar M.A, Tricolith Green GN-1, Dalamar Yellow R YT-820-D, 13-3070 Perm Red 2R, Tricolith Yellow SFY, Fast Scarlet Red BBN, 11-1400 Novoperm Yel, 12-3004 Novoperm HL-70, Ultramarine Blue EP-37, 11-2310 Hansa Brill. Yellow 4GX, Unipure Redu LC 3079, Unipure Black LC902, and combinations thereof.
16. The pearlescent color pigment of claim 1 wherein the colorant comprises a dye.
17. The pearlescent color pigment of claim 16 wherein the dye is selected from the group consisting of FD&C Yellow 5, D&C Red 28, D&C Red 22, Macrolex Violet B, Macrolex Yellow 3G, Savanna Yellow and combinations thereof.

18. A pearlescent color pigment consisting essentially of a titanium dioxide coated inorganic substrate, a thermosetting organic polymer, and a colorant.
19. The pearlescent color pigment of claim 18, wherein the inorganic substrate is selected from the group consisting of natural micas, synthetic micas, aluminum oxide, silicon dioxide, and combinations thereof.
20. The pearlescent pigment of claim 18 wherein the thermosetting organic polymer comprises an epoxy resin.
21. The pearlescent color pigment of claim 20 wherein the epoxy resin has an average molecular weight in the range from about 200 to about 6000.
22. The pearlescent color pigment of claim 21 wherein there epoxy resin has an average molecular weight in the range from about 350 to about 2000.
23. The pearlescent color pigment of claim 20 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 197.
24. The pearlescent color pigment of claim 18 wherein the colorant comprises a pigment.
25. The pearlescent color pigment of claim 18 wherein the colorant comprises a dye.
26. The pearlescent color pigment of any one of claims 1-25 further comprising a surface enhancement agent.
27. The pearlescent color pigment of claim 26 wherein the surface enhancement agent comprises an organosilane.
28. The pearlescent color pigment of any one of claims 1-27 further comprising a curing agent.
29. The pearlescent color pigment of claim 28 wherein the curing agent is an amine curing agent.
30. The pearlescent color pigment of claim 28 wherein the amine curing agent is diethylenetriamine.

31. A method for producing a pearlescent color pigment, the method comprising the steps of

- a. providing a suspension of metal oxide coated inorganic substrate in water;
- b. adding a thermosetting organic polymer to the suspension;
- c. adding a colorant to the suspension, wherein the colorant dissolves or disperses in the organic polymer; and
- d. sufficiently heating the suspension to cause the thermosetting organic polymer to coat the metal oxide coated inorganic substrate to thereby provide the pearlescent color pigment;

wherein the colorant remains dissolved or dispersed in the thermosetting organic polymer when the metal oxide coated inorganic substrate is coated with the thermosetting organic polymer.

32. The method of claim 31 wherein the inorganic substrate is selected from the group consisting of natural micas, synthetic micas, aluminum oxide, silicon dioxide, and combinations thereof.

33. The method of claim 30 wherein the inorganic substrate has a metal oxide coating selected from the group consisting of titanium dioxide, ferric oxide, zirconium dioxide, tin dioxide, and combinations thereof.

34. The method of claim 33 wherein the metal oxide coating is titanium dioxide.

35. The method of claim 34 wherein the thermosetting organic polymer is selected from the group consisting of epoxy resins, melamine, urea formaldehyde polymers, and combinations thereof.

36. The method of claim 35 wherein the thermosetting organic polymer is an epoxy resin.

37. The method of claim 36 wherein the epoxy resin has an average molecular weight in the range from about 200 to about 6000.

38. The method of claim 37 wherein the epoxy resin has an average molecular weight in the range from about 350 to about 2000.

39. The method of claim 36 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 525.
40. The method of claim 39 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 197.
41. The method of claim 31 wherein the colorant comprises a pigment.
42. The method of claim 31 wherein the colorant comprises a dye.
43. The method of claim 31 further comprising the step of adding a surface enhancement agent to the suspension prior to adding the organic polymer to the suspension.
44. The method of claim 43 wherein the surface enhancement agent comprises an organosilane.
45. The method of any one of claims 31-44 further comprising the step of adding a curing agent to the suspension after adding the thermosetting organic polymer to the suspension.
46. The method of claim 45 wherein the curing agent is an amine curing agent.
47. The method of claim 46 wherein the amine curing agent is diethylenetriamine.

AMENDED CLAIMS
received by the International Bureau on
01 December 2008 (01.12.08)

I claim:

1. A pearlescent color pigment comprising:
 - a. a metal oxide coated inorganic substrate; and
 - b. an organic polymer coated on the metal oxide coated inorganic substrate,

wherein a colorant selected from the group consisting of an organic pigment, an inorganic pigment, a nonreactive dye, and combinations thereof, is dispersed or dissolved in the organic polymer.
2. The pearlescent color pigment of claim 1 wherein the inorganic substrate is selected from the group consisting of natural micas, synthetic micas, aluminum oxide, silicon dioxide, and combinations thereof.
3. The pearlescent color pigment of claim 1 wherein the metal oxide is selected from the group consisting of titanium dioxide, ferric oxide, zirconium dioxide, tin dioxide, and combinations thereof.
4. The pearlescent color pigment of claim 3 wherein the metal oxide is titanium dioxide.
5. The pearlescent color pigment of claim 1 wherein the organic polymer comprises a thermosetting organic polymer.
6. The pearlescent color pigment of claim 5 wherein the thermosetting organic polymer is selected from the group consisting of epoxy resins, melamine, urea formaldehyde polymers, and combinations thereof.
7. The pearlescent color pigment of claim 6 wherein the thermosetting organic polymer is an epoxy resin.
8. The pearlescent color pigment of claim 7 wherein the epoxy resin has an average molecular weight in the range from about 200 to about 6000.

9. The pearlescent color pigment of claim 8 wherein there epoxy resin has an average molecular weight in the range from about 350 to about 2000.
10. The pearlescent color pigment of claim 7 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 3800.
11. The pearlescent color pigment of claim 10 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 525.
12. The pearlescent color pigment of claim 11 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 197.
13. The pearlescent color pigment of claim 12 wherein the epoxy resin has an epoxide equivalent in the range from about 185 to about 192.
14. The pearlescent color pigment of claim 1 wherein the colorant comprises an organic pigment, an inorganic pigment, or combinations thereof.
15. The pearlescent color pigment of claim 14 wherein the colorant is selected from the group consisting of Permanent Orange G, 12-1007 Permanent Orange G, 13-3042 Colanyl Red FGRG-100, 16-2034 Hostaperm Green GNX, 15-1042 Hostaperm Blue BT-617-D, 13-7012 Hostaperm Red E5B-02, 13-7016 Host. Red EG Transparent, Quindo Red R-6700, Sunbrite Red 3 216-3300, 11-3012 Novoperm Yel, Hostaperm Pink E 13-7000, 14-4007 PV Fast Viol, 15-1023 Hostaperm Blue BT-627-D, 11-2210 Colanyl Yellow 5GX-100 (82%), 12-5005 Hansa Red GG-A, 13-3061 Novoperm Red, Tricolith Yellow IYP, 13-7011 Host. Red Violet ER-02, Hansa Yellow G, Heliogen Blue L 6875, Brilliant Red 2R 30AC0186, 11-3015 F2G-A Yellow, Monolite Green GLX, Red B NRT-742-D, 11-1418 Novoperm Yellow HR-70, 248-0156 Fastogen BL, Ultramarine Blue E-37, 13-2100 Hansa Scarle, 234-6485 Perm Red 2B, Sicotan Yellow L 1012, Manox Easisperse 154, 13-3610 Novoperm Red, VCD-4272, 13-3050 Permanent Red FGR, 11-2527 Hansa Brill Y, 11-2520 Hansa Brill Y, QHD-6040, 11-3030 Hostaperm Yellow H6G, 11-1101 Permanent YE, 2915 Dianisidine Orange 16, Fast Scarlet Red BBC, YT-805-D Dalamar M.A, Tricolith Green GN-1, Dalamar Yellow R YT-820-D, 13-3070 Perm Red 2R, Tricolith Yellow SFY, Fast Scarlet Red BBN, 11-1400 Novoperm Yel, 12-3004 Novoperm HL-70, Ultramarine Blue EP-37, 11-2310 Hansa Brill. Yellow 4GX, Unipure Redu LC 3079, Unipure Black LC902, and combinations thereof.

AMENDED SHEET (ARTICLE 19)

16. The pearlescent color pigment of claim 1 wherein the colorant comprises a nonreactive dye.
17. The pearlescent color pigment of claim 16 wherein the nonreactive dye is selected from the group consisting of FD&C Yellow 5, D&C Red 28, D&C Red 22, Macrolex Violet B, Macrolex Yellow 3G, Savanna Yellow and combinations thereof.
18. A pearlescent color pigment consisting essentially of:
 - a. a titanium dioxide coated inorganic substrate; and
 - b. a thermosetting organic polymer coated on the titanium dioxide coated inorganic substrate,

wherein a colorant selected from the group consisting of an organic pigment, an inorganic pigment, a nonreactive dye, and combinations thereof is dispersed or dissolved in the thermosetting organic polymer.
19. The pearlescent color pigment of claim 18 wherein the inorganic substrate is selected from the group consisting of natural micas, synthetic micas, aluminum oxide, silicon dioxide, and combinations thereof.
20. The pearlescent pigment of claim 18 wherein the thermosetting organic polymer comprises an epoxy resin.
21. The pearlescent color pigment of claim 20 wherein the epoxy resin has an average molecular weight in the range from about 200 to about 6000.
22. The pearlescent color pigment of claim 21 wherein there epoxy resin has an average molecular weight in the range from about 350 to about 2000.
23. The pearlescent color pigment of claim 20 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 197.
24. The pearlescent color pigment of claim 18 wherein the colorant is selected from the group consisting of Permanent Orange G, 12-1007 Permanent Orange G, 13-3042 Colanyl Red FGRG-100, 16-2034 Hostaperm Green GNX, 15-1042 Hostaperm Blue BT-617-D, 13-7012 Hostaperm Red E5B-02, 13-7016 Host. Red EG Transparent, Quindo Red R-6700,

Sunbrite Red 3 216-3300, 11-3012 Novoperm Yel, Hostaperm Pink E 13-7000, 14-4007 PV Fast Viol, 15-1023 Hostaperm Blue BT-627-D, 11-2210 Colanyl Yellow 5GX-100 (82%), 12-5005 Hansa Red GG-A, 13-3061 Novoperm Red, Tricolith Yellow IYP, 13-7011 Host. Red Violet ER-02, Hansa Yellow G, Heliogen Blue L 6875, Brilliant Red 2R 30AC0186, 11-3015 F2G-A Yellow, Monolite Green GLX, Red B NRT-742-D, 11-1418 Novoperm Yellow HR-70, 248-0156 Fastogen BL, Ultramarine Blue E-37, 13-2100 Hansa Scarle, 234-6485 Perm Red 2B, Sicotan Yellow L 1012, Manox Easisperse 154, 13-3610 Novoperm Red, VCD-4272, 13-3050 Permanent Red FGR, 11-2527 Hansa Brill Y, 11-2520 Hansa Brill Y, QHD-6040, 11-3030 Hostaperm Yellow H6G, 11-1101 Permanent YE, 2915 Dianisidine Orange 16, Fast Scarlet Red BBC, YT-805-D Dalamar M.A, Tricolith Green GN-1, Dalamar Yellow R YT-820-D, 13-3070 Perm Red 2R, Tricolith Yellow SFY, Fast Scarlet Red BBN, 11-1400 Novoperm Yel, 12-3004 Novoperm HL-70, Ultramarine Blue EP-37, 11-2310 Hansa Brill. Yellow 4GX, Unipure Redu LC 3079, Unipure Black LC902, and combinations thereof.

25. The pearlescent color pigment of claim 18 wherein the colorant comprises a nonreactive dye.

26. The pearlescent color pigment of any one of claims 1-25, further comprising a surface enhancement agent.

27. The pearlescent color pigment of claim 26 wherein the surface enhancement agent comprises an organosilane.

28. The pearlescent color pigment of any one of claims 1-25 wherein a curing agent is dispersed or dissolved in the organic polymer.

29. The pearlescent color pigment of claim 28 wherein the curing agent is an amine curing agent.

30. The pearlescent color pigment of claim 28 wherein the amine curing agent is diethylenetriamine.

31. A method for producing a pearlescent color pigment, the method comprising the steps of:

- a. providing a suspension of metal oxide coated inorganic substrate in water;

AMENDED SHEET (ARTICLE 19)

- b. adding a thermosetting organic polymer to the suspension;
- c. adding a colorant selected from the group consisting of an organic pigment, an inorganic pigment, a nonreactive dye, and combinations thereof to the suspension,

wherein the colorant dissolves or disperses in the organic polymer; and

- d. sufficiently heating the suspension to coat the thermosetting organic polymer on the metal oxide coated inorganic substrate, thereby producing the pearlescent color pigment;

wherein the colorant remains dissolved or dispersed in the thermosetting organic polymer when the metal oxide coated inorganic substrate is coated with the thermosetting organic polymer.

32. The method of claim 31 wherein the inorganic substrate is selected from the group consisting of natural micas, synthetic micas, aluminum oxide, silicon dioxide, and combinations thereof.

33. The method of claim 30 wherein the metal oxide is selected from the group consisting of titanium dioxide, ferric oxide, zirconium dioxide, tin dioxide, and combinations thereof.

34. The method of claim 33 wherein the metal oxide is titanium dioxide.

35. The method of claim 34 wherein the thermosetting organic polymer is selected from the group consisting of epoxy resins, melamine, urea formaldehyde polymers, and combinations thereof.

36. The method of claim 35 wherein the thermosetting organic polymer is an epoxy resin.

37. The method of claim 36 wherein the epoxy resin has an average molecular weight in the range from about 200 to about 6000.

38. The method of claim 37 wherein the epoxy resin has an average molecular weight in the range from about 350 to about 2000.

39. The method of claim 36 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 525.

40. The method of claim 39 wherein the epoxy resin has an epoxide equivalent in the range from about 175 to about 197.
41. The method of claim 31 wherein the colorant comprises an organic pigment, an inorganic pigment, or combinations thereof.
42. The method of claim 31 wherein the colorant comprises a nonreactive dye.
43. The method of claim 31, further comprising the step of adding a surface enhancement agent to the suspension prior to adding the organic polymer to the suspension.
44. The method of claim 43 wherein the surface enhancement agent comprises an organosilane.
45. The method of any one of claims 31-44, further comprising the step of adding a curing agent to the suspension after adding the thermosetting organic polymer to the suspension.
46. The method of claim 45 wherein the curing agent is an amine curing agent.
47. The method of claim 46 wherein the amine curing agent is diethylenetriamine.
48. The pearlescent color pigment of claim 26 wherein a curing agent is dispersed or dissolved in the organic polymer.
49. The pearlescent color pigment of claim 48 wherein the curing agent is an amine curing agent.
50. The pearlescent color pigment of claim 49 wherein the amine curing agent is diethylenetriamine.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 08/70981

<p>A. CLASSIFICATION OF SUBJECT MATTER IPC(8) - C04B 14/20 (2008.04) USPC - 106/418; 106/401 According to International Patent Classification (IPC) or to both national classification and IPC</p>												
<p>B. FIELDS SEARCHED</p> <p>Minimum documentation searched (classification system followed by classification symbols) IPC(8) - C04B 14/20 (2008.04) USPC - 106/418; 106/401</p> <p>Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched IPC(8) - C04B 14/20 (2008.04) (text search only - see search history printout) USPC - 106/418; 106/401 (text search only - see search history printout)</p> <p>Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) (USPT, PGPB, EPAB, JPAB); Google Patent, Google Scholar, Dialog Web pigment, pearlescence, pearlescent, nacreous, pearl luster, mica, titanium dioxide, tio2, titanium oxide, Silicone dioxide, sio2, epoxy, epoxide, aluminum oxide, alumina, colorant, dye, thermoset, silane siloxane, silyl?, zirconium oxide, tin oxide</p>												
<p>C. DOCUMENTS CONSIDERED TO BE RELEVANT</p> <table border="1"> <thead> <tr> <th>Category*</th> <th>Citation of document, with indication, where appropriate, of the relevant passages</th> <th>Relevant to claim No.</th> </tr> </thead> <tbody> <tr> <td>X ----- Y</td> <td>US 2007/0107635 A1 (Soane et al.) 17 May 07 (17.05.2007), (para [0003], [0014], [0015], [0016], [0023], [0026], [0028], [0031], [0052] [0075], [0076], [0094], [0100]).</td> <td>1-27, 31-32, and 41-45 ----- 46-47</td> </tr> <tr> <td>Y</td> <td>US 2007/0042126 A1 (Passman et al.) 22 Feb 07 (22.02.2007), (para [0022]).</td> <td>46-47</td> </tr> </tbody> </table>			Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.	X ----- Y	US 2007/0107635 A1 (Soane et al.) 17 May 07 (17.05.2007), (para [0003], [0014], [0015], [0016], [0023], [0026], [0028], [0031], [0052] [0075], [0076], [0094], [0100]).	1-27, 31-32, and 41-45 ----- 46-47	Y	US 2007/0042126 A1 (Passman et al.) 22 Feb 07 (22.02.2007), (para [0022]).	46-47	
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X ----- Y	US 2007/0107635 A1 (Soane et al.) 17 May 07 (17.05.2007), (para [0003], [0014], [0015], [0016], [0023], [0026], [0028], [0031], [0052] [0075], [0076], [0094], [0100]).	1-27, 31-32, and 41-45 ----- 46-47										
Y	US 2007/0042126 A1 (Passman et al.) 22 Feb 07 (22.02.2007), (para [0022]).	46-47										
<p><input type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/></p>												
<p>* Special categories of cited documents:</p> <table border="0"> <tr> <td>"A" document defining the general state of the art which is not considered to be of particular relevance</td> <td>"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</td> </tr> <tr> <td>"E" earlier application or patent but published on or after the international filing date</td> <td>"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</td> </tr> <tr> <td>"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)</td> <td>"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</td> </tr> <tr> <td>"O" document referring to an oral disclosure, use, exhibition or other means</td> <td>"&" document member of the same patent family</td> </tr> <tr> <td>"P" document published prior to the international filing date but later than the priority date claimed</td> <td></td> </tr> </table>			"A" document defining the general state of the art which is not considered to be of particular relevance	"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	"E" earlier application or patent but published on or after the international filing date	"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	"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)	"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	"O" document referring to an oral disclosure, use, exhibition or other means	"&" document member of the same patent family	"P" document published prior to the international filing date but later than the priority date claimed	
"A" document defining the general state of the art which is not considered to be of particular relevance	"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											
"E" earlier application or patent but published on or after the international filing date	"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											
"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)	"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											
"O" document referring to an oral disclosure, use, exhibition or other means	"&" document member of the same patent family											
"P" document published prior to the international filing date but later than the priority date claimed												
<p>Date of the actual completion of the international search 30 Sep 2008 (30.09.2008)</p>		<p>Date of mailing of the international search report 02 OCT 2008</p>										
<p>Name and mailing address of the ISA/US Mail Stop PCT, Attn: ISA/US, Commissioner for Patents P.O. Box 1450, Alexandria, Virginia 22313-1450 Facsimile No. 571-273-3201</p>		<p>Authorized officer: Lee W. Young PCT Helpdesk: 571-272-4300 PCT OSP: 571-272-7774</p>										

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 08/70981

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search 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.: 28-30, and 33-40
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

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

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 additional fees, this Authority did not invite payment of additional fees.
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 and, where applicable, the payment of a protest fee.
- The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- No protest accompanied the payment of additional search fees.