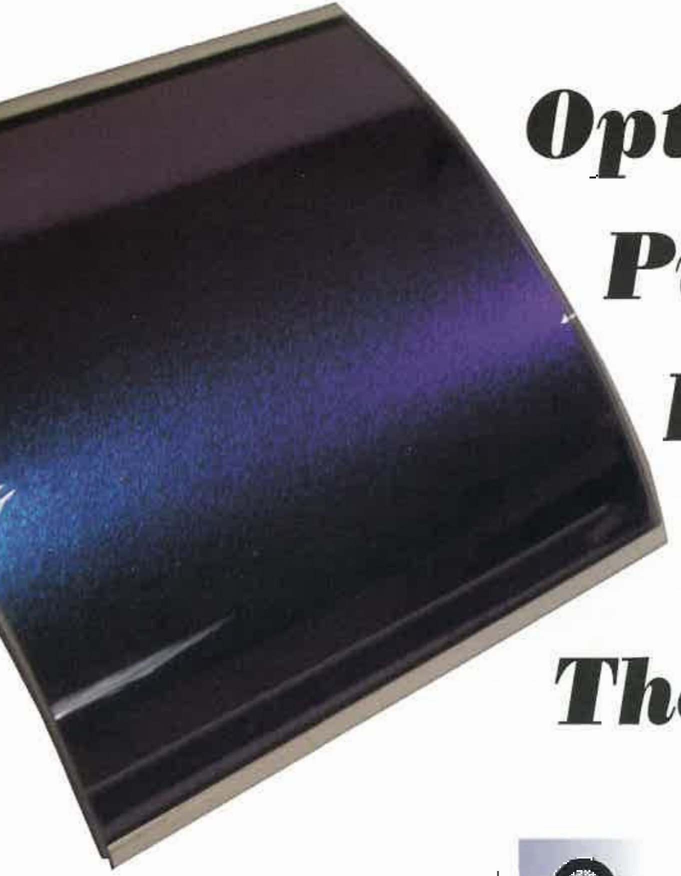

Optically Variable Pigments from Dielectric Layers and Their Applications

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Over the last decade, several new pigments made from optical layers of metals and dielectric layers, which fit the general term of optically variable pigments (OVPs), have been commercialized. These pigments generate high chroma and color travel, dramatic changes in color at different angles of illumination, and viewing to produce fascinating color effects in coatings, plastics, and cosmetics. Advances in their development have resulted in high chroma color shifting pigments, with greater utility and impact. This article will focus on dielectric layers for generating optically variable effects, present some basic structure appearance relationships, and cover some examples of application and performance in coatings.

The mechanisms of color generation and color travel of dielectric optically variable pigments are different from traditional pearlescent pigments that derive their color from the interference of light reflected from thin film of high refractive index materials.[1] These traditional effect pigments generate an intense specular, or mirror reflection color, and at aspecular angles lose their color intensity. Viewing these pigments at different specular angles produces only a small change in their hue and loss of color at near planar angles. This is demonstrated in *Figure* 1, which shows changes in a* and b* relating to red-green and yellow-blue colors, respectively, with changes in angle of illumination and viewing. The color travel for a traditional blue interference pigment is minimal, changing from a green shade to a red shade blue as shown in *Figure* 1.

Optically variable pigments, on the other hand, generate their color through the interference of light rays reflected from interfaces between multiple layers of materials differing in refractive index. Most dielectric optically variable pigments use a high-low-high refractive index layer structure referred to as an *etalon* or "stack" structure.[2]

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Pigments with this structure show the traditional chroma reduction in moving from the specular angle to aspecular angles of illumination and reflection. However, viewing the pigment at different specular angles results in intense hue shifts, or color travel, with high chroma. This is demonstrated in *Figure* 2 by the high a* and b* values, which change from yellow-green to violet. It is the low refractive index layer that provides the extended color travel for these pigments.

Color generation from these optically variable pigments is dependent on the refractive index of the materials being used as well as the thickness of the layers. The high refractive index materials used are typically metals such as aluminum, chromium, or silver, or metal oxides such as TiO_2 or Fe_2O_3. Low refractive index materials used have been SiO_2 or MgF_2. Control of the layer thicknesses determines the color range and hue, and choice of materials further determines the color travel as well as the pigment chroma. Layer thicknesses must be controlled to tolerances of several nanometers for color consistency.

The optically variable pigments fall into two broad categories: opaque and transparent.

OPAQUE OPTICALLY VARIABLE PIGMENTS

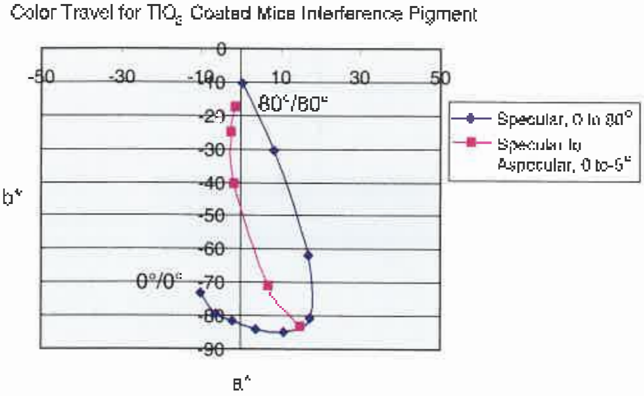
Opaque pigments are those produced using a metallic reflector that acts as a high refractive index layer centered in the pigment (*Figure* 4). Examples of these are structures with an internal layer of Al coated with MgF_2 as the low refractive index material followed by a partial mirror of Cr as the outer high refractive index material.

TRANSPARENT OPTICALLY VARIABLE PIGMENTS

The transparent class of optically variable pigment materials, defined as DOVP, includes only dielectric layers and has some distinct advantages in that they can be used in clearcoats over light colored basecoats without contributing a dirty appearance. Several different structures are used to generate pigments with high color travel. A typical structure consists of a low refractive index silica layer coated with a thin layer of highly refractive TiO_2. The silica layer is an integral component of the optical stack, therefore changes in its thickness contributes to variation in the pigment's color travel (*Figure* 5).

Another structure consists of a borosilicate substrate with a high-low-high stack of TiO_2 and SiO_2; these structures are commercially available under the tradename Firemist® Colormotion by BASF. Although the borosilicate has a low refractive index, it does not play a role in the pigment's color development as it is somewhat thick. Precise control of the SiO_2 layer thickness assures that each platelet is virtually the same, magnifying the reflection intensity of the pigment coating to increase chroma in the application (*Figure* 6).

Figure 1—Traditional effect pigment color change with illumination and viewing angle changes.



Blends of large sized pigments with high color variation create a confetti-like effect due to spatial color resolution. The substrate-based dielectric optically variable pigments with their large size and inter-platelet color consistency have a uniform color, which changes at different angles of illumination and viewing.

The color travel plot of the samples shown in *Figure* 8, measured at specular angles using an IsoColor goniospectrophotometer, demonstrates that color travel is a property of the individual pigment particles and cannot be created with blends of different pigment colors. The measured color of the interference pigment blend is the sum of the three colors that tends towards a neutral shade. The DOVP pigment shows a strong color intensity at all specular angles with large changes in hue from green-blue to red.

Figure 2—Optically variable pigment color change with illumination and viewing angle changes.

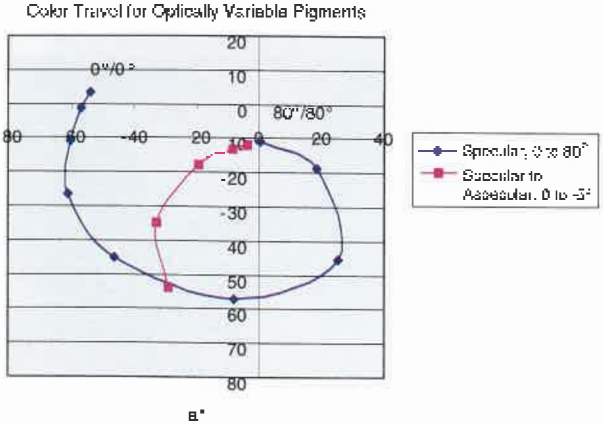


Figure 3—A typical dielectric stack structure of a $TiO_2/SiO_2/TiO_2$ used as an optically variable pigment, reflecting green at normal incidence and viewing angles, and shifting to violet at near-planar angles.

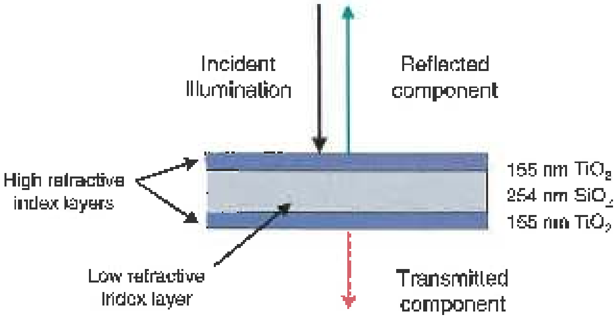
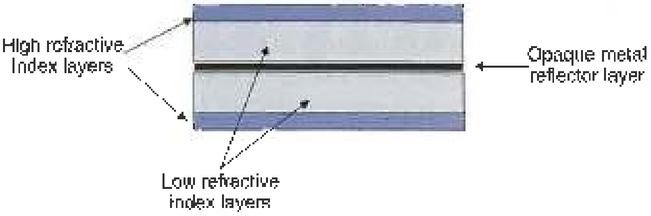


Figure 4—Stack structure surrounding a metallic reflector resulting in an opaque dielectric optically variable pigment.



APPLICATIONS AND PERFORMANCE

The platelet is thin enough to allow its use in low film build liquid coating applications, such as coatings for teletronics and packaging, yet the particle size is big enough to create a sparkle effect in high film build applications like powder coatings and screen printing. For dramatic effects, the pigment should be used over a dark background to promote only the intense interference reflection color. The transmitted color is absorbed into the background. Application over a light background produces a highlight effect due to the intense reflection color combined with full transparency to show the background color.

The pigment concentration can be varied depending on the application: for both liquid and powder coatings, a sufficient effect can be achieved by as low as 1% pigment concentration based on total solids weight; 2.5–5% will have a very intense color and color travel.

Another attribute of the borosilicate-based optically variable pigment is its ability to maintain color travel

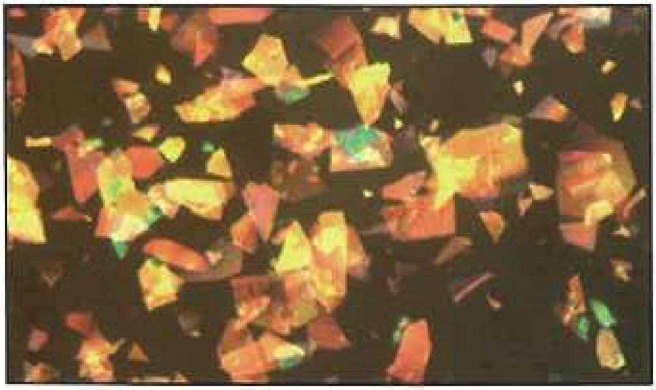


Figure 5—Dielectric layers for generating optically variable pigments based on TiO_2 coated silica platelet showing color variability due to thickness variations of the silica layer.

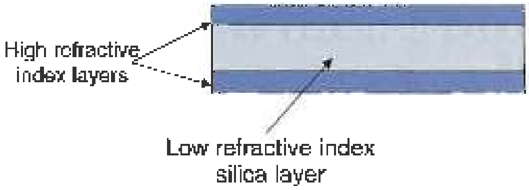


Figure 6—An optically variable pigment based on 80 micron-sized borosilicate substrate with high-low-high dielectric stack structure surrounding the substrate.

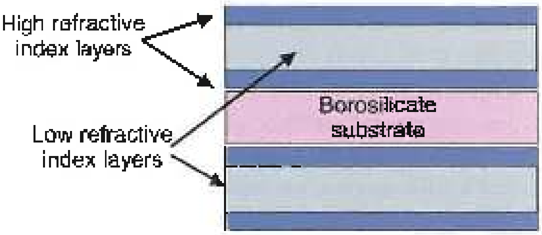


Figure 7—A DOVP (left) vs. a blend of red, blue, and gold borosilicate-based interference pigments (right).

Figure 8—DOVP compared to a blend of three borosilicate-based interference color pigments.

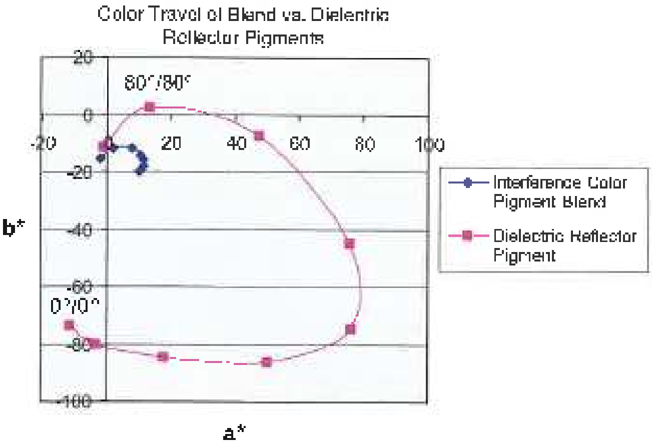


Figure 9—Coatings containing DOVP over black background.

Figure 10—Color travel for a blend of DOVP and absorption colorant.

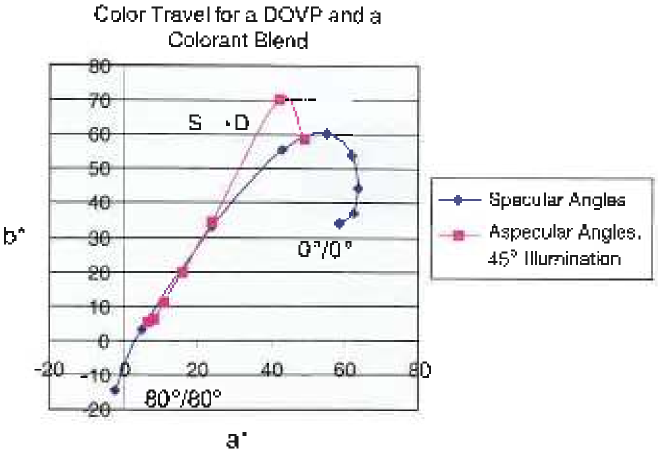
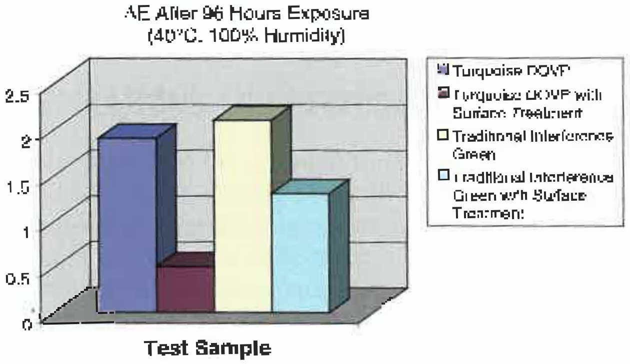


Figure 11—DOVP (red to gold) mixed with an orange.

Table 1—Color Changes for Effect Pigment Following Humidity Exposure

	ΔL*	Δa*	Δb*	ΔE
Turquoise DOVP	1.5	−0.8	−0.8	1.9
Turquoise DOVP with surface treatment	0.4	0.2	−0.1	0.5
Traditional interference green	2.1	0	0.6	2.1
Traditional interference green with surface treatment	−1.2	0.5	−0.3	1.3

Figure 12—Color changes for DOVP vs. traditional interference pigment.



when used with absorption pigments instead of becoming lost, as happens to some pigments of this type. An example shown in *Figure* 11 is a violet-red-gold dielectric optically variable pigment in use with an orange that shows excellent color travel in a coating film.

The dielectric optically variable pigment's one micron thickness allows for its use in automotive refinish coatings. The performance of the pigments can be enhanced with a surface treatment to suit exterior application requirements.

Humidity resistance is critical for platy structure material.[3] Most humidity-induced failures are due to whitening and wrinkling of the paint film, especially for waterbased coating systems. Whitening and microwrinkling in coating films can be measured by changes following exposure in L* and the dullness of the coating film, respectively. Test results from 96 hr of exposure at 40°C with 100% humidity can provide a good indication whether or not pigments will meet testing requirements for automotive applications.

Tables 1 and 2 show the color change for a surface-treated dielectric optically variable pigment versus an untreated pigment using a traditional mica-based interference green pigment widely used in automotive OEM and refinish coatings as a control. The dielectric optically variable pigmented film shows excellent color retention when tested with an appropriate treatment.

The gloss retention and appearance can also be improved by putting an exterior treatment on these types of pigments, as *Table* 2 shows. With the exterior treatment, the coating film will have higher gloss retention and less dullness increase during testing.

SUMMARY

High chroma, color changing pigments have been available for more than a decade. New borosilicate substrate-based pigments generate higher chroma, greater impact, and have good durability for a wide range of applications in high performance coatings, from teletronics to automotive refinishes.

ACKNOWLEDGMENT

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Table 2—Appearance Changes of Effect Pigments Following Humidity Exposure

	20% Gloss Retention (%)	Dullness (Initial/Final)	Appearance
Turquoise DOVP	60	55/59	Very slight wrinkling/fading
Turquoise DOVP with surface treatment	89	57/58	No change
Traditional interference green	86	22/50	Very slight wrinking
Traditional interference green with surface treatment	100	23/21	No change