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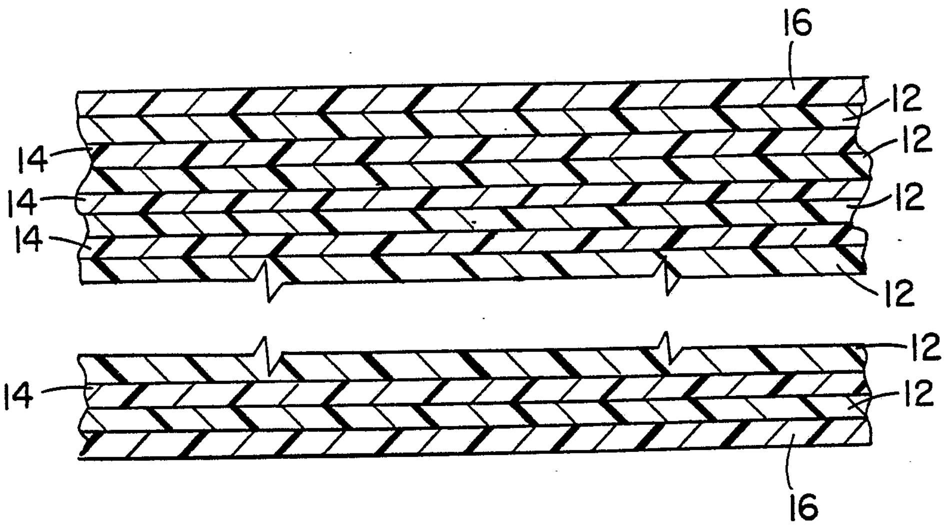

(57) Abstract

A multilayer optical interference film indicator useful as or in a protective or tamper evident, tamper resistant packaging material, offering visible evidence of strain of a selected extent or greater associated with an attempt at pampering or the like in the form of iridescence or interference colors, which comprises contiguous layers of two or more (12, 14) diverse thermoplastic materials of differing refractive indices, and wherein the yield point of at least one of the diverse materials has been at least equalled when the visible evidence is produced.

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TAMPER EVIDENT, TAMPER RESISTANT PACKAGING MATERIAL, PREPARATION AND USE

This invention relates to indicators, and more particularly to indicators providing visible evidence of strain. In one aspect, the invention relates to the use of such indicators to provide both visible evidence of tampering and a resistance to tampering with respect to articles packaged therein, whereby the indicators find use within or as a tamper evident, tamper resistant packaging material. In another aspect, the invention relates to an article such as a container which has been at least partially packaged within such a packaging material, and to a process for protecting the contents of the container from adulteration or otherwise ensuring the integrity or authenticity of the contents by wrapping the container in its entirety or in significant part with the tamper evident, tamper resistant packaging material.

Several packaging systems which purport to be tamper evident, tamper resistant, or both tamper evident and tamper resistant have been reported in the art. One packaging system said to provide visual evidence of tampering is described in United States Patent No. 4,721,217 to Phillips et al. An optically variable device is disclosed which comprises a first layer attached to a first part of the package or of a consumer article, a second layer attached to a second part of the package or article which is movable with respect to the first part, and a release layer disposed between the first and second layers of the device to permit the first and second layers to be separated. When intact, the first and second layers provide a reflection of visible color which changes according to the viewing angle. When the first and second parts are moved with respect to one another and the first and second layers separated, this optical shifting property is said to be destroyed, and cannot be restored by reassembly except as a different color shift or as a color resembling an oil slick.

One problem that is seen with the packaging material described is the requirement that consumers be able to detect the presence of a different color shift than intended by the manufacturer, or even the presence of a color shift at all.

This problem is illustrated by an example of an optically variable device provided in the patent. This device reflected a green color at normal incidence and a blue color at a 45° angle, and when tampered with reflected a grayish color in transition and a tinge of blue at an angle. Depending on the consumer, this gray to blue transition may appear to satisfy a specified green to blue transition. Moreover, that portion of the population which has blue-green color blindness may be unable to detect any color shift, as would be true of any person who insufficiently altered his or her viewing angle with respect to the device.

The present invention solves this problem by providing a multilayer optical interference film indicator comprising contiguous layers of two or more diverse thermoplastic materials of differing refractive indices. The indicator and a tamper evident packaging material made of or from the indicator are substantially colorless or of a uniform color in an initial condition. When strained to a selected extent or greater, however, the indicator and the packaging material made therefrom will produce visible evidence of such strain in the form of iridescence or interference colors, and this visible evidence will be retained by the yield point of at least one of the materials in the packaging material having been exceeded when the visible evidence is produced. Thus, the consumer is simply asked to distinguish between the absence of all color or between a single color or pattern of colors (stripes, checks, etc.) and the unstructured or unpatterned presence of many colors in the form of iridescence or interference colors generally.

In another aspect, the present invention relates to an article which has been at least partially packaged within an indicator of the present invention, as for example where the indicator has been joined to two portions of the article which are movable with respect to one another.

A method of protecting the contents of a container from adulteration or otherwise ensuring the integrity or authenticity of the contents is also disclosed, and includes the step of wrapping the container in its entirety or in significant part with a tamper evident, tamper resistant packaging material which includes an indicator of the present invention.

Other features, advantages and uses of the multilayer film indicator of the present invention will become apparent on considering the description which follows in conjunction with the appended claims and accompanying drawings.

Fig. 1 is a cross-sectional view of a preferred embodiment of the multilayer optical interference film indicator of the present invention.

Fig. 2 is a representation of a hypothetical reflectance spectrum of an indicator of the present invention in an initial condition.

Fig. 3 is a representation of the hypothetical reflectance spectrum of Fig. 2 after the corresponding indicator of Fig. 2 has been strained beyond a selected extent.

Figures 4 and 5 are simulated, computer generated reflectance spectra for various embodiments of the present invention.

In the way of background, for multilayer optical interference films having a period of P layers, it has been previously determined that the primary reflectance of normally incident light occurs at a wavelength λ_0, wherein

$\lambda_0 = 2\sum_{i=1}^{P}(n_i d_i)$, and $(n_i d_i)$ is the product of the refractive index n and physical thickness d associated with a given layer. This quantity will hereafter be referred to as the "optical thickness" of that layer.

Higher orders of reflectance occur at wavelengths determined by the equation $\lambda_M = \lambda_0/M$, where M is the order of reflectance. For example, if the primary reflectance λ_0 is at 1400 nanometers, then subsequent orders of reflectance will occur at about 700 nanometers (second order), 467 nanometers (third order), 350 nanometers (fourth order), and so on.

Referring now to the drawings, and more particularly to Fig. 1, a preferred multilayer film indicator 10 of the present invention is illustrated. The indicator 10 shown in Fig. 1 consists of stacked and coextensive or at least overlapping layers 12 and 14 of two diverse thermoplastic materials A and B in an ABAB-type layer order, wherein the layers 12 correspond to layers of a material A and the layers 14 to an optically diverse material B.

It should be noted that these "diverse thermoplastic materials" need not differ in any respect except in terms of refractive index. Thus, while the materials of the layers within an indicator may be chemically diverse, if these materials have the same refractive index then they are not "diverse" for purposes of the present disclosure. Similarly, the materials may be virtually identical in every other respect save refractive index, yet they are to be considered "diverse" for purposes of this disclosure.

The optically inactive outer skin layers 16 shown in Fig. 1 are primarily included and are sufficiently thick to prevent excessive break up of the thin layers 12 and 14 of A and B in processing and making the indicator 10 by the preferred multilayer coextrusion processes, there being in the indicators of the present invention preferably at least fifty stacked and coextensive or overlapping layers of the various diverse materials comprising the laminate. Multilayer coextrusion processes suitable for making indicators having at least this number of layers are taught, for example, in the commonly assigned United States Patent Nos. 3,565,985 and 3,773,882. The outer skin layers may also be present in a particular thickness for contributing to the structural properties of the indicator, however.

The film indicators of the present invention are designed such that in an initial condition the primary reflectance λ_0 of the preferred two-component film shown in Fig. 1, for example, is in the near infrared: $\lambda_0 = 2(n_A d_A + n_B d_B) \cong .7$ µm or greater, where n_A and n_B represent the respective refractive indices of the materials A and B, and d_A and d_B the respective average physical thicknesses of layers 12 and 14 of A and B, respectively, within a period AB of the indicator 10.

To be most effective as a tamper evident and tamper resistant film, or indeed as a strong visual indicator of strain, the visual contrast between an initial condition and a condition corresponding to the presence of a degree of strain in excess of that which might be attributable to other causes such as accidental mishandling and so forth must be sufficiently great as to be easily and unequivocally perceived.

This contrast comprises the visual evidence of tampering which is basic to the present invention, and is preferably created by achieving as nearly as possible a complete absence of color in the multilayer optical interference film indicators of the present invention in an initial condition, to be contrasted with vivid interference colors or iridescence as a sign of strain against the backdrop of an indicator's environment or against a skin layer 16 which is tinted black, for example, as will be subsequently explained.

It has now been found that in the two component indicator, where a substantial majority of the layers 12 and 14 possess an optical thickness of 0.17 micrometers or greater in the initial condition and further where the ratio $f_A = \frac{n_A d_A}{(n_A d_A + n_B d_B)}$ is approximately 1/2, an acceptable absence of color results for multilayer films having the layer thickness gradients in the film thickness direction and thus the reflectance band widths which are possible using the processing techniques described in United States Patent Nos. 3,565,985 and 3,773,882 and exemplified below.

It should be observed that ordinarily it will be desirable to have some gradient in layer thickness in the film thickness direction and thus a fairly broad bandwidth for the primary reflectance of an indicator, as an indicator possessing such a bandwidth will give a more vivid indication of strain than a film indicator having a narrow primary reflectance bandwidth. Preferably, then, as respects the bandwidth of the primary reflectance peak the reflectance spectrum will have more of the character of Figures 2 and 3 than that shown in computer-generated Figures 4 and 5.

Obviously, the extent of accidental strain for which allowance must be made to avoid an unacceptable number of false-positive answers may vary according to the environment and use of the indicator, and given a primary reflectance of a particular magnitude and bandwidth, other accommodations may need to be made in the optical thicknesses of the various layers, in the layer thickness gradients produced in the thickness direction of an indicator by a given manufacturing process, and/or in the number of layers in the indicators to preserve an acceptable lack of color in these other circumstances.

While some degree of nonuniformity is thus desirable in the layer thickness direction, layer thickness nonuniformities throughout the length and width of the indicator in contrast are preferably small enough so that the indicator 10 is not colorless at one location and already showing visible color at another location. What nonuniformities are "small enough" for a given application again depends on the context and environment in which the indicator is used, but a particular example of the limits implied by this term may be found in the examples which follow.

Essentially the amount of strain which is induced in routine handling or which is otherwise accidentally induced is of concern because as the film is stretched either locally or on a larger scale, the actual physical thicknesses d_A and d_B of film layers A and B are reduced. In turn, as the thicknesses d_A and d_B grow smaller, so do the corresponding optical thicknesses and the sum of the optical thicknesses, and thus so too the wavelengths corresponding to the primary and higher order reflectances of the film.

At a selected extent of strain, the optical thicknesses of a sufficient number of layers are reduced in the indicator so that visible evidence of the strain is produced in the form of interference colors, and the yield point of at least one of the materials A and B or of the materials in the skin layers 16 if present is exceeded. This selected extent of strain will preferably be only slightly in excess of that which might be accidentally produced, so as to give prompt visual evidence of even the most abortive attempts at tampering without the rejection of an undue percentage of unadulterated products.

Ordinarily it will also be desirable, and for tamper evident/tamper resistant packaging materials in particular, if the materials which comprise the multilayer indicators of the present invention are selected so that the evidence of strain is still visible when the film indicators rupture and break, rather than having the optical thicknesses of the various layers decline with additional tampering to an extent whereby the primary reflectance wavelength passes into the ultraviolet range. In this manner, the purchaser of an article which is wrapped or packaged in the tamper evident/tamper resistant packaging material can know an article has been tampered with if the material is still intact and evidences a signal color change, or if the material has been removed altogether.

Where the visual evidence of strain generally or of an attempt at tampering will be produced in the indicator will depend on the construction of the indicator, and on how the indicator is connected to an article as a tamper evident packaging material, for example whether the ends or edges of the indicator are attached to the article. In terms of the construction of the indicator, it may be desirable in some applications to reduce the cross-sectional area of a film indicator at regular intervals while not altering the thickness of the indicator, as by locally narrowing the width of the indicator or by perforating the indicator. These locally narrowed portions of the indicator can then be preferentially strained so that characteristic intermittent bands of interference colors or iridescence are shown in the indicator.

The shift in the reflectance profiles of the strained and unstrained film samples which corresponds to the production of the visible evidence is illustrated in Figures 2 and 3 for a hypothetical film.

Preferably the materials A and B in the two component embodiment of our invention should differ in refractive index by 0.03 or greater, and the number of stacked and coextensive or overlapping layers sufficiently great (in the area of overlap) to make the reflectance of the interference colors or iridescence unmistakable from the distances at which the indicator would likely be observed in use; where immediately adjacent layers of the various diverse materials generally differ in refractive index by the same 0.03 or greater, this should preferably correspond for the uses presently contemplated for the present invention to having at least fifty layers within the indicators. Typically, a hundred or more alternating coextensive or overlapping layers of the A and B materials will be created in the two component embodiment of the present invention. The use of this number of layers, in addition to lending emphasis to the signal color change produced on achieving a degree of strain, is advantageous also in that it makes counterfeiting the indicators very difficult if not impossible.

In order for the color change to be effective evidence of strain generally or of an attempt at tampering in the more particular case of our tamper evident/tamper resistant films, the skin layers 16 at the least should not interfere with the perception of this change. Accordingly, at least one of the skin layers 16 adjacent a first surface 18 of the indicator 10 should be transparent to visible light.

Where the background provided by the environment of the indicator 10 presents an adequate contrast to enable the color change to be reliably perceived by an observer thereof, both skin layers 16 may be transparent. Where an adequate contrast is not thus presented, however, the second skin layer 16 may be pigmented as suggested earlier to provide the contrast necessary to have the signal color change recognized.

Where processing equipment and techniques permit and an adequate percentage of layers can be said to have an initial optical thickness to allow the indicators to be substantially colorless absent an attempt at tampering, the skin layers 16 may be omitted altogether. Because of the very small physical thicknesses of the hundred or more layers which have thus far been used in developing the present tamper evident/tamper resistant packaging materials, however, it has been thought necessary to employ skin layers in actual practice which comprise from 5 to 10 percent by volume of the indicators of the present invention at least, although as noted earlier it may be desirable for structural rather than optical purposes to have thicker skin layers also.

The two component embodiment which has thus far been primarily particularly described is generally considered satisfactory for purposes of the present invention, but even where the second order reflection has been suppressed (by the selection of optical thicknesses so that f_A is approximately 1/2), third, fourth and higher order reflections may exist which would give the appearance of color to some observers in the initial condition or after accidental mishandling and the like. Accordingly, a three component system has been developed which enables the suppression of not only the second order reflection but in a particularly preferred embodiment also the third and fourth order reflections.

In this three component system, the multilayer film indicator consists of at least one period ABCB of stacked and coextensive or overlapping layers of three diverse thermoplastic materials A, B and C wherein A possesses a higher refractive index n_A, B an intermediate refractive index n_B, and C a lesser refractive index n_C. The optical thicknesses and refractive indices of the various layers are preferably selected so that:

$$f_A = \frac{n_A d_A}{(n_A d_A + n_B d_B + n_C d_C + n_B d_B)} \cong 1/3;$$

$$f_B = \frac{n_B d_B}{(n_A d_A + n_B d_B + n_C d_C + n_B d_B)} \cong 1/6;$$

$$f_C = \frac{n_C d_C}{(n_A d_A + n_B d_B + n_C d_C + n_B d_B)} \cong 1/3; \text{ and}$$

$$n_B \cong \sqrt{n_A n_C}\ .$$

To start out again with a primary initial reflectance at a wavelength of at least 700 nanometers, preferably at least a substantial majority of the layers A, B and C should have an optical thickness of 0.09 micrometers or greater throughout the thickness of the indicator when the indicator is in the initial condition. The optical thicknesses of the layers may be adjusted, as before, where considerations of handling strains and the like require.

And, again as before, preferably the materials A, B and C will be selected so that the highest and lowest refractive indices of layers within a period of P layers will differ by .03 or greater, with a sufficient number of coextensive or overlapping layers to make the signal color change produced on achieving a selected extent of strain readily detectable. Most preferably, the refractive indices of adjacent layers of materials within a period of P layers will differ by 0.03 or greater. Preferably, the indicators of the present invention in this regard will comprise at least fifty layers, and most preferably, at least one hundred layers.

The two and three component embodiments which have been described in detail above are preferred because the two and three component embodiments of specified f-ratios provide a multilayer film indicator which has the least amount of higher order reflectance in the visible wavelengths that is presently deemed achievable. This results generally in greater control over the reflectance of the films, and permits the initial reflectance to be designed for a higher wavelength without higher order reflections bringing color to the films.

The greater separation between the wavelength corresponding to the primary reflectance in an initial condition and the upper end of the visible range provides a processing window for making the multilayer film indicators of the present invention which is generally desirable. Also, one may not know precisely how much "innocent" or accidental strain might be encountered by the indicators of the present invention in a given application, so that a safety factor could be built into the devices, if desired, to prevent excessive false-positive readings without an excessive loss in detection capabilities.

This is not to suggest, however, that other f-ratios and arrangements, employing any number of thermoplastic materials with different refractive indices, might not also exist which would function adequately in making the indicators of the present invention initially substantially colorless, particularly in a given environment or use. In this regard, a number of other arrangements and materials have been reported in the dielectric vacuum deposition or sputtering art which might be suitable, see for example United States Patent No. 3,247,392 to Thelen.

With regard to achieving and preserving evidence of strain, the multilayer film indicators may in summary be constructed in a number of ways. One possible configuration would place at least one material whose yield point is at least equalled when the evidence is produced within the core layers of a multilayer film indicator having skin layers.

Another configuration would encapsulate core layers comprised of elastomeric materials within skin layers whose yield point would be at least equalled when the color change occurs, thus preventing the elastomeric inner or core layers from returning to an unstretched or unstrained condition. This configuration is exemplified below in Example 1.

As noted earlier, the film indicators of the present invention may not employ skin layers at all. Or, if skin layers are employed, one or both may be transparent. However constructed, the multilayer film indicators of the present invention are well adapted for a number of uses, and are particularly well suited for a tamper evident, tamper resistant packaging material.

When used in or as a tamper evident, tamper resistant packaging material, the multilayer film indicators of the present invention may preferably be heat-shrinkable and used as a shrink band joined to two portions of an article which are movable with respect to one another, such as the neck and screw cap of a prescription bottle. Alternatively, the films may be used in the form of a membrane seal across an opening in a container. Or, the films may be used to overwrap an article entirely. It may also be desirable to employ the films of the present invention in two or all three of these capacities to improve the tamper evidence or tamper resistance properties of an article packaged therein.

The multilayer film indicators of the present invention may be further illustrated by the examples which follow.

Example 1

In this example a multilayer film indicator including 197 alternating layers of Pebax® 2533 SA00 polyether amide (Atochem Company, Paris, France) with a refractive index of 1.49, and of Pellethane® 2355-95 AEF polyurethane produced by The Dow Chemical Company, Midland, Michigan, and having a refractive index of 1.56, was made by multilayer coextrusion according to the processes and using equipment and techniques such as described in commonly-assigned United States Patents No. 3,759,647, 3,773,882, and 3,884,606. These 197 layers were encapsulated within skin layers of transparent and unpigmented Profax® 6131 polypropylene (Hercules, Inc.) by coextrusion therewith, with each of the cap or skin layers comprising approximately 10 percent of the total 0.002 inch thickness of the film. A one inch wide strip of this multilayer film was wrapped around the neck and screw cap of a bottle, and a hot air gun was used to cause the film to shrink to fit tightly around the neck and cap. Unscrewing of the cap caused the film to reflect vivid colors of red, green, yellow and blue.

Further twisting of the cap caused the film to rupture. The cap could be rotated less than approximately 15° until the color change appeared and approximately 45° before the film ruptured.

Example 2

This example simulated the reflectance spectrum of a multilayer film indicator consisting of layers of three diverse thermoplastic materials A, B and C in an ABCBA layer order as an ABCB repeating unit, with layers of the A material being immediately adjacent opposing outer surfaces of the indicator. The A, B and C materials selected for the simulation were a polystyrene of a refractive index of 1.59, a styrene-methylmethacrylate copolymer with a refractive index of 1.539, and a polymethylmethacrylate of a refractive index of 1.49. These materials were chosen to satisfy the relationship $n_B \cong \sqrt{n_A n_C}$. The optical thicknesses of materials within a series of such layers were chosen so that:

$$f_A = \frac{n_A d_A}{(n_A d_A + n_B d_B + n_C d_C + n_B d_B)} \cong 1/3;$$

$$f_B = \frac{n_B d_B}{(n_A d_A + n_B d_B + n_C d_C + n_B d_B)} \cong 1/6;$$

$$f_C = \frac{n_C d_C}{(n_A d_A + n_B d_B + n_C d_C + n_B d_B)} \cong 1/3; \text{ and}$$

$$\lambda_0 = 2(n_A d_A + n_B d_B + n_C d_C + n_B d_B) = 1.35\ \mu m.$$

Using the materials and relationships set forth above a reflectance spectrum was simulated for this three component multilayer film indicator having ABCB repeating units, according to a program developed using a MATHCAD 2.0 software package from Mathsoft, Inc., Cambridge, Mass., and run on an IBM AT personal computer with a 640K base memory and a 1 MB expanded memory.

The computer programmer relied on the matrix method discussed extensively in A. Vasicek, "Optics of Thin Films", North Holland Publishing Co., Amsterdam (1960), and used the equations developed therein and in H. A. MacLeod, "Thin-Film Optical Filters", Adam Hilger, London (1969), and in M. Born and E. Wolf, "Principles of Optics", Pergamon Press (1965). The refractive indices of the A, B and C materials were supplied to the model, as was the number of repeating ABCB series or periods of the film to be modeled. The angle of incidence of incoming light, the various optical thickness ratios f_A, f_B and f_C, the wavelength corresponding to a desired primary reflectance, and the wavelength range for simulation were also supplied.

Fig. 4 shows the above-mentioned simulated reflectance spectrum constructed by this method with the polystyrene, styrene-methylmethacrylate and polymethylmethacrylate system, and confirms that a selection of the f-ratios and refractive indices for a three component indicator which are set forth above will suppress the second, third and fourth orders of reflection and result in an indicator which in an initial condition is substantially colorless.

The same model also simulated the reflectance spectra of two component film indicators constructed as in Example 1 to confirm the general accuracy of the model's simulated spectra by comparison to those seen in actual practice. The simulated spectra corresponded well with those seen in actual practice and show a suppression of the second order reflection where f_A is approximately equal to 0.5, see Fig. 5.

Examples 1 and 2 demonstrate that the two and three component multilayer film indicators which have been described herein as preferred are suitable for accomplishing the purposes and realizing the advantages of the present invention.

WHAT IS CLAIMED IS:

1. A multilayer optical interference film indicator which in an initial condition is substantially colorless, but which when strained to a selected extent will produce visible evidence of such strain in the form of iridescence or interference colors, comprising contiguous layers of two or more diverse thermoplastic materials of differing refractive indices, wherein the yield point of at least one of the diverse materials has been at least equalled when the visible evidence is produced.

2. An indicator as defined in Claim 1, wherein the film indicator will rupture with a degree of additional strain while the evidence is still visible.

3. An indicator as defined in Claim 1, wherein the refractive index corresponding to a layer of each of the two or more materials differs from the refractive indices of immediately adjacent layers by 0.03 or greater.

4. An indicator as defined in Claim 1, consisting of at least one period AB of layers of two diverse thermoplastic materials A and B, and having

average layer thicknesses d_A and d_B, respectively, wherein A possesses a refractive index n_A and B a refractive index n_B, and further wherein the ratio

$$f_A = \frac{n_A d_A}{(n_A d_A + n_A d_A)}$$ is approximately 1/2.

5. An indicator as defined in Claim 4, wherein a substantial majority of said layers of said two diverse thermoplastic materials A and B have an optical thickness of 0.17 µm or greater when the indicator is in the initial condition.

6. An indicator as defined in Claim 1, further including outer skin layers which are transparent to visible light.

7. An indicator as defined in Claim 1, consisting of at least one series ABCB of layers of three diverse thermoplastic materials A, B and C having average layer thicknesses d_A, d_B and d_C, respectively, wherein A possesses a higher refractive index n_A, B an intermediate refractive index n_B, and C a lesser refractive index n_C, and wherein:

the ratio $$f_A = \frac{n_A d_A}{(n_A d_A + n_B d_B + n_C d_C + n_B d_B)}$$

has a value of approximately 1/3;

the ratio $$f_B = \frac{n_B d_B}{(n_A d_A + n_B d_B + n_C d_C + n_B d_B)}$$

has a value of approximately 1/6;

the ratio $f_C = \frac{n_C d_C}{(n_A d_A + n_B d_B + n_C d_C + n_B d_B)}$ has a value of approximately 1/3; and

$$n_B \cong \sqrt{n_A n_C} .$$

8. An indicator as defined in Claim 7, wherein a substantial majority of the layers of A, B and C have an optical thickness of .09 µm or greater when the indicator is in the initial condition.

9. An indicator as defined in Claim 7, wherein A, B and C respectively include a polystyrene of refractive index n_A of 1.59, a styrene-methylmethacrylate copolymer of refractive index n_B of 1.54, and a polymethylmethacrylate with a refractive index n_C of 1.49.

10. An indicator as defined in Claim 1, wherein the cross-sectional area of the indicator is reduced at one or more locations.

11. An indicator as defined in Claim 10, wherein the indicator has been perforated at said one or more locations.

12. An indicator as defined in Claim 1, wherein the highest and lowest refractive indices corresponding to the contiguous layers of the two or more diverse thermoplastic materials differ by 0.03 or greater.

13. A tamper evident, tamper resistant packaging material which includes an indicator as defined in any of Claims 1-12.

14. A packaging material as defined in Claim 13 which is heat-shrinkable.

15. An article which has been at least partially packaged within an indicator as defined in any of Claims 1-12, and wherein the indicator has been joined to two portions of the article which are movable with respect to one another.

16. An article which comprises a membrane seal, wherein the membrane seal includes a multilayer film indicator as defined in any of Claims 1-12.

17. A multilayer film indicator which when strained to a selected extent will produce a signal color change evidencing such strain, including contiguous layers of two or more diverse thermoplastic materials of differing refractive indices encapsulated within opposing outer skin layers, wherein the skin layers adjacent at least a first side of the indicator are transparent, and further wherein the yield point of at least one of the diverse materials and the skin layers has been at least equalled when the color change is produced.

18. An indicator as defined in Claim 17, consisting essentially of at least one series AB of layers of a polyurethane of a refractive index n_A of 1.56 and of a polyether amide of a refractive index n_B of 1.49 with layers of the polyurethane immediately adjacent the opposing outer skin layers, and having average layer thicknesses d_A and d_B, respectively, corresponding to the layers of the polyurethane and of the polyether amide, wherein the ratio

$$f_A = \frac{n_A d_A}{(n_A d_A + n_B d_B)}$$ is approximately 1/2 and

wherein the skin layers are comprised of a material whose yield point has been at least equalled when the color change is produced.

19. A process for protecting the contents of a container from adulteration and for otherwise ensuring the integrity and authenticity of such contents, including the step of wrapping the container in its entirety or in significant part with the tamper evident, tamper resistant packaging material of claim 13.

FIG. 1

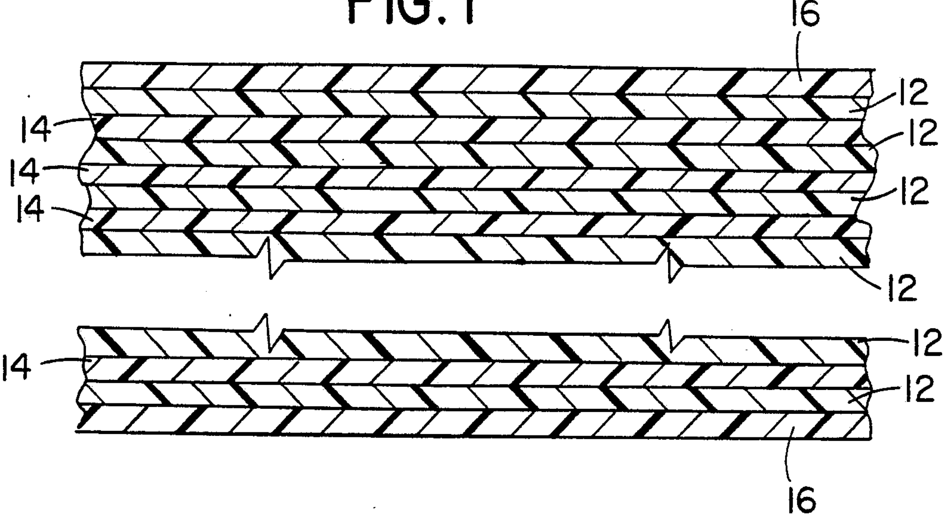


FIG. 2

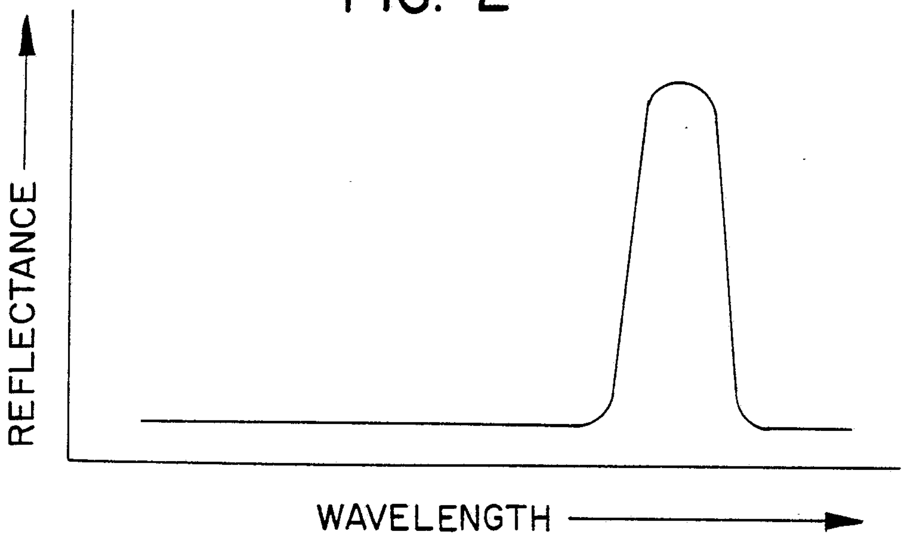
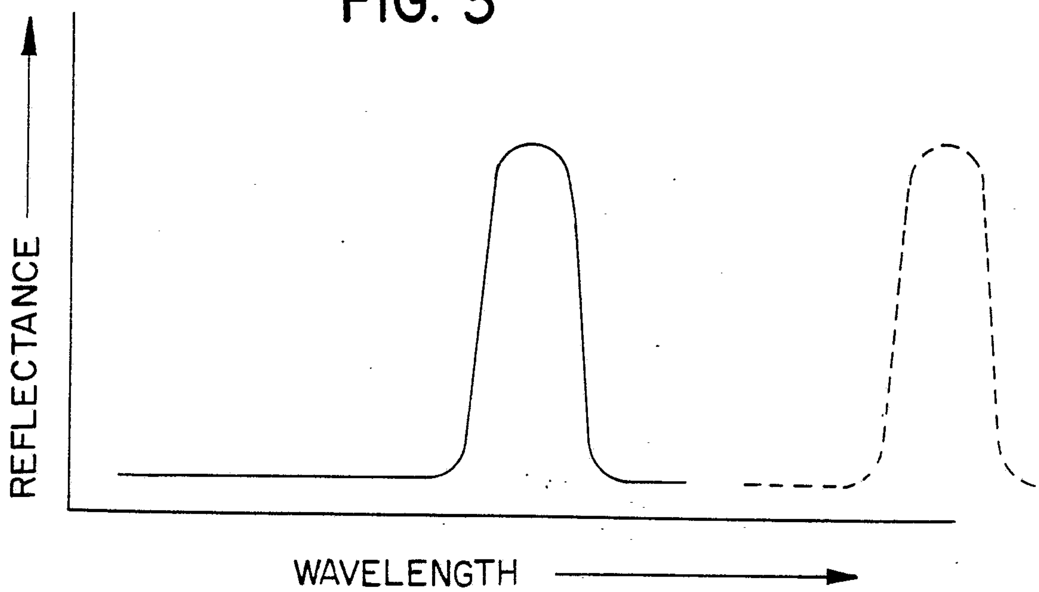
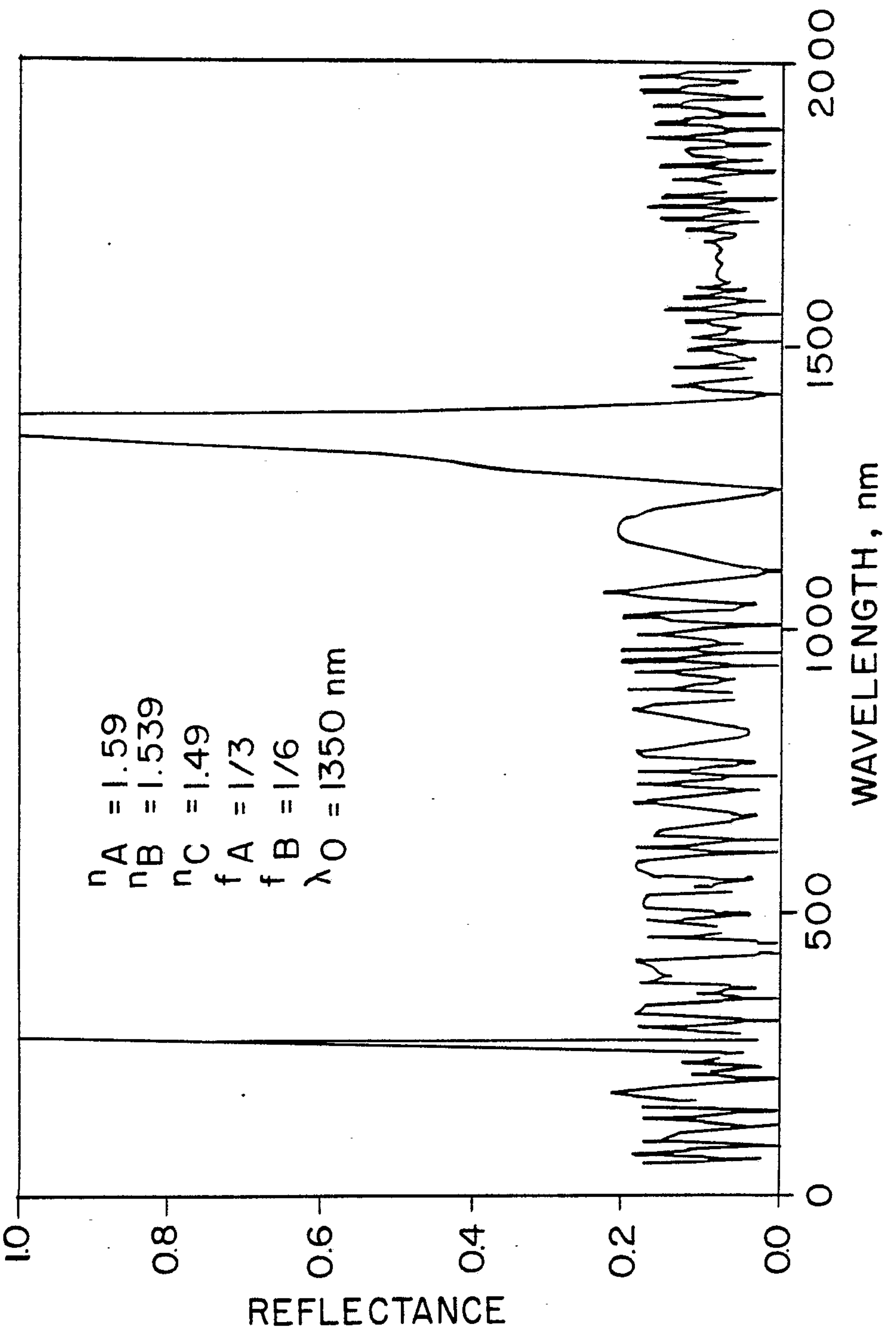


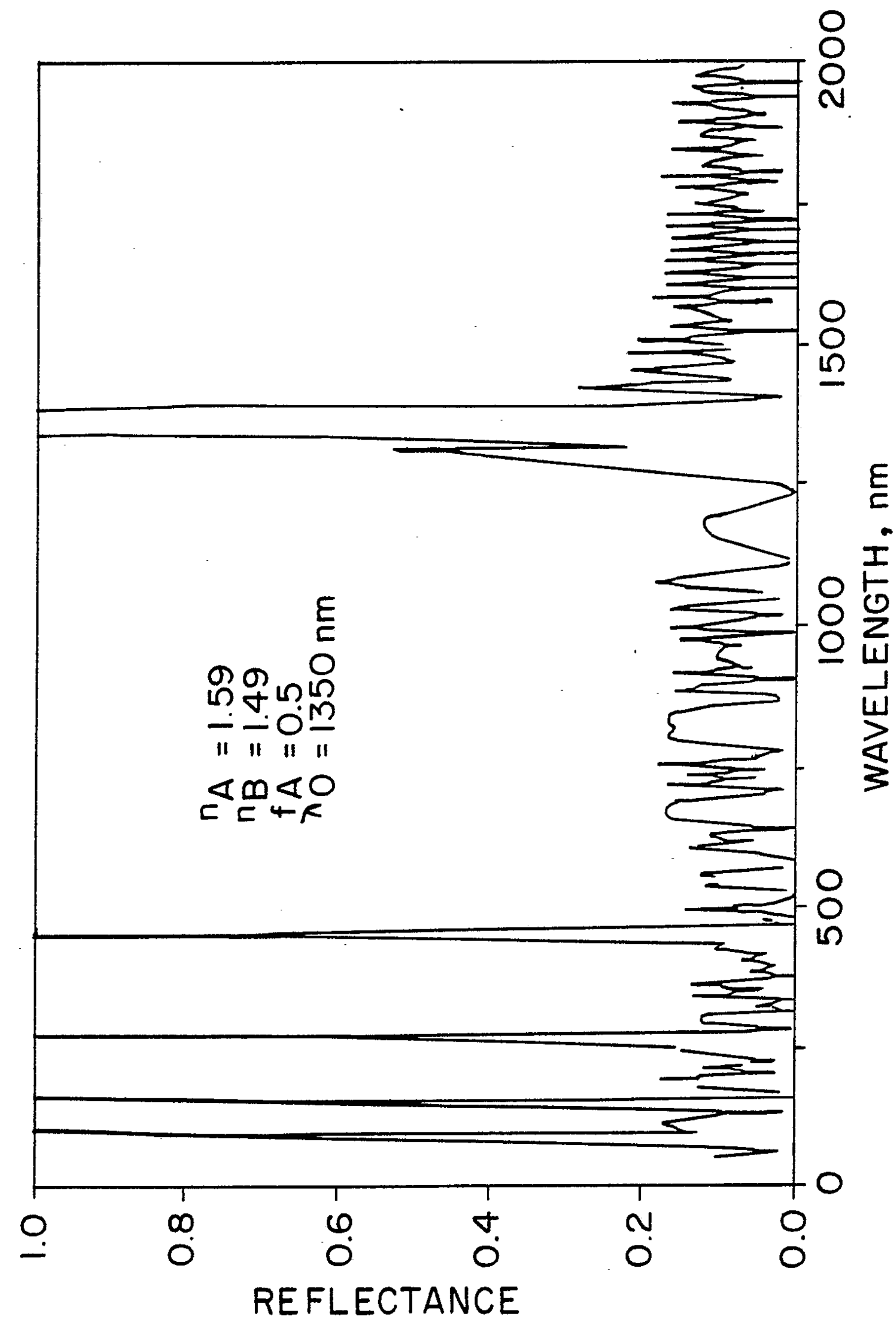
FIG. 3





Reflectance Spectrum of Three-Component 401-Layer Film

FIG. 4



Reflectance Spectrum of 201-Layered Two-Component Film

FIG. 5

INTERNATIONAL SEARCH REPORT

International Application No. PCT/US90/07523

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [6]

According to International Patent Classification (IPC) or to both National Classification and IPC

IPC (5): B32B 7/02, 27/08; B65D 53/02; B65B 11/02; G02B 5/28
U.S. CL. 428/131, 156, 212

II. FIELDS SEARCHED

Minimum Documentation Searched [7]

Classification System	Classification Symbols
U.S.	428/131, 156, 212, 213, 215, 423.1, 480, 913, 916 428/918; 350/404, 1.6, 164, 165; 215/246 206/807; 53/442

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [8]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [9]

Category *	Citation of Document, [11] with indication, where appropriate, of the relevant passages [12]	Relevant to Claim No. [13]
Y	US, A, 4,310,584 (COOPER) 12 JANUARY 1982 See column 1, lines 53-70 and column 2, lines 1-45.	7, 8 & 9
X,P	US, A, 4,937,134 (SCHRENK) 26 JUNE 1990 See column 9, lines 15-35 and column 2, lines 42-48.	1-6, 12, 13, 17, 18 & 19
A	US, A, 3,247,392 (THELEN) 19 APRIL 1966	
A	US, A, 4,018,640 (AMBERG) 19 APRIL 1977	
A	Journal of the Optical Society of America Vol. 53, pages 1266-1270 (1963), THELEN, "Multilayer Filters With Wide Transmittance Bands".	
A	US, A, 4,516,679 (SIMPSON) 14 MAY 1985	
A	US, A, 4,837,061 (SMITS) 06 JUNE 1989	
A	US, A, 4,721,217 (PHILLIPS) 26 JANUARY 1988	

* Special categories of cited documents: [10]

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"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)

"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

"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

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"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.

"&" document member of the same patent family

IV. CERTIFICATION

Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report
11 FEBRUARY 1991	04 MAR 1991
International Searching Authority	**Signature of Authorized Officer**
ISA/US	William Watkins

International Application No. PCT/US90/07523

III DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category *	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No
A	POLYMER ENGINEERING AND SCIENCE, MAY 1973, Vol. 13, No. 3, pages 216-221 (ALFREY, JR.), "Reflectivity of Iridescent Coextruded Multilayered Plastic Films".	