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(54) Title: LIGHT STABLE COLOR COMPOSITIONS (57) Abstract A composition comprising colorant-containing particles having a coating comprising propylene glycol and a carrier component selected from the group consisting of carriers suitable for food, drug and cosmetic preparations and mixtures thereof.		

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Description

Light Stable Color Compositions

Specification

This patent application is a continuation-in-part of United States Patent Application Serial No. 08/018,089, filed February 16, 1993, the disclosure of which is incorporated by reference herein.

5 The present invention relates to color compositions which have improved light stability and sun protection factor (SPF) properties. More specifically, the compositions of the present invention have an outer protective layer which absorbs ultraviolet light and
10 thereby prevents the fading or discoloration of the colorant-containing particles. The outer layer further acts to impart an improved sunburn protection factor to the novel compositions of the invention.

15 Background of the Invention

Color compositions are widely used in the cosmetic industry principally for aesthetic purposes. Accordingly, it is important that color compositions remain stable, i.e., not fade or discolor. One
20 principal cause for discoloration of color compositions used in the cosmetic field is the tendency of colorants to absorb light, principally ultraviolet light, which results in the degradation of the colorant molecules.

In the past, efforts to improve the light
25 stability of color compositions have involved generally the use of ultraviolet light absorbing compositions, such as benzophenones, and/or antioxidants, such as butylated-hydroxytoluene (BHT) or butylated hydroxyanisol (BHA).

30 An object of the present invention is to provide compositions comprising colorant-containing particles

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with improved light stability properties so that these compositions as well as cosmetic products which include them are less susceptible to fading or discoloration.

Another object of the present invention is to
5 provide color compositions having an improved sunburn protection factor (SPF).

Yet a further object of the invention is to provide a process for making color compositions which have improved light stability and SPF properties.

10 These and further objects and advantages of the present invention will, upon reading the following specification, become apparent to those skilled in the field of color compositions.

15 Summary of the Invention

It has been found that the objects and advantages described above are realized by compositions comprising colorant-containing particles which are coated with a composition comprising a suitable carrier component,
20 such as titanium dioxide (TiO_2), and a propylene glycol component. This coating renders colorant-containing substrate particles resistant to degradation by ultraviolet light and is therefore particularly useful for light sensitive colorants. Although carriers other
25 than TiO_2 may be used as a coating component, the invention will be primarily described with reference to TiO_2 . Accordingly, one aspect of the present invention is the novel color compositions comprising colorant-containing particles coated with titanium dioxide and
30 propylene glycol. Another aspect of the invention is the novel process for preparing the coated color containing particles. A further aspect of the invention is the novel use of carriers such as TiO_2 , in combination with propylene glycol to impart the
35 property of improved light stability to the colorant-containing particles. And yet a further aspect of the

invention is the novel use of TiO_2 and propylene glycol to impart an improved SPF property to compositions comprising colorant-containing particles. A still further aspect of the invention is the use of novel
5 substrate molecules, including but not limited to starch and mica, in coated colorant-containing particles. Yet another aspect of the invention is the use of propylene glycol to improve the SPF of micronized TiO_2 .

10

Brief Description of the Drawings

Figure 1 shows SPF results for micronized TiO_2 .

Figure 2 shows SPF results for micronized TiO_2 and 5% by weight of propylene glycol.

15

Figure 3 shows color spectrophotometric measurements for FD&C blue #1 dye laked onto starch (B1) or aluminum hydrate (B2).

Figure 4 shows color spectrophotometric measurements for FD&C green #5 laked onto starch (G1) or
20 aluminum hydrate (G2).

Figure 5 shows color spectrophotometric measurements for D&C red #28 laked onto starch (R1) or aluminum hydrate (R2).

25

Detailed Description of the Invention

The present invention has been found to significantly improve both light stability and SPF properties of compositions comprising colorant-containing particles which are unstable in ultraviolet light.

30

While such color compositions are used in a wide number of fields, e.g., paints and cosmetics, the following discussion will primarily be addressed to compositions for use in the cosmetic field; notwithstanding the fact that application of this invention embraces any and all
35 fields where the stability of color compositions is desirable.

In accordance with the present invention, colored compositions are prepared by coating colorant-containing particles with a composition comprising propylene glycol and carrier particles.

5 As used herein, the term colorant-containing particles refers to any particle which imparts color useful in cosmetic, paint or similar compositions and particularly those which are light sensitive, i.e., susceptible to fading or discoloration. While
10 colorant-containing particles include particles of inorganic pigments, e.g., iron oxides, the preferred colorant-containing particles comprise organic dyes laked onto a suitable substrate such as TiO_2 , and other such FD&C and D&C approved substrates. Specifically
15 the dyes include, but are not limited to, the well-known FD&C and D&C dyes, for example, FD&C Blue No. 1, FD&C Yellow No. 5, FD&C Green No. 5, Brown 13693, carmoisine edicol, FD&C Red No. 40, D&C Red 21, D&C Red 27 and D&C Red 28.

20 Hereinafter the colorant, unless indicated otherwise, will refer to FD&C and D&C dyes laked on to a suitable substrate in accordance with conventional dilution or slurry methods.

 FD&C, D&C and External D&C dyes may be used to
25 make insoluble lake pigments. The formation of a lake pigment requires three basic conditions; namely, (i) a water soluble dye; (ii) a suitable substrate; and (iii) a suitable precipitating agent. The laking process involves the following steps: (1) addition of a
30 substrate; (2) addition of a suitable dye; (3) absorption of the dye by the substrate; (4) addition of a metal salt; and (5) formation and precipitation of a lake pigment. Once the laking process is completed, the water soluble dyes are converted to water insoluble
35 pigments. The water soluble dye will impart the basic

color hue. The substrate will influence the transparency, opacity, brightness, etc.

While TiO_2 is a preferred substrate onto which the dye or suitable mixtures thereof is absorbed, other
5 substrates include all FD&C and D&C approved substrates e.g., alumina, zinc oxide, aluminum benzoate, calcium carbonate, and those listed in the Code of Federal Regulations, Food and Drugs, paragraph 82.1051, subpart C - Drugs and Cosmetics, or combinations of such
10 substrates, as well as, according to the present invention, substrates which have not yet been approved but which exhibit suitable physical properties, such as mica or starch.

It has been found that when FD&C or D&C dyes were
15 laked on to a starch substrate, pigment laked with more transparency, better light reflectance, higher color value, better feel and good light stability. Suitable starches include, but are not limited to, food grade starches such as starch derived from cornmeal and/or
20 tapioca. Suitable dyes include, but are not limited to, FD&C Blue No. 1, D&C Red No. 28, FD&C Green No. 5, and FD&C Yellow No. 5. Although not yet an FDA approved substrate, the favorable physical properties of starch indicate that it may be desirable to petition
25 the FDA to approve its use in cosmetics and drugs. Alternatively, mica may be used as substrate.

Colorant-containing particles can be formed by absorbing an organic dye onto substrate particles which are preferably in the form of micronized particles
30 (average particle size of .05 to .4 microns). The resulting particles are then advantageously precipitated, dried and crushed to form a fine powder. The particle size depends on the type of pulverizing equipment used; however a particle size of about .2 to
35 .3 microns is preferred and is obtained by using a Model No. 2D4 Pulverizer. TiO_2 is a preferred substrate

for the colorant because of its superior sunscreen properties. However, since TiO_2 absorbs ultraviolet radiation, its use contributes to the degradation of the absorbed colorant (dye) which results in its fading and/ or discoloration.

The light stability of colorants, particularly those absorbed on a TiO_2 substrate, is remarkably improved by coating the colorant particle with a coat comprising a carrier component, such as TiO_2 , and propylene glycol. While the carrier TiO_2 is a preferred component of the coating composition, other suitable coating components include carriers such as zinc oxide and aluminum hydroxide. The carrier component is preferably micronized, i.e., having a particle size in the range of about .1 to about .3 microns. While these are the generally preferred parameters for the particle size of the carrier component, non-micronized particles may be used; however, the size of particles in the coating must, of course, be substantially smaller than the size of the colorant-containing particle onto which it is to be coated.

In addition to the carrier component, the coating composition further comprises propylene glycol. As will be apparent from the examples set forth below, the propylene glycol is preferably sprayed onto a homogeneous mixture of the colorant-containing particles and the micronized carrier. Nevertheless, however the propylene glycol is added to the mixture, it is desirable that the homogeneous mixture is uniformly contacted with propylene glycol and that the particles do not agglomerate when contacted with the propylene glycol. By adding to the colorant-containing particles a coating comprising a micronized carrier or mixtures thereof and the propylene glycol component, the coating (i) acts as a protective barrier against radiation (which causes fading and discoloration)

reaching the colorant-containing particles and (ii) increases the sun protection factor (SPF) of compositions containing the particles.

The coating composition used in the present invention preferably contains the propylene glycol and the carrier component in a ratio of from about 1:4 to about 1:5 by weight. However, other suitable ratios of propylene glycol and carrier may be used. The ratio of coating composition to colorant-containing particle used to coat colorant-containing particles depends on the shade of the colorant which is desired, i.e., the more coating that is used the paler the shade of the colorant. The size of the resulting coated colorant-containing particles depends on both the initial particle size and the coating thickness. Nevertheless, generally final particle sizes of about 1 micron or less are suitable and micronized particle sizes are preferred especially for cosmetic uses, i.e., particularly for use as a sunscreen factor. The present invention also provides for a method of improving the sun protection factor of a composition comprising sunscreen grade TiO_2 (micronized TiO_2 with a particle size of 1 micron or less) comprising coating the TiO_2 particles with propylene glycol as well as providing for compositions produced by this method.

In specific, non-limiting embodiments of the present invention, the photostability of FD&C and D&C dyes laked onto a substrate particle can be significantly improved by contacting the laked substrate particle with a coating containing propylene glycol as well as particles of the compound used as substrate, in uncolored form and at a suitable size. For example, and not by way of limitation, the photostability of dye laked on TiO_2 substrate particles may be improved by coating the dye-laked TiO_2 particles with uncolored TiO_2 particles and propylene glycol; the

photostability of dye laked on zinc oxide substrate particles may be improved by coating the dye-laked zinc oxide particles with uncolored zinc oxide particles and propylene glycol; and the photostability of dye laked
5 on starch or mica, may be improved by coating the dye-laked starch or mica particle with uncolored starch or mica particles and propylene glycol.

The following examples are provided in order to disclose the invention in the fullest possible extent
10 and in the best mode known. Nevertheless they are to be construed as purely illustrative and not limiting of the present invention.

Example 1

15 Procedure

In order to make 908 gms. of uncoated light stable micronized TiO_2 colored with FD&C Blue No. 1, the following procedure was followed:

The color load chosen was 1% by weight.

20 The 9.08 gms. of FD&C Blue No. 1 were dissolved in 3,500 ml of water. The solution was mixed well, until all the dye was completely dissolved. In a separate container, 10,000 ml of water was added and vigorously mixed, so as to slowly disperse 726 gms. of micronized
25 TiO_2 having an average particle size of .1 microns. The dispersion was mixed for 15 minutes until dispersed completely using Yamato LR-41A Labo-Stirrer. Thereafter the dye solution (FD&C Blue No. 1) was added to the TiO_2 dispersion while mixing, and then continued
30 to be mixed for 30 minutes to allow all the color to be absorbed by the TiO_2 particles. The volume was thereafter increased to 16,000 mls by the addition of tap water. The colorant-particle solution was then precipitated out by using an AlCl_3 solution, 32° Bé,
35 diluted 1:1 with water.

After adding the AlCl_3 solution, the pH was checked and if necessary adjusted to be 3.0 ± 0.3 with AlCl_3 . Also, the bleed was checked, i.e., to determine the absence of unprecipitated dye, and once it was
5 determined to be negative, the precipitant was filtered using a Buchner filtration apparatus with a Whatman qualitative #5 filter. The filtered product was dried in a Lindberg/Bleum laboratory oven Model No. SW-17TA-1 at a temperature between $70^\circ\text{--}80^\circ\text{C}$. When fully dried,
10 i.e., all moisture absent, the product was crushed and pulverized in a pulverizer Model No. 204 to a fine particle size having an average of about .2 to .3 microns.

The uncoated color particles were then placed in a
15 Waring CB6 blender (Model No. 34 BL 22) and 182 gms. of micronized TiO_2 were added. The mixture was blended well until homogeneous and uniform. While blending, 136.2 gms of propylene glycol were uniformly sprayed (using a True-Temper pressure sprayer Model No. HS-200)
20 onto the batch carefully so as to avoid any agglomeration of the particles and the batch was well blended until homogenous and uniform.

Example 2

25 An alternative method of preparing uncoated particles of the colored TiO_2 lake is the slurry method. According to this method, 310 gms. of micronized TiO_2 (having an average particle size of about .1 microns) are dispersed in 500 gms. of water using a homogenizer.
30 This mixture is homogenized until a low to medium viscosity suspension is obtained.

2.5 g. of a suitable FD&C (or D&C) dye is dissolved in 50 gms. of water, added to the blend, and mixed using a paddle mixer for 30 minutes until all the
35 dye is absorbed by the TiO_2 .

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The dye is then precipitated with an AlCl_3 solution. Once the bleed is determined to be negative, the slurry is transferred and dried at $70^\circ\text{--}80^\circ\text{C}$. When all the water has evaporated, the product is crushed and pulverized to a small particle size (having an average size of about .2 to .3 microns). Thereafter a micronized TiO_2 /propylene glycol coat is added, first by adding 55.14 g. micronized TiO_2 (average particle size .1 microns) and, once homogeneous and uniform, 18.38 g. propylene glycol is uniformly sprayed onto the batch and the batch is mixed well until uniform. The light stability of the product made with the slurry method is similar to that obtained in Example 1.

15 Example 3

Samples of both the uncoated colorant particles and coated colorant particles prepared in accordance with Example 1 were placed in 1 oz. clear plastic vials and exposed for 24 hours to the following sources of radiation:

- shortwave ultraviolet
- longwave ultraviolet
- white light 3600° Kelvin (approximate)
- north sky light 7000° K (approximate)

25 After the 24 hour exposure, samples of the uncoated colorant particles showed, in all cases, complete fading and developed an off-white to gray-white color. The samples of the coated colorant particles, i.e., those which were coated with the micronized TiO_2 and propylene glycol showed none to slight color loss, maintaining the characteristics of their original color.

35 Additional tests were performed using other dyes such as FD&C Yellow No. 5, Brown 13693, and carmoisine edicol, and all showed the same results as the FD&C Blue No. 1.

Example 4

The SPF effectiveness of the micronized TiO_2 /propylene glycol coated titanium dioxide particles was established by comparing the following two samples:

- 5 - micronized TiO_2 (Sample 1)
- micronized TiO_2 and 5% by weight of propylene glycol (Sample 2)

The latter sample was prepared by spraying 95 g. of micronized TiO_2 (average particle size of .1 micron) with 5 g. of propylene glycol and mixing the sprayed TiO_2 particles well to insure a homogeneous mixture.

A 20% dispersion of each of the above samples in isopropyl myristate was prepared. Each sample was thereafter measured to determine its SPF (sun protection factor) using a SPF-290 spectrophotometer (Optometrics USA). The results are shown in Figures 1 and 2. Figure 1 shows SPF results for sample 1. Figure 2 shows SPF results for sample 2.

By comparing the two graphs, it was concluded that the addition of 5% propylene glycol to the TiO_2 particles significantly increased the SPF value of the TiO_2 when used as a sunscreen agent. It should be noted that during the measurement, six runs were taken on each sample. Because of the fact that for the TiO_2 /propylene glycol (Sample 2) the average was off the instrument's scale, runs 2, 4, and 6 were excluded in order to plot a curve within the instrument's capability (maximum 99.99).

Example 5

The following experiments were performed in order to evaluate the usefulness of starch as a substrate for laked pigment.

Blue, green, or red dye was laked onto either starch or aluminum hydrate substrates, and the quality

of color associated with the laked pigments were compared spectrophotometrically.

In the data presented in Tables I, II and III and Figures 3, 4 and 5, the following designations are used for pigment/substrate combinations. FD&C blue No. 1 laked on starch is designated B1 and FD&C blue No. 1 laked on aluminum hydrate is designated B2. FD&C green No. 5 laked on starch is designated G1 and FD&C green No. 5 laked on aluminum hydrate is designated G2. D&C red No. 28 laked on starch is designated R1 and D&C red No. 28 laked on aluminum hydrate is designated R2.

Dyes were laked on substrate at 10% load. Using a color spectrophotometer, measurements were taken of a color film made by dispersing 10% laked colors in an acrylic polymer solution.

Table IA, Table IIA and Table IIIA present the spectrophotometer measurements for blue, green, and red dyes, respectively, laked onto either starch or aluminum hydrate. These results are graphically depicted in Figs. 3A, 4A and 5A respectively.

Tables IB, IIB and IIIB show the differences in color value for the various pigments laked onto starch or aluminum hydrate. For example, in Table IB the Delta E value 7.11 indicates that blue dye laked onto starch exhibited 7.11 units more color than blue dye laked onto aluminum hydrate. Similarly, the Delta E value in Table IIB indicates that green dye laked onto starch exhibited 5.15 units more color than green dye laked onto aluminum hydrate, and the Delta E value in Table IIIB indicated that red dye laked onto starch exhibited 6.15 units more color than red dye laked onto aluminum hydrate. The values presented in Tables IB, IIB and IIIB are graphically depicted in Figs. 3B, 4B and 5B respectively.

In summary, these experiments demonstrated the following. Blue dye laked onto aluminum hydrate was

darker, less green, and less blue than blue dye laked onto starch. Green dye laked onto aluminum hydrate was lighter, less green and less blue than green dye laked onto starch. Red dye laked onto aluminum hydrate was lighter, less red and bluer than red dye laked onto starch. Accordingly, pigments laked onto starch appeared to be brighter and more intense than those same pigments laked onto aluminum hydrate.

1

TABLE I.

2	(a)	d/8°	B1	B2	(b)	B1	B2
3		400	5.62	5.01	X	9.13	7.84
4		420	6.04	5.25	Y	12.62	10.20
5		440	24.03	18.88	Z	33.11	27.92
6		460	44.19	38.56	X	0.1664	0.1706
7		480	41.67	35.58	Y	0.2300	0.2219
8		500	31.89	25.17			
9		520	19.13	13.47	L*	42.18	38.20
10		540	8.66	6.31	a*	-21.63	-15.77
11		560	4.72	4.35	b*	-34.83	-34.23
12		580	4.11	3.95			
13		600	3.99	3.81	D10°	ΔL^*	-3.98
14		620	3.97	3.73		Δa^*	5.86
15		640	4.10	3.77		Δb^*	0.60
16		660	4.35	3.97		ΔC^*	-3.31
17		680	4.98	4.32		ΔE^*	7.11
18		700	19.68	9.61	*REJECT*		

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TABLE II.

2	(a)	d/8°	G1	G2		G1	G2
3		400	16.63	16.73	X	19.19	19.87
4		420	18.29	18.11	Y	28.32	28.49
5		440	24.70	23.19	Z	35.38	32.34
6		460	38.12	33.97	x	0.2315	0.2462
7		480	56.04	44.53	y	0.3416	0.3530
8		500	50.55	47.06			
9		520	41.58	40.70	L*	60.18	60.33
10		540	31.32	32.02	a*	-34.77	-32.00
11		560	22.94	24.62	b*	-6.83	-2.49
12		580	17.48	19.27			
13		600	13.12	15.26	D 10°	ΔL^*	0.15
14		620	12.17	13.50		Δa^*	2.77
15		640	12.05	13.14		Δb^*	4.34
16		660	12.48	13.57		ΔC^*	-3.34
17		680	19.15	16.97		ΔE^*	5.15
18		700	36.57	26.03	*REJECT*		

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TABLE III.

2	(a)	d/8°	R1	R2	(b)	R1	R2
3		400	30.59	36.37	X	39.08	44.91
4		420	37.91	46.95	Y	22.05	26.61
5		440	39.97	48.91	Z	34.69	44.18
6		460	32.81	42.16	x	0.4078	0.3882
7		480	17.07	26.04	y	0.2302	0.2300
8		500	8.03	13.41			
9		520	4.82	7.22	L*	54.08	58.62
10		540	4.34	5.43	a*	70.02	68.15
11		560	4.44	5.04	b*	-16.43	-20.14
12		580	16.99	26.62			
13		600	65.44	74.18	D 10°	ΔL^*	4.53
14		620	78.88	83.90		Δa^*	-1.87
15		640	82.78	86.30		Δb^*	-3.71
16		660	84.73	87.08		ΔC^*	-0.86
17		680	85.47	87.20		ΔE^*	6.15
18		700	86.13	87.50	*REJECT*		

Claims

- 1 1. A composition comprising colorant-containing
2 particles having a coating comprising propylene
3 glycol and a carrier component selected from the
4 group consisting of carriers suitable for food,
5 drug and cosmetic preparations and mixtures
6 thereof.
- 1 2. A composition according to claim 1 wherein the
2 colorant-containing particles comprise organic
3 dyes laked on a suitable substrate.
- 1 3. A composition according to claim 1 wherein the
2 dyes are suitable for use in food, drug and
3 cosmetic preparations.
- 1 4. A composition according to claim 1 wherein the
2 colorant in the colorant-containing particles is
3 susceptible to ultraviolet degradation.
- 1 5. A composition according to claim 1 wherein the
2 colorant-containing particles comprise an
3 inorganic pigment or mixtures thereof.
- 1 6. A composition according to claim 1 wherein the
2 carrier component of the coating is titanium
3 dioxide.
- 1 7. A composition according to claim 6 wherein the
2 average size of the carrier component of the
3 coating ranges from about .1 to about .3 microns.
- 1 8. A composition according to claim 1 wherein the
2 average particle size of the coated colorant-

3 containing particle is about 0.5 to about .4
4 microns.

1 9. A process for preparing a composition comprising
2 colorant-containing particles, said process
3 comprising the steps of: mixing colorant-
4 containing particles with particles of a carrier
5 selected from the group consisting of carriers
6 suitable for food, drug and cosmetic preparations
7 and mixtures thereof; and contacting the resulting
8 mixture of particles with propylene glycol whereby
9 colorant-containing particles coated with the
10 carrier and propylene glycol are formed.

1 10. A process according to claim 9 wherein the
2 colorant-containing particles are an inorganic
3 pigment or mixtures thereof.

1 11. A process according to claim 9 wherein the
2 colorant is laked onto a substrate.

1 12. A process according to claim 9 wherein the
2 colorant is suitable for use in food or drug or
3 cosmetic preparations.

1 13. A process according to claim 9 wherein the
2 colorant-containing particles have an average
3 particle size ranging from about .05 to .4 prior
4 to coating.

1 14. A process according to claim 9 wherein the
2 colorant-containing particles are mixed with
3 micronized particles of the carrier to form a
4 substantially homogenous blend.

- 1 15. A process according to claim 15 wherein the
2 carrier is micronized titanium dioxide having a
3 particle size ranging from about 0.1 to 0.3
4 microns.
- 1 16. A process according to claim 15 wherein the
2 propylene glycol is sprayed onto the mixture of
3 colorant-containing particles and micronized
4 carrier.
- 1 17. A method of improving the stability of colorant-
2 containing particles which are degraded by
3 ultraviolet light, comprising contacting said
4 particles with a coating comprising propylene
5 glycol and a carrier component selected from the
6 group consisting of carriers suitable for food,
7 drug and cosmetic preparations and mixtures
8 thereof.
- 1 18. A method of improving the sun protection factor
2 (SPF) of a cosmetic composition comprising a
3 colorant-containing particle, said method
4 comprising coating said colorant-containing
5 particle with propylene glycol and a carrier
6 selected from the group consisting of carriers
7 suitable for food, drug and cosmetic preparations
8 and mixtures thereof.
- 1 19. A method according to claim 19, wherein the
2 carrier is titanium dioxide.
- 1 20. A sunscreen comprising micronized TiO_2 and
2 propylene glycol.
- 3 21. A method of improving the photostability of a dye
4 suitable for food, cosmetic or drug preparations,

5 laked onto a particle consisting essentially of a
6 substrate compound suitable for food, cosmetic, or
7 drug preparations in a dye-laked particle, com-
8 prising contacting the dye-laked particle with a
9 coating comprising propylene glycol and uncolored
10 particles of the substrate compound.

11 22. The method of claim 21 in which the substrate
12 compound is titanium dioxide.

13 23. The method of claim 21 in which the substrate
14 compound is zinc oxide.

15 24. The method of claim 21 in which the substrate
16 compound is mica.

17 25. The method of claim 21 in which the substrate
18 compound is starch.

19 26. A method of improving the sun protection factor of
20 a composition comprising micronized titanium
21 dioxide particles comprising coating the titanium
22 dioxide particles with propylene glycol.

23 27. The composition of claim 2 in which the substrate
24 is selected from the group consisting of starch
25 and mica.

26 28. The process of claim 11 in which the substrate is
27 selected from the group consisting of starch and
28 mica.

29 29. The method of claim 17 in which the particles
30 comprise a substrate selected from the group
31 consisting of starch and mica.

-21-

32 30. The method of claim 18 in which the particles
33 comprise a substrate selected from the group
34 consisting of starch and mica.

35 31. The method of claim 21 in which the substrate is
36 selected from the group consisting of starch and
37 mica.

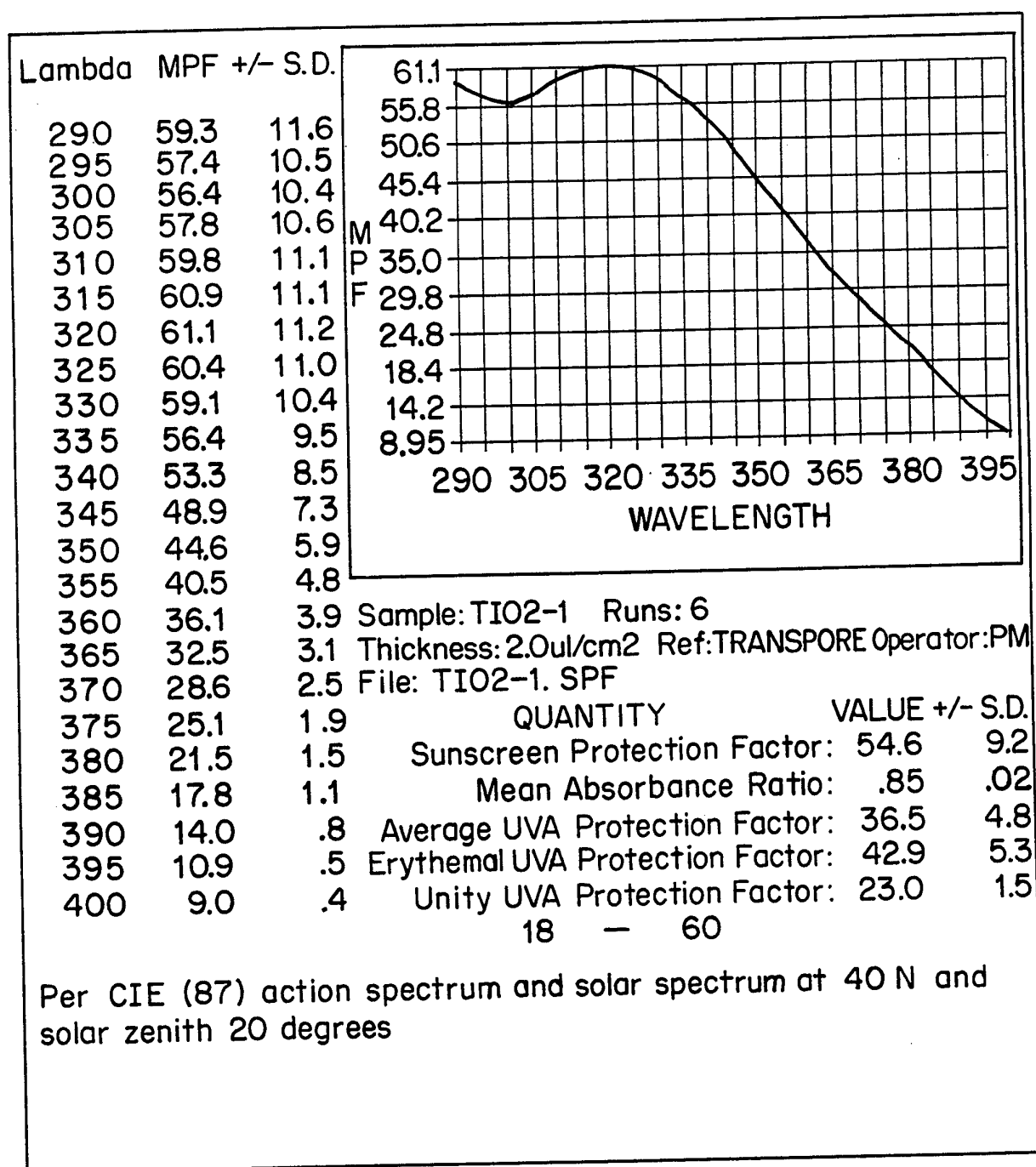


FIG. 1

2 / 5

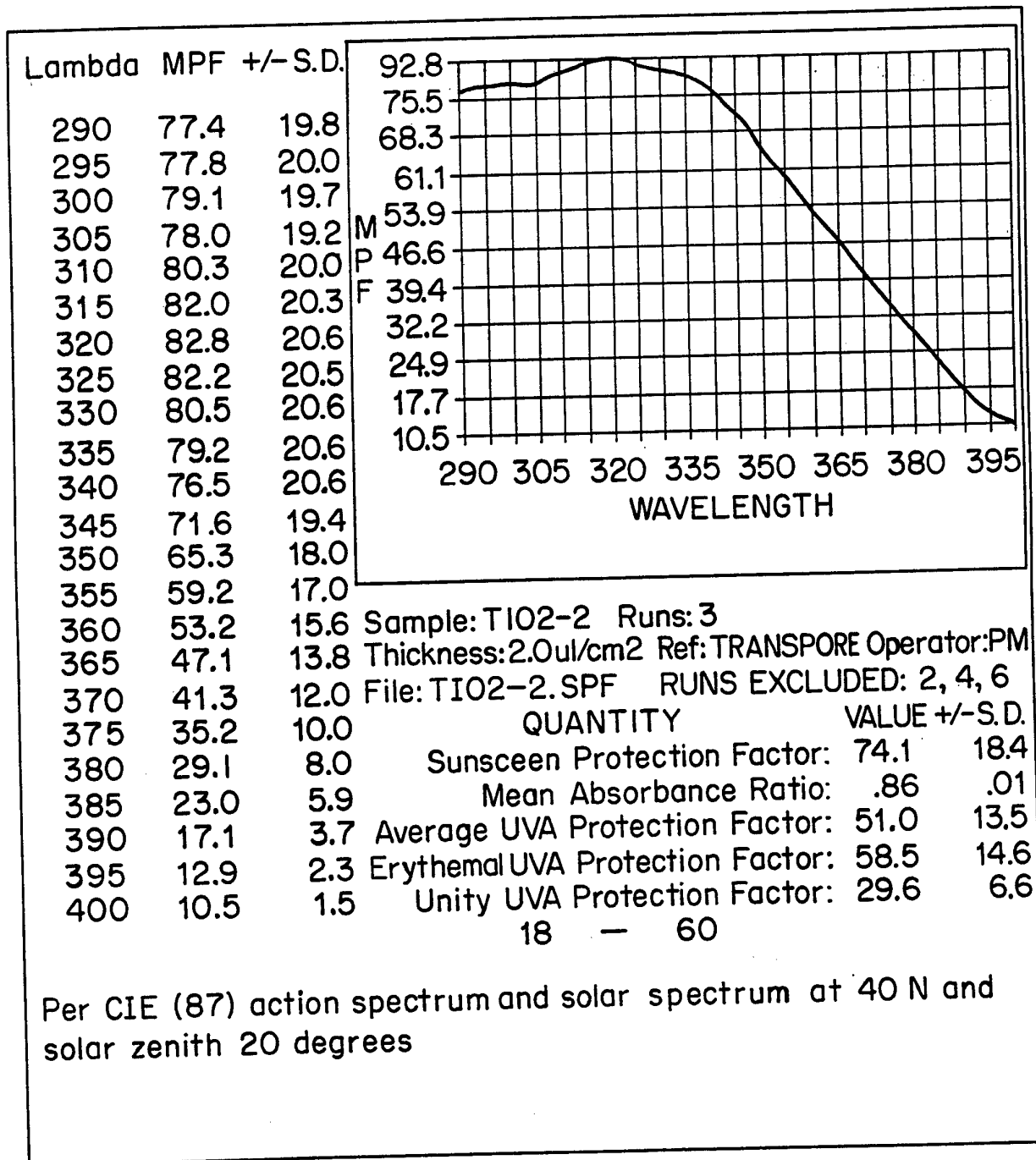


FIG. 2

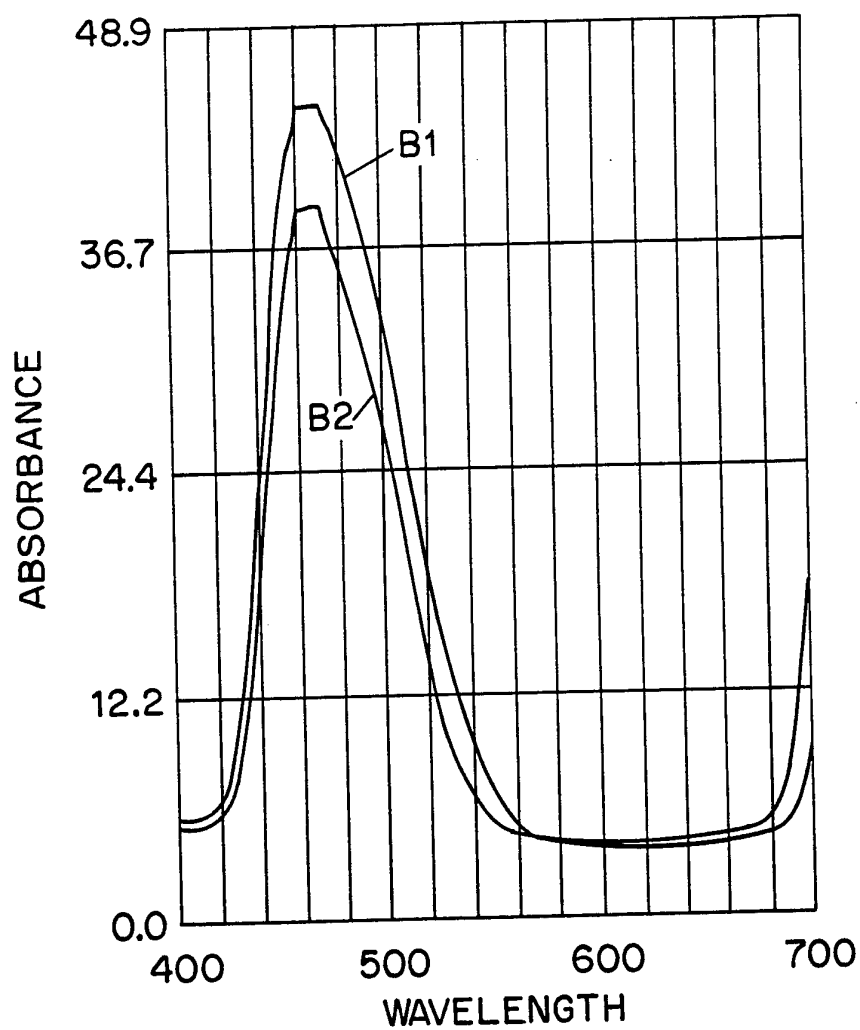


FIG. 3(a)

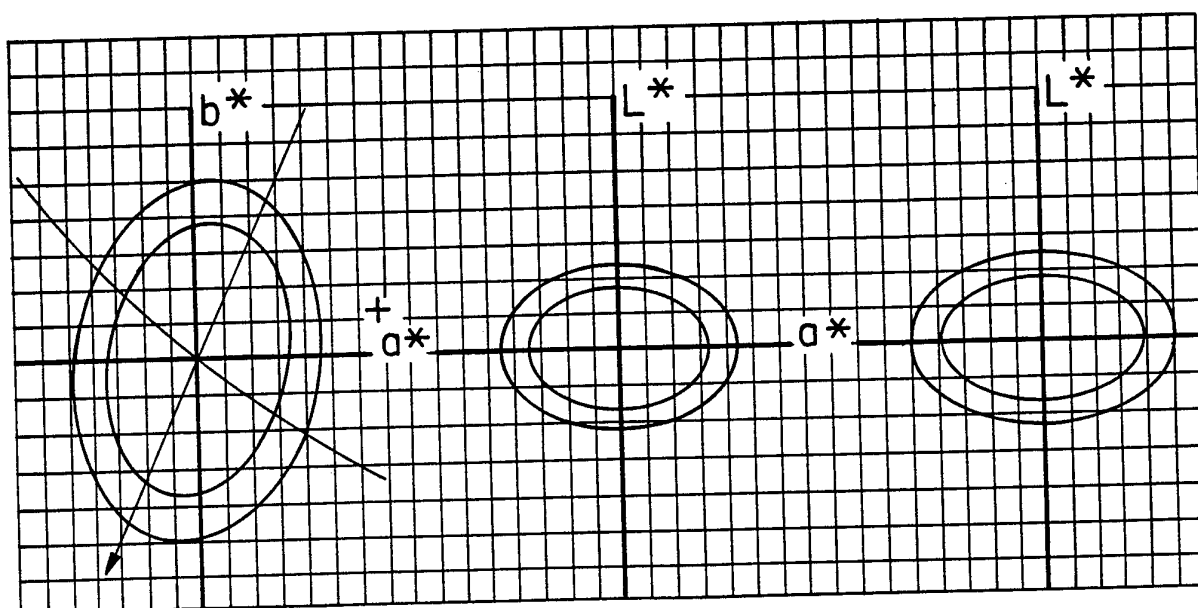


FIG. 3(b)

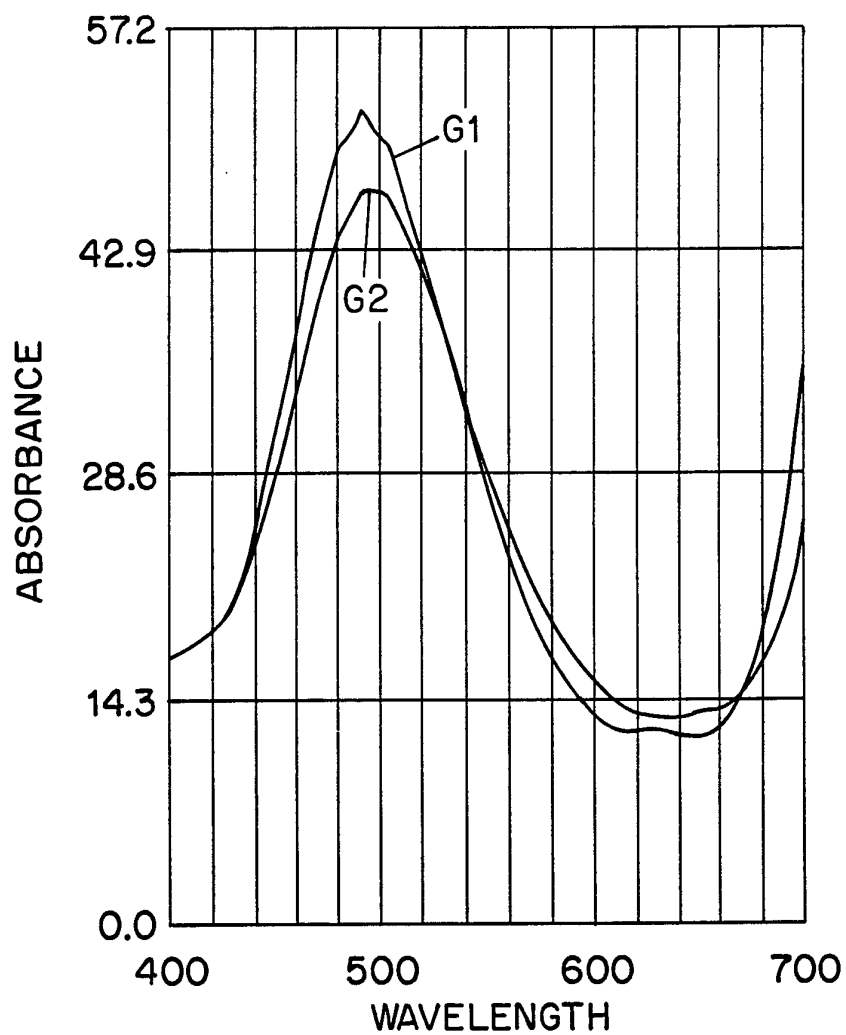


FIG. 4(a)

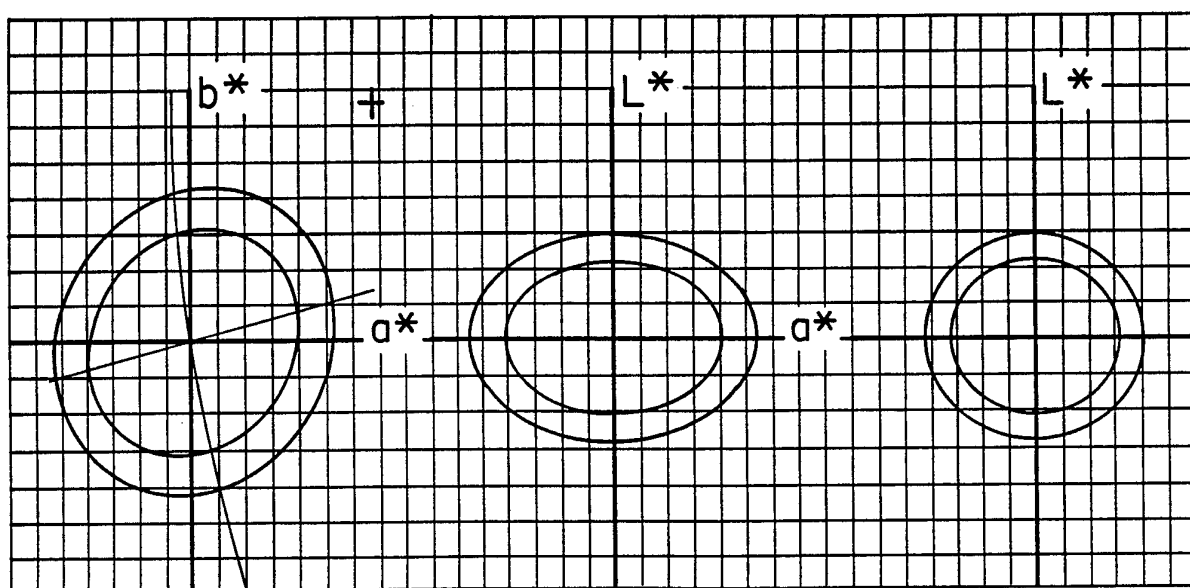


FIG. 4(b)

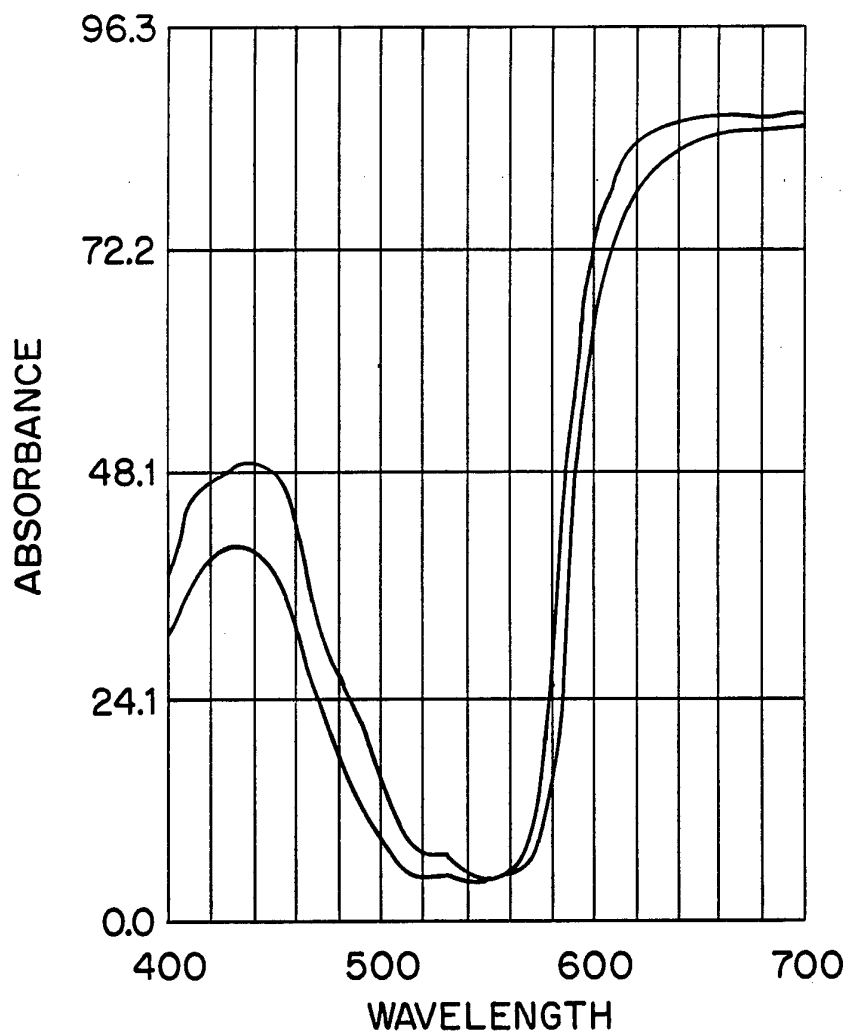


FIG. 5 (a)

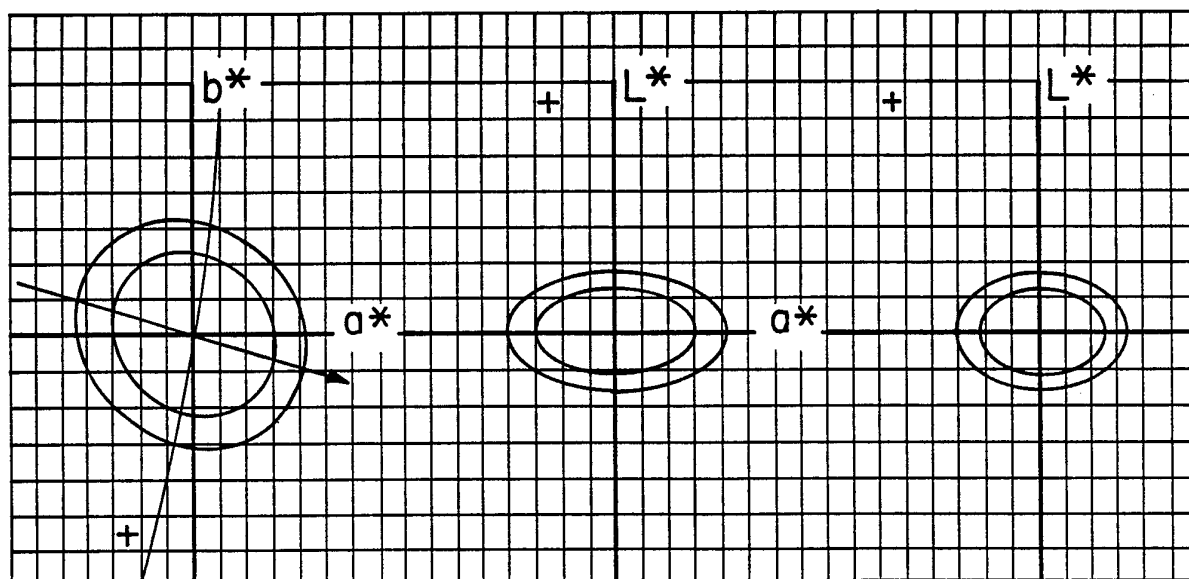


FIG. 5 (b)

INTERNATIONAL SEARCH REPORT

Intern. Appl. No.

PCT/US 94/01929

A. CLASSIFICATION OF SUBJECT MATTER
 IPC 5 A61K7/00 A61K7/42 C09B67/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 5 A61K C09B

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

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	FR,A,2 134 621 (BRITISH TITAN LTD) 8 December 1972 see page 1, line 13 - line 26 see page 2, line 1 - line 26 see page 2 - page 3; example 1 see claims 1-3,5,6,9,10 ---	1,5,6,9, 12
X	PATENT ABSTRACTS OF JAPAN vol. 13, no. 84 (C-572)27 February 1989 & JP,A,63 270 618 (KANEBO LTD) 8 November 1988 see abstract --- -/--	20



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

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Date of the actual completion of the international search

5 July 1994

Date of mailing of the international search report

12.07.94

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Boulois, D

INTERNATIONAL SEARCH REPORT

Intern. Application No

PCT/US 94/01929

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE WPI Section Ch, Week 7715, Derwent Publications Ltd., London, GB; Class D11, AN 77-26389Y & JP,A,52 009 442 (SHISEIDO KK) 16 March 1977 see abstract ---	1,5,6
A	WO,A,86 04598 (ATKINSON G.) 14 August 1986 see page 16; example 2 see page 25 - page 26; example 10 see claims 1-3,25,36,37 ---	1-31
A	US,A,4 648 908 (TAKASUKA Y. ET AL) 10 March 1987 see column 5 - column 6; examples 2-4 see column 17; table 4 see claims 1,6,7 ---	1-31
A	EP,A,0 328 906 (NIPPON OIL AND FATS COMPANY LTD) 23 August 1989 see claim 1 ---	1-31
A	WO,A,90 06103 (THE BOOTS COMPANY LTD) 14 June 1990 see page 12; example 1 ---	1-31
A	EP,A,0 220 617 (MERCK PATENT GESELLSCHAFT MIT BESCHRÄNKTER HAFTUNG) 6 May 1987 see page 2, line 21 - page 3, line 11 see page 4, line 9 - page 5, line 9 -----	1-31

INTERNATIONAL SEARCH REPORT

Information on patent family members

Intern. Application No

PCT/US 94/01929

Patent document cited in search report	Publication date	Patent family member(s)	Publication date	
FR-A-2134621	08-12-72	AU-B-	456328	12-12-74
		AU-A-	4139772	25-10-73
		CA-A-	962003	04-02-75
		DE-A-	2220822	09-11-72
		GB-A-	1327833	22-08-73
		US-A-	3808022	30-04-74

WO-A-8604598	14-08-86	US-A-	4599114	08-07-86
		AU-A-	5540486	26-08-86
		DE-T-	3690042	04-06-87
		EP-A-	0211941	04-03-87
		GB-A, B	2182028	07-05-87
		US-A-	4909852	20-03-90
		US-A-	4741780	03-05-88

US-A-4648908	10-03-87	US-A-	4710375	01-12-87

EP-A-0328906	23-08-89	JP-A-	2000669	05-01-90
		DE-D-	68909634	11-11-93
		DE-T-	68909634	07-04-94
		US-A-	5091010	25-02-92
		US-A-	5213618	25-05-93

WO-A-9006103	14-06-90	AU-B-	630406	29-10-92
		AU-A-	4821690	26-06-90
		EP-A, B	0446290	18-09-91
		JP-T-	4502326	23-04-92

EP-A-0220617	06-05-87	JP-B-	6011871	16-02-94
		JP-A-	62101662	12-05-87
		JP-B-	5005267	21-01-93
		JP-A-	62101664	12-05-87
		AU-B-	596378	03-05-90
		AU-A-	6344986	30-04-87
		CA-A-	1279962	12-02-91
		DE-A-	3686544	01-10-92
		FI-B-	89453	30-06-93
		US-A-	4772331	20-09-88
