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(54) **Electroluminescent device**

(57) Cyanoalkylated pullulan polymeric resins and their admixtures with other cyanoalkylated resins are used as dielectric binders in an electroluminescent device 10. The resins can be used in the phosphor/binder layer 16 and/or the barium titanate/binder layer 14. The resultant electroluminescent devices have improved brightness.

FIG. 1

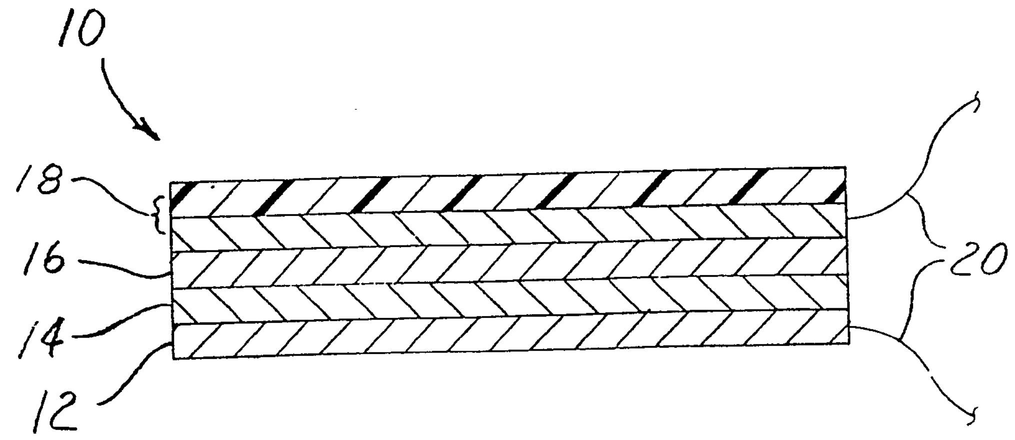

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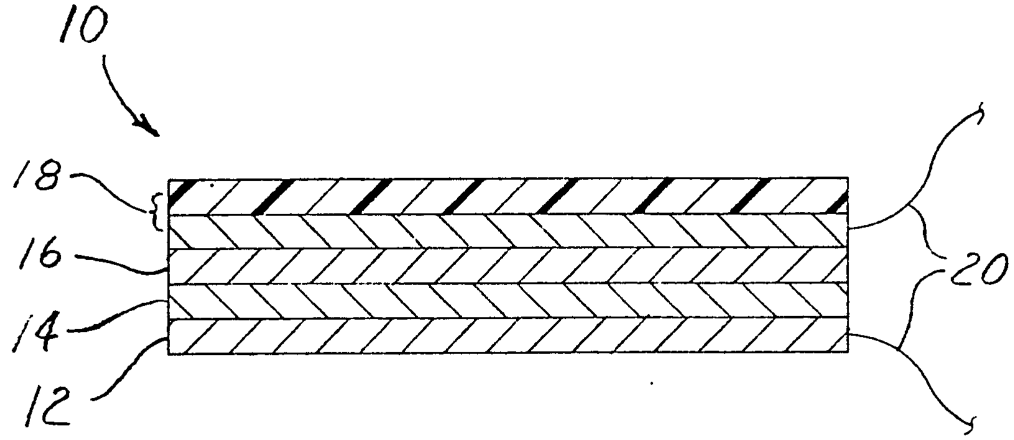
FIG. 1



IMPROVED ELECTROLUMINESCENT DEVICE

This invention relates to improved electroluminescent devices. More particularly, this invention relates to electroluminescent devices having improved luminescence and lifetimes.

Electroluminescent devices are comprised of two electrodes, one of which is transparent, with a phosphor layer therebetween. When a source of electric current is applied to the electrodes, the phosphors become activated and emit visible photons or light. The emission spectrum and wavelength generated by the phosphors is controlled by the activator element in the phosphor. The amount of light emitted by the device is controllable by the amount of current applied. Generally the more current that is applied, the more intense the light that is emitted, within certain limits.

The phosphors are highly sensitive to moisture and to heat, including the heat they themselves generate during emission. Thus in order to protect the phosphors, it has become the practice to mix them in a solvent-based high dielectric constant binder, such as an organic polymeric matrix, dissolved in a solvent. The polymer encapsulates the phosphor particles and binds them together. The organic solvent is then evaporated. A separate insulating layer of a high dielectric constant material such as barium titanate also acts as protection for the phosphor layer. The barium titanate can also be mixed with a high dielectric constant polymeric binder medium in similar fashion.

The manufacture of these electroluminescent devices has also been improved over the years. The barium titanate/binder and phosphor/binder layers are deposited separately as thin layers onto flexible electrodes made of metal foils and plastic films coated with a transparent conductive layer respectively. The layers are assembled and the assembly is laminated together by inserting between plastic sheets as, for example, sheets of polyester or polycarbonate, and these are sealed using heat and pressure to form a final flexible hermetically sealed package. Leads can be attached to the electrodes either before or after assembly of the device.

Several high dielectric constant polymeric binder materials have been suggested as phosphor encapsulants heretofore. In U.S. Patent 3,238,407, Jaffe discloses cyanoethyl cellulose as the polymeric binder material. However, this material diminishes the intensity of emitted light in electroluminescent devices and thus is less than satisfactory. Takahashi in U.S. Patent 3,389,286 describes the use of cyanoethylated polyvinyl alcohol as a dielectric binder layer; however, this material partially dissolves the phosphor and also diminishes the light intensity of the electroluminescent devices.

In U.S. Patent 4,560,902, Kardon describes a thermoplastic epoxy as the polymeric binder layer. This epoxy is prepared by reacting a resin, such as a urethane or epoxy resin, with the reaction product of an epihalohydrin and a bisphenol in certain proportions to form a resin having a molecular weight of about 450-75,000.

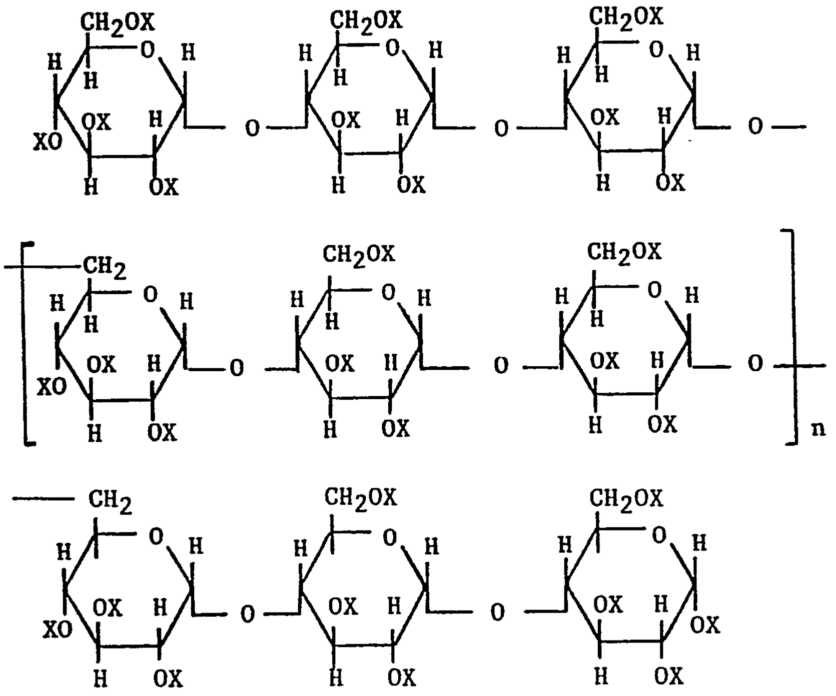
The above electroluminescent devices, while satisfactory, like all electroluminescent devices are limited in the amount of light they produce. It would be highly desirable to increase the brightness of the light emitted by these devices without increasing the thickness of the layers or the amount of relatively expensive phosphors required.

We have found that particular cyanoalkylated pullulan polymers, when employed as a high dielectric constant polymeric binder in electroluminescent devices, significantly increases the amount of light emitted by the devices.

Preferred embodiments of the present invention will now be described, by way of example only, with reference to the accompanying drawing which is a perspective view of a multilayered electroluminescent device of the present invention.

Electroluminescent devices comprising a phosphor layer on a transparent electrode and a dielectric layer on a metallic electrode wherein pullulan polymers are present as a binder in one or both layers, have significantly increased brightness or light emission over conventional electroluminescent devices. These polymers can be employed in conventional coating and lamination processes without modification and provide excellent adhesion to either or both of the phosphor and dielectric layers.

The cyanoalkylated pullulan polymer binder materials which may be employed in this invention include the following:

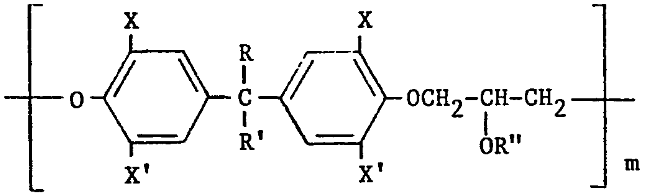


wherein n is an integer of from about 20 to about 4000 and X in each occurrence is hydrogen or cyano lower alkyl as, for example straight or branched chain lower alkyl of 1-5 carbon atoms such as methyl, ethyl, n-propyl, isopropyl, n-butyl, tertiary butyl, n-pentyl and the like. Preferably, X represents hydrogen, cyanomethyl or cyanoethyl.

The cyanoalkylated pullulan polymers are known compounds which are described in U.S. Patent No. 4,322,524. They are naturally occurring products in which the recurring maltotriose units are interactive and may be readily cyanoethylated when reacted with an acrylonitrile in the presence of an alkali catalyst. The degree of cyanoethylation is controllable by the reaction conditions and can reach 90% or higher.

Mixtures of the above cyanoalkylated pullulan polymers with other cyanoalkylated resins such as cyanoalkylatedpolyvinyl alcohol, cyanoalkylcellulose, cyanoalkylsucrose, other cyanoalkylated sugars and starches, and cyanoalkylhydroxycellulose can also be employed. Certain mixtures of these cyanoalkylated resins with cyanoalkylated pullulan are available from Shin Etsu of Japan under the trademark Cyepl resins. Cyanoethyl pullulan, CR-S, has a nitrogen content of from 11.8-13.0% by weight, a specific gravity of 1.25 and a softening temperature of about 90-110°C. A mixture of cyanoethyl pullulan and cyanoethylcellulose, CR-C, has a nitrogen content of about 11.8-13.0% by weight, a specific gravity of 1.27 and a softening temperature greater than 270°C. A mixture of cyanoethyl pullulan and cyanoethylpolyvinyl alcohol, CR-V, has a nitrogen content of about 12.2-13.5% by weight, a specific gravity of 1.27 and a softening temperature of about 20-40°C. A mixture of cyanoethyl pullulan and cyanoethyl sucrose, CR-U, has a nitrogen content of about 14.4% by weight, a specific gravity of about 1.24 and is a liquid at room temperature. A mixture of cyanoethyl pullulan and cyanoethyl hydroxycellulose, CR-E, has a nitrogen content of about 9.0-10.9% by weight, a specific gravity of about 1.30 and a softening temperature of about 55-65°C. The relative amount of cyanoalkylation can be varied by the reaction conditions.

Cyanoalkylated polymers derived from a urethane or epoxy reacted with the reaction product of a bisphenol such as bisphenol A and an epihalohydrin in known manner are also useful as the cyanoalkylated resins in admixture with cyanoalkylated pullulan polymers. These resins have the formula



wherein R and R' can be independently hydrogen, lower alkyl and mononuclear aryl; X and X' are independently hydrogen, lower alkyl, or halogen; R" can be hydrogen or a cyanoloweralkyl, for example straight or branched chain lower alkyl of 1-5 carbon atoms, including methyl, ethyl, n-propyl, isopropyl, n-butyl, tertiary butyl, n-pentyl and the like; and m is an integer greater than 1. The degree of cyanoalkylation can be varied by varying the amount of the acrylonitrile reactant.

The pullulan polymers and their admixture with other cyanoalkylated resins are made by reaction of the hydroxy-containing precursors of the pullulan polymers with an acrylonitrile in known manner.

Suitable acrylonitrile reactants include acrylonitrile, crotonacrylonitrile, 2,2-dimethylacrylonitrile, 2-methyl-2-

ethylacrylonitrile and the like. The cyanoalkyl groups react with the hydroxyl groups of the polymeric precursors described above.

The amount of acrylonitrile reactant can vary depending upon the degree of cyanoalkylation desired. In order to obtain cyanoalkylation of about 50% of the available hydroxyl groups, about 3-5 moles of acrylonitrile is used per mol of hydroxy units in the starting hydroxy-containing compounds. If an excess of acrylonitrile is employed, it also acts as a solvent for the starting material.

Although the following described process is directed to the preparation of cyanoalkylated pullulan resins, cyanoalkylation of other hydroxy-containing polymers is carried out in similar manner.

The reaction between a pullulan natural product which can vary in molecular weight from about 50,000 to about 2,000,000, and an acrylonitrile occurs either by dissolving the pullulan starting material in an aqueous alkali solution and adding an acrylonitrile in an organic solvent, whereby the reaction proceeds readily at room temperature; or by dispersing the starting material pullulan in a solution of an acrylonitrile in an organic solvent and adding an aqueous solution of an alkali and heating to a temperature of about 40-60°C. The relative amounts of the reactants and alkali can be varied to control the amount of cyanoalkylation of the pullulan starting material. For example, when pullulan or other starting material is dissolved in a sodium hydroxide solution, the concentration of starting material can be about 2-20% by weight in a sodium hydroxide solution of about 2-20% by weight in water. When sodium hydroxide is added to an organic solvent mixture, the hydroxide suitably has a concentration of about 10-50% by weight in water, and from about 1-10% by weight of sodium hydroxide based on the pullulan starting material can be employed. The cyanoalkylated pullulan resin is collected by neutralizing the reaction mixture with acid and diluting with water to precipitate the cyanoalkylated pullulan. The product can be further purified by washing with water, or can be dissolved in a ketone such as acetone and reprecipitated with water. The material is then dried.

The cyanoalkylated pullulan resin is soluble in organic solvents and thus materials such as barium titanate and phosphors can be readily admixed therewith. Suitable cyanoalkylated pullulan polymers have softening points of from about 20-110°C, and is preferably about 90°C. The nitrogen content can vary from about 4-20 weight % and is preferably about 12 weight %.

The cyanoalkylated pullulan resin can be admixed in various proportions with other cyanoalkylated resins such as those described above.

The cyanoalkylated polymers are soluble in various organic solvents including glycol ethers, alkyl ketones or aromatic solvents. Suitable ethers include glycolalkyl ethers such as propylene glycol methyl ether, dipropylene glycol methyl ether, tripropylene glycol methyl ether, ethylene glycol butyl ether, diethylene glycol methyl ether, diethylene glycol ethyl ether and diethylene glycol butyl ether. These ethers are colorless liquids which combine the properties of alcohols, ethers and hydrocarbons. They are miscible with most organic solvents and they are excellent solvents for mixing with the phosphor and barium titanate components of electroluminescent devices.

Suitable alkyl ketones and aromatic solvents which can also be employed as solvents include lower alkyl ketones such as acetone, methyl ethyl ketone, ethyl ketone and methylisobutyl ketone, toluene, xylene and the like.

In making the barium titanate layer, the barium titanate solid is first admixed with a suitable solvent as described above. The amount of barium titanate which is added to the glycol ether or other solvent is preferably about 70-90% by weight. The barium titanate and the solvent are stirred together to form a homogeneous slurry. To this slurry is added from 10 to 30 parts of a binder, including a cyanoalkylated polymer resin as described above which can also be in a like solvent. After mixing thoroughly, the barium titanate-binder-solvent slurry is deposited onto a metal foil or other electrode and dried. Suitably a barium titanate/resin binder layer thickness when dried is from about 2-6 mils.

The phosphor is also first admixed with a suitable solvent and mixed well. Suitable phosphors are luminescent under the influence of an electric current, as for example zinc sulfide, zinc oxide or zinc sulfide activated with manganese, copper, copper/lead or copper/manganese mixtures. The amount of phosphor added to the solvent can range suitably from about 60-95% by weight of the mixture, and is preferably from about 75-85% by weight. The light emitted by the electroluminescent cell is at least partially dependent upon the particle size and concentration of phosphor in the cyanoalkylated polymer. The maximum amount of phosphor is added consistent with there being sufficient amount of binder present to encapsulate the phosphor to protect it from moisture. After mixing well, from 5 to 40 parts of a binder is also added to the phosphor slurry. The resultant phosphor slurry is deposited onto a transparent electrode in known manner. A suitable phosphor/binder layer thickness when dried is from about 2-6 mils.

The phosphor-containing and barium titanate-containing slurries can be deposited onto their respective electrodes by spray coating, roller coating, painting or hot presssing techniques known in the art. The thickness of the deposited layers may be varied to provide optimum light emitting effects.

The electrodes of the present electroluminescent devices include a transparent electrode, which can be of transparent plastic film or sheet such as polyethylene, polypropylene, polyethylene terephthalate, polyvinyl chloride, polymethylmethacrylate and the like, or of glass. The transparent film or glass is coated with a transparent conductive film, such as an indium-tin oxide film, as is known. The other electrode is of a conductive metal, such as aluminum, gold, silver, copper, in a variety of thicknesses, from a thick sheet to a thin foil.

The two parts of the electroluminescent cell as described above, i.e., the barium titanate/binder layer on a metal electrode and the phosphor/binder layer on a transparent electrode, are assembled by hot pressing at temperatures in the range of about 150-400°F at a pressure of from about 5-100 psi for a period of about 0.1-2.0 seconds. Electrical leads or connections are made to the electrodes and then connected to a source of electrical current. When an electrical current is passed across the electrodes, the phosphor particles are activated and light is emitted from the cell. In accordance with the invention, at least one of the phosphor layers or barium titanate layers contains a cyanoalkylated pullulan resin or resin mixture as the binder.

The assembled electroluminescent device 10 is shown in the Figure wherein a barium titanate/binder layer 14 is deposited onto an aluminum foil 12 and a phosphor/binder layer 16 is deposited onto an indium-tin oxide coated polymer film 18. Leads 20 complete the device 10.

The electroluminescent devices of the invention, having at least one of the phosphor/binder and barium titanate/binder layers containing as a binder a cyanoalkylated pullulan resin or mixture thereof, unexpectedly have high intensity light emission, much higher than that of conventional electroluminescent devices. The presence of the high dielectric constant cyanoethylated pullulan polymers and mixtures in the present devices increases the brightness achievable with these devices. Increased brightness is achieved with the binders of the invention providing pullulan polymer is added to one or more of the layers of the electroluminescent device even when conventional binders, such as epoxy resins, are used as a binder in one of the layers of the electroluminescent device, or as a portion of the binder in either layer. This result was highly unexpected. Further, these resins are able to be used in commercial coating and lamination processes without modification, and provide adequate adhesion to both the phosphor and barium titanate components.

The invention will be further illustrated by the following examples, but the invention is not meant to be limited to the details described therein. In the Examples, parts and percentages are by weight.

EXAMPLE 1

Pullulan-Phosphor Modified Lamp

A mixture was made from 10.24 grams of a binder comprised of a mixture of cyanoethyl pullulan resin and cyanoethylpolyvinyl alcohol available as Cyepl 02 from Shin Etsu of Japan. This resin as a 20% solution in dimethylformamide had a viscosity of 240-260 centipoises; a nitrogen content of between 12.2 and 13.5; a specific gravity of 1.27 and a softening temperature of 20-40°C. The binder was mixed in a 50:50 weight ratio of about one part binder to two parts of solvent. To this mixture was added 43.64 grams of zinc sulfide phosphor containing a copper activator.

The resultant slurry was cast onto a series of polyethylene terephthalate tapes having an indium/tin oxide conductive layer thereon in varying thickness.

A control series of tapes was made using a standard phosphor slurry containing the resin D.E.R. 684-EK40, as described in U.S. Patent 4,560,902 to Kardon. D.E.R.-EK40 is an ultra high molecular weight epoxy resin derived from the reaction of bisphenol A and epichlorohydrin, and is a registered trademark of the Dow Chemical Company, Midland, Michigan.

Both series of tapes were dried and laminated to an aluminum foil having a layer of barium titanate in standard epoxy binder thereon. The layers were passed several times through a heat sealing laminator and brightness measured in foot-lamberts at 120 Volts, 400 Hz. The data are summarized in Table I below:

TABLE 1

Film Thickness, mils	Brightness, foot-lamberts				
	Pass 1	Pass 2	Pass 3	Pass 4	Pass 5
Control					
1.45	7.0 7.5	8.5 8.8	9.6 9.6	10.3 10.1	10.3 10.1
1.60	9.4 9.4	10.4 10.4	11.3 11.5	11.9 12.2	12.0 11.0
1.90	9.6 9.7	10.4 10.5	11.0 10.5	______	______
2.25	8.9 9.6	9.7 10.0	--------	______	______
Example 1					
1.65	11.3 12.9	13.1 14.4	15.1 16.0	16.2 16.2	17.9 17.2
2.00	12.9 11.7	13.9 15.2	15.7 16.9	16.9 17.6	17.9 18.3
2.35	14.4 15.4	16.3 16.5	16.7 16.9	______	______
2.75	14.8 14.9	______	______	______	______
3.10	12.7 13.2	14.2 14.9	14.6 15.2	14.6 13.8	______

Thus it is apparent that when the resin of the invention was mixed with the phosphor layer alone, the brightness of the electroluminescent cells improved significantly.

EXAMPLE 2

Pullulan-Barium Titanate Modified Lamp

Barium titanate (18.84 grams) was admixed with the cyanoethyl pullulan/cyanoethylpolyvinyl alcohol binder of Example 1 (31.06 grams) in 15 grams of a 50:50 blend of acetone and methyl ethyl ketone solvent. The resultant slurry was deposited onto aluminum foil at varying thickness, dried and laminated to a conventional phosphor coating 4 mils thick using an epoxy binder on an indium/tin oxide polyethylene tape.

The electroluminescent cells were passed through the laminator three times, and the brightness measured after each pass as in Example 1. The results are tabulated below in Table II.

TABLE II

Layer Thickness, mils	Brightness, foot-lamberts					
	Pass I		Pass 2		Pass 3	
2	15.5	16.2	15.9	16.6	18.0	17.2
3	17.4	17.5	19.4	18.8	19.8	18.8
4	19.0	18.7	21.7	20.2	21.8	20.3
5	18.8	18.0	20.7	19.5	20.8	19.7
6	17.9	17.9	20.0	20.5	20.3	20.8

It is apparent that improved brightness is obtained when the cyanoalkylated pullulan binder of the invention is present in the barium titanate layer alone as well.

EXAMPLE 3

Pullulan-Phosphor and Pullulan-Barium Titanate Modified Lamp

The binder employed in Examples 1 and 2 was used to make both the phosphor layers and the barium titanate layers of electroluminescent cells. In the first group of cells, the layer thickness of the barium titanate layer was varied and the phosphor layer was maintained at 4 mils; in the second group of cells, the barium titanate was maintained at 5 mils and the phosphor layer thickness was varied. The cells were passed through a laminating process three times and the brightness measured after each pass as in Example 1. The results are summarized in Table III below.

TABLE III

Layer Thickness, Mils	Brightness, foot-lamberts					
	Pass 1		Pass 2		Pass 3	
Group 1						
3	29.4	30.0	29.4	30.0	30.2	29.3
4	30.0	29.6	33.3	32.8	35.8	34.7
5	31.0	30.5	32.6	34.0	34.0	35.4
6	30.8	30.6	32.2	32.7	34.8	34.9
Group 2						
2	30.7	31.4	34.8	34.5	38.2	37.5
3	34.1	33.4	35.6	35.2	36.1	35.4
4	31.4	32.0	32.1	33.9	35.7	34.1
5	31.3	31.7	33.0	33.5	34.6	34.4

It is apparent that the brightness of the above electroluminescent cells is far superior to conventional cells, and the brightness is increased when both the barium titanate and phosphor layers use the binder of the invention.

EXAMPLE 4

Several electroluminescent cells were made varying the phosphor and barium titanate layers and the laminating conditions. A phosphor layer of Example 1 was deposited to a thickness of 2.3 mils, and a control standard phosphor layer using epoxy binder was deposited in a thickness of 1.75 mils. The barium titanate/binder mixture of Example 2 was deposited onto 1.5 mil thick aluminum foil in a thickness of 1.3 mils, and a control barium titanate/conventional epoxy binder was deposited onto aluminum foil in a thickness of 1.3 mils. Cells A were laminated using a standard phosphor/epoxy resin binder and the barium titanate/binder mixture of Example 2; thus the pullulan polymer was present in only one layer, the barium titanate layer. Cells B were laminated using pullulan in both the phosphor/binder and barium titanate/binder layers of Examples 1 and 2. The control used the standard epoxy binder containing phosphor and barium titanate layers. The brightness was measured as in Example 1. The results are summarized in Tables IV and V below.

TABLE IV *

Temperature, °F	Brightness, foot-lamberts		
	Control	Cells A	Cells B
200	11.2	22.0	29.7
240	12.5	24.5	26.8
275	12.4	26.8	39.3

* Pressure was constant at 60 lbs.

TABLE V **

Pressure, lbs	Brightness, foot-lamberts		
	Control	Cells A	Cells B
20	9.5	18.9	27.2
40	11.1	24.3	26.8
60	27.2	31.9	39.3

** Temperature was constant at 175°F.

Thus the electroluminescent devices containing pullulan had increased brightness over conventional electroluminescent devices containing binders of conventional epoxy resins. Electroluminescent devices containing pullulan in both the phosphor layer and the dielectric layer had the greatest brightness.

EXAMPLE 5

An electroluminescent cell was made using 87 grams of lead sulfide phosphor admixed with 15 grams of cyanoalkylated pullulan having a nitrogen content of about 12% and a softening temperature in the range 90-110°F in dimethylformamide. The resultant solution at 20°C had a viscosity of 240-360 centipoises. The specific gravity of the solution was 1.25. Propylene glycol methyl ether was used as a solvent. Layers varying in thickness were applied to a transparent indium/tin oxide on polyethylene terephthalate sheet. The mixture was dried in an oven at 120°C.

A layer of barium titanate mixed with cyanoethylated pullulan as above (85 grams of barium titanate per 15 grams of pullulan resin) in propylene glycol methyl ether and deposited on an aluminum foil and dried in an oven at 120°C.

The two layers were compressed together at 150 psi pressure at 150°C, leads attached, and the cell joined to a source of 120 volt AC current. The cells were tested as follows:

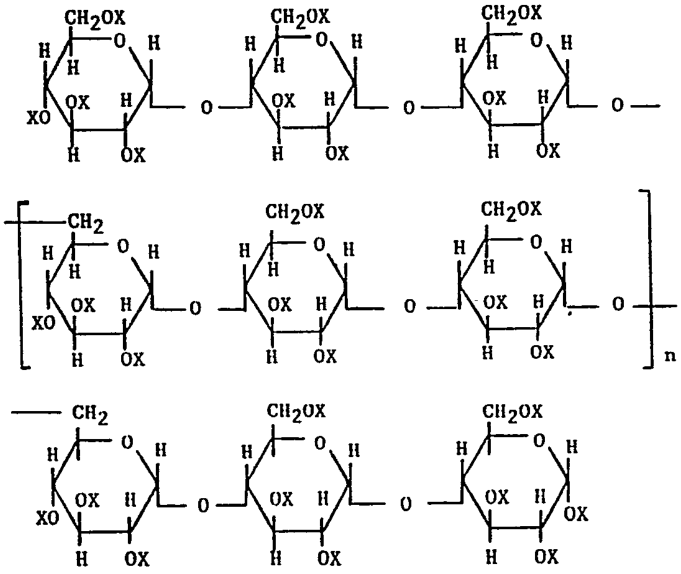
Thickness, mils	Power	Foot-Lamberts
1.60	120V,400Hz	30.0
	140V,600Hz	44.5
1.45	120V,400Hz	29.6
	75V,600Hz	22.7
1.35	120V,400Hz	33.0
	75V,600Hz	20.9
1.05	120V,400Hz	30.9
	75V,600Hz	21.9
0.80	120V,400Hz	23.1
	75V,600Hz	20.5

The above electroluminescent devices all had excellent brightness. Thus the addition of pullulan polymers and polymer mixtures unexpectedly improves the brightness of electroluminescent devices.

Claims

1. An electroluminescent device comprising a phosphor layer and a dielectric layer disposed between two electrodes, one of which is transparent, wherein there is incorporated into at least one of said layers as a binding agent a pullulan selected from among cyanoalkylated pullulan polymers or a mixture of cyanoalkylated polymers.

2. The device according to claim 1 wherein said polymer is a compound of the formula



wherein X is hydrogen or cyanoloweralkyl and n is an integer having a value of from about 20-4000.

3. The device according to claim 1 or 2, wherein said polymer has nitrogen content of from about 4-20% by weight.

4. The device according to claim 2 or 3 wherein the degree of cyanoalkylation in said polymer is at least about 50%.

5. The device according to claim 1 wherein said cyanoalkylated pullulan is admixed with a cyanoalkylated polymer of the group consisting of cyanoalkylcellulose, cyanoalkylpolyvinyl alcohol, cyanoalkylsucrose, cyanoalkylhydroxy cellulose and cyanoalkylated epoxy resin.

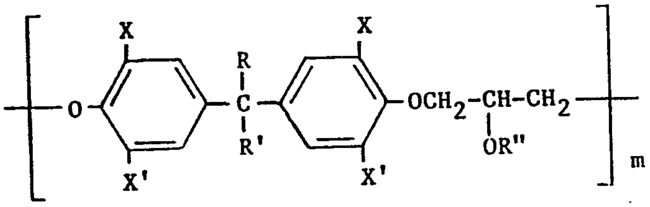
6. The device according to claim 5 wherein said mixture comprises cyanoalkylated pullulan and cyanoalkylpolyvinyl alcohol having a nitrogen content of about 12.2-13.5% by weight and a softening temperature of from 20-40°C.

7. The device according to claim 5 wherein said mixture comprises cyanoalkylated pullulan and cyanoalkylcellulose having a nitrogen content of about 11.8-13.0% by weight and a softening temperature greater than 270°C.

8. The device according to claim 5 wherein said mixture is a liquid comprised of cyanoalkylated pullulan and cyanoalkylsucrose having a nitrogen content of about 14% by weight.

9. The device according to claim 5 wherein said mixture comprises cyanoalkylated pullulan and cyanoalkylhydroxy cellulose having a nitrogen content of about 9.0-10.9% by weight and a softening temperature of about 55-65°C.

10. The device according to claim 5 wherein said mixture comprises cyanoalkylated pullulan and a cyanoalkylated epoxy resin of the formula



wherein R and R′ are the same or different members selected from the group consisting of hydrogen, lower alkyl and mononuclear aryl; R" is hydrogen or cyanoloweralkyl; X and X′ are the same or different members selected from the group consisting of hydrogen, lower alkyl and halogen; and m is an integer greater than 1.

11. The device according to any preceding claim wherein said cyanoloweralkyl is cyanoethyl.

12. The device according to any preceding claim wherein said pollulan is incorporated into said phosphor layer.

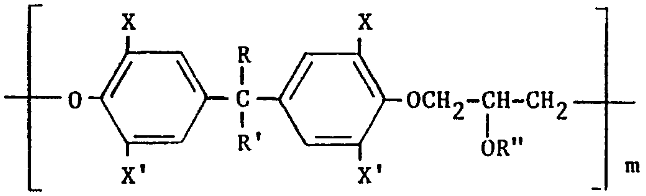
13. The device according to any of claims 1 to 11 wherein said pollulan is incorporated into said dielectric layer.

14. The device according to claim 13 wherein said dielectric layer is barium titanate.

15. The device according to any of claims 1 to 11 wherein said pullulan is incorporated into said phosphor layer and said dielectric layer.

16. An electroluminescent device comprising a transparent electrode having a layer of a phosphor and a cyanoalkylated polymer of pullulan or polymer mixtures thereof thereon, an adjacent layer containing a dielectric and a cyanoalkylated polymer of pullulan or polymer mixtures thereof thereon, and a metal electrode contacting said dielectric layer.

17. The device according to claim 16 wherein said mixture comprises cyanoalkylated pullulan and a cyanoalkylated epoxy resin of the formula



wherein R and R' are the same or different members selected from the group consisting of hydrogen, lower alkyl and mononuclear aryl; R" is hydrogen or cyanoloweralkyl; X and X' are the same or different members selected from the group consisting of hydrogen, lower alkyl and halogen; and m is an integer greater than 1.

18. The device according to claim 16 or 17 wherein said cyanoalkylated pullulan has a nitrogen content of about 4-20% by weight and a softening point of from about 20°C.

19. The device according to any of claims 16 to 18 wherein said cyanoalkyl is cyanoethyl.

20. The device according to any of claims 16 to 19 wherein said dielectric layer is barium titanate.

21. The device according to any of claims 16 to 20 wherein said cyanoalkylated pullulan is admixed with a cyanoalkylated polymer of the group consisting of cyanoalkylcellulose, cyanoalkylpolyvinyl alcohol, cyanoalkylsucrose, cyanoalkylhydroxy cellulose and cyanoalkylated epoxy resin.

22. The device according to claim 21 wherein said mixture comprises cyanoalkylated pullulan and cyanoalkylpolyvinyl alcohol having a nitrogen content of about 12.2-13.5% by weight and a softening temperature of from 20-40°C.

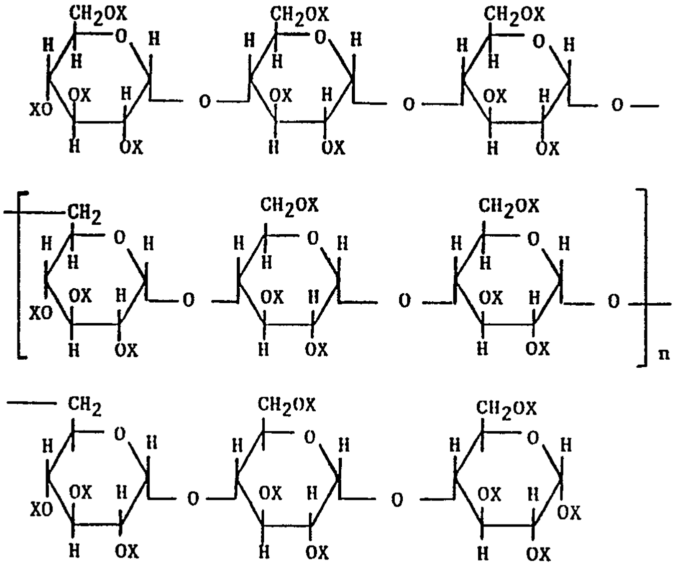
23. The device according to claim 21 wherein said mixture comprises cyanoalkylated pullulan and cyanoalkylcellulose having a nitrogen content of about 11.8-13.0% by weight and a softening temperature greater than 270°C.

24. The device according to claim 21 wherein said mixture is a liquid comprised of cyanoalkylated pullulan and cyanoalkylsucrose having a nitrogen content of about 14% by weight.

25. The device according to claim 21 wherein said mixture comprises cyanoalkylated pullulan and cyanoalkylhydroxy cellulose having a nitrogen content of about 9.0-10.9% by weight and a softening temperature of about 55-65°C.

26. A composition useful as a binder in an electroluminexcent device comprising a phosphor/binder layer and a dielectric/binder layer which comprises a cyanoalkylated pullulan polymer or a mixture of cyanoalkylated polymers.

27. A composition according to claim 26 wherein said cyanoalkylated pullulan has the formula



wherein X is hydrogen or cyanoloweralkyl and n is an integer having a value of from about 20-4000.

28. A composition according to claim 26 or 27 wherein said cyanoalkylated pullulan is admixed with a member selected from the group consisting of cyanoalkylcellulose, cyanoalkylpolyvinyl alcohol, cyanoalkylsucrose, cyanoalkylhydroxycellulose and cyanoalkylated epoxy resin.

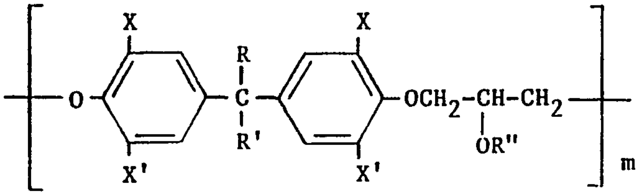
29. A composition according to claim 28 wherein said cyanoalkylpolyvinyl alcohol has a nitrogen content of about 12.2-13.5% by weight and a softening temperature of from 20-40°C.

30. A composition according to claim 28 wherein said cyanoalkylcellulose/cyanoalkylpullulan mixture has a nitrogen content of from about 11.8-13.0% by weight and has a softening point over 270°C.

31. A composition according to claim 28 wherein said cyanopolyalkylsucrose/cyanoalkylpullulan mixture is a liquid and has a nitrogen content of about 14% by weight.

32. A composition according to claim 28 wherein said cyanoalkylhydroxycellulose/cyanoalkylpullulan mixture has a nitrogen content of from about 9.0-10.9% by weight and has a softening temperature of about 55-65°C.

33. A composition according to claim 28 wherein the epoxy resin of said cyanoalkylated epoxy/cyanoalkylpullulan mixture has the formula



wherein R and R' are the same or different members selected from the group consisting of hydrogen, lower alkyl and mononuclear aryl; R" is hydrogen or cyanoloweralkyl; X and X' are the same or different members selected from the group consisting of hydrogen, lower alkyl and halogen; and m is an integer greater than 1.

34. The composition according to any of claims 26 to 33 wherein said cyanoloweralkyl is cyanoethyl.

35. An electroluminescent device substantially as herein described with reference to the accompanying drawing.

36. A binder composition substantially as herein described with reference to the accompanying drawing.

Patents Act 1977
Examiner's report to the Comptroller under Section 17 (The Search Report)

Application number

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Relevant Technical fields

(i) UK Cl (Edition K) H1K (KEAM); C3K (KDA); C3V (VACH, VDF, VDX, VEJ, VEX, VEB)

(ii) Int Cl (Edition 5) H01L; C09K

Search Examiner

W A MORRIS

Databases (see over)

(i) UK Patent Office

(ii) ONLINE DATABASES: WPI

Date of Search

7 OCTOBER 1992

Documents considered relevant following a search in respect of claims 1-36

Category (see over)	Identity of document and relevant passages		Relevant to claim(s)
X	GB 1072285	(HITACHI) Whole document	1, 26
X	GB 1013846	(AMERICAN CYANAMID) Whole document	1, 4, 11-12, 26, 34
X	EP 0328120 A2	(SHIN-ETSU CHEMICAL) Whole document	1, 11, 13, 26
X	EP 0076698 A2	(SUMITOMO) See pages 8-9	26
X	US 3342848	(LEVETAN) Whole document	1, 12, 26
X	JP 03037291	(NICHIA KAGAKU) Whole document	1, 12, 26
X	JP 02309593	(SHINETSU) Whole document	1, 12-16, 20, 26
X	JP 02223192	(HITACHI MAXELL) Whole document	1, 12, 26

Category	Identity of document and relevant passages	Relevant to claim(s)

Categories of documents

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