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ELECTROLUMINESCENT LAMP HAVING AN ALUMINUM ELECTRODE WITH AN ALUMINUM OXIDE LAYER DISPOSED BETWEEN THE ALUMINUM ELECTRODE AND THE ELECTROLUMINESCENT MATERIAL

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Fig. 1.

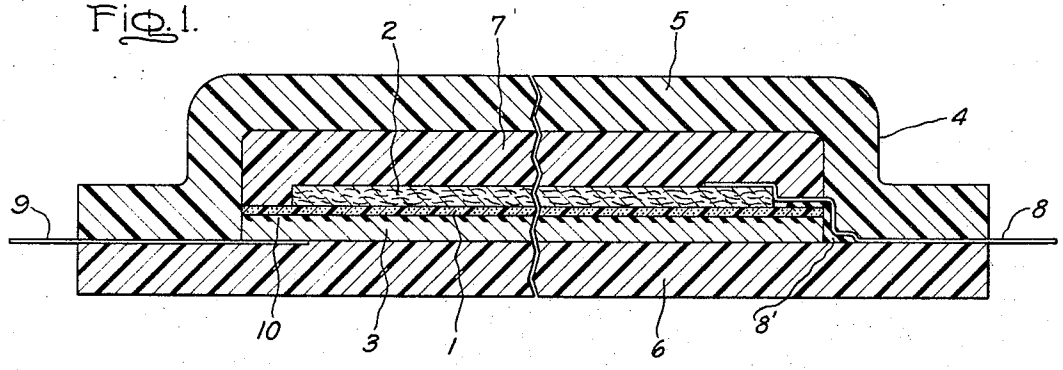
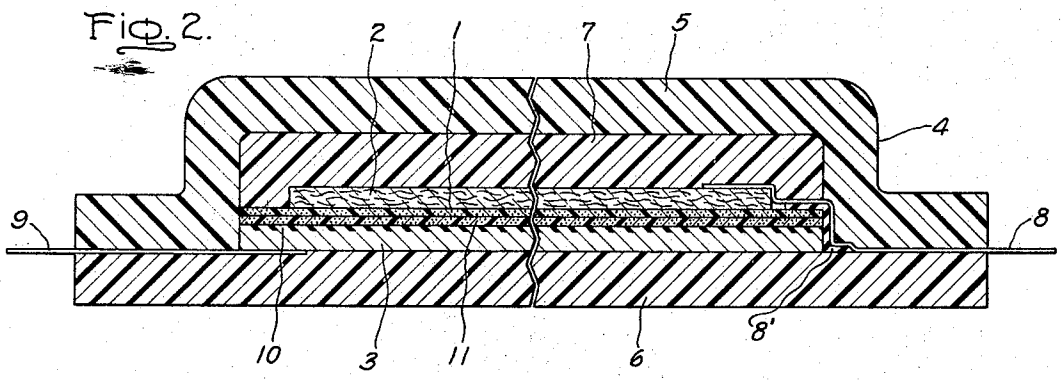


Fig. 2.



Inventor:
Paul A. Dell
by James J. Lagna
His Attorney

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ELECTROLUMINESCENT LAMP HAVING AN ALUMINUM ELECTRODE WITH AN ALUMINUM OXIDE LAYER DISPOSED BETWEEN THE ALUMINUM ELECTRODE AND THE ELECTROLUMINESCENT MATERIAL

Paul A. Dell, Highland Heights, Ohio, assignor to General Electric Company, a corporation of New York

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The present invention relates, in general, to electroluminescent devices such as electroluminescent cells or lamps, and more particularly to an insulating antibreakdown or barrier layer therefor.

As known at present, electroluminescent cells or lamps, sometimes referred to as luminous capacitors, comprise in general a thin layer of a field-responsive phosphor sandwiched between a pair of electrically conductive layers or electrodes at least one of which is transparent or is, at least, light-transmitting. When an alternating voltage is applied across the electrodes, the phosphor emits visible light which then escapes through the transparent electrode.

In order to obtain superior operating characteristics, in particular, less susceptibility to arcing and greater efficiency, such electroluminescent cells or lamps have been customarily provided with a thin anti-breakdown or barrier layer of a dielectric or insulating material between the phosphor-bearing layer and one of the electrically conductive electrode layers, usually the non-light-transmissive back electrode layer. By employing for such insulating or barrier layer a material of improved permittivity and electrical breakdown strength as compared with the phosphor layer, the electroluminescence of the cell or lamp is increased since a greater voltage stress can be applied to the electroluminescent phosphor without breakdown. In addition, where the insulating layer employed is of light-reflecting character, good reflection of the light produced by the phosphor is also effected by the insulating layer, thereby producing greater lamp brightness at the light-transmitting conducting surface of the cell or lamp.

It is an object of the invention to provide an improved electroluminescent device having a simple, inexpensive and effective insulating barrier layer for permitting the application to the device of electric fields of greater intensity, without likelihood of electrical breakdown, to thereby increase the light-emitting efficiency thereof.

Another object of the invention is to provide an electroluminescent device having a solvent-impermeable and mechanically stable insulating barrier layer affording greater stability of electrical characteristics with time and also affording temperature stability such as permits use of higher-temperature component materials and processing procedures.

Briefly stated, in accordance with one aspect of the invention, at least one of the electrode layers of an electroluminescent device, between which the phosphor-bearing layer is sandwiched, is comprised of a thin sheet or foil of aluminum at least that surface of which, at the interface with the phosphor-bearing layer of the device, is anodized to transform it into a thin insulating layer of aluminum oxide. In accordance with a further aspect of the invention, the anodized surface layer of the aluminum sheet or foil electrode is constituted of a coarsely crystalline and comparatively porous primary layer portion, and a dense secondary layer portion of relatively fine crystallinity filling the pores of the primary layer portion.

Further objects and advantages of the invention will

appear from the following detailed description of species thereof and from the accompanying drawing.

In the drawing, FIG. 1 is a sectional view, on a greatly enlarged scale, of an electroluminescent device incorporating the invention, and

FIG. 2 is a sectional view, likewise on a greatly enlarged scale, of a modified form of such an electroluminescent device.

Referring to the drawing, the electroluminescent cell or lamp there shown is comprised, in general, of the usual layer 1 of phosphor-impregnated dielectric material sandwiched between a pair of electrically conductive layers 2 and 3 which form the front and back electrodes, respectively, of the device. At least one of the electrode layers, e.g., the front electrode layer 2, is transparent or, at least, capable of transmitting light so as to permit the light generated by the phosphor-bearing layer 1 to pass out through the said front electrode layer. The assembly of the phosphor-bearing layer 1 and the two electrode layers 2 and 3 together constitute the electrically active elements of the electroluminescent cell or lamp.

To prevent the ingress of moisture from the atmosphere into the interior of the cell or lamp such as would cause the rapid deterioration thereof, the electrically active assembly of the cell is preferably encased in a thin outer encapsulating envelope 4 of thermoplastic material of high moisture impermeability such as, for example, polychlorotrifluoroethylene film, known as Kel F. The encapsulating envelope 4 may be formed of two sheets 5 and 6 of such highly water-impermeable thermoplastic material between which sheets the electrically active assembly of the cell is laminated under heat and pressure, while the space between the sheets is evacuated, to thereby effect the softening and heat-sealing together of the plastic sheets around their marginal edges so as to encapsulate the electrically active cell assembly. A thin, flexible film 7 of a suitable thermoplastic material such as low density polyethylene, or nylon 6 such as that known as Caplene, is preferably laminated into the cell between the front electrode layer 2 and the plastic top sheet 5 of the encapsulating envelope 4 for the purpose, among other things, of firmly holding the front electrode layer 2 in place and cementing it to both the underlying phosphor layer 1 and the overlying layer 5 of the encapsulating envelope 4. The electroluminescent cell or lamp is energized by applying a suitable potential such as an alternating voltage, for example, 120 volts 60 cycles A.C., to electrical conductors 8 and 9 connected to the respective electrodes 2 and 3 and sealed through and projecting laterally from the edges of the outer envelope 4. The conductors 8 and 9 are preferably formed of relatively fine mesh wire cloth, for example, 200 to 300 mesh, of suitable electrically conductive material such as copper or Phosphor-bronze, for instance, and they may be sealed between the two plastic sheets 5 and 6 at the time they are heat-sealed together to form the outer encapsulating envelope 4.

The back electrode layer 3, which customarily is of non-light-transmissive or opaque character, is composed of metal sheet or foil as described more fully hereinafter The other or light-transmitting front electrode layer 2 may consist of any of the well known types of light-transmitting electrode layers conventionally employed for such purpose in electroluminescent cells or lamps. For example, it may consist of a thin, light-transmitting coating of tin oxide or other metallic oxide such as indium oxide on a suitable substrate such as glass or light-transmitting plastic sheet, or it may be in the form of electrically conductive paper comprised of glass fibers which are coated with a thin light-transmitting film of electrically conductive material such as, for example, a metal or a metallic oxide, or an indium compound as described more fully in

U.S. Patent 2,849,339, Jaffe, dated Aug. 26, 1958, and assigned to the assignee of this invention. As shown, the front electrode layer 2 is of smaller surface area size than the back electrode 3 and the phosphor layer 1 thereon so as to leave a narrow margin of the phosphor layer 1 exposed all around its marginal edges. The phosphor and electrode layers 1, 2 and 3 as well as the plastic layer 7, have a configuration generally corresponding to that of the outer encapsulating envelope 4 which may be of any desired configuration such as rectangular, square, circular, elliptical, or any other shape.

The phosphor-bearing layer 1 may be any of the usual types employed for such purpose in electroluminescent cells or lamps, such phosphor-bearing layers comprising, in general, an electroluminescent phosphor such as, for example, zinc sulfide-zinc oxide with suitable activators such as copper, manganese, lead or silver, dispersed in a suitable dielectric matrix material. As is well known in the art, the dielectric matrix material of the phosphor layer 1 may consist either of a glass frit, or of a suitable organic polymeric material, preferably one having a high dielectric constant such as cyanoethyl cellulose plasticized with cyanoethyl phthalate as disclosed in U.S. Patent 2,951,865, Jaffe et al., issued Sept. 6, 1960, and assigned to the same assignee as the present invention.

In accordance with well known practice, an insulating barrier layer **10** of dielectric material is interposed between the back electrode layer **3** and the phosphor inpregnated layer **1** for the purpose, among other things, of imparting higher breakdown strength to the electroluminescent device in order to permit the application thereto of electric fields of greater strength, without electrical breakdown of the device, to thereby produce increased light emission therefrom. Heretofore, such insulating barrier layers **10** have generally consisted of a coating of dielectric material applied to the metal sheet or foil back electrode layer **3**. In accordance with the invention, such barrier layer coatings are obviated by forming the metal sheet or foil back electrode layer **3** of aluminum, preferably full-soft aluminum foil of a thickness of around 2 mils for example, and constituting the dielectric barrier layer **10** by an anodized surface layer portion of the aluminum sheet or foil electrode **3**. As is well known in the art, an anodized aluminum surface layer consists of aluminum oxide which is inherently electrically insulating in character and possesses comparatively good dielectric properties. Moreover, because the anodized layer **10** is formed by a chemical transformation of the surface layer of the aluminum sheet or foil **3** itself, it therefore constitutes an integral part of the aluminum sheet or foil electrode 3 in the form of a tightly adhering film or layer thereon. As a result, it is not subject to separation from the aluminum back electrode layer **3** such as may occur with the prior type separate barrier coatings, with resulting deterioration and ultimate failure of the electroluminescent cell or lamp. Anodized aluminum in sheet or foil form is available commercially at relatively low cost, as compared to the expense of applying a separate coating or layer of dielectric material onto the back electrode layer **3** of an electroluminescent cell in accordance with prior general practice. Also, because of the high temperature stability of anodized aluminum and its nonsusceptibility to being softened by heat, and further because of its solvent-impermeable character such as renders it nonsusceptible to being softened by the common solvents customarily employed in the coating suspensions which are commonly used to form the phosphor-bearing layers **1** of electroluminescent devices, the use of an anodized aluminum surface layer as a dielectric barrier layer **10** in an electroluminescent device is of particular added advantage. Among other things, a wider latitude in the choice of manufacturing procedures for the fabrication of electroluminescent devices is afforded through the use of an anodized aluminum surface layer for the dielectric barrier layer **10** of the device.

To increase the dielectric strength of the barrier layer beyond that of the anodized aluminum surface layer **10**, an additional layer or coating **11** of high dielectric constant material, such as the conventional type coating of barium titanate dispersed in an organic polymeric matrix material such as that employed for the phosphor-bearing layer **1**, may be provided, if desired, between the phosphor-bearing layer **1** and the anodized aluminum surface layer **10** of the aluminum back electrode **3**, as shown in the modification illustrated in FIG. 2. Such a modified form of electroluminescent device forms the subject matter of copending application Ser. No. 232,699, Gunther H. Dierssen, filed of even date herewith and assigned to the same assignee as the present application. The anodized aluminum surface layer **10** in such a modified form of electroluminescent device affords improved adherence of the barium titanate coating **11** to the back electrode **3**, thereby minimizing the likelihood of any separation occurring therebetween such as would result in deterioration and ultimate failure of the device.

Where the anodized aluminum surface layer **10** of the aluminum back electrode **3** alone serves as the dielectric barrier layer between the phosphor layer **1** and the back electrode layer **3** of the electroluminescent device, as in the form of the invention shown in FIG. 1, then the anodized aluminum surface layer **10** in such case is necessarily made of substantially greater thickness than the very thin natural skin of aluminum oxide which forms on bare aluminum when exposed to the atmosphere, such a natural skin of aluminum oxide not possessing a sufficiently high dielectric strength for use as the barrier layer **10**. For the purposes of the invention, the anodized aluminum surface layer **10**, particularly where it serves as the only dielectric barrier layer in an electroluminescent lamp as in FIG. 1, should have a thickness of at least ½ to 1 micron or so, for lamps of maximum brightness operated at 120 volts and 60 cycles. However, for higher voltage lamps it may be desirable to employ anodized aluminum layers **10** of much greater thicknesses even ranging up to as high as 25 microns or so. Where the anodized aluminum surface layer **10** is provided primarily for the purpose of affording improved adherence of another or primary dielectric layer **11** such as a conventional barium titanate layer to the back electrode **3**, as in the case of the modified form of electroluminescent device shown in FIG. 2, then in such case the anodized aluminum surface layer **10** can be of considerably thinner character than the minimum thickness specified above for an anodized aluminum surface layer serving as the only dielectric barrier layer in an electroluminescent device. For example, the anodized aluminum surface layer **10** in such case may have a thickness as little as 0.1 micron or less.

While for the purposes of the invention the anodization of the aluminum sheet or foil back electrode layer **3** to form the aluminum oxide surface layer **10** thereon may be carried out by the use of most any of the well known processes for anodizing aluminum, for best results it is preferred to form the anodized surface layer **10** by a process which results in the formation on the aluminum sheet or foil electrode **3** of a composite aluminum oxide layer comprised of a comparatively porous promary layer portion of relatively coarse crystallinity and a comparatively dense aluminum oxide secondary layer portion of relatively fine crystallinity which fills the pores of the porous layer portion to thereby produce a composite aluminum oxide film of good dielectric strength and substantially free of pinholes such as would ordinarily lead to electrical short circuits in the electroluminescent device or greater electrical losses in the operation thereof. Because of their good dielectric properties, barrier layers **10** consisting of such composite anodized aluminum films can be kept to a minimum thickness, i.e., as thin as the aforementioned thicknesses

of ½ to 1 micron or so, while still affording sufficient protection against electrical breakdown. This, of course, is of distinct advantage since the thinner the insulating layer **10**, the greater will be the capacitance of the electroluminescent device and the lower will be the voltage loss across the insulating layer **10**. As a result, a greater proportion of the operating voltage applied to the electroluminescent device will be impressed across the phosphor-bearing layer **1**, thus producing maximum light output therefrom.

A composite anodized aluminum film or layer **10** of the character referred to above may be produced by a two-stage electrolytic anodizing process wherein the bare aluminum sheet or foil **3** the surface of which may, if not sufficiently clean, be suitably cleaned as by immersion in a chromic acid bath for example, is first electrolytically anodized in an electrolyte consisting of an approximately 3% aqueous solution of oxalic acid to thereby produce the porous primary layer of aluminum oxide of relatively coarse crystallinity which constitutes the major portion of the total thickness of the final composite aluminum oxide film. The so-anodized aluminum sheet or foil **3** is then subjected to a secondary electrolytic anodization process in an electrolyte consisting of an approximately 10% aqueous solution of boric acid, to thereby form the dense secondary layer of aluminum oxide of relatively fine crystallinity which fills and seals off the pores of the porous aluminum oxide layer. The secondary anodization step is continued until the desired overall thickness of the composite aluminum oxide film **10** is attained on the aluminum sheet or foil electrode **3**.

Inasmuch as the anodization of the aluminum sheet or foil electrode **3** by most any of the known aluminum anodizing processes normally results in the formation of an aluminum oxide film (which is inherently electrically insulating) on both sides of the aluminum sheet or foil **3**, the electrical connection of the inlead conductor **9** to the anodized aluminum sheet or foil electrode **3** may be made by suitably removing, as by abrading, a portion of the insulating film of aluminum oxide from the outwardly facing side of the aluminum electrode **3** so as to expose the bare aluminum and then positioning the conductor **9** opposite the so abraded surface area of the aluminum electrode so as to be pressed and held thereagainst by the thermoplastic encapsulating sheet **6** to thus effect the electrical connection therewith. As shown, the other conductor **8** is electrically insulated from the back electrode **3** by a small strip **8'** of suitable insulating material.

What I claim as new and desire to secure by Letters Patent of the United States is:

1. An electroluminescent device comprising a layer of aluminum forming a first electrode, a thin transparent layer of conductive material forming a second electrode, a layer of electroluminescent phosphor-impregnated dielectric material between said first and second electrode layers forming an electroluminescent layer, and a layer of aluminum oxide tightly adhered to said aluminum layer and disposed between said aluminum layer and said electroluminescent layer, said layer of aluminum oxide being of a thickness greater than the thickness of the natural skin of aluminum oxide which forms on aluminum when exposed to the atmosphere.

2. An electroluminescent device according to claim **1** in which said layer of aluminum oxide is of a thickness within the range of approximately ½ to 1 micron.

3. An electroluminescent device comprising a back electrode layer of sheet aluminum, a light-transmitting electrically-conductive front electrode layer, and a phosphor-bearing layer disposed between the said electrode layers, the said aluminum sheet electrode having an anodized insulating surface layer of aluminum oxide at its interface with said phosphor-bearing layer, said layer of aluminum oxide being of a thickness greater than the thickness of the natural skin of aluminum oxide which forms on aluminum when exposed to the atmosphere and ranging up to approximately 25 microns.

4. An electroluminescent device as specified in claim **3** wherein the said anodized surface layer of the aluminum sheet electrode is constituted of a comparatively porous primary layer of relatively coarse crystallinity and a dense secondary layer of relatively fine crystallinity filling the pores of the said porous layer.

5. An electroluminescent device as specified in claim **3** wherein the said anodized surface layer of the aluminum sheet electrode is constituted of a comparatively porous primary layer of relatively coarse crystallinity and a dense secondary layer of relatively fine crystallinity filling the pores of the said porous layer, the total thickness of the composite anodized aluminum surface layer being within the range of approximately ½ to 1 micron.

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JAMES W. LAWRENCE, *Primary Examiner.*

GEORGE N. WESTBY, *Examiner.*

R. JUDD, C. R. CAMPBELL, *Assistant Examiners.*