

March 29, 1966

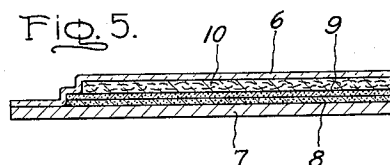
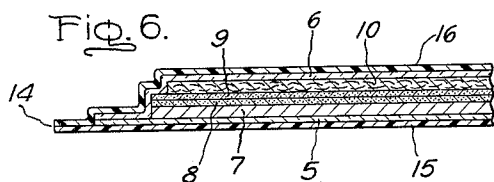
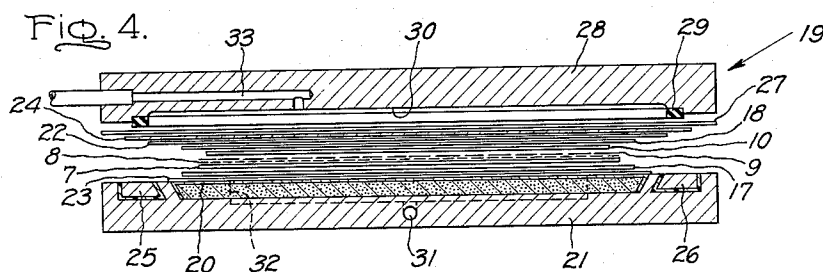
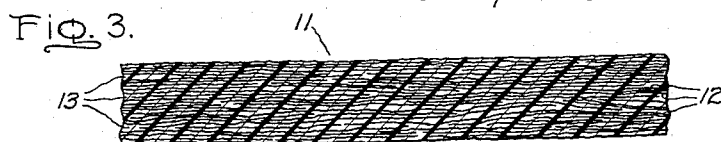
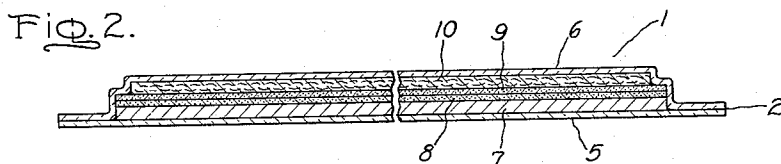
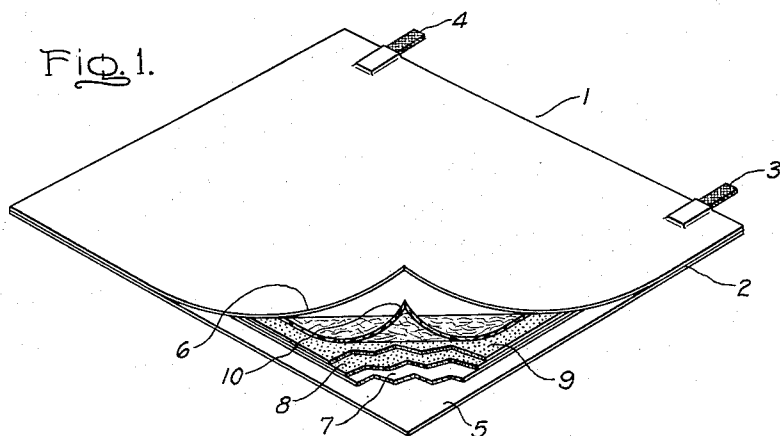
R. V. LEVETAN

3,243,629

ELECTROLUMINESCENT LAMP AND MANUFACTURE THEREOF

Filed June 19, 1961

2 Sheets-Sheet 1



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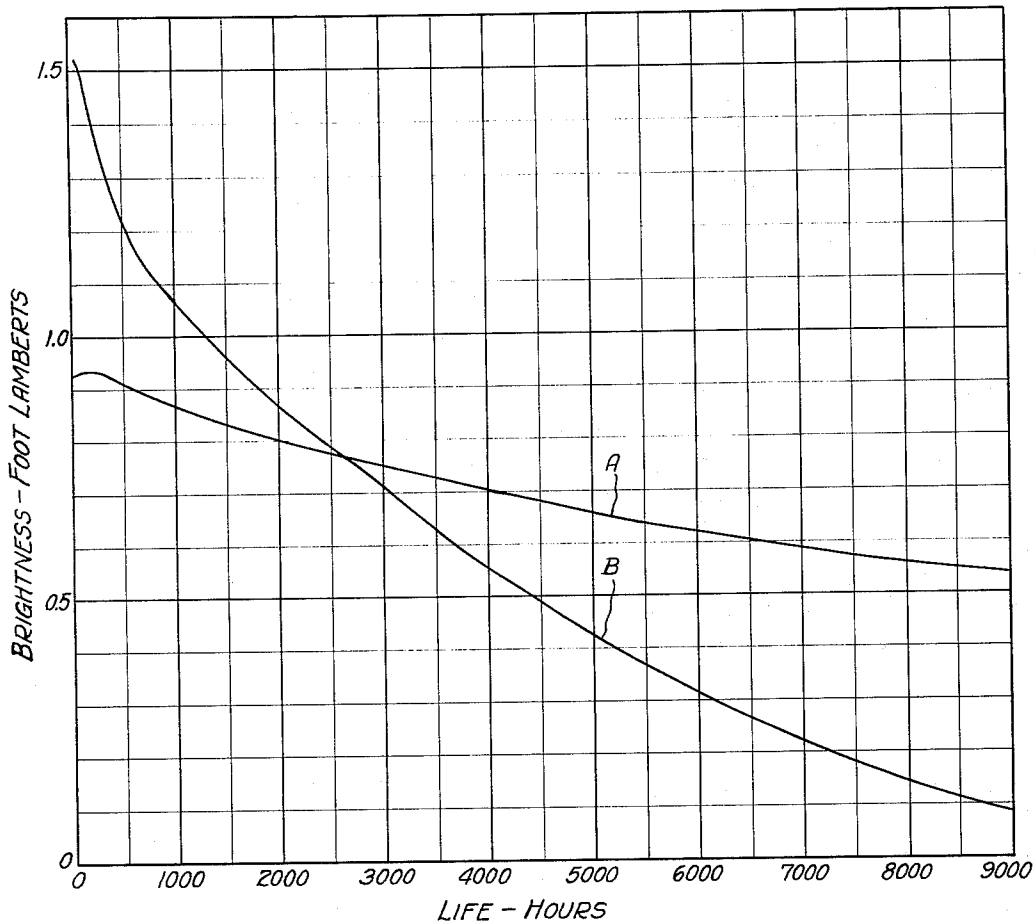
3,243,629

ELECTROLUMINESCENT LAMP AND MANUFACTURE THEREOF

Filed June 19, 1961

2 Sheets-Sheet 2

FIG. 7.



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3,243,629

ELECTROLUMINESCENT LAMP AND MANUFACTURE THEREOF

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Filed June 19, 1961, Ser. No. 118,112
15 Claims. (Cl. 313-108)

This invention relates in general to electroluminescent lamps or cells and more particularly to a flexible laminated electroluminescent lamp. The invention is also concerned with a process for the manufacture of such lamps.

An electroluminescent lamp or cell generally comprises a layer of phosphor capable of generating light under the action of an electric field and sandwiched between a pair of conducting plates or films at least one of which is transparent or light transmitting. One form of electroluminescent lamp now well-known is described and claimed in U.S. Patent No. 2,774,004, Jaffe, assigned to the same assignee as the present invention and comprising a flexible laminated assembly of electrically active layers or elements each of which is of flexible character. The electrically active elements comprise an aluminum foil coated with a layer of high dielectric constant material which in turn is overcoated with a layer of electroluminescent phosphor and finally overlaid with a transparent flexible conductive sheet such as a thin sheet of compacted glass fibers in the form of conducting glass cloth or paper. The aluminum foil and the conducting glass paper form the electrodes of the lamp to which an alternating potential is supplied to cause the electroluminescent phosphor to generate light which is then emitted through the transparent conductive glass paper.

The known electroluminescent phosphors commonly employed at present for electroluminescent lamps are extremely sensitive to water vapor and depreciate and darken rapidly in its presence with corresponding loss of light output from the lamp. Without the provision of some form of protective means against the ingress of moisture into the lamp from the surrounding atmosphere, the brightness of the lamp falls off very rapidly as the period of lamp usage extends past a few hours. In many instances, the brightness of the electroluminescent lamp falls to less than one-half its initial brightness in a period as short as ten hours. For this reason, therefore, practical forms of electroluminescent lamps have generally included the provision of some form of protective water vapor barrier for encapsulating or sealing the electroluminescent phosphor from the atmosphere in order to exclude moisture and prevent its coming into contact with the phosphor.

Such water vapor barrier may, of course, be constituted of glass such as a non-porous glass matrix for the phosphor particles in which they are suspended or imbedded, or a glass overlay, as a layer of glass or a glass sheet or glass film which is sealed over the phosphor layer and which may be also provided with a transparent coating of electrically conductive material such as tin oxide to constitute the glass overlay as the light-transmitting conductive member of the lamp. While such glass-type water vapor barriers effectively prevent the passage of water vapor therethrough, nevertheless their use for such purpose in an electroluminescent lamp normally results in an attendant sacrifice of flexibility and ruggedness for the lamp because of the substantially non-flexible and fragile character of the glass itself. Thus, electroluminescent lamps with such a glass imbedded phosphor layer or with a glass sheet type transparent conductor cannot be flexed or formed into any desired shape, nor can they be subjected to any more rugged applications than any other large piece of fragile glass without fracturing the

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glass. Sheet mica may also be employed as a water vapor barrier layer for electroluminescent lamps, but it is far too expensive, in the larger sizes needed for such application, to render it economically practical for such purpose.

The only materials, therefore, which have been proposed and employed heretofore as water vapor barriers for flexible type electroluminescent lamps have been certain organic plastic transparent sheet or film materials of comparatively low water vapor permeability possessing the necessary flexibility and ruggedness to permit their incorporation in the lamp without any undesirable sacrifice in the flexibility and ruggedness thereof. However, even the most water-vapor impermeable of the known organic plastic sheet or film materials are not sufficiently impermeable to exclude water vapor completely over the life of an electroluminescent lamp. It has been observed, for instance, that even where a laminated electroluminescent lamp is provided with a thermoplastic encapsulating envelope consisting of high-density polyethylene, or of yet more water vapor impermeable polychlorotrifluoroethylene (Kel-F), deterioration of the lamp and loss of light output therefrom sets in after around one hundred hours or so of operation in a maximum humidity environment. Of course, the advent of such deterioration may be further delayed by resorting to the use of thicker sheets for the encapsulating envelope, but this is not an economically attractive expedient.

It is an object of the present invention, therefore, to provide an improved electroluminescent lamp of long life and having good brightness maintenance even under highly humid operating conditions.

Another object of the invention is to provide electroluminescent lamps of flexible and rugged construction and possessing improved brightness maintenance characteristics.

Still another object of the invention is to provide a construction for improving the resistance to water vapor depreciation, and therefore the life and maintenance, of electroluminescent lamps encapsulated in flexible water vapor barrier materials.

A further object of the invention is to provide an electroluminescent lamp of initial moisture-free character and highly resistant to the penetration of water vapor therein from the surrounding atmosphere.

A still further object of the invention is to provide a novel method of making an electroluminescent lamp with an effective water vapor barrier incorporated therein.

Briefly stated, in accordance with one aspect of the invention, an electroluminescent lamp or cell is provided with a transparent flexible water vapor barrier laminated thereto and comprised of resin-impregnated micaceous sheet material such as that described and claimed in my co-pending U.S. application Serial No. 118,113, filed of even date herewith and assigned to the same assignee as the present invention. The said micaceous sheet material comprises mica flake paper such as, for example, that commonly known as mica mat impregnated with from 33 to 55% by volume of a suitable resin which substantially completely fills all the interstitial space within the mica paper between the compacted coplanar and overlapping individual mica flakes thereof to firmly bond the mica flakes together and transform the mica paper into a tough, flexible and transparent sheet material of exceptionally low water vapor permeability and of high tensile strength and moisture resistance. The resin impregnant of the mica flake sheet material forming the water vapor barrier of the lamp may be either a polyester or an epoxy resin, but is preferably constituted by an anhydride-cured epoxy resin such as, for example, an almost pure diglycidal ether of Bisphenol-A or a dicyclo-diepoxy carboxylate.

According to a further aspect of the invention, the formation of an electroluminescent lamp with an effective water vapor barrier is performed by assembling the electrically active elements of the lamp, comprised of the electroluminescent phosphor layer sandwiched between the two electrically conductive layers or electrodes one of which may consist of conductive microfibre glass paper, between top and bottom sheets of mica flake paper overreaching the marginal edges of the electrically active assembly and impregnated with fluid resin-forming material or resin solution, and then laminating the layered assembly of the said elements under high pressure and heat in a hydrostatic press such as that described in my aforesaid co-pending application Serial No. 118,113 to effect the firm bonding together of the various layers or elements and the coalescing and sealing together of the marginal edges of the mica flake paper sheets so as to encapsulate the assembly, and simultaneously cure the resin impregnant in the mica flake paper in situ to effect an adhesive bonding of the latter to the adjacent layers of the assembly throughout the interfaces therewith and at the same time transform the mica flake paper into a tough, flexible and transparent sheet material of exceptionally low water vapor permeability such as to constitute it a highly efficient water vapor barrier for the lamp. By impregnating the mica paper with an excess of resin solution over the amount required to completely fill all the interstitial space within the mica paper, the excess resin solution is squeezed out of the mica paper, during the pressing and heating cycle, so as to flow down into and between the fibres of the conducting glass paper electrode and thereby hold it in intimate surface contact with the immediately adjacent phosphor layer in the finished lamp.

In accordance with a still further aspect of the invention, the fluid resin impregnant of the mica flake paper is constituted of a mixture of an epoxy resin with an anhydride hardening agent a small part of which, during the pressure and heat laminating of the lamp, chemically combines with any residual water vapor remaining in the lamp to thereby effectively remove it therefrom and thus produce a substantially moisture-free or dry lamp exhibiting very little initial depreciation in its light output over the first several hundreds of hours of its operation such as would be caused by a small amount of entrapped moisture.

Further objects and advantages of my invention will appear from the following detailed description of species thereof and from the accompanying drawing.

In the drawing, FIG. 1 is a pictorial view of a flexible laminated electroluminescent lamp according to my invention with the various constituent layers thereof delaminated or peeled open at one corner to show the internal construction of the lamp.

FIG. 2 is a fragmentary cross-sectional view on an enlarged scale of the electroluminescent lamp comprising my invention.

FIG. 3 is a fragmentary cross-sectional view, on a greatly magnified scale, of the water vapor barrier element of the electroluminescent lamp comprising my invention.

FIG. 4 is a vertical sectional view through the hydrostatic press which is employed to fabricate the electroluminescent lamp in accordance with the method of my invention, with the pressure and vacuum plates of the press shown in place and the lay-up of the electroluminescent lamp components stacked in place on the bottom or vacuum plate of the press.

FIG. 5 is a fragmentary sectional view, on an enlarged scale, of a modified form of electroluminescent lamp according to the invention.

FIG. 6 is a fragmentary sectional view, likewise on an enlarged scale, of another modified form of electroluminescent lamp according to the invention, and

FIG. 7 is a graph illustrating the brightness characteristic of the electroluminescent lamp comprising my

invention in comparison with that of prior type electroluminescent lamps.

Referring to FIG. 1, there is shown an electroluminescent lamp 1 having one corner delaminated or peeled open to show the constituent layers and the internal construction of the lamp. The thickness of the layers has been exaggerated for the sake of illustration. The lamp or cell 1 is flat and rectangular and is made up of flexible components laminated together and entirely sealed or encapsulated in an enclosure or envelope 2 composed of transparent flexible sheet material of low water-vapor permeability so as to constitute it a water-vapor barrier for the lamp. The lamp is energized by applying an alternating voltage, for instance, 120 volts, 60 cycle A.C., to copper screen terminals 3, 4 projecting laterally from the edge of the encapsulating envelope 2 and electrically connected to the conductive electrode layers thereof. The underside and the topside of the lamp envelope 2, consisting of sheets 5 and 6 of the transparent water-vapor barrier sheet material, are laminated or sealed together along their margins, by means of the resin impregnant therein, to thereby encapsulate the lamp.

The electrically active elements of the electroluminescent lamp, i.e., the light-producing components thereof, comprise a rectangular sheet of metal foil 7, for instance full-soft annealed aluminum of .0022" thickness, coated with an insulating layer 8 of high dielectric constant material, the latter overcoated with a light-producing layer 9 of electroluminescent phosphor. The aluminum foil sheet 7 constitutes one of the conductive electrode layers of the lamp and is placed over the lowermost sheet 5 of the encapsulating envelope 2, leaving a clear margin all around, as shown in FIG. 1. The insulating layer 8 may consist of barium titanate dispersed in an organic polymeric matrix of high dielectric constant such as cyanoethyl cellulose plasticized with cyanoethyl phthalate according to U.S. Patent No. 2,951,865, Jaffe et al., issued Sept. 6, 1960. The electroluminescent layer 9 may consist of any known electroluminescent phosphors such as zinc sulfide-zinc oxide with suitable activators such as copper, chlorine and manganese, likewise dispersed in an organic polymeric matrix such as that used in connection with the insulating layer 8. A thin fibrous transparent electrically conductive sheet 10, constituting the other electrode of the lamp, is laid over the phosphor coating 9 on the foil 7, leaving a narrow margin of the phosphor-coated foil uncovered all around. The fibrous sheet 10 may consist of commercially available microfibre glass paper approximately .0001" thick, which is made electrically conductive by dipping in a solution of indium basic trifluoroacetate in an organic solvent and baking at an elevated temperature, according to U.S. Patent No. 2,849,339, Jaffe, issued Aug. 26, 1958. For a more complete description of the construction of the electrically active components of the electroluminescent lamp 1, reference may be made to U.S. Patent No. 2,945,976, Fridrich et al., issued July 19, 1960.

In accordance with the invention, the transparent flexible sheet material employed for the encapsulating envelope 2 to serve as a water vapor barrier for the lamp 1 is constituted of thin micaceous sheet material 11 (FIG. 3) as described and claimed in my co-pending U.S. application Serial No. 118,113 previously referred to. Such micaceous sheet material 11 is comprised, in general, of mica flake paper such as that commonly known as mica mat, for example, and composed of many layers of extremely thin mica flakes or platelets 12 which have been compacted together, by conventional paper making techniques, into the form of a sheet in which the individual mica platelets 12 are oriented in a generally coplanar and overlapping or shingled arrangement and are bonded together by their own cohesive forces, the mica paper being additionally provided with a filling 13 of a controlled amount of a suitable organic plastic or resin filler material, in the manner disclosed in my afore-

said co-pending application Serial No. 118,113, to thereby further compact and firmly bond together all the individual mica flakes 12 and substantially completely fill all the interstitial space within the mica paper. By virtue of such impregnation of the mica paper with the plastic or resinous filler material 13, the mica paper is transformed from a fragile and brittle sheet material of silvery and translucent appearance and of high water-vapor permeability, into a tough, flexible, transparent sheet material of high light transmissivity, amounting to in excess of 80% and ranging up to as high as 87% or so, and of exceptionally low water-vapor permeability. In fact, for equivalent thicknesses, the water-vapor permeability of the resin-impregnated micaceous sheet material 11 is many times, i.e., from 5 to 10 times, less than that of the least water-vapor permeable plastic sheet material known at present, i.e., polychlorotrifluoroethylene plastic film known as Kel-F.

For the purposes of the invention, the resin filler material 13 present in the finished sheet material 11 is controlled so as to more or less correspond in volume to all the interstitial space within the mica paper between the mica flakes 12 thereof and, in the finished product, is uniformly distributed throughout such interstitial space so as to completely fill the same. In any case, however, the amount of resin filler material 13 should be somewhat greater, but not less than the minimum amount required to completely fill all the said interstitial space within the mica paper. To this end, therefore, it has been determined that the resin filler content in the finished sheet material 11 should range from around 33 to 55% by volume, and preferably between 40 to 50% by volume, of the composite sheet material.

In accordance with the invention, the resinous substance employed for the impregnation of the mica paper is comprised of a low-viscosity resin system or resin solution capable of readily penetrating the pores of the mica paper and readily wetting the surface of the mica flakes and forming a good bond thereto, and consisting of a solution of totally inter-reactive resin-forming substances and curing agent materials which, during the heat-curing thereof preferably in the presence of a catalyst, chemically react completely with one another without the formation of any volatile byproducts or any liberation of volatile solvents such as would require subsequent removal thereof from the impregnated mica paper and would be likely to produce gas bubbles or voids in the final cured resin-filled sheet material. Any such gas bubbles, or any trapped air pockets in the final cured resin filled sheet material 11 would produce multiple light reflections therein (due to the high difference in the refractive index at the interface between the air or gas of the bubble and the mica or resin surface) such as would act to impart a high internal reflection characteristic to the sheet material and render it translucent and pearly in appearance, rather than transparent in accordance with the objects of the invention. Resinous materials of such characteristics, therefore, which are suitable for use as impregnants for the particular purposes of the invention are solutions of low molecular weight polyester resins dissolved in styrene, methylstyrene or mixtures of these monomers and preferably containing, in addition, a small amount of a suitable catalyst such as benzoyl peroxide, or solutions of epoxy resins and a suitable curing or hardening agent such as an amine or an organic anhydride and preferably containing in addition, in the case of the anhydride, a small amount of a suitable catalyst such as N,N-dimethyl-benzylamine or trimethylol propane. A specific example of a suitable low molecular weight polyester resin type of impregnating material is composed of a solution or mixture, in equal parts by weight, of a polyester resin such as that commercially known as AR-943, manufactured by applicant's assignee, General Electric Company, dissolved in styrene, the resin solution containing in addition a small amount of around

0.8% or so by weight of a suitable catalyst such as benzoyl peroxide.

The resinous filler materials 13 which have been found to be the most suitable and are preferred, however, for the purposes of the invention are anhydride-cured epoxy resins or diepoxides, and particularly those formed from liquid type epoxy resins or diepoxides by curing with an organic anhydride, preferably in the presence of a suitable catalyst. Epoxy resins are known to give good adhesive bonds. Also, they "wet" readily to polar surfaces such as the surface of the mica flakes 2 of which mica paper is normally composed. Because of this property, therefore, the use of such epoxy resins as the impregnating material for the mica paper assures the complete displacement or driving out of all the air present in the interstices thereof, and in the surface irregularities or adsorbed on the surfaces of the mica flakes, during the resin-impregnating treatment of the mica paper in accordance with the invention. As a result, no trapped air pockets are left in the final sheet product, after the pressure compacting and heat-curing thereof, such as would produce surface reflections therein and consequent impairment of the transparency of the resin-impregnated mica paper. The low molecular weight, low-viscosity liquid type epoxy resins are preferred for use as the impregnating material because of the greater ease and assuredness of obtaining complete penetration of the mica paper with such liquid type epoxy resin impregnants. Liquid epoxy resins which have been found particularly suitable for the purposes of the invention are, for example, the Bisphenol-A diglycidal ethers such as those commercially known as D.E.R.-332 and D.E.R.-331, manufactured by the Dow Chemical Company and that commercially known as Epoxide 828 manufactured by Shell Chemical Company. Other suitable liquid epoxy resins are the aliphatic diepoxides such as those commercially known as UNOX-201 and UNOX-207 manufactured by Union Carbide Chemicals Company and comprising, respectively, the dicyclo diepoxy carboxylate identified as 3,4-epoxy-6-methylcyclohexylmethyl - 3,4 - epoxy-6-methylcyclohexanecarboxylate, and the dicyclodiepoxy identified as dicyclopentadiene dioxide.

For the impregnation of the mica paper, the epoxy resin is mixed with any suitable organic anhydride type curing or hardening agent in the approximate proportions of around 0.8 to 1 mole of the anhydride per mole of the resin, for example, 87½ parts (or approximately 45 to 50%) by weight of hexahydro phthalic anhydride to 100 parts (or approximately 50 to 55%) by weight of the D.E.R.-332 resin, or 49½ parts (or approximately 30 to 35%) by weight of hexahydro phthalic anhydride to 100 parts (or approximately 65 to 70%) by weight UNOX-201 resin. This produces a clear, nearly colorless oily liquid which is stable at room temperature for in excess of several days. If desired, in the case of D.E.R.-332 epoxy resin, from 10 to 40% of the epoxide may be replaced with a like amount by weight of a silicone diepoxide monomer such as that known as Sylkem-90, manufactured by the Dow Chemical Company. Hexahydro phthalic anhydride is preferred as the organic anhydride curing agent because of its advantage in forming a fluid and mobile formulation of stable and homogeneous character at room temperature, and also because it affords a rapid rate of cure at higher temperatures. However, other organic anhydrides may be employed as well, such as phthalic, tetrahydro phthalic, methyl tetrahydro phthalic, methyl nadic, succinic, dodecenyl succinic, dichloromaleic, chlorendic, glutaric and pyromellitic dianhydride. Immediately before the impregnation of the mica paper with the fluid epoxy resin and anhydride mixture, a suitable accelerator or catalyst is preferably added to the mixture, in a small amount of around 1 to 3.5% by weight of the epoxy resin (or around 0.5 to 2.0% of the total admixture) for the purpose of increasing the curing rate of the epoxy resin. In the case of D.E.R.-332 epoxy

resin, a suitable catalyst is N,N-dimethyl-benzylamine or other tertiary amine in an amount of about 0.5% by weight of the total admixture, while in the case of the UNOX-201 and UNOX-207 epoxy resins a suitable catalyst is trimethylol propane or other aliphatic polyol in an amount of about 2% by weight of the total admixture.

The resin filled mica flake sheet material **11** employed for the encapsulating envelope **2** of the electroluminescent lamp **1** as a water vapor barrier therefor may be prepared separately in the manner disclosed and claimed in my aforesaid co-pending application Serial No. 118,113 after which the finished sheet material **11** is then assembled together with the other components of the electroluminescent lamp **1** and laminated together therewith under pressure and heat in a manner similar to that described hereinafter in connection with FIG. 4. In such case, however, the lamp is baked out before the lamination thereof, in order to drive out any moisture present therein, i.e., to dry the lamp, after which the lamp is then preferably enclosed within a sealed outer envelope **14**, as shown in FIG. 6, formed of two sheets **15** and **16** of suitable thermoplastic film material such as cellulose acetate butyrate which, during the lamination of the lamp, softens and heat-seals together around the margins of the water-vapor barrier envelope **2** and around the projecting lamp terminals **3**, **4** so as to form a sealed enclosure completely encapsulating the electroluminescent lamp **1** including the water-vapor barrier envelope **2**. The use of such a sealed outer encapsulating envelope **14** is desirable in such case for the reason that, because of the thermosetting character of the resin impregnant which is preferably employed in the micaceous sheet material **11**, the resin impregnant is in a cured state at the time of the lamp laminating operation and therefore is no longer capable of softening under heat, during such lamp laminating operation, to thereby effect the sealing together of the two micaceous sheets **5** and **6** around the electrically active elements of the electroluminescent lamp to form a sealed encapsulating envelope therearound. The use of such an outer envelope **14** of thermoplastic film material also affords a convenient way of holding the assembled layers or elements of the lamp together for ease of handling prior to the final pressing or laminating thereof into the finished lamp assembly **1**, the two sheets **15** and **16** of thermoplastic material being heat-tacked together for such purpose at several spaced points around their outer margins outwardly of the assembled lamp-forming elements to thereby hold the latter together in proper relation.

In accordance with the preferred method of making the electroluminescent lamp **1** comprising my invention, the micaceous sheet material **11** is not preformed separately and then assembled and laminated together with the other elements of the lamp to form the completed lamp assembly **1**, but instead is pressed and heat-cured into its final form simultaneously with the pressure and heat laminating together of the electrically active elements **7**, **8**, **9** and **10** of the lamp. Besides reducing the number of manufacturing operations and thus simplifying lamp manufacture, the use of such a method of fabricating the electroluminescent lamp **1** also enables the production, where an anhydride-cured resin is employed as the impregnant for the micaceous sheet material **11** forming the lamp encapsulating envelope **2**, of a lamp which is initially of substantially moisture-free character and which maintains its initial light output, even under adverse moisture conditions, for an appreciable length of time up to as much as 800 hours or so of life at 85% relative humidity. This initial moisture-free character of such a lamp is due to the fact that a small portion of the anhydride which is present in the resinous impregnant of the mica paper acts as a getter, during the lamp laminating operation, to pick up and chemically combine with any residual moisture remaining in the lamp at such time, thus eliminating such residual moisture from the completed lamp assembly

which otherwise would cause early depreciation in lamp light output. The product of this reaction of the anhydride with the residual moisture is the corresponding acid which is also capable of reacting with the epoxide present in the fluid resin impregnant in the mica paper to effect the curing or hardening of the resin during the pressure and heat laminating of the lamp.

In producing an electroluminescent lamp **1** by the improved process according to the invention, the electrically active elements of the lamp, comprising the aluminum foil conductor layer **7** coated with the insulating layer **8** and overcoated phosphor layer **9** and overlaid with the conductive glass fiber paper **10**, is sandwiched between two sheets **17** and **18** of mica flake paper or mica mat which have been impregnated with a controlled amount of a fluid resin impregnant according to the invention in the manner such as disclosed in my aforesaid co-pending application Serial No. 118,113, i.e., by immersing or laying the mica paper or mica mat in a controlled thickness layer or coating of the fluid resin impregnant on a suitable backing sheet, such as a glass plate or a sheet of polyethylene terephthalate film such as Mylar or similar plastic material, to cause the mica paper to pick up or absorb enough of the fluid resin coating to correspond in volume to, and preferably slightly exceed the amount required to completely fill, in the finished lamp, all the interstitial space within the mica paper including all the surface irregularities of the mica flakes of which the mica paper is composed. Where the fluid resin impregnant is of the type employing an anhydride as the curing agent, the mica paper is covered with a protective sheet of suitable moisture-impervious plastic material such as Mylar or polyethylene during the fluid resin impregnation operation in order to preclude the water vapor present in the surrounding atmosphere from coming in contact with, and chemically reacting with the anhydride component of the impregnant. For this reason, also, after the stripping or removal of the fluid resin impregnated mica paper off the backing sheet or support, it is sandwiched between two such protective sheets of moisture-impervious material and stored in a desiccator, preferably for a period of at least 24 hours or so before use in the fabrication of the lamp.

The assemblage of the electrically active elements **7**, **8**, **9** and **10** of the electroluminescent lamp sandwiched between the two sheets **17** and **18** of mica paper impregnated with resin solution is then subjected to pressure and heat, in a manner similar to that disclosed in co-pending U.S. application Serial No. 748,537, Fridrich, filed July 14, 1958, now Patent No. 3,047,052, and by the use of a hydrostatic press **19** such as shown in FIG. 4 and described more fully in the said Fridrich application and in my co-pending application Serial No. 118,113, to thereby laminate the various layers or elements of the lamp together into a unitary sealed lamp structure **1** from which the residual moisture has been eliminated by the gettering action of the anhydride present in the fluid resin impregnant of the mica paper sheets **17** and **18** and, at the same time, perform the necessary further processing operations on the fluid resin impregnated mica paper sheets **17** and **18** corresponding to those further processing operations described in my said co-pending application Serial No. 118,113, in order to thereby complete the processing of the mica paper sheets into the finished micaceous sheet material **11** and transform them into an effective water vapor barrier of transparent and flexible character and exceptionally low water vapor permeability. As shown in FIG. 4, the lamp-forming assemblage of the electrically active elements **7**, **8**, **9** and **10** sandwiched between the two fluid resin-impregnated mica paper sheets **17** and **18**, is laid flat on the flat stainless steel upper surface **20** of the bottom or vacuum plate **21** of the hydrostatic press **19**, the lamp-forming assemblage preferably being sandwiched between two release sheets **22** and **23** of electrically non-conductive thin flexible sheet material which may con-

veniently consist of polytetrafluoroethylene film such as Teflon, in order to permit ready separation of the finished lamp from the immediately adjacent pressure-applying members of the hydrostatic press following the compressing and heating operation. A thin flexible sheet 24 of electrically conductive material such as soft annealed aluminum foil around 0.001 inch thick, and serving as the heater element of the press 19, is laid over the top Teflon release sheet 22 and in contact with copper bus bars 25 and 26 recessed in the upper side of, but insulated from, the stainless steel upper side 20 of the vacuum plate 21. An electrically non-conductive gas impervious sheet 27 of thin, flexible material, which may suitably consist of polyethylene terephthalate film such as Mylar and which serves as a conformable diaphragm for applying pressure to the lamp-forming assemblage through the flexible aluminum foil heater element 24 and the top Teflon release sheet 22, is laid over the aluminum foil sheet 24. A top or pressure plate 28 is then closed against the bottom or vacuum plate 21 by suitable means, as by hydraulic pressure-operated platens (not shown), to clamp and hermetically seal the conformable diaphragm 27 between the two plates 21 and 28 through the medium of a compression sealing gasket 29 of rubber which extends around the marginal portions of the underside of the pressure plate 28. The pressure plate 28 is provided with a shallow recess or chamber 30 in its underside of sufficient depth to fully accommodate therein the lamp-forming assemblage together with the Teflon release sheets 22 and 23, aluminum foil heater element 24 and conformable diaphragm 27 without contacting the latter when the press is completely closed. For a more complete description of the construction and manner of operation of the hydrostatic press 19, reference may be made to the previously mentioned Fridrich application Serial No. 748,537 and my co-pending application Serial No. 118,113.

With the two plate members 21 and 28 closed tightly together to hermetically seal the conformable diaphragm 27 therebetween, vacuum is applied to the portion of the closed chamber 30 beneath the diaphragm 27 for a short period of one or two minutes or thereabouts, to purge the said lower portion of the chamber, and the various elements therein, of air and any gases trapped therein. The vacuum is applied to the chamber 30 through passageway 31 in the vacuum plate 21 and communicating porous inserts 32 in the upper side of the vacuum plate. Pressurized gas, at a pressure of from 200 to 600 lbs. per square inch, is then introduced into the upper portion of the chamber 30 above the conformable diaphragm 27 through a passageway 33 in the pressure plate 28, the vacuum applied to the lower portion of the chamber 30 beneath the diaphragm 27 being continued. The pressurized gas causes the flexible and therefore conformable diaphragm 27, through the correspondingly flexible aluminum foil heater element 24 and upper Teflon release sheet 22 to apply uniform pressure to the various component layers 7, 8, 9, 10, 17 and 18 of the lamp, and completely around the peripheral edges of the fluid resin impregnated mica paper sheets 17, 18. The resulting high compression of the mica paper sheets 17, 18 not only acts to further compact the mica flakes thereof more closely together to densify the mica paper but also acts to forcefully drive or squeeze the fluid resin impregnant present in the mica paper into and completely throughout the interstitial space within the mica paper between the compacted mica flakes thereof. The pressure exerted by the conformable diaphragm 27 also acts to force the excess resin impregnant in the top mica paper sheet 6 down into the pores of the conductive glass paper layer or electrode 10 of the lamp (where such a fibrous type electrode is employed) so as to firmly hold it down against and in intimate surface contact with the phosphor layer 9. In addition, the pressure exerted by the conformable diaphragm 27 compresses the marginal edge portions of the mica paper sheets 5 and 6, which ex-

tend beyond the margins of the active elements 7, 8, 9 and 10 of the lamp, into firm and intimate contact with one another and tightly around the conductive terminal leads 3 and 4 of the lamp so as to coalesce and become united and sealed together through the action of the impregnating resin, thereby forming a sealed enclosure 2 completely encapsulating the electrically active elements of the lamp 1.

After the concurrent application of the pressurized gas and vacuum to the pressure and vacuum plates 28 and 21 for a short period of around one to two minutes or so, the aluminum foil sheet 24 is then heated, while the pressure and vacuum is continued, by the passage of a low voltage high-density current therethrough from the bus bars 25, 26, to raise the temperature of the lamp components over a period of around one minute or so to an elevated temperature, of around 200° C. to 210° C. for example, corresponding to the curing temperature of the resin component of the fluid impregnant in the mica paper sheets 17 and 18. The lamp components are then maintained at such temperature, while held under the aforementioned pressure by the diaphragm 27, for a sufficient length of time, for example, from 2 to 6 minutes or so, in order to effect the proper curing of the resin in situ and complete the lamination of the lamp assembly 1. Thereafter, the laminated lamp assembly 1 is cooled to a temperature of around 75° C. or so while still held under pressure in the chamber 30 by the diaphragm 27, after which the pressure is then released, thereby completing the manufacture of the lamp 1 in accordance with the novel method of my invention. During the laminating of the lamp components as described above, the overlapping marginal edges of the mica paper sheets 17, 18 coalesce and become sealed together by the resin impregnant therein to form a sealed envelope encapsulating the electrically active components of the lamp. At the same time, the mica paper sheets 17, 18 become adhesively bonded by the cured resin impregnant to the adjacent layers of the assembly completely throughout the interfaces therewith, and they are simultaneously transformed into a tough and flexible transparent sheet material to form the encapsulating sheets 5 and 6 of exceptionally low water-vapor permeability such as to constitute them a highly efficient water-vapor barrier for the lamp.

In producing the electroluminescent lamp 1 comprising my invention by the above-described preferred method of manufacture wherein the micaceous sheet material 11 is pressed and heat cured into its final form simultaneously with the pressure and heat laminating together of the electrically active elements 7, 8, 9 and 10 of the lamp, the use of an anhydride as the resin-curing agent in the resinous impregnant of the mica paper sheets 17 and 18 obviates the need for any preliminary heating and baking out of the electrically active elements 7, 8, 9 and 10 of the lamp, prior to the final pressure and heat laminating operation, such as has been customary heretofore for the purpose of driving out any residual moisture in these lamp elements, although such preliminary heating and baking out of the said lamp elements may still be performed if desired. The elimination of such a preliminary heating and baking out of the lamp elements 7, 8, 9 and 10 is permissible in such case because of the fact that the anhydride itself acts as a getter or desiccant during the lamp laminating operation to pick up and chemically combine with any residual moisture remaining in the electrically active elements 7, 8, 9 and 10 of the lamp at such time. The elimination of such preliminary heating and baking out of the electrically active elements of the lamp thus greatly simplifies and reduces the time required for the manufacture of the electroluminescent lamp. Where a non-desiccant type of resin-curing agent is employed, however, in the resinous impregnant of the mica paper sheets 17, 18, the electrically active elements 7, 8, 9 and 10 in such case are then subjected to the heretofore customary preliminary heating and baking out operation, prior to the final pressure and heat laminating together of the lamp forming ele-

ments. For such purpose, the electrically active elements of the lamp may be stacked together in their final positional relation in the finished lamp, and the assembly then heated to a suitable temperature of, for example, 110° C. or so for a period of around one-half hour in an oven. To facilitate the handling of the stacked assembly of electrically active elements 7, 8, 9 and 10 for such a bake out operation, the assembly of such elements may, as described hereinafter, be enclosed and held together in proper stacked relation within an envelope or holder bag consisting of a suitable transparent flexible thermoplastic film material such as that known as nylon 6 or Caplene, for example.

Because of its comparatively soft character such as renders it prone to becoming surface scratched or marred, with resulting loss of its transparency, the sheets 5 and 6 of micaceous sheet material 11 forming the encapsulating envelope 2 of the lamp may be provided with protective or so-called scuff sheets such as the outer sheets 15, 16 shown in FIG. 6. The scuff sheets are made of a suitable hard-surfaced plastic material such as Mylar or cellulose acetate butyrate laminated and bonded to the outer side of the mica flake sheets 5, 6. In addition to their function as scuff sheets, these plastic outer laminates serve to distribute the forces of stress generated in the laminated assembly when it is flexed. Therefore, they reinforce the laminate and make it more flexible and resistant to cracking.

If desired, an inner sheet or lining of a plastic material having an affinity for water or a disposition to retain water, that is, a hydrophilic sheet, may be laminated into the lamp assembly 1 between the outer resin-impregnated mica flake sheet 6 and the glass fiber conducting sheet 10, as disclosed and claimed in co-pending U.S. application Serial No. 80,613, now Patent No. 3,148,299, Devol, filed January 1, 1961 and assigned to the assignee of the present invention. The preferred material for such a hydrophilic inner sheet is either of the linear polyamides commonly known as nylon 6, 6 and nylon 6, for example, that commercially known as Caplene. Such an inner layer or lining of hydrophilic sheet material may be conveniently provided in the finished lamp assembly 1 by assembling the electrically active elements 7, 8, 9 and 10 between two such hydrophilic sheets and heat-tacking the latter together at spaced points around their marginal edges to form, in effect, a bag which serves as a convenient holder means for facilitating the handling of the assembly of the electrically active elements of the lamp (as during the baking out thereof) prior to the laminations of such elements together with the resin-impregnated mica flake sheets 5, 6 or 17, 18.

Instead of employing an envelope 2 comprised of the bottom and top sheets 5 and 6 of the micaceous sheet material 11 completely encapsulating and sealing the electrically active elements 7, 8, 9 and 10 of the lamp from the atmosphere, only the top sheet 6 of the micaceous sheet material 11 may be provided in the finished lamp, as shown in the modification illustrated in FIG. 5. In such form of the invention, the metal foil electrode 7 is extended beyond the marginal edges of the other electrically active elements 8, 9 and 10 of the lamp, and is suitably sealed to the overlapping marginal edge portions of the top micaceous sheet 6, as by means of the resin impregnant 13 thereof during the pressure and heat-laminating of the lamp.

The marked improvement in brightness maintenance of flexible electroluminescent lamps constructed in accordance with the invention as compared to prior type flexible electro luminescent lamps is clearly illustrated by the curves A and B in FIG. 7 which show the lamp brightness as a function of hours of life in lamps operated in an atmosphere of 80° F. temperature and adverse humidity conditions of 85% relative humidity. The curves A and B are for yellow light emitting lamps of identical construction except for the encapsulating envelope 2

thereof which, in the case of curve A is constituted of 2 mil thick resin-impregnated mica mat in accordance with the invention, and in the case of curve B is constituted of 5 mil thick Kel-F in accordance with the prior art. It will be observed that while the brightness of the lamps according to the invention (curve A) is somewhat lower than that of prior type lamps (curve B) during the early portion of lamp life due to the slight light-absorbing effect of the mica mat encapsulating envelope 2, nevertheless the lamp according to the invention does not exhibit the early rapid depreciation in light output which is characteristic of the prior art lamps but instead maintains its light output at a much more uniform level throughout lamp life and, at an early period around 2600 hours or so of its total life, actually exhibits thereafter a much higher brightness throughout the remaining, and by far the greater portion of its total life. Thus, at around 6000 hours life, the brightness of the lamp according to the invention is over 60% of its early peak brightness as compared to around 20% for that of the prior art lamp.

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

1. A laminated evacuated electroluminescent lamp comprising an electrically active assembly including an electroluminescent phosphor layer and a layer of conductive light-transmitting material over said phosphor layer, said phosphor layer and conductive light-transmitting layer being enclosed within a sealed evacuated envelope comprised of a pair of flexible encapsulating sheets of low water-vapor permeability sealed together around their marginal edges and serving as water-vapor barriers to delay substantially the onset of water depreciation in the lamp, at least that one of said barrier sheets overlying the said conductive light-transmitting layer being transparent and consisting of mica flake paper having its interstitial spaces substantially completely filled with a light-transmitting organic plastic constituting approximately 33 to 55% by volume of the total barrier sheet, the said organic plastic adhesively bonding the mica flake paper to the said other encapsulating sheet and to said electrically active assembly throughout its interface therewith.

2. A laminated evacuated electroluminescent lamp comprising an electrically active assembly including a conducting metal layer with an overlying electroluminescent phosphor layer and a layer of conductive light-transmitting material over said phosphor layer, said electrically active assembly being enclosed within a sealed evacuated envelope composed of top and bottom sheets of transparent and flexible micaceous sheet material of low water vapor permeability sealed together around their overlapping marginal edges to serve as a water vapor barrier for the lamp, said micaceous sheet material consisting of mica flake paper having its interstitial spaces substantially completely filled with a light-transmitting resin impregnant constituting approximately 33 to 55% by volume of the total micaceous sheet material, the said resin impregnant coalescing the said sheets of mica flake paper together at their overlapping marginal edges and adhesively bonding the said sheets to said electrically active assembly throughout their interfaces therewith.

3. An electroluminescent lamp as specified in claim 2 wherein the said resin impregnant comprises a resin from the group consisting of polyester resins, epoxy resins and diepoxides.

4. An electroluminescent lamp as specified in claim 2 wherein the said resin impregnant comprises a resin from the group consisting of epoxy resins and diepoxides.

5. An electroluminescent lamp as specified in claim 2 wherein the said resin impregnant comprises a resin from the group consisting of anhydride-cured epoxy resins and diepoxides.

6. An electroluminescent lamp as specified in claim 2 wherein the said resin impregnant comprises an anhydride-cured Bisphenol-A diglycidyl ether.

7. An electroluminescent lamp as specified in claim 2 wherein the said resin impregnant comprises an aliphatic diepoxide.

8. An electroluminescent lamp as specified in claim 2 wherein the said resin impregnant comprises the dicyclo diepoxy carboxylate identified as 3,4 - epoxy - 6 - methyl-
cyclohexylmethyl - 3,4 - epoxy - 6 - methylcyclohexane-
carboxylate.

9. An electroluminescent lamp as specified in claim 2 wherein the said impregnant comprises the dicyclo diepoxy identified as dicyclopentadiene dioxide.

10. A laminated evacuated electroluminescent lamp comprising an electrically active assembly including a conducting metal layer with an overlying electroluminescent phosphor layer and a layer of conductive light-transmitting material over said phosphor layer, said phosphor layer and conductive light-transmitting layer being enclosed within a sealed evacuated envelope comprised in part of said conducting metal layer and in part of a transparent and flexible sheet of low water vapor permeability overlying the said conductive light-transmitting layer and sealed around its marginal edges to said metal layer to serve as a water vapor barrier to delay substantially the onset of water depreciation in the lamp, said water vapor barrier sheet consisting of mica flake paper having its interstitial spaces substantially completely filled with a light-transmitting organic plastic constituting approximately 33 to 55% by volume of the total barrier sheet, the said organic plastic adhesively bonding the mica flake paper to the said metal sheet and to said electrically active assembly throughout its interface therewith.

11. A laminated evacuated electroluminescent lamp comprising an electrically active assembly including a conducting metal foil with an overlying insulating layer of high dielectric constant material dispersed in a plastic matrix and a layer thereover of an electroluminescent phosphor dispersed in a plastic matrix, and a light-transmitting layer of conductive glass fibers over said phosphor layer, said assembly being enclosed within a sealed evacuated envelope composed of top and bottom sheets of transparent and flexible micaceous sheet material of low water vapor permeability, said micaceous sheet material consisting of mica flake paper having its interstitial spaces substantially completely filled with a light-transmitting resin filler material constituting approximately 33 to 55% by volume of the total sheet material, the said sheets of resin-filled micaceous sheet material over-reaching at their marginal edges the said electrically active assembly and having their said over-reaching marginal edges sealed together by the said resin impregnant thereof, said resin filler material adhesively bonding the said sheets to said electrically active assembly throughout their interfaces therewith and penetrating said glass fiber sheet and holding it down in intimate surface contact with said phosphor layer.

12. A method of making an electroluminescent lamp

comprising the steps of impregnating mica flake paper with a fluid resin-forming material, sandwiching an electrically active assembly comprising an electrically conductive layer having a layer of an electroluminescent phosphor on one side thereof and a light-transmitting layer thereover of electrically conductive material between sheets of the said fluid resin-impregnated mica flake paper with the marginal edges thereof overreaching the said electrically active assembly, and then laminating the said electrically active assembly and the said mica flake paper sheets together under pressure and heat to effect a coalescing and sealing together of the said overreaching marginal edge portions of the said mica flake paper sheets by the said resin impregnant thereof, and also simultaneously cure the said resin impregnant in situ and transform the said mica flake paper sheets into a flexible and transparent sheet material of low water vapor permeability to thereby effectively serve as a water vapor barrier for the lamp.

13. A method of making an electroluminescent lamp comprising the steps of impregnating mica flake paper with a solution of a thermosetting resin of the group consisting of epoxy resins and diepoxides mixed with an organic anhydride, sandwiching an electrically active assembly, comprising an electrically conductive layer having a layer of an electroluminescent phosphor on one side thereof and a light-transmitting layer thereover of electrically conductive material, between sheets of the said resin-impregnated mica flake paper with the marginal edges thereof overreaching the said electrically active assembly, and then laminating the said electrically active assembly and the said mica flake paper sheets together under pressure and heat to effect a coalescing and sealing together of the said overreaching marginal edge portions of the said mica flake paper sheets by the said resin impregnant thereof and also simultaneously cure the said resin impregnant in situ and transform the said mica flake paper sheets into a flexible and transparent sheet material of low water vapor permeability to thereby effectively serve as a water vapor barrier for the lamp.

14. The method of making an electroluminescent lamp as specified in claim 13 wherein the impregnant of the said mica flake paper sheets comprises a solution of a Bisphenol-A diglycidal ether and hexahydro phthalic anhydride.

15. The method of making an electroluminescent lamp as specified in claim 13 wherein the impregnant of the said mica flake paper sheets comprises a solution of an aliphatic diepoxide and hexahydro phthalic anhydride.

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