

CERTIFICATE OF TRANSLATION ACCURACY

I, Benjamin Barrett, declare:

1. I am a native speaker of English and am well versed in both the Japanese and English languages and have over 25 years of experience translating Japanese patents and other technical documents into English.
2. The following translation of the corresponding source text from Japanese into English is accurate and complete to the best of my knowledge.

JP H10-41069 METHOD FOR MANUFACTURING ORGANIC ELECTROLUMINESCENT ELEMENT

I declare under penalty of perjury under the laws of the United States of America that the foregoing is true and accurate.

Statements made herein are to the best of my knowledge true and are based on information that I believe to be true and further these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the patent application in the United States of America or any patent issuing thereon.

Executed this 29th day of March 2024 in the Skagit County of the State of Washington.



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(19) Japan Patent Office (JP)

(12) Patent Application Gazette (A)

(11) Published Application No.

JP H10-41069

(43) Appl. Published: Feb. 13, 1998

(51) Int. Cl. ⁶	Identification code	File number	F1	Technical indication
H05B	33/10		H05B	33/10

Request for examination: Not yet requested; Number of claims: 17 FD (Total of 11 pages)

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(33) Priority Country:	Japan (JP)		

(54) (Title of the Invention) **METHOD FOR MANUFACTURING ORGANIC ELECTROLUMINESCENT ELEMENT**

(57) (ABSTRACT)

(PROBLEM) To provide a method for manufacturing an organic electroluminescent element with favorable pattern processing accuracy without the need to use a wet process.

(MEANS TO SOLVE THE PROBLEM) An organic electroluminescent element in which a light-emitting layer comprising an organic compound is present between a positive electrode and a negative electrode, wherein a method for manufacturing the organic electroluminescent element is such that a pattern processing portion contained in the element is formed through a mask attached to a substrate with magnetic force.

(SCOPE OF PATENT CLAIMS)

(CLAIM 1) An organic electroluminescent element in which a light-emitting layer comprising an organic compound is present between a positive electrode and a negative electrode, wherein a method for manufacturing the organic electroluminescent element is such that a pattern processing portion contained in the element is formed through a mask attached to a substrate with magnetic force.

(CLAIM 2) The method for manufacturing the organic electroluminescent element described in claim 1, wherein the mask on which a magnetic field exerts an attractive force is arranged on a front side of a substrate that is a manufacturing face of the organic electroluminescent element, and a magnetic force generation source is arranged on a rear side of the substrate.

(CLAIM 3) The method for manufacturing the organic electroluminescent element described in claim 1 or 2, wherein the mask contains at least 1% of at least one element among Fe, Co and Ni.

(CLAIM 4) The method for manufacturing the organic electroluminescent element described in claim 1 or 2, wherein the mask contains a total of at least 20% of at least one element among Fe, Co and Ni.

(CLAIM 5) The method for manufacturing the organic electroluminescent element described in claim 1 or 2, wherein the mask thickness is 2 mm or less.

(CLAIM 6) The method for manufacturing the organic electroluminescent element described in claim 1 or 2, wherein the mask thickness is 500 μm or less.

(CLAIM 7) The method for manufacturing the organic electroluminescent element described in claim 1 or 2, wherein a minimum pattern pitch of the mask is 2 mm or less.

(CLAIM 8) The method for manufacturing the organic electroluminescent element described in claim 1 or 2, wherein a minimum pattern pitch of the mask is 500 μm or less.

(CLAIM 9) The method for manufacturing the organic electroluminescent element described in claim 2, wherein a magnetic field generation source is an electromagnet.

(CLAIM 10) The method for manufacturing the organic electroluminescent element described in claim 2, wherein a magnetic field generation source is a permanent magnet.

(CLAIM 11) The method for manufacturing the organic electroluminescent element described in claim 1 or 2, wherein a positive electrode and/or a negative electrode is transparent.

(CLAIM 12) The method for manufacturing the organic electroluminescent element described in claim 1 or 2, wherein a transparent positive electrode is provided on a transparent substrate.

(CLAIM 13) The method for manufacturing the organic electroluminescent element described in claim 1 or 2, wherein a pattern processing portion is a light-emitting layer.

(CLAIM 14) The method for manufacturing the organic electroluminescent element described in claim 13, wherein the light-emitting layer is formed with a deposition method.

(CLAIM 15) The method for manufacturing the organic electroluminescent element described in claim 1 or 2, wherein a pattern processing portion is a positive electrode and/or a negative electrode.

(CLAIM 16) The method for manufacturing the organic electroluminescent element described in claim 15, wherein the positive electrode and/or the negative electrode is formed with a deposition method or a sputtering method.

(CLAIM 17) The method for manufacturing the organic electroluminescent element described in claim 1 or 2, wherein a non-light-emitting portion of the organic electroluminescent element is provided with a spacer.

(DETAILED DESCRIPTION OF THE INVENTION)

(0001)

(TECHNICAL FIELD OF THE INVENTION) The present invention relates to a method for manufacturing an organic electroluminescent element that can convert electrical energy into light for use in fields such as display elements, flat panel displays, backlights, illumination, interior lighting, signs, signage, and electrophotography.

(0002)

(CONVENTIONAL ART) In recent years, there has been active research into organic electroluminescent elements in which electrons injected from a negative electrode and positive holes injected from a positive electrode recombine in the organic fluorescent material sandwiched between the two electrodes to emit light. These elements have attracted much attention because of having a thin profile, high luminance light emission under low drive voltage, and multicolor light emission as a result of fluorescent material selection.

(0003) The high luminance emitted by the organic electroluminescent element was first demonstrated by C.W. Tang *et al.* at Kodak (Appl. Phys. Lett. 51(12), 21, p. 913, 1987). The typical configuration of the organic electroluminescent element presented by Kodak consisted of a positive hole-transporting diamine compound, an 8-hydroxyquinoline aluminum light-emitting layer, and a Mg:Ag negative electrode provided successively on an ITO glass substrate using the vapor deposition method, and green light emission of 1000 cd/m² was possible with a drive voltage of about 10 V. Current organic electroluminescent elements basically follow Kodak's configuration, although the configuration may be modified such as by providing an electron transport layer in addition to the element constituent elements described above. The use of these organic electroluminescent elements, which are capable of high luminance and multicolor light emission, is being actively investigated for use in display devices and other applications. (0004) However, as pointed out in *Nikkei Electronics* on January 29, 1996 (no. 654, p. 102), one major problem is the pattern processing of the element.

(0005) For example, in the case of a full-color display, it is necessary to form R, G, and B light-emitting layers at predetermined locations. Such pattern processing is usually achieved with a wet process typified by the photolithography method, but the organic films that form organic electroluminescent elements are not durable against moisture, organic solvents, and chemicals. As shown by, for example, JP Patent Appl. Publication No. JP 6-234969, wet-processable elements can be obtained by devising organic materials, but this method limits the organic materials used in the elements.

(0006) Furthermore, electrode patterning is essential for display elements. Damage to the organic film is also a problem when pattern processing the electrode above the organic layer with the wet process.

(0007) For these reasons, organic electroluminescent elements have been conventionally manufactured with dry processes such as vapor deposition, and pattern processing has often been achieved using masks. In further detail, a mask is arranged in front of the substrate on which the element is to be manufactured, and the organic layer or the electrode is deposited only at the mask opening.

(0008)

(PROBLEM TO BE SOLVED BY THE INVENTION)

However, the mask must be made thin to accommodate a fine pattern. Since sufficient strength cannot be obtained, the adhesion between the substrate and mask deteriorates due to warping and other factors. Therefore, with conventional methods, the finer the pattern is, the more the position of the deposited material wrapping around the shadow of the mask cannot be ignored, making it difficult to pattern the element with adequate accuracy. This is a major problem that can cause a short in the elements and crosstalk due to increased leakage current. For example, according to Patent Appl. Publication No. JP H2-66873, a pattern pitch of less than 300 μm is a much smaller pixel size than the range where pattern processing using a mask is possible.

(0009) It is an object of the present invention to solve this problem and to provide a manufacturing method that can achieve favorable pattern processing accuracy in an organic electroluminescent element for which wet processes cannot be used. In other words, the object is not to simply affix the mask to the substrate, but to provide a method to achieve adequate adhesion between the mask and the substrate, which is necessary to achieve fine pattern processing with the mask, which has heretofore been impossible.

(0010)

(MEANS TO SOLVE THE PROBLEM) In order to solve the problem, the manufacturing method of the present invention is such that in "an organic electroluminescent element in which a light-emitting layer comprising an organic compound is present between a positive electrode and a negative electrode, ... a pattern processing portion contained in the element is formed through a mask attached to a substrate with magnetic force."

(0011)

(EMBODIMENTS TO PRACTICE THE INVENTION) The organic electroluminescent element in the present invention is an element in which a light-emitting layer comprising an organic compound is present between a positive electrode and a negative electrode and emits light using electrical energy, and at least one of the layers composing the element, such as the positive electrode, the negative electrode, or the light-emitting layer, is patterned.

(0012) The positive electrode and the negative electrode should be conductive enough to supply adequate current for the element to emit light, but at least one of the electrodes is desirably transparent to allow light to be extracted.

(0013) If there is a visible light transmittance of at least 30%, there is no major obstacle in using transparent electrodes, but ideally a visible light transmittance close to 100% is favorable. Basically, the transmittance is favorably almost the same throughout the visible light range, but if the color is to be changed, it is possible to actively add absorption. In this case, it is technically easier to use a color filter or an interference filter to change the color.

Transparent electrodes often comprise at least one element selected from indium, tin, gold, silver, zinc, aluminum, chromium, nickel, oxygen, nitrogen, hydrogen, argon, and

carbon, but inorganic conductive materials such as copper iodide and copper sulfide and conductive polymers such as polythiophene, polypyrrole, and polyaniline can also be used as the material for the transparent electrode without any particular limitation.

(0014) Examples of particularly favorable positive electrodes in the present invention include tin oxide, zinc oxide, indium oxide, and indium tin oxide (ITO) on a transparent substrate. ITO, which has excellent processing properties, is particularly suitable for display applications and the like in which pattern processing and the like is carried out. ITO may contain small amounts of metal such as silver or gold to reduce surface resistance and curb voltage drop, and tin, gold, silver, zinc, indium, aluminum, chromium, and nickel can be used as the ITO guide electrode. Among these, chromium can function as both a black matrix and a guide electrode and therefore is a particularly suitable metal. From the viewpoint of element power consumption, it is desirable for the ITO to have low resistance. For example, an ITO substrate of 300 Ω/sq or less can function as an element electrode, but since it is now possible to supply substrates of about 10 Ω/sq , it is especially desirable to use low-resistance ITO. The thickness of the ITO can be selected arbitrarily according to the resistance value but is usually between 100 and 300 nm. The material of the transparent substrate is not particularly limited and can be made of a plastic sheet or a film comprising polyacrylate, polycarbonate, polyester, polyimide, or aramid, but a glass substrate is also a suitable example. Soda lime glass, alkali-free glass, and the like can be used, and a thickness of 0.7 mm or more is adequate as long as the thickness is adequate to maintain the mechanical strength. Alkali-free glass is preferred as the glass material due to the low level of ions eluted from the glass, but commercially available soda lime glass with a barrier coating of SiO_2 or the like can also be used. The ITO film formation method is not particularly limited and the electron beam technique, the sputtering technique, the chemical reaction technique and the like may be used.

(0015) The negative electrode is not particularly limited as long as the negative electrode is a substance that can efficiently inject electrons into the light-emitting layer of the present element. Therefore, low work-function metals such as alkali metals can be used, but considering electrode stability, metals such as platinum, gold, silver, copper, iron, tin, aluminum, magnesium, indium, alloys of such metals and low work-function metals, and the like are preferred examples. In addition, by doping a small amount of a low work-function metal into the organic layer in advance and then depositing a relatively stable metal as a negative electrode, a stable electrode can be obtained while maintaining high electron injection efficiency. The electrode can be fabricated by any dry process, such as resistance heating evaporation, electron beam evaporation, sputtering, and ion plating, but the present invention uses resistance heating evaporation, which is easy to use.

(0016) The organic layers contained in the organic electroluminescent element can be any of: 1) positive hole transport layer/light-emitting layer, 2) positive hole transport layer/light-emitting layer/electron transport layer, 3) light-emitting layer/electron transport layer, and 4) light-emitting layer that combines the above materials in one layer. In other words, if a light-emitting layer comprising an organic compound is present as the element configuration, it is possible to provide the multilayer laminate structure in any of 1) to 3) or, as in 4), a light-emitting layer containing a light-emitting material alone or a light-emitting material and a positive hole transport material and/or an electron transport material.

(0017) The positive hole transport layer is formed with a positive hole transport material alone or with a positive hole transport material and a polymer binding agent, and as the positive hole transport substance, triphenylamines such as typified by N,N'-diphenyl-N,N'-di(3-methylphenyl)-1,1'-diphenyl-4,4'-diamine (TPD) and N,N'-diphenyl-N,N'-dinaphthyl-1,1'-diphenyl-4,4'-diamine (NPD), tertiary amines such as N-isopropylcarbazole, heterocyclic compounds such as pyrazoline derivatives, stilbene compounds, hydrazone compounds, oxadiazole derivatives, and phthalocyanine derivatives, and, in polymers, polycarbonates and styrene derivatives having the aforementioned monomers in a side chain, polyvinylcarbazole, polysilane, and the like are preferred but the positive hole transport substance is not particularly limited to these substances.

(0018) In addition to anthracene and pyrene, which have been primarily known as light-emitting layer materials for some time, and the 8-hydroxyquinoline aluminum mentioned above, bis-styrylanthracene derivatives, tetraphenylbutadiene derivatives, coumarin derivatives, oxadiazole derivatives, distyrylbenzene derivatives, pyrolopyridine derivatives, perinone derivatives, cyclopentadiene derivatives, and thiadiazolopyridine derivatives, for example, can be used as the light-emitting material, as can polymers such as polyphenylenevinylene derivatives, polyparaphenylene derivatives, and polythiophene derivatives, and the like. As dopants added to the light-emitting layer, the aforementioned rubrene, quinacridone derivatives, Phenoxazone 660, DCM1, perinone, perylene, Coumarin 540, diazaindacene derivatives, and the like can be used.

(0019) Electron transport materials need to efficiently transport electrons from the negative electrode between electrodes subjected to an electric field, and it is desirable to have high electron injection efficiency and efficient transport of injected electrons. To achieve this, the material must have a high electron affinity, high electron mobility, excellent stability, and resistance to the generation of impurities that are trapped during manufacture and use. Substances that satisfy these conditions include 8-hydroxyquinoline aluminum, hydroxybenzoquinoline beryllium, oxadiazole derivatives such as 2-(4-biphenyl)-5-(4-t-butylphenyl)-1,3,4-oxadiazole (t-BuPBD), and, as oxadiazole dimer derivatives that improve thin film stability, 1,3-bis(4-t-butylphenyl)-1,3,4-oxadiazolyl)biphenylene (OXD-1) [and] 1,3-bis(4-t-

butylphenyl-1,3,4-oxadiazolyl)phenylene (OXD-7), triazole derivatives, phenanthroline derivatives, and the like.

(0020) The materials used for the positive hole transport layer, the light-emitting layer, and the electron transport layer can form each layer independently but can also be used dispersed in a solvent soluble resin such as polyvinyl chloride, polycarbonate, polystyrene, poly(N-vinylcarbazole), polymethyl methacrylate, polybutyl methacrylate, polyester, polysulfone, polyphenylene oxide, polybutadiene, hydrocarbon resin, ketone resin, phenoxy resin, polysulfone, polyamide, ethylcellulose, vinyl acetate, ABS resin, or polyurethane resin or in a curable resin such as phenol resin, xylene resin, petroleum resin, urea resin, melamine resin, unsaturated polyester resin, alkyd resin, epoxy resin, or silicone resin.

(0021) The formation methods of the organic layers such as the positive hole transport layer, the light-emitting layer, and the electron transport layer are not particularly limited to resistance heating evaporation, electron beam evaporation, and sputtering, but resistance heating evaporation and electron beam evaporation are usually preferred as deposition methods in terms of characteristics. The thickness of the layer depends on the resistance of the organic layer and therefore cannot be limited but is empirically selected between 10 and 1000 nm.

(0022) Electrical energy primarily refers to DC current, but pulse and alternating currents can also be used. There are no particular limitations on current and voltage values, but considering the power consumption and lifetime of the element, the maximum luminance should be obtained at the lowest possible energy.

(0023) Patterned portions in the present invention basically signify the portions involved in the light emission of the organic electroluminescent element, but may also be non-light-emitting portions formed for the purpose of improving the contrast, pattern accuracy, and electrical insulation of the portions involved in light emission according to need, and therefore are not particularly limited. The light-emitting layer, the positive electrode, and the negative electrode are preferably patterned.

(0024) The light-emitting layer and the electrode are particularly important as portions involved in light emission. The shape and size are not particularly limited and should be selected as appropriate for use. For example, in display elements and display applications, one or more light-emitting regions defined by the light-emitting layer, the electrode, and the pattern shape of the non-emitting region form one pixel. In other words, the pattern shape and size of the processed light-emitting layer and the electrode according to the present invention do not need to match the shape and size of the pixel. Although the shape of the pixel varies depending on the display method, such as segmental or dot matrix, a pixel size of 2 mm square or smaller is preferable. For high-definition display applications, a pixel size of 500 μm square or less is desirable, and an example of a suitable pixel size is 100 $\times$ 300 μm , which is the size of a single-color pixel in full-color liquid crystal displays currently used in practical applications.

(0025) In display applications, it is not uncommon for the pattern processing shape of the light-emitting layer and the electrode to have a repetitive cycle. In such cases, the minimum pitch of the pattern processing size is favorably 2 mm or less, and 500 μm or less is even more favorable.

These displays may be simple matrix or active matrix, such as TFT, and the present invention does not limit the structure or drive system of the display.

(0026) The electrodes described above refer to the positive electrode and/or the negative electrode in an organic electroluminescent element, and both of the electrodes may be patterned according to the present invention according to need. The electrode also includes elements such as a guide electrode added to improve electrode conductivity.

Basically, one of the electrodes on the substrate is patterned by a wet process first, and after an organic layer is formed on the electrode, the other electrode is favorably patterned with the method of the present invention. A more suitable example use the previously patterned transparent electrode as the positive electrode on a transparent substrate, form an organic layer thereon, and then pattern the negative electrode at the upper part of the organic layer according to the present invention.

(0027) The non-light-emitting portions to be patterned according to the present invention are not particularly limited, but suitable examples include a black matrix and an insulating layer that defines the light-emitting region. The black matrix blackens the gaps in the light-emitting region to improve contrast. The insulating layer protects the electrode edges, defines the light-emitting region, and improves the insulation between electrodes. There is no single material, shape, or size for these non-light-emitting regions, and the most appropriate specifications are used according to application.

(0028) The mask in the present invention is a plate or film-like surface with one or more openings. Organic material or electrode material flying toward the substrate in vapor deposition or the like is deposited on the substrate through the mask opening placed in front of the substrate to achieve the patterning of the organic electroluminescent element corresponding to the mask opening shape.

(0029) An important feature of the present invention is that the adhesion between the mask and the substrate is improved by magnetic force. This effect reduces the proportion of the area where the deposited material or other material wraps around, i.e., the portion deposited from the mask opening that bleeds to the shadow portion of the mask, achieving fine pattern processing not conventionally possible. Since it is not a direct object of the present invention to affix the position of the mask to the substrate or to support the weight of the mask itself, the methods of doing so are not particularly limited and may be done by magnetic means, mechanical contact, or the like.

(0030) In the present invention, the relationship between the mask and at least one other element is not particularly limited as long as a mutual magnetic force is exerted between the mask and the other element(s). For example, if the mask and the substrate of an organic electroluminescent

element exert an attractive force on each other by means of magnetic force, the substrate may be a magnet, the mask itself may be a magnet, or both may be magnets. Of course, it is not necessary for it to be the substrate that mutually exerts a magnetic force with the mask, and there are no particular limitations on whether the magnetic force is an attraction or a repulsion. A preferred example in the present invention is a method in which a mask on which a magnetic field exerts an attractive force, i.e., a mask made of a material attracted to a magnet, is placed on the front side of the substrate, which is the production surface of the organic electroluminescent element, and the mask is attached to the substrate by means of a magnet arranged at the back side of the substrate. The magnitude of the magnetic force required depends on the shape, size, weight, strength, and other factors of the mask opening, so a general indication cannot be given, but when the mask is affixed with mechanical contact, the magnetic force should be adequate to obtain the mask adhesiveness required, and if the mask is affixed with magnetic force, the magnetic force should be adequate for mask adhesion and positional affixing.

(0031) Suitable examples of the magnet include a permanent magnet and an electromagnet. The shape and size are not limited, but the magnet is desirably at least as large as the mask opening so that the attractive force perpendicular to the substrate surface works uniformly across the entire mask opening. Also, while a single magnet may be used to generate a magnetic field over the required area, a plurality of magnets may also be used by pasting the magnets together or arranging them at predetermined intervals. The distance from the substrate is not particularly limited as long as an adequate magnetic force works on the mask.

(0032) Suitable materials for the permanent magnet include, but are not limited to, quench-hardened magnetic materials such as pure iron, carbon steel, W steel, Cr steel, Co steel, and KS steel, precipitation hardened magnetic materials such as MK steel, Alnico steel, NKS steel, and Cunico steel, sintered magnetic materials such as OP ferrite and Ba ferrite, and various rare earth magnetic materials typified by Sm-Co and Nd-Fe-B. Although these materials are often used in bulk, a powder may be mixed with rubber or resin, or a powder may be pressurized and molded into a magnet.

(0033) An electromagnet can electrically control the switching of magnetic field generation as well as the magnetic force and therefore is particularly suitable in the present invention. The structure is not particularly limited, but a magnetic core inserted into a solenoid coil wound a plurality of times around a conductor is particularly desirable. In addition to the permanent magnet materials listed above, magnetic core materials include metal core materials such as silicon steel sheets, Al-Fe alloys, Ni-Fe alloys (permalloys), ferrite core materials such as Mn-Zn, Ni-Zn, and Cu-Zn, and pressed core materials made by compressing a fine powder of carbonyl iron, Mo permalloy, sendust, or the like with a binder. Such an electromagnet can be used alone or a plurality of electromagnets can be combined to generate a magnetic field.

(0034) These magnetic materials are also suitable materials for the mask. It is favorable to fabricate the mask from these materials molded into thin sheets, but it is also possible to use rubber or resin mixed with magnetic powder and molded into a film shape, so the material composition of masks cannot be generally specified. In order to efficiently generate magnetic attraction, the mask desirably contains a total of at least 1% of at least one of the magnetic elements Fe, Co, and Ni (at least 1% if an element is used alone) and more desirably a total of at least 20%. The mask itself may or may not be magnetized from the start, although the magnetic field will exert an attractive force on the fabricated mask. If necessary, the mask can be fabricated from magnetized material or magnetized after fabrication.

(0035) The shape and size of the mask opening are basically equivalent to the shape and size of the pattern processing of the organic electroluminescent element. However, the pattern processing of the organic electroluminescent element may be achieved using a single mask, or a plurality of masks may be combined and an approach taken such as shifting the relative position of each mask to the substrate over a plurality of times, so the pattern of the organic electroluminescent element and the pattern of the mask do not need to match. When a pattern with a repetitive cycle is achieved in an element with a single mask, as in the display application described above, a pattern pitch of 2 mm is a suitable example for the mask, and 500 μm is even more favorable.

(0036) The mask thickness depends on the strength, size, and pattern size, and therefore cannot be generally indicated. If the mask is thicker and stronger than needed, a large magnetic force is required to improve adhesion to the substrate, and it becomes difficult for the mask to follow the warping of the substrate. As a general rule, a thickness of 2 mm or less is suitable. The present invention is particularly effective for fine patterning, and 500 μm is an example of a more favorable pattern pitch. In this case, for example, if the opening is 300 μm , the remaining width of the mask is 200 μm . To achieve favorable pattern accuracy, the thickness of the mask is desirably no greater than 5 times the remaining width (200 μm in this case), and it is particularly desirable that the thickness be no greater than the remaining width. Therefore, a more suitable mask thickness for fine pattern processing is 500 μm or less.

(0037) When the surface of the substrate used in the organic electroluminescent element is flat, it is advantageous to use a mask with high planarity in order to achieve uniform adhesion between the mask and the substrate. However, if the mask is made thinner as described above to accommodate fine patterns, the mask is more susceptible to deformation during the mask fabrication process, resulting in undulations in the mask and loss of planarity. In such a case, methods such as annealing may be used to improve the planarity of the mask. Furthermore, masks are often used in a state where affixed to an appropriately shaped frame, but even in such cases, the mask planarity should be brought closer to the ideal state by means such as affixing the mask to the frame while applying tension.

(0038) In the manufacturing method of the present invention, it is favorable to arrange a spacer that acts as a cushion on the non-light-emitting portion of the organic electroluminescent element to prevent the mask from damaging the organic layer and the like deposited on the substrate when the manufacturing surface of the organic electroluminescent element and the mask are adhered to each other. This spacer can also serve as all or a part of the black matrix. Although there is no particular limitation on the fabrication method or material of the spacer, it is easy to use the photolithography method or the like and arrange the spacer on the substrate in advance so that the light-emitting area can be effectively protected from contact with the mask. The height of the spacer must exceed the thickness of the light-emitting portion of the organic electroluminescent element to the extent that the pattern processing accuracy does not deteriorate due to wraparound of the vapor-deposited material, and formation is normally in the range of 0.5 to 100 μm , but there are no particular limitations thereto, and the size should be optimized according to conditions. (0039) The function of the spacer can also be achieved by placing protrusions on portions of the substrate contact surface of the mask and bringing the protrusions into contact with the non-light-emitting regions of the organic electroluminescent element or by coating the entire surface of the mask with a relatively flexible material such as a resin. The materials for the protrusions and coating are not particularly limited, but it is easy to form them using a photoresist or the like.

(0040)

(EMBODIMENTS)

The present invention is described below with embodiments and comparative examples but is not limited thereby.

(0041) Embodiment 1

A 28 x 36 mm, 50- μm thick Kovar (composition ratio of Fe: 54%, Ni: 29%, Co: 17%) plate was prepared by wet etching a mask with openings. The main portion of one opening was rectangular with lengths of 200 μm and 15 mm in the short and long direction, respectively. These openings were repeated 16 times with a pitch of 300 μm in the short side direction. In other words, the center portion was a mask with 16 stripes at a pitch of 300 μm (opening width of 200 μm width and 100 μm width for the rest). In order to facilitate an external electrical connection, both sides of the rectangular opening in the longitudinal direction were continuously widened to a pitch of 1.27mm (opening width of 600 μm) for another 7.5 mm. The mask was affixed by laser fusion to a stainless steel frame of 2 mm width with the same outer shape. The mask was affixed inside the vacuum evaporation apparatus so that the stainless steel frame side faced the evaporation source (lower side), and on the opposite Kovar plate side (upper side), a glass substrate 38 x 46 mm and 1.1 mm thick and a plate magnet 34 x 42 mm and 3 mm thick were placed in this order and affixed. The plate magnet was a rare earth magnet (H-23CV) manufactured by Hitachi Metals, Ltd. After evacuating until the vacuum in the apparatus was no greater than 5×10^{-4} Pa, 150 nm of metal Al was deposited at a rate of 1 nm/s with the resistance heating deposition method using an alumina crucible. The substrate was rotated during the deposition to make the film thickness distribution even.

(0042) After deposition, the glass substrate was removed, and FIG. 1 is a photograph in which the Al electrode deposited on the substrate surface was observed with a microscope. The width of the electrode at the center in the longitudinal direction was 200 μm . The resistance was measured with a tester using the portion where the electrode pitch widened, and it was found that there were no electrical short circuits between the 16 electrodes, and the resistance between adjacent electrodes was at least 20 M Ω , which was the limit of measurement.

(0043) Comparative Example 1

Al electrodes were patterned in the same manner as in Embodiment 1, except that no magnet was used during deposition, and as shown in FIG. 2, the average electrode width was 270 μm . However, large wrapping of deposited Al was observed at a number of locations, and 10 pairs of adjacent electrodes out of the 16 electrodes were electrically short-circuited.

(0044) Embodiment 2

The Al electrode was patterned in the same manner as in Embodiment 1, except that the opening width was 250 μm and the remaining width was 50 μm (the pitch was the same at 300 μm). FIG. 3 is a photograph of the deposited Al electrode observed with a microscope. The electrode width was 255 μm . There were no electrical shorts between the electrodes, and the resistance between adjacent electrodes was at least 20 M Ω , which was the limit of measurement.

(0045) Comparative Example 2

The Al electrodes were patterned in the same way as in Embodiment 2, except that no magnet was used during deposition, and as shown in FIG. 4, all of the adjacent electrodes overlapped each other, and the electrode width could not be measured. In the resistance measurement, all of the adjacent electrodes were electrically short-circuited.

(0046) Embodiment 3

A 1.1-mm-thick glass substrate with an ITO transparent electrode film (electron beam evaporation product, 15 Ω/sq) was cut into a size 38 $\times$ 46 mm, and the ITO was patterned at a 300- μm pitch (ITO remaining width of 270 μm) $\times$ 32 stripes using the photolithography method. One side of the ITO stripe in the longitudinal direction was widened to a 1.27-mm pitch (800- μm opening width) to facilitate an external electrical connection. The substrate was cleaned and treated with UV-ozone. The substrate was affixed inside a vacuum evaporation apparatus and evacuated until the vacuum inside the apparatus was no greater than 2×10^{-4} Pa. Bis(m-methylphenylcarbazole), a positive hole transport material, was deposited from a tantalum boat at a rate of 0.3 nm/sec to 120 nm using the resistance heating method, and 30 nm of 8-hydroxyquinoline aluminum (Alq3), doped with 0.35 wt% quinacridone, was deposited at a rate of 0.3 nm/sec, and 70 nm of Alq3 was sequentially deposited at a rate of 0.3 nm/sec. The above organic materials were deposited

on the entire surface of the substrate. Next, a magnet was used as in Embodiment 1 and the mask set so that the ITO and electrode stripe patterns were orthogonal to each other. In the vacuum, the organic layer that had already been deposited was exposed to lithium vapor and doped (film thickness equivalent of 1 nm), and then Al was deposited at a rate of 0.5 nm/sec to a thickness of 150 nm. (0047) When the resulting 32 $\times$ 16-pixel simple matrix organic electroluminescent element was caused to emit light with line sequential driving, it was possible to clearly display characters. FIG. 5 is a photograph showing the light-emitting region observed with a microscope. Each pixel is 270 $\mu\text{m} \times$ 255 μm and the length of each side corresponds to the actual patterned ITO and Al electrode widths.

(0048) Embodiment 4

A mask was prepared with openings in a Ni plate that was 120 $\times$ 84 mm in outer dimensions and 25 μm in thickness as a mask for the light-emitting layer. The basic shape of the opening was a stripe 67.2 mm long and 100 μm wide, with a pitch of 300 μm and 272 stripes in the width direction. As shown schematically in FIG. 6, each stripe-shaped opening 2a has a crossing line 3 with a width of 20 μm every 4.8 mm to prevent deformation of the opening. As a negative electrode mask, a mask was prepared with openings in a Kovar plate 120 $\times$ 84 mm in outer dimensions and 100 μm in thickness by wet etching. As shown schematically in FIG. 7, each stripe-shaped opening 2b is 100 mm long and 270 μm wide, with a pitch of 600 μm and 100 stripes in the width direction. The two masks were affixed to a stainless steel frame 2 mm wide with the same outer shape by electron beam welding, taking care not to damage the planarity of the masks.

(0049)

A glass substrate 10 with the same ITO transparent electrode film as in Embodiment 3 was cut into a size of 120 $\times$ 100 mm, and the ITO was patterned into 816 stripes 20 of 100 μm pitch (ITO remaining width of 90 μm) using the conventional photolithography method as shown in FIG. 8. Next, a paste of non-sensitive polyimide mixed with a black pigment whose primary component was phthalocyanine oxide condensate was applied to the substrate using the spin-coating method, and semi-curing was carried out at 140°C for 10 minutes in a nitrogen atmosphere in a clean oven. After the polyimide layer was patterned with a conventional photolithography method using a positive photoresist, the result was cured at 280°C for 30 minutes to form a black layer 1.0 μm thick that served as both a spacer and a black matrix. As shown in FIG. 9, a black layer 30 had an opening 31 that was 70 $\times$ 250 μm and exposed the center of the underlying ITO stripe 20. The volume resistivity of the black layer was at least $10^8 \Omega\text{cm}$ and had favorable insulation properties.

(0050) After cleaning and UV-ozone treatment, the substrate was affixed to a vacuum evaporation apparatus, which was evacuated until the internal vacuum was no more than 2×10^{-4} Pa. First, 20 nm of copper phthalocyanine and 100 nm of bis(m-methylphenylcarbazole), as positive hole transport materials, were deposited on the entire surface of the substrate to form a positive hole transport layer 32. Next, the mask for the light-emitting layers was arranged in front of the substrate, and a ferrite plate magnet 110 x 100 mm, 10 mm thick, and having a residual magnetic flux density of 0.3 T was placed behind the substrate, and all the elements were fastened. At this time, the positioning was arranged such that the center lines of the stripe-shaped opening 2a of the mask and the ITO stripes 20 of the substrate were aligned and the crossing line 3 did not interfere with the opening 31 of the black layer. In the vacuum, 30 nm of bis(2-methyl-8-hydroxyquinolinolato)-paraphenylphenolato aluminum (BALq_3) doped with 0.3 wt% perylene and then 70 nm of BALq_3 were deposited as a blue emitting layer 40. The mask for the light-emitting layers was shifted by one third of a pitch relative to the substrate, then the substrate and magnet were fastened, and 30 nm of Alq_3 doped with 0.3 wt% quinacridone and then 90 nm of Alq_3 were sequentially deposited on the adjacent ITO stripe as a green light-emitting layer 41. Similarly, 30 nm of Alq_3 doped with 0.3 wt% 4-(dicyanomethylene)-2-methyl-6-(paradimethylaminostyryl)-4-pyran (DCM) and then 80 nm of Alq_3 were deposited sequentially on the adjacent ITO stripe as a red light-emitting layer 42. Each light-emitting layer was arranged at every three ITO stripes 20 as shown in FIG. 10, completely covering the ITO-exposed area. Subsequently, the mask for the light-emitting layers was removed and 20 nm of Alq_3 was deposited on the entire surface of the substrate as a light-emitting layer covering layer 43. Next, the mask for the negative electrode was arranged in front of the substrate, and the same magnet as described above was placed behind the substrate, and these elements were fastened. At this time, the stripe-shaped openings 2b of the mask were positioned so as to be orthogonal to the ITO stripe 20 on the substrate and also to be aligned with the center of a black layer opening 31. In a vacuum, the organic layer that had already been deposited was exposed to lithium vapor and doped (film thickness equivalent of 1 nm), and then Al was vapor-deposited to a thickness of 200 nm to serve as a negative electrode. At this stage, there was a negative electrode 50a with a pitch of 600 μm (electrode width of 275 μm) as shown in FIG. 11. Furthermore, the negative electrode deposition mask was shifted by one half of a pitch relative to the substrate, then the substrate and the magnet were fastened, and Li doping and Al negative electrode deposition were carried out in a similar manner to form a negative electrode 50b with the same electrode width in the gap with the negative electrode that was deposited first. As shown schematically in FIG. 12 and FIG. 13, a simple matrix color display was obtained at the end in which red, green, and blue (RGB) light-emitting layers alternated on 816 ITO stripes and 200 negative electrodes with a pitch of 300 μm were arranged orthogonal

to the ITO stripes. Three light-emitting regions comprising RGB form one pixel, so the display had 272×200 pixels with a pitch of 300 μm . In FIG. 13, the thickness of the black layer 30 is depicted as thinner than the light-emitting portion for ease of explanation, but in reality, the black layer 30 is thicker than the light-emitting portion and fully functioned as a spacer to protect the light-emitting portion from contact with the mask.

(0051)

When the fabricated display was caused to emit light, each light-emitting region emitted light in independent RGB colors, and no bleeding or the like of emission colors caused by evaporation material going into other light-emitting regions during light-emitting layer evaporation was found. Similarly, there were no shorts between the stripe-like negative electrodes. The size of the light-emitting region was $70 \times 250 \mu\text{m}$, which was consistent with the opening size of the black layer. Line-sequential driving enabled clear pattern display and multi-coloring.

(0052)

(EFFECT OF THE INVENTION) According to the present invention, an organic electroluminescent element with favorable pattern processing accuracy can be produced without using a wet process.

(BRIEF DESCRIPTION OF THE DRAWINGS)

(FIG. 1) FIG. 1 is a photomicrograph of the pattern of the Al electrode patterned according to Embodiment 1.

(FIG. 2) FIG. 2 is a photomicrograph of the pattern of the Al electrode patterned according to Comparative Example 1.

(FIG. 3) FIG. 3 is a photomicrograph of the pattern of the Al electrode patterned according to Embodiment 2.

(FIG. 4) FIG. 4 is a photomicrograph of the pattern of the Al electrode patterned according to Comparative Example 2.

(FIG. 5) FIG. 5 is a photomicrograph of the pattern of the pixel portion of the organic electroluminescent element patterned according to Embodiment 3.

(FIG. 6) FIG. 6 is a plan view describing an overview of the light-emitting layer mask used in Embodiment 4.

(FIG. 7) FIG. 7 is a plan view describing an overview of the negative electrode mask used in Embodiment 4.

(FIG. 8) FIG. 8 is a plan view describing an overview of the organic electroluminescent element of Embodiment 4 (ITO pattern processing step).

(FIG. 9) FIG. 9 is a plan view describing an overview of the organic electroluminescent element of Embodiment 4 (black layer formation step).

(FIG. 10) FIG. 10 is a plan view describing an overview of the organic electroluminescent element of Embodiment 4 (RGB light-emitting layer formation step).

(FIG. 11) FIG. 11 is a plan view describing an overview of the organic electroluminescent element of Embodiment 4 (first negative electrode formation step).

(FIG. 12) FIG. 12 is a plan view describing an overview of the organic electroluminescent element fabricated in Embodiment 4.

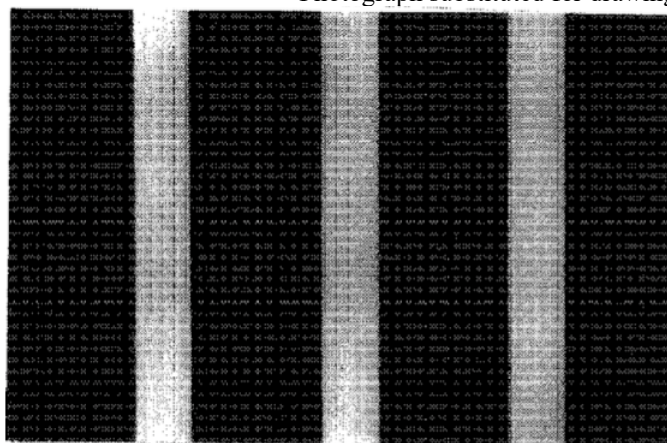
(FIG. 13) FIG. 13 is a cross-sectional view of AA' in FIG. 12.
(DESCRIPTION OF THE REFERENTIAL NUMBERS)

- 1a, 1b mask portions
- 2a, 2b stripe-like openings
- 3 cross line
- 10 glass substrate
- 20 ITO stripe

- 30 black layer
- 31 black layer opening
- 32 positive hole transport layer
- 40 blue light-emitting layer
- 41 green light-emitting layer
- 42 red light-emitting layer
- 43 light-emitting layer cover layer
- 50a, 50b negative electrodes

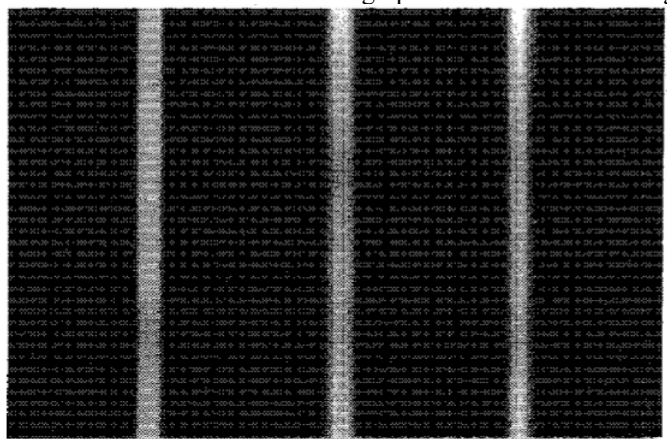
(FIG. 1)

Photograph substituted for drawing

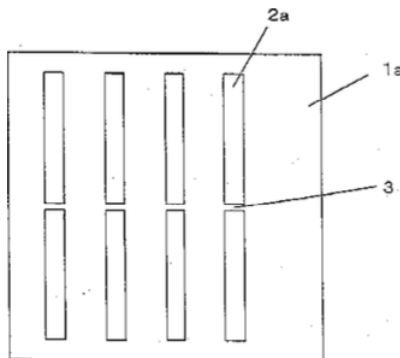


(FIG. 2)

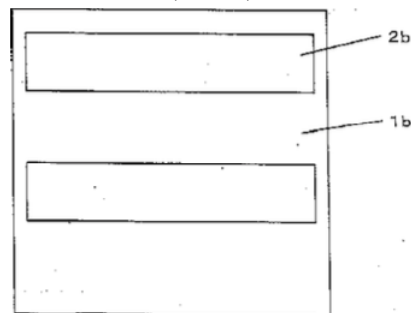
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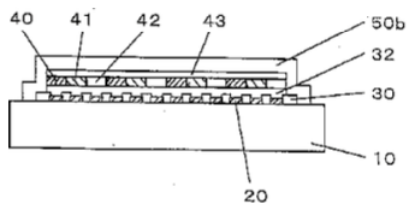
(FIG. 6)



(FIG. 7)

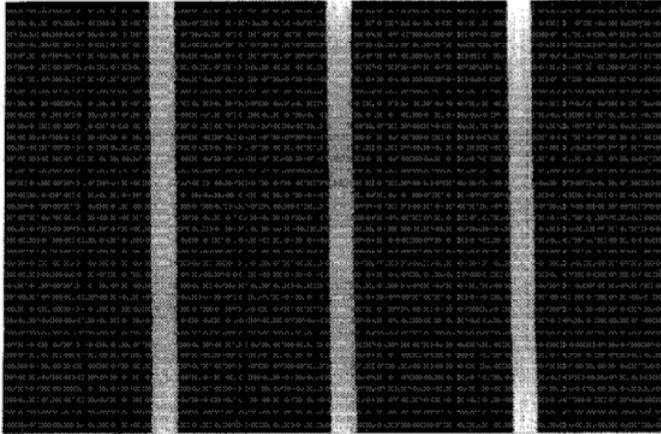


(FIG. 13)



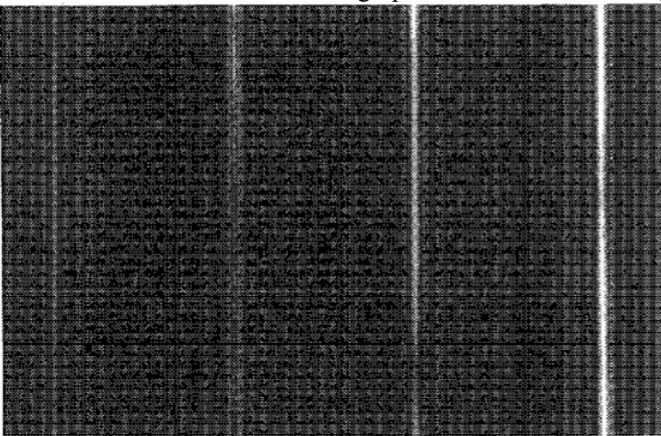
(FIG. 3)

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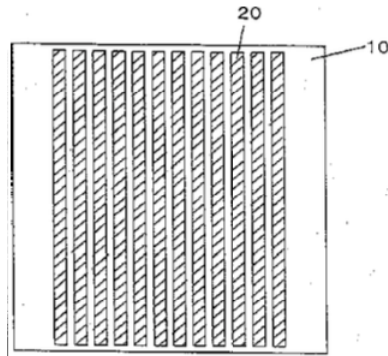


(FIG. 4)

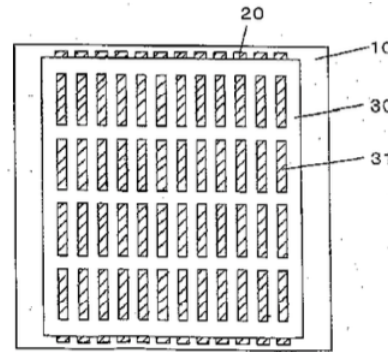
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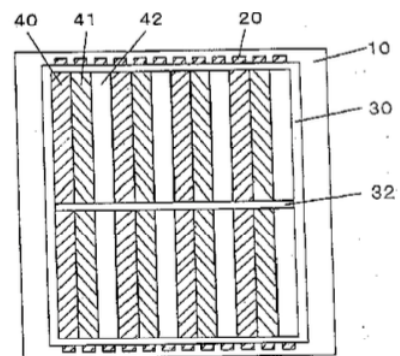
(FIG. 8)



(FIG. 9)

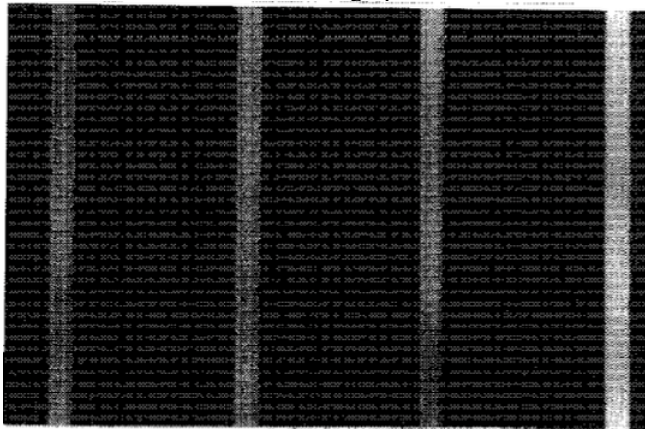


(FIG. 10)



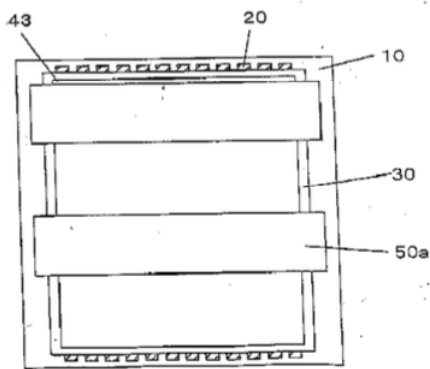
(FIG. 5)

Photograph substituted for drawing



300 μ m

(FIG. 11)



(FIG. 12)

