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(54) **METHOD FOR PREPARING UNEVEN PARTICLE LAYER, ORGANIC ELECTROLUMINESCENT DEVICE, AND DISPLAY DEVICE**

(57) The present invention provides a method for preparing an uneven particle layer, an organic light emitting diode device and a display device. The method for preparing an uneven particle layer includes the following steps: forming a nanoparticle layer on a substrate; heating the substrate to fuse nanoparticles that are in contact with the substrate, whereas the nanoparticles on the surface keep a solid state; and cooling the substrate to form a nanoparticle layer with an uneven surface. The method

of the present invention is simple in process, and industrial production is easy to achieve. The substrate including the uneven particle layer is applied to the OLED device, so the propagation direction of rays from the organic light emitting layer of the OLED device can be changed so as to avoid total reflection on an interface, emit more light into the air and improve the light extraction efficiency of the OLED device. The OLED device prepared in the present invention is suitable for various display devices.

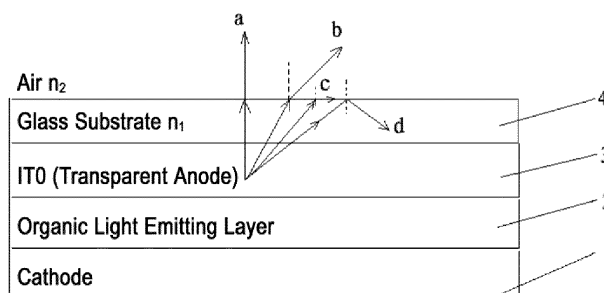


Fig. 1

Description

FIELD OF THE INVENTION

[0001] The present invention relates to the field of display technology, and particularly relates to a method for preparing an uneven particle layer, an organic light emitting diode device and a display device comprising the uneven particle layer.

BACKGROUND OF THE INVENTION

[0002] An OLED (Organic Light Emitting Diode) device simultaneously has the excellent characteristics of being self-luminous, free of backlight, high in contrast, small in thickness, wide in viewing angle, high in response speed, available for flexible panels, wide in using temperature range, simple in constructing and manufacturing procedures and the like, thereby being deemed as an emerging technology of the next-generation flat display devices.

[0003] The existing OLED has a structure as shown in schematic diagrams Fig. 1 and Fig.2, and it includes a cathode, an organic light emitting layer, an anode and a glass substrate.

[0004] The inventor finds that the prior art at least has the following problems: when a photon emitted by the organic light emitting layer irradiates the air layer, the photon is reflected and refracted on an interface of an incident medium (the incident medium in Fig. 1 is a glass substrate, and the incident in Fig.2 medium is a semi-transparent cathode) and the air, and an incidence angle and a refraction angle meet a relational expression: $n_1 \cdot \sin \theta_1 = n_2 \cdot \sin \theta_2$, wherein n_1 represents a refractive index of the incident medium, n_2 represents a refractive index of the air. When the incidence angle is greater than or equal to a critical angle, total reflection occurs, and rays cannot be emitted to the air. In Fig. 1 and Fig.2, rays a and b are emitted into the air after being refracted, while rays c and d perform total reflection on the interface and thus cannot escape from the surface of the glass substrate, since the incidence angle thereof is greater than or equal to the critical angle, namely an "escape cone" of a certain angle exists, which reduces a light extraction efficiency of the OLED device.

SUMMARY OF THE INVENTION

[0005] In view of the above problem of low light extraction efficiency of the existing OLED device, the present invention provides a method for preparing an uneven particle layer, an organic light emitting diode device and a display device containing the uneven particle layer.

[0006] The technical solution for solving the technical problem in the present invention is as follows: a method for preparing an uneven particle layer including the following steps:

forming a nanoparticle layer on a substrate;
heating the substrate to fuse nanoparticles that are in contact with the substrate, whereas the nanoparticles on the surface keep a solid state; and
cooling the substrate to form a nanoparticle layer with an uneven surface.

[0007] Preferably, the step of forming a nanoparticle layer on a substrate is to coat a mixed solution of the nanoparticles which includes the nanoparticles, a solvent used for dispersing the nanoparticles and a dispersant, on the substrate.

[0008] Preferably, the nanoparticle layer is composed of a single layer of nanoparticles.

[0009] Preferably, the step of heating the substrate is carried out on a side of the substrate with no nanoparticle layer.

[0010] Preferably, the nanoparticle layer is made of a transparent polymer material.

[0011] Preferably, the transparent polymer material includes polystyrene and/or polymethacrylate.

[0012] Preferably, the glass transition temperature of the transparent polymer material is T_1 , and the heating temperature is T_1 to $T_1 + 50^\circ\text{C}$.

[0013] Preferably, the heating time is 1-20min.

[0014] Preferably, the shape of the nanoparticles is a sphere.

[0015] Preferably, the particle size of the nanoparticles is 400-700 nm.

[0016] Preferably, the thickness of the uneven particle layer is less than 1000 nm.

[0017] Preferably, before coating the mixed solution of the nanoparticles, an electrode is arranged on the substrate in advance.

[0018] The present invention further provides an OLED device, including a substrate, an anode, an organic light emitting layer and a cathode, wherein the substrate includes an uneven particle layer prepared according to the above method.

[0019] The present invention further provides a display device, including the aforementioned OLED device.

[0020] According to the method for preparing the uneven particle layer in the present invention, only coating and heating steps need to be added, so the method is simple and industrial production is easy to achieve. A level of the unevenness of the surface of the uneven particle layer prepared by the method is uniform, and the periodicity thereof is good. The substrate including the uneven nanoparticle layer is applied to the OLED device, the performance of the device is stable, and the propagation direction of rays from the organic light emitting layer of the OLED device can be changed to avoid total reflection on an interface so as to emit more light into the air, thereby improving the light extraction efficiency of the OLED device and improving the brightness and the viewing angle of the OLED device. The OLED device prepared by the method in the present invention is suitable for various display devices.

BRIEF DESCRIPTION OF THE DRAWINGS

[0021]

Fig.1 is a structural schematic diagram of an existing OLED device;

Fig.2 is a structural schematic diagram of another existing OLED device;

Fig.3 is a schematic diagram of a method for preparing an uneven particle layer according to the present invention;

Fig.4 is a structural schematic diagram of an OLED device according to the third embodiment of the present invention;

Fig.5 is a schematic diagram of light emission of the OLED device according to the third embodiment of the present invention;

DESCRIPTION OF THE PREFERRED EMBODIMENT

[0022] In order that those skilled in the art can better understand the technical solutions of the present invention, a further detailed description of the present invention will be given below in combination with the accompany drawings and specific embodiments.

[0023] According to an embodiment of the present invention, a method for preparing an uneven particle layer is provided, including the following steps:

forming a nanoparticle layer on a substrate;
heating the substrate to fuse nanoparticles that are in contact with the substrate, whereas the nanoparticles on the surface keep a solid state; and
cooling the substrate to form a nanoparticle layer with an uneven surface.

[0024] As show in Fig.3, the method specifically includes the following steps:

S1, forming a nanoparticle layer on a substrate, which is realized by:

mixing nanoparticles with a solvent used for dispersing the nanoparticles and a dispersant, to form a mixed solution of the nanoparticles; and then, coating the mixed solution of the nanoparticles on the substrate 4.

[0025] The nanoparticles can be made of an inorganic material or an organic material. Preferably, the nanoparticles are made of a transparent polymer material. The transparent polymer material includes, but not limited to, polystyrene, polymethylacrylic acid, etc.

[0026] As the transparent polymer material is adopted, when the substrate 4 with the uneven particle layer is applied to an OLED device, the light extraction efficiency can be improved.

[0027] Preferably, the nanoparticles can be polystyrene and/or polymethacrylate. Preferably, the nanoparticle layer is composed of a single layer of nanoparticles.

[0028] The nanoparticles are mixed with the solvent and the dispersant, and the nanoparticles can be uni-

formly dispersed by the solvent and the dispersant, so as to obtain a nanoparticle layer with a uniform thickness. The obtained nanoparticle layer can reduce total reflection and improve the light extraction efficiency of an OLED device. Particularly, the dispersant is helpful to improve the arrangement of the nanoparticles on the surface of the substrate 4 and prevent the accumulation of the nanoparticles, so it is helpful to form a single layer of nanoparticles.

[0029] Those skilled in the art can select a suitable solvent and a suitable dispersant according to the type of the specifically selected nanoparticles. When the nanoparticles are polystyrene, methanol or toluene and the like organic solvents can be adopted; and when the nanoparticles are polymethacrylate, chloroform, acetic acid, ethyl acetate, acetone and the like organic solvents can be adopted. Preferably, a solvent with strong volatility is adopted. Those skilled in the art can select the dispersant adaptive to the nanoparticles according to experience, for example, PVP (polyvinyl pyrrolidone), etc.

[0030] Preferably, the shape of the nanoparticles is a sphere.

[0031] When the shape of the nanoparticles is a sphere, the unevenness of the surface of the uneven particle layer 5 prepared by the method is uniform, and the periodicity thereof is good. When the uneven particle layer is applied to an OLED device, the rays do not perform total reflection on the spherical surfaces, and the emission direction of the rays is the same as the normal direction, as shown in Fig.5.

[0032] Preferably, the particle size of the nanoparticles is 400-700 nm.

[0033] That is to say, if the particle size of the nanoparticles is too large or too small, it is unbeneficial to controlling the thickness of the formed uneven particle layer 5, and when the particle size of the nanoparticles is 400-700nm, the coating operation and the heating formation operation are easy to carry out.

[0034] The mixed solution of the nanoparticles can be coated on the substrate 4 by adopting a conventional coating method. Preferably a spin coating mode is applied by which it can be guaranteed that a film layer with a uniform thickness is formed by the mixed solution on the substrate 4 and a monomolecular layer of polystyrene nanoparticles is formed.

[0035] S2, heating the substrate 4 to fuse nanoparticles that are in contact with the substrate, whereas the nanoparticles on the surface keep a solid state;

[0036] Preferably, the heating is carried out on a side of the substrate 4 not coated with nanoparticles. That is to say, the back surface of the substrate 4 is heated, by which the nanoparticles on the substrate 4 are heated, so the molecular motion thereof accelerates and a part of nanoparticles are gradually fused, thereby changing the surface morphology of the nanoparticles so as to form an uneven surface.

[0037] Preferably, the heating time is 1-20 min, preferably 1-10 min and more preferably 2-5 min. Those

skilled in the art can change the specific heating time for different nanoparticle materials according to experience.

[0038] Preferably, if the glass transition temperature of the transparent polymer is T_1 , the heating temperature is T_1 to $T_1+50^\circ\text{C}$, preferably T_1 to $T_1+30^\circ\text{C}$, and more preferably T_1 to $T_1+15^\circ\text{C}$. That is, the heating temperature is between the glass transition temperature of the transparent polymer and the glass transition temperature thereof plus 50°C . Upon exceeding the glass transition temperature, polymer macromolecules start to be unfastened and are gradually fused. According to the time-temperature equivalence principle, the heating time necessary for a higher heating temperature is shorter, and on the contrary, if the heating temperature is lower, the necessary heating time is longer. Specifically, if the glass transition temperature of the polystyrene nanoparticles is 80°C , then the heating temperature is not greater than 130°C and is between 80 - 130°C . The heating time is longer at 80°C , and the heating time is shorter at 130°C .

[0039] S3, cooling the substrate 4 to form a nanoparticle layer 5 with an uneven surface

[0040] Preferably, the thickness of the uneven particle layer 5 is less than 1000nm , so as to guarantee better performance of an OLED device prepared by the method.

[0041] Preferably, before coating the mixed solution on the nanoparticles, an anode 3 is arranged on the substrate 4. In such case, the mixed solution of the nanoparticles coated on the substrate 4 is actually coated on the anode 3.

[0042] In another specific embodiment of the present invention, an OLED device is provided, including a substrate, an anode, an organic light emitting layer and a cathode, wherein the substrate is prepared according to the above method and includes an uneven particle layer. Apparently, the OLED device in the specific embodiment can be in a form of top light emission and can also in the form of bottom light emission.

[0043] Another solution of the present invention provides a display device, including the aforementioned OLED device. The display device can be any product or component having a display function, such as electronic paper, a mobile phone, a tablet computer, a television, a display, a notebook computer, a digital picture frame, a navigator, etc.

Example 1

[0044] As shown in Fig.4, an OLED device A is provided, including a substrate 4, an anode 3, an organic light emitting layer 2 and a cathode 1, wherein the substrate 4 includes an uneven particle layer prepared according to the above method.

[0045] The nanoparticles are polystyrene nanoparticles with a particle size of 600 nm , the solvent is methanol, and the dispersant is polyvinylpyrrolidone (PVP).

[0046] The substrate 4 is made of a polyethylene terephthalate (PET) material. The anode 3 is made of aluminum, and the thickness of the anode 3 is 150 nm .

[0047] The specific preparation steps include:

adding the polystyrene nanoparticles with a particle size of 600 nm and the dispersant PVP into the solvent methanol, and mixing them uniformly to form a mixed solution;

coating the mixed solution on the PET substrate 4; heating a surface not coated with nanoparticle of the substrate 4 at 110°C for 2min to fuse the nanoparticles close to the substrate 4, and keep the nanoparticles away from the substrate 4 in a solid state to form a hemisphere;

cooling the substrate 4 to form an uneven particle layer 5 on the substrate;

forming the anode 3 on the substrate 4 after the above steps are completed;

forming the organic light emitting layer 2 on the substrate 4 after the above steps are completed; and forming the cathode 1 on the substrate 4 after the above steps are completed.

Comparative example 1

[0048] An OLED device B is manufactured by the same method as Example 1, and the difference lies in that no uneven particle layer 5 is formed.

[0049] A brightness test is carried out on the OLED device A and the OLED device B. Under the same voltage, the brightness of the OLED device A is 1.2 times as large as the brightness of the OLED device B at a front viewing angle ($\theta=0^\circ$, $\Phi=0^\circ$), and the brightness of the device A is 1.5 times as large as the brightness of the device B at a side viewing angle ($\theta=45^\circ$, $\Phi=45^\circ$).

Example 2

[0050] As shown in Fig.4, an OLED device C is provided, including a substrate 4, an anode 3, an organic light emitting layer 2 and a cathode 1, wherein the substrate 4 includes an uneven particle layer prepared according to the above method.

[0051] The nanoparticles are polymethylacrylic acid nanoparticles with a particle size of 600 nm , the solvent is methanol, and the dispersant is PVP.

[0052] The substrate 4 is made of a glass material, the anode 3 is made of aluminum, and the thickness of the anode 3 is 150 nm .

[0053] The specific preparation steps include:

adding the polymethylacrylic acid nanoparticles with a particle size of 600 nm and the dispersant PVP into the solvent methanol, and mixing them uniformly to form a mixed solution;

coating the mixed solution on the PET substrate 4; heating a surface not coated with nanoparticle of the substrate 4 at 120°C for 2 min to fuse the nanoparticles close to the substrate 4, and keep the nanoparticles away from the substrate 4 in a solid state

to form a hemisphere;
cooling the substrate 4 to form an uneven particle layer 5 on the substrate;
forming the anode 3 on the substrate 4 after the above steps are completed;
forming the organic light emitting layer 2 on the substrate 4 after the above steps are completed; and
forming the cathode 1 on the substrate 4 after the above steps are completed.

Comparative example 2

[0054] An OLED device D is manufactured by the same method as the Example 2, and the difference lies in that no uneven particle layer 5 is formed.

[0055] A brightness test is carried out on the OLED device C and the OLED device D. Under the same voltage, the brightness of the OLED device A is 1.3 times as large as the brightness of the OLED device B at a front viewing angle ($\theta=0^\circ$, $\Phi=0^\circ$), and the brightness of the device A is 1.6 times as large as the brightness of the device B at a side viewing angle ($\theta=45^\circ$, $\Phi=45^\circ$).

[0056] It can be understood that, the above embodiments are merely exemplary embodiments used for illustrating the principle of the present invention, but the present invention is not limited hereto. Those of ordinary skill in the art can make a variety of modifications and improvements without departing from the spirit and essence of the present invention, and these modifications and improvements are deemed as the protection scope of the present invention.

Claims

1. A method for preparing an uneven particle layer, comprising the following steps:

forming a nanoparticle layer on a substrate;
heating the substrate to fuse nanoparticles that are in contact with the substrate, whereas the nanoparticles on the most surface keep a solid state; and
cooling the substrate to form a nanoparticle layer with an uneven surface.

2. The method for preparing the uneven particle layer according to claim 1, wherein the step of forming a nanoparticle layer on the substrate is to coat a mixed solution of the nanoparticles comprising the nanoparticles, a solvent used for dispersing the nanoparticles and a dispersant on the substrate.
3. The method for preparing the uneven particle layer according to claim 1, wherein the step of heating the substrate is carried out on a side of the substrate with no nanoparticle layer.

4. The method for preparing the uneven particle layer according to claim 1, wherein the nanoparticle layer is made of a transparent polymer material.
5. The method for preparing the uneven particle layer according to claim 4, wherein the transparent polymer material comprises polystyrene and/or polymethacrylate.
6. The method for preparing the uneven particle layer according to claim 4, wherein the glass transition temperature of the transparent polymer material is T_1 , and the heating temperature is T_1 to $T_1+50^\circ\text{C}$.
7. The method for preparing the uneven particle layer according to claim 6, wherein the heating time is 1-20min.
8. The method for preparing the uneven particle layer according to claim 1, wherein the nanoparticle has a shape of sphere.
9. The method for preparing the uneven particle layer according to claim 1, wherein the nanoparticle has a particle size of 400-700 nm.
10. The method for preparing the uneven particle layer according to claim 1, wherein the uneven particle layer has a thickness of less than 1000nm.
11. The method for preparing the uneven particle layer according to claim 1, wherein the nanoparticle layer is composed of a single layer of nanoparticles.
12. The method for preparing the uneven particle layer according to claim 2, wherein before coating the mixed solution of the nanoparticles, an electrode is arranged on the substrate.
13. An organic light emitting diode device, comprising a substrate, an anode, an organic light emitting layer and a cathode, wherein the OLED device further comprises an uneven particle layer prepared by the method of any one of claims 1-11.
14. A display device, comprising the OLED device of claim 12.

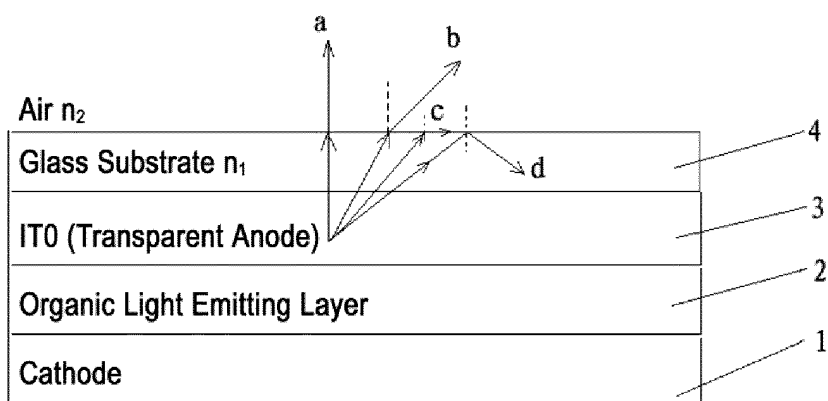


Fig. 1

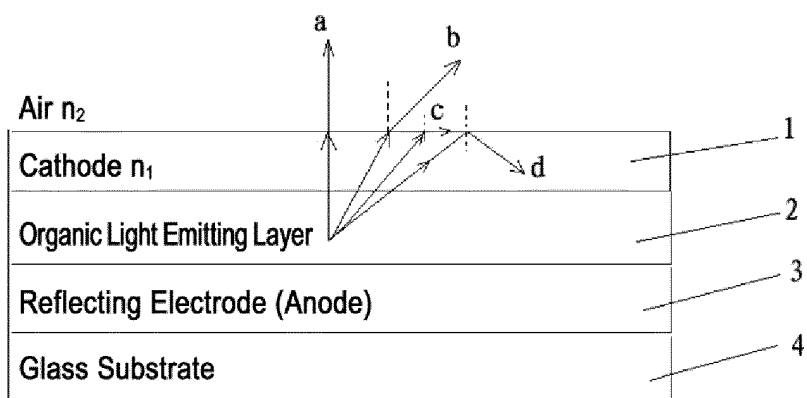


Fig. 2

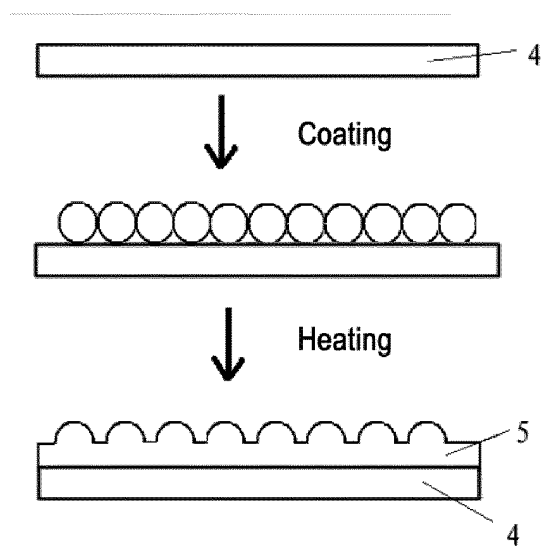


Fig. 3

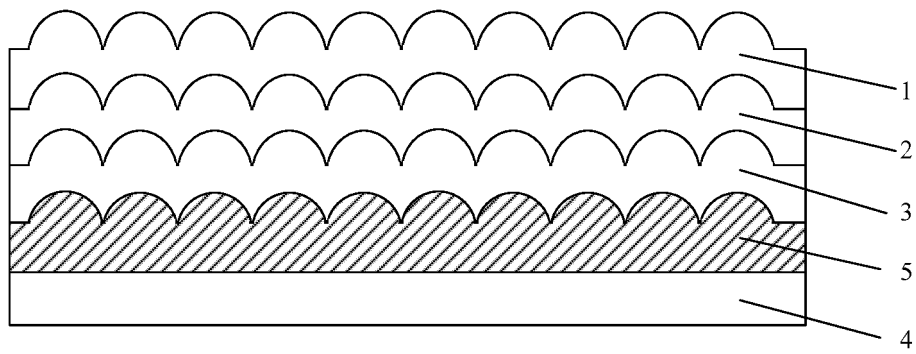


Fig. 4

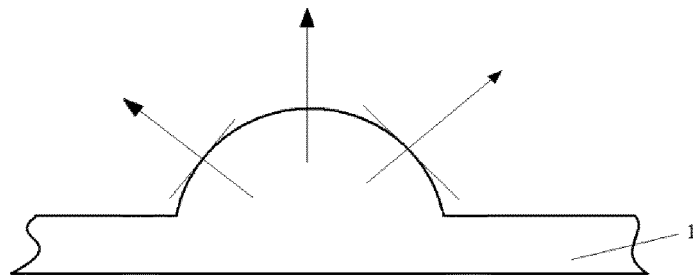


Fig. 5

INTERNATIONAL SEARCH REPORT

International application No.

PCT/CN2016/073987

A. CLASSIFICATION OF SUBJECT MATTER

H01L 51/56 (2006.01) i; H01L 51/52 (2006.01) i

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC: H01L 51/-

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

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

CNPAT CNKI WPI EPODOC: organic light emitting diode, organic electroluminescence, light extraction, scattering, particle, OLED, organic, electroluminescent, heat+, nano+, light, out, diffus+, melt+

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
PX	CN 105118934 A (BOE TECHNOLOGY GROUP CO., LTD. et al.), 02 December 2015 (2015-12 -02), description, paragraphs [0030]-[0080], and figures 1-5	1-14
Y	CN 103943738 A (INSTITUTE OF SEMICONDUCTORS, CHINESE ACADEMY OF SCIENCES), 23 July 2014 (23.07.2014), description, paragraphs [0023]-[0032], and figures 3, 7 and 8	1-14
Y	CN 103503571 A (MATSUSHITA ELECTRIC INDUSTRIAL CO., LTD.), 08 January 2014 (08.01.2014), description, paragraphs [0047]-[0064] and [0131]	1-14
X	CN 102701143 A (WU, Kui et al.), 03 October 2012 (03.10.2012), description, paragraph [0023], and figure 2	1-2, 4-5, 8, 10-11
A	CN 103688385 A (HITACHI LTD.), 26 March 2014 (26.03.2014), the whole document	1-14
A	US 2015179977 A1 (PANASONIC INTELLECTUAL PROPERTY MANAGEMENT CO., LTD.), 25 June 2015 (25.06.2015), the whole document	1-14

☐ Further documents are listed in the continuation of Box C.
 ☒ See patent family annex.

* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A" document defining the general state of the art which is not considered to be of particular relevance	
"E" earlier application or patent but published on or after the international filing date	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"O" document referring to an oral disclosure, use, exhibition or other means	
"P" document published prior to the international filing date but later than the priority date claimed	"&" document member of the same patent family

Date of the actual completion of the international search 14 April 2016 (14.04.2016)	Date of mailing of the international search report 05 May 2016 (05.05.2016)
Name and mailing address of the ISA/CN: State Intellectual Property Office of the P. R. China No. 6, Xitucheng Road, Jimenqiao Haidian District, Beijing 100088, China Facsimile No.: (86-10) 62019451	Authorized officer WANG, Jue Telephone No.: (86-10), 01062413509

Form PCT/ISA/210 (second sheet) (July 2009)

INTERNATIONAL SEARCH REPORT
 Information on patent family members

 International application No.
PCT/CN2016/073987

Patent Documents referred in the Report	Publication Date	Patent Family	Publication Date
CN 105118934 A	02 December 2015	None	
CN 103943738 A	23 July 2014	None	
CN 103503571 A	08 January 2014	WO 2013001891 A1	03 January 2013
		US 2014027753 A1	30 January 2014
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		JP 2013026300 A	04 February 2013
		JP 5789439 B2	07 October 2015
US 2015179977 A1	25 June 2015	JP 2015144110 A	06 August 2015
		US 9214649 B2	15 December 2015

专利名称(译)	制备不均匀颗粒层的方法，有机电致发光器件和显示装置		
公开(公告)号	EP3352238A4	公开(公告)日	2019-04-24
申请号	EP2016763187	申请日	2016-02-18
[标]申请(专利权)人(译)	京东方科技集团股份有限公司 合肥京东方光电科技有限公司		
申请(专利权)人(译)	京东方科技集团股份有限公司. 合肥京东方光电科技有限公司.		
当前申请(专利权)人(译)	京东方科技集团股份有限公司. 合肥京东方光电科技有限公司.		
[标]发明人	GUO YUANHUI ZANG YUANSHENG LIU ZHI LI XIAOHE WANG HUI		
发明人	GUO, YUANHUI ZANG, YUANSHENG LIU, ZHI LI, XIAOHE WANG, HUI		
IPC分类号	H01L51/56 H01L51/52		
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其他公开文献	EP3352238A1		
外部链接	Espacenet		

摘要(译)

本发明提供一种制备不均匀颗粒层的方法，有机发光二极管装置和显示装置。制备不均匀颗粒层的方法包括以下步骤：在基板上形成纳米颗粒层；加热底物以融合与基底接触的纳米颗粒，而表面上的纳米颗粒保持固态；冷却基板以形成具有不平坦表面的纳米颗粒层。本发明的方法工艺简单，易于实现工业生产。将包括不均匀颗粒层的基板施加到OLED器件，因此可以改变来自OLED器件的有机发光层的光线的传播方向，以避免在界面上的全反射，向空气中发射更多的光。并提高OLED器件的光提取效率。在本发明中制备的OLED器件适用于各种显示器件。