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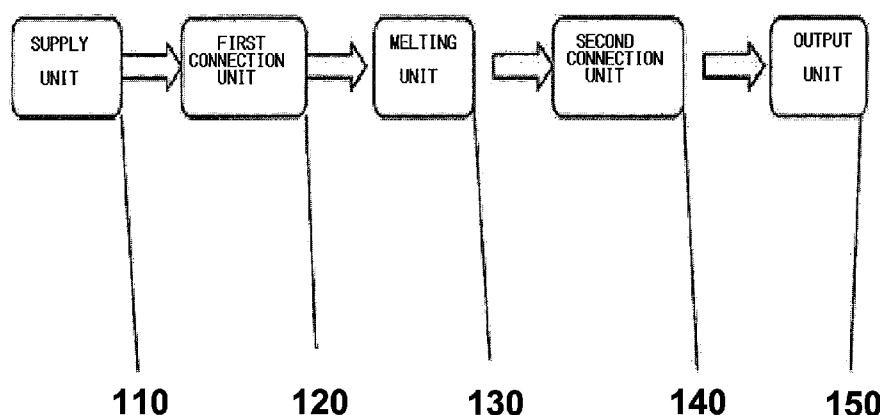
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(54) Title: EQUIPMENT AND METHOD FOR PRODUCING OLED USING PHASE-CHANGED MATERIAL



(57) Abstract: The present disclosure relates to an ink jet device for manufacturing an organic electroluminescent device, comprising a melting unit for phase-changing a phase-change material in a solid state including an organic electroluminescent compound into a liquid state; and an output unit for spraying the phase-change material in a liquid state together with an induction gas. Accordingly, it is not necessary to use a solvent for producing an organic electroluminescent device, and a low-molecular / monomolecular-based material as well as a polymeric material can be used as the organic electroluminescent compound.



Description

Title of Invention: EQUIPMENT AND METHOD FOR PRODUCING OLED USING PHASE-CHANGED MATERIAL

Technical Field

- [1] The present disclosure relates to an apparatus for manufacturing an organic electroluminescent device (OLED) using phase-change material and a method for manufacturing an organic electroluminescent device.

Background Art

- [2] A method for manufacturing an organic electroluminescent device comprises vacuum deposition (evaporation), sputtering, plasma, ion plating, ink jet printing, nozzle printing, slot coating, spin coating, immersion coating, flow coating, screen printing, microgravure printing, spreading, stamp printing, spraying, paint, laser transfer, etc.
- [3] The vacuum deposition method is a method of manufacturing an organic electroluminescent device using a low molecular organic material of a single compound type, which is advantageous in terms of lifetime and luminous efficiency of the device. However, since the method requires vacuum deposition of an organic material in powder form, it is difficult to maintain a big size and uniformity; thus it is necessary to improve the cost in terms of waste of material.
- [4] Among the methods for forming a film after an organic material is dissolved in a solvent, an ink jet printing method is mainly used for a polymer organic material. Generally, since the liquid state light emitting material is finely jetted through each of the nozzles, the ink jet printing technique discards only a small amount of polymer organic material. Further, because of a process in the liquid phase, for example, in the large production line of 8 generations or more, it is not necessary to cut the ledge glass substrate, so that the process can be simplified somewhat. As a result, not only the investment cost of equipment can be minimized, but also the overall process time can be saved. Korean Patent Publication Nos. 546921 and 1020240 disclose soluble inkjet printing technology.
- [5] Meanwhile, the process of forming a film after an organic material is dissolved in a solvent must include a cost of the solvent and the purification for producing the soluble material, and reduction of the cost is required. In addition, such a solubilized solution process must include a drying time of the solvent, and there is a need to improve the time with regard to shortening manufacturing time. Furthermore, polymeric copolymer materials, which are predominantly used in soluble ink jet systems, are likely to have an uneven layer or non-polymerized area, which may act as a trap position; thus which may degrade the performance of the device or reduce the uniformity of an

interface. In addition, a large amount of material may be lost during the removal of the organic solvent. Also, after the process, if the organic solvent for dissolving the material is not completely removed, the residual solvent may act as a trap and impurities contained in the solvent may degrade the luminescent properties of the device. That is, the solubility ink jet method using a solvent has a lower uniformity than the vapor evaporation method, and it requires an additional apparatus for evaporating or sublimating the solvent.

Disclosure of Invention

Technical Problem

- [6] The object of the present disclosure is to provide a method and apparatus for manufacturing an organic electroluminescent device without depending on dissolution, particularly, a method of jetting a predetermined gas together with a material of an organic electroluminescent device, and an ink jet apparatus which executes the method.

Solution to Problem

- [7] As a result of intensive studies to solve the technical problem above, the present inventor found that the above objective can be achieved by an ink jet device for manufacturing an organic electroluminescent device, comprising: a melting unit for phase-changing a phase-change material in a solid state including an organic electroluminescent compound into a liquid state; and an output unit for spraying the phase-change material in a liquid state together with an induction gas.
- [8] According to one embodiment of the present disclosure, the output unit may spray the induction gas in a form surrounding the phase-change material in a liquid state.
- [9] According to one embodiment of the present disclosure, the output unit may start the spraying of the phase-change material in a liquid state after starting the spraying of the induction gas.
- [10] According to one embodiment of the present disclosure, the induction gas may comprise at least one of nitrogen or argon.
- [11] According to one embodiment of the present disclosure, the phase-change material may comprise at least one kind of organic electroluminescent compound, and the melting unit includes a heater for phase-changing the phase-change material to a liquid state by applying heat corresponding to a temperature equal to or lower than a melting point of any one of the organic electroluminescent compound of the at least one kind of organic electroluminescent compound.
- [12] According to one embodiment of the present disclosure, the phase-change material may comprise at least two kinds of organic electroluminescent compounds, and the melting unit includes a heater for phase-changing the phase-change material to a liquid

state by applying heat corresponding to a temperature lower than an arithmetic average value of melting points of each of the two or more organic electroluminescent compounds.

- [13] The present disclosure also provides a method for manufacturing an organic electroluminescent device, comprising steps of: phase-changing a phase-change material in a solid state including an organic electroluminescent compound into a liquid state; and spraying the phase-change material in a liquid state together with an induction gas.
- [14] According to one embodiment of the present disclosure, the step of spraying the phase-change material in a liquid state together with the induction gas may comprise: spraying the induction gas in a form surrounding the phase-change material in a liquid state.
- [15] According to one embodiment of the present disclosure, the step of spraying the phase-change material in a liquid state together with the induction gas may comprise: starting the spraying of the phase-change material in a liquid state after starting the spraying of the induction gas.
- [16] According to one embodiment of the present disclosure, the induction gas may comprise at least one of nitrogen or argon.
- [17] According to one embodiment of the present disclosure, the phase-change material may comprise at least one kind of organic electroluminescent compound, and the step of spraying the phase-change material in a liquid state together with the induction gas may comprise: phase-changing the phase-change material to a liquid state by applying heat corresponding to a temperature equal to or lower than a melting point of any one of the organic electroluminescent compound of the at least one kind of organic electroluminescent compound.

Advantageous Effects of Invention

- [18] Since a commonly used organic solvent in a soluble ink jet method is not used in the apparatus and method of present disclosure, the cost of solvent and its purification can be reduced, and the evaporation time for removing the solvent is not required, which can shorten the manufacturing process time.
- [19] According to the apparatus and method of the present disclosure, liquid material can be applied closer to the desired shape by spraying the liquid material with induction gas and/or carrier gas.
- [20] According to the apparatus and method of the present disclosure, there is no need to develop new material used in the apparatus and the method since conventional low molecular / monomolecular vapor deposition materials can be used as they are, instead of a soluble polymeric material that can cause problems of non-uniformity.
- [21] According to the apparatus and method of the present disclosure, damage to the

organic electroluminescent compound contained in the phase-change material can be prevented by melting the phase-change material at a relatively low temperature, which also can reduce cost and time in terms of heat energy usage.

Brief Description of Drawings

- [22] Fig. 1 schematically shows an ink jet apparatus for manufacturing an organic electroluminescent device according to one embodiment of the present disclosure.
- [23] Fig. 2 illustrates a method of manufacturing an organic electroluminescent device according to one embodiment of the present disclosure.
- [24] Fig. 3 illustrates a configuration of an ink jet apparatus for manufacturing an organic electroluminescence device according to an embodiment of the present disclosure.
- [25] Fig. 4 illustrates a configuration of an ink jet apparatus for manufacturing an organic electroluminescence device according to an embodiment of the present disclosure.
- [26] Fig. 5 illustrates a configuration of an ink jet apparatus for manufacturing an organic electroluminescence device according to an embodiment of the present disclosure.
- [27] Fig. 6 schematically shows a structure of an organic electroluminescent device manufactured according to an embodiment of the present disclosure.

Mode for the Invention

- [28] Hereinafter, the present disclosure will be described in detail. However, the following description is intended to explain the disclosure, and is not meant in any way to restrict the scope of the disclosure.
- [29] The term "an organic electroluminescent compound" in the present disclosure means a compound that may be used in an organic electroluminescent device, and may be comprised in any layers constituting an organic electroluminescent device, if necessary.
- [30] The term "an organic electroluminescent material" in the present disclosure means a material that may be used in an organic electroluminescent device, and may comprise at least one compound. If necessary, the organic electroluminescent material may be comprised in any layers constituting an organic electroluminescent device. For example, the organic electroluminescent material may be a hole injection material, a hole transport material, a hole auxiliary material, a light-emitting auxiliary material, an electron blocking material, a light-emitting material, an electron buffer material, a hole blocking material, an electron transport material, an electron injection material, etc.
- [31] The term "a phase-change material" in the present disclosure means a material that may change from one phase (for example, a solid state, a liquid state, or a gaseous state) to another depending on certain external conditions such as temperature, pressure, magnetic field, and the like. The phase-change material may be an organic electroluminescent compound or may include an organic electroluminescent

compound. The phase-change material includes material whose structure and/or performance is not changed before and after the phase change. If the phase-change material changes its structure and/or performance after a phase change, it may retain the original structure and/or performance or it may have a new structure and/or performance in case that it returns to the original phase again.

- [32] The phase-change material includes an organic electroluminescent material capable of undergoing at least one phase change among melting, sublimation, vaporization, solidification and liquefaction due to temperature, pressure, or the like. The phase-change material may correspond to a material whose phase transition from a solid state to a liquid state or from a solid state to a gaseous state does not require much energy, but whose phase change from a liquid state or a gas phase to a solid state proceeds slowly. For example, the phase-change material may be an amorphous solid material at room temperature while having a relatively low melting point. For materials with low melting points, the probability that the chemical structure or physical properties are changed due to the heating during melting becomes low.
- [33] The terms "a carrier gas" and/or "an induction gas" in the present disclosure refer to a gas used for transporting, moving, spreading, blocking, guiding, or retransporting a predetermined substance. For example, the carrier gas and the induction gas may each comprise at least one of nitrogen, which is inactive at room temperature, and argon, which is generally inert. Since the nitrogen and/or argon has low reactivity with the phase-change material and air, the rate of causing a chemical reaction is low even when mixed with the phase-change material or injected into the air. Also, it is not necessary to provide a separate device for removing nitrogen and/or argon after being sprayed with the phase-change material. The carrier gas and the induction gas may be the same material, but at least some of the components may be different from each other.
- [34] The term "an ink jet apparatus" in the present disclosure generally refers to an apparatus for spraying or ejecting a substance containing a substance in a liquid state. For example, the ink jet apparatus may use at least one of a piezo method, a thermal bubble method, a bubble-jet method, and an aerosol-jet method.
- [35] Fig. 1 schematically shows an ink jet apparatus for manufacturing an organic electroluminescent device according to one embodiment of the present disclosure. Hereinafter, an ink jet device for manufacturing an organic electroluminescent device will be described with reference to Fig 1.
- [36] Referring to Fig. 1, an ink jet apparatus for manufacturing an organic electroluminescent device may include a supply unit 110, a first connection unit 120, a melting unit 130, a second connection unit 140, and an output unit 150. The ink jet apparatus for manufacturing an organic electroluminescent device of the present invention is not

limited to the embodiment shown in Fig. 1. For example, the ink jet apparatus for manufacturing an organic electroluminescent device may only include the melting unit 130 and the output unit 150. Alternatively, the ink jet apparatus for manufacturing an organic electroluminescent device may include at least one of a display unit, a communication unit, a sensing unit, and a control unit in addition to the units shown in Fig. 1.

[37] One ink jet apparatus for manufacturing an organic electroluminescent device may include one or more supply units 110. Each of the supply units 110 may include one or more inlets (injection ports). An organic electroluminescent compound or a phase-change material containing an organic electroluminescent compound from outside may be injected through the supply unit 110 in a liquid state or a solid state. The supply unit 110 may be referred to as a loading chamber or an ink loader or the like. When the supply unit 110 includes a plurality of inlets, different materials may be injected through each inlet. Meanwhile, the supply unit 110 may be included in the melting unit 130 or the ink jet apparatus may not include the supply unit 110.

[38] The first connection unit 120 may control transfer of the phase-change material from the supply unit 110 to the melting unit 130. Under specific conditions, the first connection unit 120 may retain the phase-change material without transferring it. The specific condition may include at least one of a volume (mass) of the phase-change material being held in the supply unit 110 or the first connection unit 120, the user's input, a predetermined pressure range, a predetermined temperature range, or a predetermined time.

[39] The first connection unit 120 may accommodate or may convey the phase-change material from the supply unit 110 in a solid state. When transporting different phase-change materials, the first connection unit 120 may be configured to transport the phase-change materials respectively through a plurality of paths. According to one embodiment of present disclosure, if the phase-change material provided from the supply unit 110 is not in a solid state, the first connection unit 120 may change a state of the phase-change material to a solid state through cooling. The first connection unit 120 transfers the phase-change material from the supply unit 110 to the melting unit 130 according to whether the conditions of the volume (mass) of the transferred phase-change material, temperature, pressure, time, or user input are satisfied. The first connection unit 120 may include a gate valve that can be opened and closed, and may control the transfer of the phase-change material by the gate valve.

[40] In the present disclosure, the first connection unit 120 is an optional component and may be omitted when implementing the invention. Or, the first connection unit 120 may be included in the melting unit 130.

[41] The melting unit 130 heats phase-change material in a solid state containing an organic electroluminescent compound to change it to a liquid state. The phase-change

material further includes a material that does not act as a solvent for the organic electroluminescent compound, thereby the organic electroluminescent compound may be phase-changed to a liquid state at a temperature lower than the melting point of the organic electroluminescent compound. For example, when the phase-change material includes one kind of organic electroluminescent compound, the heat applied to the phase-change material may be configured to be equal to or lower than the melting point of any one of the organic electroluminescent compounds. Or, when the phase-change material includes two or more kind of organic electroluminescent compounds, the heat applied to the phase-change material may be configured to correspond to a temperature lower than an arithmetic average value of melting points of the two or more kind of organic electroluminescent compounds. The melting unit 130 may also be referred to as a melting chamber or a heater. The melting unit 130 may transfer the melted phase-change material to the output unit 150 through the second connection unit 140 or directly.

[42] The second connection unit 140 connects the melting unit 130 and the output unit 150. The second connection unit 140 controls the phase-change material in the second connection unit 140 to be maintained in a liquid state. The second connection unit 140 may be configured to allow the phase-change material to be transferred from the melting unit 130 to the output unit 150 when conditions such as volume (mass) of the phase-change material, temperature, pressure, time, or user input are satisfied. The second connection unit 140 may include a gate valve that can be opened and closed, and the second connection unit 140 may control the transfer of the phase-change material by the gate valve. The second connection unit 140 and/or the first connection unit 120 may be opened and closed by a separate control unit.

[43] In the present disclosure, the second connection unit 140 is an optional component, and may be omitted to implement the invention. The second connection unit 140 may be included as a part of the melting unit 130 or the output unit 150.

[44] The output unit 150 outputs the phase-change material melted in the melting unit 130 in the form of a spray or the like. The output unit 150 may be referred to as a printer head, a radiation unit, or the like. The output unit 150 may continuously heat the melted phase-change material to prevent the melted phase-change material from solidifying until phase-change material is output. The output unit 150 may include one or more nozzles for spraying the phase-change material in a molten state onto a substrate or the like. The one or more nozzles may be configured to be attached to the output unit 150, respectively.

[45] The at least one nozzle may mix the molten phase-change material in the carrier gas to form an aerosol mist to be sprayed. The carrier gas may also be utilized as a means for applying pressure to transfer the phase-change material into the nozzle within the

output unit 150.

- [46] The one or more nozzles may spray the molten phase-change material and the induction gas together. The one or more nozzles may spray the induction gas in a form surrounding the phase-change material in a liquid state. The induction gas may be referred to as a sheath gas, and may induce the sprayed phase-change material to be uniformly applied to the substrate.
- [47] For sophisticated application, each nozzle may be configured to initiate spraying of the liquid phase phase-change material after a predetermined time (e.g., 0.5 seconds) has elapsed since the start of the spraying of the induction gas. Alternatively, each nozzle may simultaneously initiate spraying of the induction gas and the phase-change material in a liquid state.
- [48] Preferably, the induction gas and the carrier gas are low in reactivity and do not require a removal step. For example, the induction gas and the carrier gas may be a material containing at least one of nitrogen and argon, respectively. The induction gas and the carrier gas may be the same or may correspond to a material in which at least some of the constituent components are different from each other.
- [49] Fig. 2 illustrates a method of manufacturing an organic electroluminescent device according to one embodiment of the present disclosure. Hereinafter, a method of manufacturing an organic electroluminescent device will be described with reference to Fig. 2.
- [50] In step 210, a phase-change material is provided from the outside. For example, the supply unit may have one or more inlets, and the same or different compound may be provided through each inlet. The supplied phase-change material may be in a solid state or in a liquid state. According to another embodiment of the present disclosure, step 220 can be performed without step 210 on the phase-change material at a particular location.
- [51] In step 220, the solid state phase-change material containing the organic electroluminescent compound is heated to phase change to a liquid state. The heating may be performed on the phase-change material present at a particular location without going through the supply of step 210. The phase-change material is phase-changed into a liquid state even at a temperature lower than the melting point of the organic electroluminescent compound. When the phase-change material comprises at least one kind of organic electroluminescent compound, the applied heat may be configured to correspond to a temperature equal to or lower than the melting point of any one of the at least one organic electroluminescent compound. Alternatively, when the phase-change material includes two or more organic electroluminescent compounds, the applied heat may be configured to correspond to a temperature lower than an arithmetic mean value of melting points of the two or more organic electroluminescent

compounds.

- [52] The apparatus for performing an organic electroluminescent device manufacturing method can heat a phase-change material when a predetermined condition is satisfied. The predetermined condition includes an environment at a specific position of the apparatus. The environment may include, for example, at least one of the volume (mass) of the phase-change material, temperature, pressure, time, and user input.
- [53] In step 230, the phase-change material in a liquid state is sprayed together with the induction gas. For example, the phase-change material may be sprayed through a nozzle (aerosol type). The phase-change material may be implemented to be released from the nozzle along with the induction gas. The induction gas may be sprayed to surround the phase-change material.
- [54] The sprayed phase-change material may be applied to a substrate of the organic electroluminescent device or one or more layers constituting the organic electroluminescent device to form a film.
- [55] Unlike step 230 in which the phase-change material is sprayed together with the induction gas, a carrier gas may be sprayed together with the phase-change material. In this case, the carrier gas may be mixed with the phase-change material and injected. Or, when the phase-change material is sprayed, both the induction gas and the carrier gas may be sprayed.
- [56] Fig. 3 illustrates a configuration of an ink jet apparatus for manufacturing an organic electroluminescence device according to an embodiment of the present disclosure. Hereinafter, an operation of the ink jet apparatus for manufacturing an organic electroluminescence device will be described with reference to Fig. 3.
- [57] Referring to Fig. 3, the ink jet apparatus for manufacturing an organic electroluminescent device may include a supply unit 310 and a first connection unit 350.
- [58] The supply unit 310 temporarily stores phase-change materials 320, 330, and 340 supplied through an injection port (inlet) 360. The cross section of the inner wall of the supply unit 310 may have a shape of a circle, an ellipse, a rectangle, a polygon, or the like. The phase-change materials 320, 330, and 340 supplied to the supply unit 310 may have a predetermined shape such as a rectangular parallelepiped, a cylinder, and the like.
- [59] In Fig. 3, the supply unit 310 has only one injection port. However, the supply unit 310 may have two or more injection ports. Unlike Fig. 3, the injection port may be open in a direction other than the vertical direction. In Fig. 3, only one unit of phase-change material is stacked at the same height, but two or more phase-change materials may be stacked at the same height. Meanwhile, the phase-change material may be supplied in powder form.
- [60] The first connection unit 350 transfers the phase-change material from the supply

unit 310 to the melting unit 370 according to the conditions such as temperature, pressure, time, or user input. Before being transferred to the melting unit, the phase-change material is stored in the supply unit 310. The first connection unit 350 of Fig. 3 moves in the horizontal direction, thereby opening and closing the passage to the melting unit. By the movement of the first connection unit 350, the phase-change material is sequentially transferred from the lowermost phase-change material 340 to the melting unit 370. In Fig. 3, the first connecting unit 350 is shown reciprocally moving in the horizontal direction, but the present disclosure is not limited thereto. Meanwhile, the first connection unit 350 may be implemented as a form including a valve that can be opened and closed. In this case, the phase-change material may pass through the first connection unit 350 in a liquid state. Fig. 4 shows a first connection including a valve.

- [61] The ink jet apparatus for manufacturing an organic electroluminescent device of the present invention is not limited to the embodiment shown in Fig. 3. Unlike Fig. 3, the phase-change material may be transferred in a direction other than the vertical direction. The supply unit 310 may also include one or more protrusions for securing the phase-change material to a specific position when it is not in motion.
- [62] According to another embodiment of the present invention, the supply unit 310 and/or the first connection unit 350 may be further simplified or omitted. When both the supply unit 310 and the first connection unit 350 are omitted, the ink jet device for manufacturing an organic electroluminescent device of the present invention directly heats the phase-change material at a specific position before movement of the phase-change material.
- [63] Fig. 4 illustrates the structure of an ink jet apparatus for manufacturing an organic electroluminescent device according to an embodiment of the present invention. Hereinafter, the operation of the ink jet apparatus for fabricating the organic electroluminescent device will be described with reference to Fig. 4.
- [64] Referring to Fig. 4, the ink jet apparatus for manufacturing an organic electroluminescent device may include a first connection unit 410, a melting unit 420, and a second connection unit 450. The ink jet apparatus for fabricating an organic electroluminescent device of the present invention is not limited to the embodiment shown in Fig. 4. For example, the first connection unit 410 or the second connection unit 450 may be simplified or omitted. Unlike Fig. 4, the phase-change material may be implemented to be transmitted in a direction other than a vertical direction.
- [65] The phase-change material 405 transferred from the supply unit is transferred to the melting unit 420 through the first connection unit 410. The first connection unit 410 may include a valve 415 capable of opening and closing. The first connection unit 410 allows the phase-change material 405 to be transferred from the supply unit to the

melting unit 420 according to whether the conditions such as temperature, pressure, time, or user input are satisfied.

[66] The melting unit 420 includes a heater 425 for melting the phase-change material in a solid state into a liquid state. According to an embodiment, the melting unit 420 includes an agitator 430 for stirring the phase change ink, a hole 435 through which molten ink escapes, a screening film 440 for allowing the ink to be fixed until the ink is melted, and a collector 445 for collecting the phase-change material to transfer to the second connection. When different kinds of materials are delivered through the first connection unit 410, the plurality of materials may be mixed at the agitator 430 to form a blend. In Fig. 4, the second connection unit 450 is described as a bottleneck type, but is not limited thereto.

[67] The phase-change material transferred from the melting unit 420 is transferred to the output unit through the second connection unit 450. The second connection unit 450 may include a valve 455 capable of opening and closing.

[68] The hole 435 may be opened or closed depending on whether the conditions of volume (mass) of the melted phase-change material in the melting unit 420, temperature, pressure, time, or user input are satisfied. The collector 445 and/or the second connection 450 may include equipment to continuously heating the molten phase-change material so that it prevents the molten phase-change material from becoming a solid state.

[69] Fig. 5 shows a configuration of an ink jet device for manufacturing an organic electroluminescent device according to an embodiment of the present invention. Hereinafter, the operation of the ink jet apparatus for manufacturing an organic electroluminescent device will be described with reference to Fig. 5.

[70] Referring to Fig. 5, an ink jet apparatus for manufacturing an organic electroluminescent device may include a second connection unit 510, an output unit 520, and nozzles 545 and 550. The ink jet apparatus for manufacturing an organic electroluminescent device of the present invention is not limited to the embodiment shown in Fig. 5. Unlike Fig. 5, the phase-change material may be implemented to be transmitted and injected in a direction other than the vertical direction. On the other hand, the nozzles 545 and 550 may be defined as being part of the output unit.

[71] The phase-change material 505 transferred from the melting unit is transferred to the output unit 520 through the second connection unit 510. The second connection unit 510 may include a valve 515 capable of opening and closing.

[72] The output unit 520 includes a heater 530 for maintaining the temperature of the molten phase-change material so that the molten phase-change material does not become a solid, a bottom surface 535 for storing the phase-change material until the phase-change material is discharged through the nozzle, and a first injection port 525

for injecting gas. The carrier gas injected through the first injection unit 525 can increase the pressure in the output unit 520 so that the phase-change material can be sprayed through the nozzle. In addition, the carrier gas may be blended with the phase-change material and sprayed with the phase-change material through the nozzles 545 and 550 in a state that the carrier gas is not dissolved in the phase-change material.

[73] The nozzles may include a nozzle upper end 545 having a relatively narrow passage and a nozzle lower end 550 having a relatively wide passage. In Fig. 5, the nozzles are divided into two sections according to the area of the cross section, but the present invention is not limited thereto. For example, the cross section of each nozzle may be the same in all sections or may have three or more cross sectional areas.

[74] According to an embodiment, the nozzles 545 and 550 may further include a second injection port 540 for injecting the induction gas 555. The induction gas 555 injected through the second injection port 540 may be sprayed together with the phase-change material 560 in a liquid state.

[75] Meanwhile, an ink jet apparatus according to an embodiment of the present invention may include a plurality kinds of nozzles. In this case, the widths of the cross sections of the respective nozzles may be equal to or different from each other. For example, the ink jet apparatus may include a first nozzle having a relatively large cross section and located in a middle portion, and a plurality of second nozzles having a relatively narrow cross section and located in a peripheral portion. According to another embodiment of the present invention, part of the first nozzle or the plurality of second nozzles may be configured not to spray the induction gas.

[76] In Fig. 5, the spray angle of the induction gas 555 and the spray angle of the phase-change material 560 is the same, but spray angles of them may be configured as different from each other. For example, while the phase-change material 560 is being injected perpendicular to the substrate, the induction gas 555 may be sprayed at an angle of 15 degrees to the direction of the phase-change material 560, respectively.

[77] In Fig. 5, the cross section of the nozzle is circular, and the induction gas 555 is sprayed in a form surrounding the phase-change material 560. According to another embodiment of the present invention, the cross section of the nozzle may have the shape of an ellipse or a polygon. According to another embodiment of the present invention, the induction gas 555 may be sprayed while in contact with the phase-change material 560 only partially. For example, in a nozzle having a rectangular cross section, the induction gas 555 may be injected while contacting the phase-change material only at each vertex of the quadrangle. Alternatively, in the nozzle having the elliptical cross section, the induction gas 555 may be injected while being in contact with the phase-change material only in a region where the curvature value of the ellipse is within a predetermined range.

- [78] In Fig. 5, the carrier gas introduced through the first injection port 525 may be used to apply pressure to inject the phase-change material through the nozzle, and the induction gas introduced through the second injection port 540 may be a phase-change material and can be used for finely coating the substrate.
- [79] According to one embodiment of the present invention a method of manufacturing an organic electroluminescent device and a phase-change material used in an ink jet apparatus using the same is provided. The phase-change material comprises at least one organic electroluminescent compound. The phase-change material may be composed of, for example, one organic electroluminescent compound, or may be a compound in which two or more organic electroluminescent compounds are physically simply mixed without causing a chemical reaction.
- [80] The phase-change material may include a compound ordinarily included in the organic electroluminescent device, and may include a material that does not affect the melting characteristics of the phase-change material. The phase-change material may also include a substance that returns to its original properties at the time of film formation depending on the melting state even if it has changed. According to one embodiment of the present invention, the melting point of the phase-change material can be lowered by mixing an organic electroluminescent compound with another specific material, or by blending specific organic electroluminescent compounds to form a phase-change material.
- [81] According to one embodiment of the present invention, in a solvent-free environment, the solid state phase-change material in the melting unit of the ink jet device for manufacturing an organic electroluminescence device can be changed into a liquid state due to temperature and/or pressure. According to one embodiment of the invention, the melted phase-change material can be maintained in the melted state by being continuously heated in the melting unit until it is discharged by the nozzle.
- [82] According to an embodiment of the present invention, the injected phase-change material may be included in an organic electroluminescent device, and the organic electroluminescent device includes at least one of a light-emitting layer, a hole injection layer, a hole transport layer, a hole assisting layer, light-emitting assisting layer, an electron transport layer, an electron buffer layer, an electron injection layer, an interface layer, a hole blocking layer, and an electron blocking layer, but is not limited thereto.
- [83] According to one embodiment of the present invention, the melting point of the phase-change material can be lowered by mixing the organic electroluminescent compound with another specific material or blending specific organic electroluminescent compounds to form a phase-change material. As a result, it is possible to provide an organic electroluminescent compound which maintains the molten state for

a long time and does not change the material properties and does not crystallize during the manufacturing process. Specifically, the melting point of the phase-change material comprising at least one kind of organic electroluminescent compound is lower than a melting point of any one of the at least one kind of organic electroluminescent compound. The melting point of the phase-change material comprising two or more kinds of organic electroluminescent compounds may be lower than the arithmetic mean value of the melting point of each of the two or more kinds of organic electroluminescent compounds. According to one embodiment of the present application, by melting the phase-change material of the present invention at a relatively low temperature, it is possible to prevent the damage of the organic electroluminescent compound contained in the phase-change material and to reduce costs and/or time by energy savings.

- [84] According to one embodiment of the present invention, the organic electroluminescent compound used in the ink jet apparatus and/or the method of manufacturing an organic electroluminescent device of the present invention may be at least one high molecular weight compound and/or at least one low molecular weight compound, preferably, may be a low molecular weight compound. In addition, the organic electroluminescent compound may be phase-changed by itself regardless of whether the organic electroluminescent compound is included in the phase-change material. According to one embodiment of the present invention, the melting point of the phase-change material can be lowered by mixing an organic electroluminescent compound with another specific material or by blending specific organic electroluminescent compounds to form the phase-change material.
- [85] The organic electroluminescent compound may be an alkyl-based compound, a cycloalkyl-based compound, an aryl-based compound, a heteroaryl-based compound, an amine-based compound, a metal complex, or a combination thereof, but the present disclosure is not limited thereto.
- [86] Herein, the term "(C1-C30)alkyl" is meant to be a linear or branched alkyl having 1 to 30 carbon atoms constituting the chain, in which the number of carbon atoms is preferably 1 to 20, and more preferably 1 to 10. The above alkyl may include methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl, etc. The term "(C3-C30)cycloalkyl" is a mono- or polycyclic hydrocarbon having 3 to 30 ring backbone carbon atoms, in which the number of carbon atoms is preferably 3 to 20, and more preferably 3 to 7. The above cycloalkyl may include cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, etc. The term "(3- to 7- membered) heterocycloalkyl" is a cycloalkyl having 3 to 7, preferably 5 to 7, ring backbone atoms, and including at least one heteroatom selected from the group consisting of B, N, O, S, Si, and P, and preferably the group consisting of O, S, and

N. The above heterocycloalkyl may include tetrahydrofuran, pyrrolidine, thiolan, tetrahydropyran, etc. The term "(C6-C30)aryl(ene)" is a monocyclic or fused ring radical derived from an aromatic hydrocarbon having 6 to 30 ring backbone carbon atoms, in which the number of the ring backbone carbon atoms is preferably 6 to 25, more preferably 6 to 18. The above aryl(ene) may be partially saturated, and may comprise a spiro structure. The above aryl may include phenyl, biphenyl, terphenyl, naphthyl, binaphthyl, phenylnaphthyl, naphthylphenyl, fluorenyl, phenylfluorenyl, benzofluorenyl, dibenzofluorenyl, phenanthrenyl, phenylphenanthrenyl, anthracenyl, indenyl, triphenylenyl, pyrenyl, tetracenyl, perylenyl, chrysenyl, naphthacenyl, fluo-ranthenyl, spirobifluorenyl, etc. The term "(5- to 30-membered)heteroaryl(ene)" is an aryl having 5 to 30 ring backbone atoms, and including at least one, preferably 1 to 4 heteroatoms selected from the group consisting of B, N, O, S, Si, and P. The above heteroaryl(ene) may be a monocyclic ring, or a fused ring condensed with at least one benzene ring; may be partially saturated; may be one formed by linking at least one heteroaryl or aryl group to a heteroaryl group via a single bond(s); and may comprise a spiro structure. The above heteroaryl may include a monocyclic ring-type heteroaryl such as furyl, thiophenyl, pyrrolyl, imidazolyl, pyrazolyl, thiazolyl, thiadiazolyl, isothiazolyl, isoxazolyl, oxazolyl, oxadiazolyl, triazinyl, tetrazinyl, triazolyl, tetrazolyl, furazanyl, pyridyl, pyrazinyl, pyrimidinyl, and pyridazinyl, and a fused ring-type heteroaryl such as benzofuranyl, benzothiophenyl, isobenzofuranyl, dibenzofuranyl, dibenzothiophenyl, benzimidazolyl, benzothiazolyl, benzoisothiazolyl, benzoisoxazolyl, benzoxazolyl, isoindolyl, indolyl, benzoindolyl, indazolyl, benzothiadiazolyl, quinolyl, isoquinolyl, cinnolyl, quinazolinyl, quinoxalinyl, naphthyridinyl, carbazolyl, benzocarbazolyl, dibenzocarbazolyl, phenoxazinyl, phenothiazinyl, phenanthridinyl, benzodioxolyl, and dihydroacridinyl.

[87] The phosphorescent dopant material applied to the organic electroluminescent device of the present disclosure is not particularly limited, but may be preferably selected from the metallated complex compounds of iridium (Ir), osmium (Os), copper (Cu), and platinum (Pt), more preferably selected from ortho-metallated complex compounds of iridium (Ir), osmium (Os), copper (Cu), and platinum (Pt), and even more preferably ortho-metallated iridium complex compounds.

[88] Although various examples in present disclosure describe various layers as comprising a single material, a combination of materials such as a mixture of host and dopant, or more generally a mixture, may be used. In addition, the layer may have various sublayers. In addition, the OLED can be described as having "an organic layer" disposed between a cathode and an anode. The organic layer may be composed of a single layer or a plurality of layers made of different organic materials.

[89] Fig. 6 schematically shows a structure of an organic electroluminescent device manu-

factured according to an embodiment of the present disclosure. Hereinafter, referring to Fig. 6, the structure of an organic electroluminescent device and a method for preparing it, will be described in detail.

[90] Fig. 6 shows an organic electroluminescent device 600 comprising a substrate 610, a first electrode 620 formed on the substrate 610, an organic layer 630 formed on the first electrode 620, and a second electrode 640 formed on the organic layer 630 and facing the first electrode 620.

[91] The organic layer 630 comprises a hole injection layer 631, a hole transport layer 632 formed on the hole injection layer 631, a light-emitting layer 633 formed on the hole transport layer 632, an electron buffer layer 634 formed on the light-emitting layer 633, and an electron transport zone 637 formed on the electron buffer layer 634; and the electron transport zone 637 comprises an electron transport layer 635 formed on the electron buffer layer 634, and an electron injection layer 636 formed on the electron transport layer 635.

Claims

- [Claim 1] An ink jet device for manufacturing an organic electroluminescent device, comprising:
a melting unit for phase-changing a solid state phase-change material including an organic electroluminescent compound into a liquid state;
and
an output unit for spraying the phase-change material in a liquid state together with an induction gas.
- [Claim 2] The ink jet device according to claim 1,
wherein the output unit sprays the induction gas in a form surrounding the phase-change material in a liquid state.
- [Claim 3] The ink jet device according to claim 1,
wherein the output unit starts the spraying of the phase-change material in a liquid state after starting the spraying of the induction gas.
- [Claim 4] The ink jet device according to claim 1,
wherein the induction gas comprises at least one of nitrogen or argon.
- [Claim 5] The ink jet device according to claim 1,
wherein the phase-change material comprises at least one kind of organic electroluminescent compound, and
wherein the melting unit includes a heater for phase-changing the phase-change material to a liquid state by applying heat corresponding to a temperature equal to or lower than a melting point of any one of the organic electroluminescent compound of the at least one kind of organic electroluminescent compound.
- [Claim 6] A method for manufacturing an organic electroluminescent device, comprising steps of:
phase-changing a solid state phase-change material including an organic electroluminescent compound into a liquid state; and
spraying the phase-change material in a liquid state together with an induction gas.
- [Claim 7] The method according to claim 6,
wherein the step of spraying the phase-change material in a liquid state together with the induction gas comprises: spraying the induction gas in a form surrounding the phase-change material in a liquid state.
- [Claim 8] The method according to claim 6,
wherein the step of spraying the phase-change material in a liquid state together with the induction gas comprises: starting the spraying of the

phase-change material in a liquid state after starting the spraying of the induction gas.

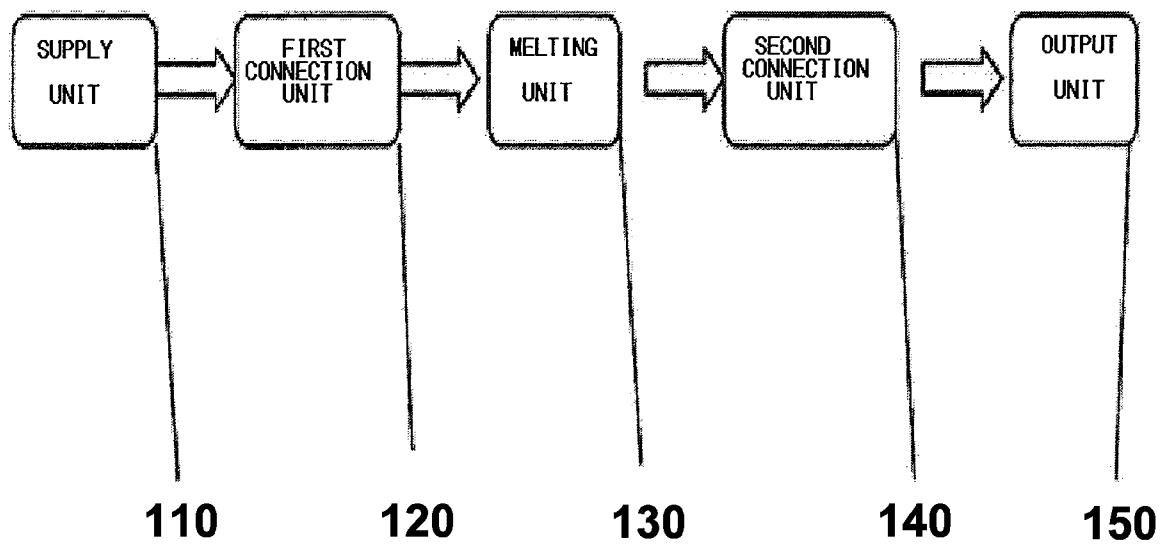
[Claim 9]

The method according to claim 6,
wherein the induction gas comprises at least one of nitrogen or argon.

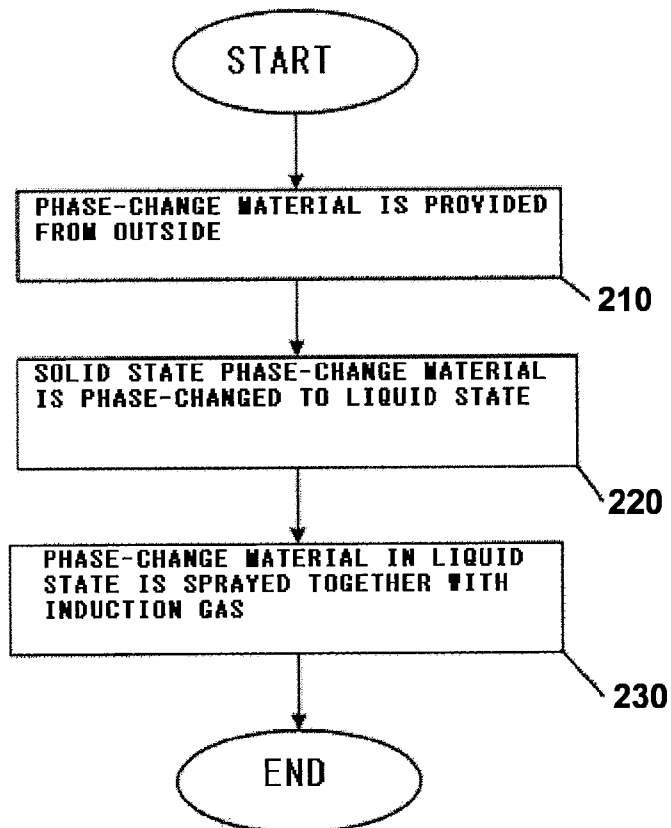
[Claim 10]

The method according to claim 6,
wherein the phase-change material comprises at least one kind of organic electroluminescent compound, and
wherein the step of phase-changing a phase-change material in a solid state including an organic electroluminescent compound into a liquid state comprises: phase-changing the phase-change material to a liquid state by applying heat corresponding to a temperature equal to or lower than a melting point of any one of the organic electroluminescent compound of the at least one kind of organic electroluminescent compound.

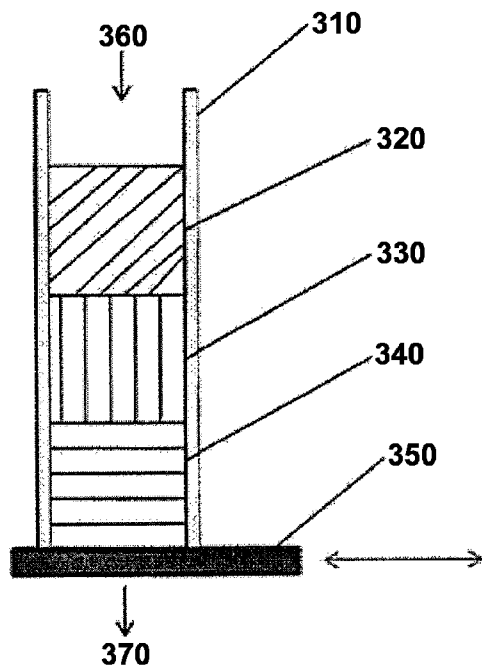
[Fig. 1]



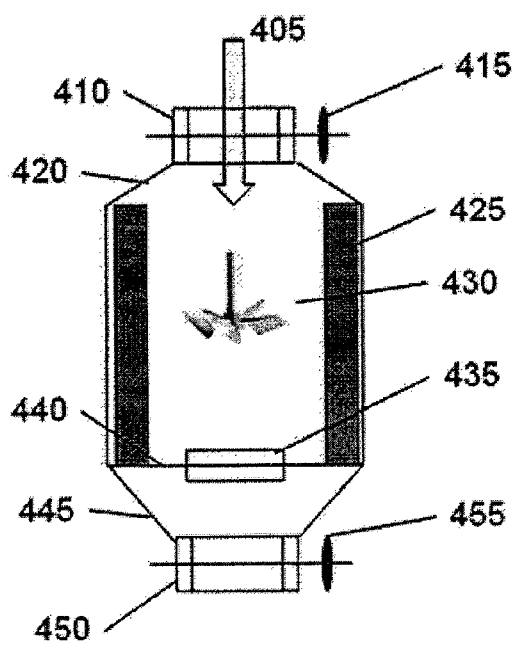
[Fig. 2]



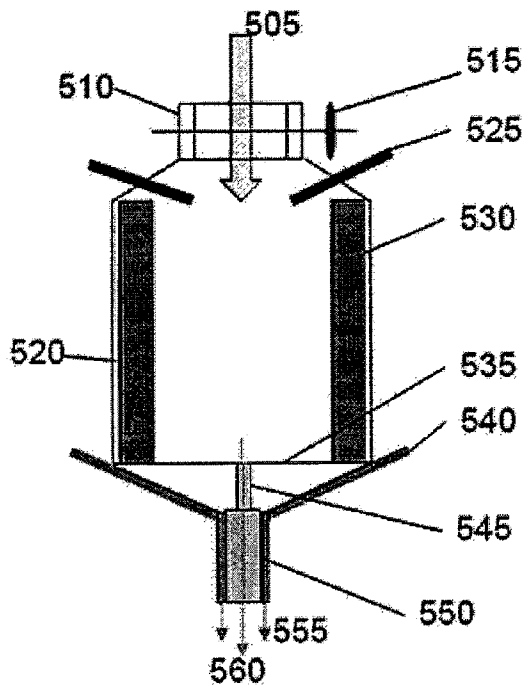
[Fig. 3]



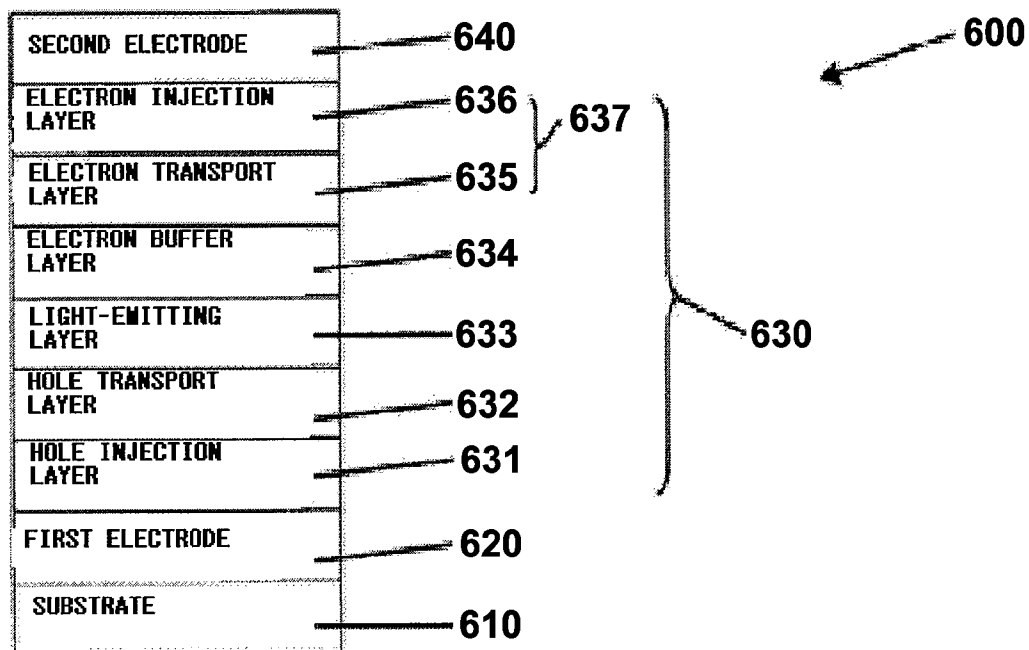
[Fig. 4]



[Fig. 5]



[Fig. 6]



INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2018/000314

A. CLASSIFICATION OF SUBJECT MATTER

H01L 51/56 (2006.01) H01L 51/00 (2006.01) H01L 51/50 (2006.01)

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)

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)

PATENW: IPC/CPC H01L51, H01L51/0002, H01L51/0005 and Keywords(oled, inkjet, phase, change, material, transition, vapour, electroluminescent, induction gas, carrier gas) and like terms; Applicant/Inventor search
Google patents/scholar: oled, organic, electroluminescent, phase change, melt, induction gas, carrier gas, spray and like terms.
Applicants/Inventors names searched in internal databases provided by IP Australia

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
	Documents are listed in the continuation of Box C	

☒ Further documents are listed in the continuation of Box C

☒ See patent family annex

* "A"	Special categories of cited documents: document defining the general state of the art which is not considered to be of particular relevance	"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
"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
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"O"	document referring to an oral disclosure, use, exhibition or other means	"&"	document member of the same patent family
"P"	document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search
29 March 2018

Date of mailing of the international search report
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INTERNATIONAL SEARCH REPORT		International application No.
C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		PCT/KR2018/000314
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2005/0009231 A1 (KARG et al.) 13 January 2005 Abstract; Paragraphs 20, 21, 24-26; Figures	1-10
A	US 7431968 B1 (SHTEIN et al.) 07 October 2008 Abstract; Figures; Columns 1-6	
A	WO 2012/175126 A1 (AIXTRON SE) 27 December 2012 Abstract; Figures	

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

INTERNATIONAL SEARCH REPORT		International application No.	
Information on patent family members		PCT/KR2018/000314	
This Annex lists known patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.			
Patent Document/s Cited in Search Report		Patent Family Member/s	
Publication Number	Publication Date	Publication Number	Publication Date
US 2005/0009231 A1	13 January 2005	US 2005009231 A1	13 Jan 2005
		US 7282430 B2	16 Oct 2007
		CN 1574214 A	02 Feb 2005
		CN 1771614 A	10 May 2006
		JP 2004363104 A	24 Dec 2004
		KR 20040104368 A	10 Dec 2004
		KR 100613955 B1	18 Aug 2006
		KR 20060018830 A	02 Mar 2006
		KR 100773014 B1	05 Nov 2007
		TW 200509737 A	01 Mar 2005
		TW I285514 B	11 Aug 2007
		TW 200511622 A	16 Mar 2005
		TW I306678 B	21 Feb 2009
		US 2008220561 A1	11 Sep 2008
		WO 2004107472 A1	09 Dec 2004
US 7431968 B1	07 October 2008	US 2008233287 A1	25 Sep 2008
		US 7431968 B1	07 Oct 2008
		CN 1568377 A	19 Jan 2005
		CN 1883060 A	20 Dec 2006
		CN 1883060 B	15 Sep 2010
		EP 1423552 A1	02 Jun 2004
		EP 1423552 B1	21 Oct 2015
		EP 1676331 A1	05 Jul 2006
		EP 1676331 B1	27 Aug 2008
		EP 1978570 A1	08 Oct 2008
		EP 1978570 B1	06 Dec 2017
		EP 2275587 A1	19 Jan 2011
		EP 3144988 A1	22 Mar 2017
		JP 2007520854 A	26 Jul 2007
		JP 5000302 B2	15 Aug 2012
		JP 2005502779 A	27 Jan 2005
		JP 5052737 B2	17 Oct 2012
		JP 2012043799 A	01 Mar 2012
		KR 20040044534 A	28 May 2004
		KR 100998059 B1	03 Dec 2010
Due to data integration issues this family listing may not include 10 digit Australian applications filed since May 2001.			
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INTERNATIONAL SEARCH REPORT		International application No.	
Information on patent family members		PCT/KR2018/000314	
This Annex lists known patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.			
Patent Document/s Cited in Search Report		Patent Family Member/s	
Publication Number	Publication Date	Publication Number	Publication Date
		KR 20060108681 A	18 Oct 2006
		KR 101128144 B1	26 Mar 2012
		TW I223840 B	11 Nov 2004
		TW I284155 B	21 Jul 2007
		TW 200526793 A	16 Aug 2005
		TW I363805 B	11 May 2012
		US 2003087471 A1	08 May 2003
		US 6716656 B2	06 Apr 2004
		US 2004048000 A1	11 Mar 2004
		US 7404862 B2	29 Jul 2008
		US 2008299311 A1	04 Dec 2008
		US 7682660 B2	23 Mar 2010
		US 2008311296 A1	18 Dec 2008
		US 7722927 B2	25 May 2010
		US 2005087131 A1	28 Apr 2005
		US 7744957 B2	29 Jun 2010
		US 2011027481 A1	03 Feb 2011
		US 7897210 B2	01 Mar 2011
		US 2011045196 A1	24 Feb 2011
		US 8535759 B2	17 Sep 2013
		US 2013337173 A1	19 Dec 2013
		WO 03020999 A1	13 Mar 2003
		WO 03034471 A1	24 Apr 2003
		WO 2005043641 A1	12 May 2005
WO 2012/175126 A1	27 December 2012	WO 2012175126 A1	27 Dec 2012
		CN 103930588 A	16 Jul 2014
		CN 103930588 B	17 Aug 2016
		EP 2723914 A1	30 Apr 2014
		JP 2014522909 A	08 Sep 2014
		JP 5877245 B2	02 Mar 2016
		KR 20140041794 A	04 Apr 2014
		KR 101711502 B1	02 Mar 2017
		TW 201311924 A	16 Mar 2013
		TW I560305 B	01 Dec 2016
End of Annex			
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Patent Document/s Cited in Search Report		Patent Family Member/s	
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