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(71) Applicant (for all designated States except US): **EASTMAN KODAK COMPANY** [US/US]; 343 State Street, Rochester, NY 14650 (US).

(72) Inventors; and

(75) Inventors/Applicants (for US only): **LONG, Michael** [US/US]; 10 Black Tern Terrance, Hilton, NY 14468 (US). **GRACE, Jeremy, Matthew** [US/US]; 132 Hollybrook Drive, Penfield, NY 14526 (US). **KOPPE, Bruce, Edward** [US/US]; 702 Cooney Road, Calendonía, NY 14423 (US).

(74) Common Representative: **EASTMAN KODAK COMPANY**; 343 State Street, Rochester, NY 14650 (US).

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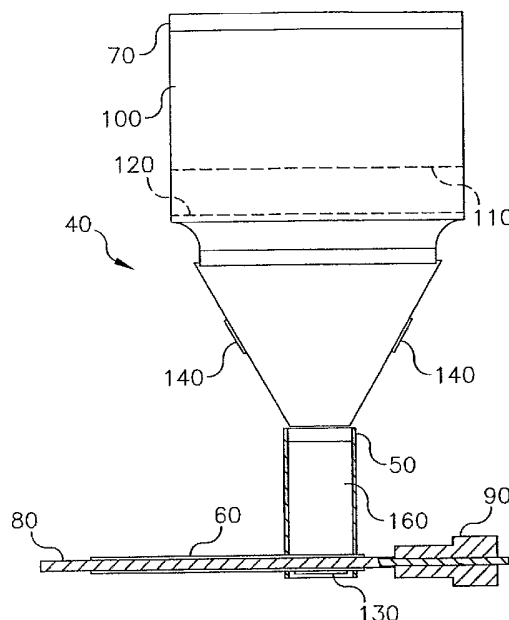
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(54) Title: DELIVERING PARTICULATE MATERIAL TO A VAPORIZATION SOURCE



(57) Abstract: A method for vaporizing particulate material and condensing it onto a surface to form a layer provides a quantity of first particulate material in a first container and a quantity of second particulate material in a second container spaced apart from the first container, the first and second containers respectively having first and second openings. The first particulate material is transferred through the first opening in the first container into a manifold and vaporized in the manifold. The second particulate material is transferred through the second opening in the second container into the manifold and vaporized in the manifold, whereby the first and second vaporized particulate materials are mixed. The mixed vaporized materials are delivered from the manifold to the surface to form the layer.

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**DELIVERING PARTICULATE MATERIAL**  
**TO A VAPORIZATION ZONE**

**FIELD OF THE INVENTION**

The present invention relates to the field of physical vapor  
5 deposition of particulate material.

**BACKGROUND OF THE INVENTION**

An OLED device includes a substrate, an anode, a hole-  
transporting layer made of an organic compound, an organic luminescent layer  
with suitable dopants, an organic electron-transporting layer, and a cathode.  
10 OLED devices are attractive because of their low driving voltage, high luminance,  
wide-angle viewing and capability for full-color flat emission displays. Tang et  
al. described this multilayer OLED device in their U.S. Patent Nos. 4,769,292 and  
4,885,211.

Physical vapor deposition in a vacuum environment is the principal  
15 means of depositing thin organic material films as used in small molecule OLED  
devices. Such methods are well known, for example Barr in U.S. 2,447,789 and  
Tanabe et al. in EP 0 982 411. The organic materials used in the manufacture of  
OLED devices are often subject to degradation when maintained at or near the  
desired rate dependant vaporization temperature for extended periods of time.  
20 Exposure of sensitive organic materials to higher temperatures can cause changes  
in the structure of the molecules and associated changes in material properties.

To overcome the thermal sensitivity of these materials, only small  
quantities of organic materials have been loaded in sources and they are heated as  
little as possible. In this manner, the material is consumed before it has reached  
25 the temperature exposure threshold to cause significant degradation. The  
limitations with this practice are that the available vaporization rate is very low  
due to the limitation on heater temperature, and the operation time of the source is  
very short due to the small quantity of material present in the source. In the prior  
art, it has been necessary to vent the deposition chamber, disassemble and clean  
30 the vapor source, refill the source, reestablish vacuum in the deposition chamber  
and degas the just-introduced organic material over several hours before resuming  
operation. The low deposition rate and the frequent and time consuming process

associated with recharging a source has placed substantial limitations on the throughput of OLED manufacturing facilities.

A secondary consequence of heating the entire organic material charge to roughly the same temperature is that it is impractical to mix additional organic materials, such as dopants, with a host material unless the vaporization behavior and vapor pressure of the dopant is very close to that of the host material. This is generally not the case and as a result, prior art devices frequently require the use of separate sources to co-deposit host and dopant materials.

A consequence of using single component sources is that many sources are required in order to produce films containing a host and multiple dopants. These sources are arrayed one next to the other with the outer sources angled toward the center to approximate a co-deposition condition. In practice, the number of linear sources used to co-deposit different materials has been limited to three. This restriction has imposed a substantial limitation on the architecture of OLED devices, increases the necessary size and cost of the vacuum deposition chamber and decreases the reliability of the system.

Additionally, the use of separate sources creates a gradient effect in the deposited film where the material in the source closest to the advancing substrate is over represented in the initial film immediately adjacent the substrate while the material in the last source is over represented in the final film surface. This gradient co-deposition is unavoidable in prior art sources where a single material is vaporized from each of multiple sources. The gradient in the deposited film is especially evident when the contribution of either of the end sources is more than a few percent of the central source, such as when a co-host is used. FIG. 1 shows a cross-sectional view of such a prior-art vaporization device 5, which includes three individual sources 6, 7, and 8 for vaporizing organic material. Vapor plume 9 is preferably homogeneous in the materials from the different sources, but in fact varies in composition from side to side resulting in a non-homogeneous coating on substrate 15.

A further limitation of prior art sources is that the geometry of the vapor manifold changes as the organic material charge is consumed. This change requires that the heater temperature change to maintain a constant vaporization

rate and it is observed that the overall plume shape of the vapor exiting the orifices can change as a function of the organic material thickness and distribution in the source, particularly when the conductance to vapor flow in the source with a full charge of material is low enough to sustain pressure gradients from non-uniform vaporization within the source. In this case, as the material charge is consumed, the conductance increases and the pressure distribution and hence overall plume shape improve.

### SUMMARY OF THE INVENTION

It is an object of the present invention to provide an effective way of transferring particulate material from a container to a vaporization zone.

This object is achieved by 1. A method for vaporizing particulate material and condensing it onto a surface to form a layer, comprising:

- (a) providing a quantity of first particulate material in a first container and a quantity of second particulate material in a second container spaced apart from the first container, the first and second containers respectively having first and second openings;
- (b) transferring the first particulate material through the first opening in the first container into a manifold and vaporizing the first particulate material in the manifold;
- (c) transferring the second particulate material through the second opening in the second container into the manifold and vaporizing the second particulate material in the manifold, whereby the first and second vaporized particulate materials are mixed; and
- (d) delivering the mixed vaporized materials from the manifold to the surface to form the layer.

It is an advantage of the present invention that the continuous heating of material during operation of prior art devices is eliminated in that only a small portion of particulate material is heated, for a short period of time and at a controlled rate. The bulk of particulate material is maintained at a temperature that can be as much as 300°C cooler than the desired rate-dependant vaporization temperature. This can be particularly advantageous when vaporizing organic materials.

It is a further advantage of the present invention that it can maintain a steady vaporization rate with a continuously replenished charge of particulate material and with a steady heater temperature. The device thus allows extended operation of the source with substantially reduced risk of degrading even  
5 very temperature-sensitive organic materials.

It is a further advantage of the present invention that it permits materials having different vaporization rates and degradation temperature thresholds to be co-sublimated in the same source.

It is a further advantage of the present invention that it permits  
10 linear vaporization rate control by controlling the volumetric metering rate or controlling the feed pressure of the compacted particulate material.

It is a further advantage of the present invention that it can rapidly stop and reinitiate vaporization and achieve a steady vaporization rate quickly by controlling the metering rate of the particulate material, minimizing contamination  
15 of the deposition chamber walls and conserving the particulate materials when a substrate is not being coated.

It is a further advantage that the present device achieves substantially higher vaporization rates than in prior art devices with substantially reduced material degradation. Further still, no heater temperature change is  
20 required as the source material is consumed.

It is a further advantage of the present invention that it can provide a vapor source in any orientation, which is frequently not possible with prior-art devices.

It is a further advantage of some embodiments of this invention  
25 that it can remove adsorbed gases from the particulate material through the use of heat and vacuum as a much smaller quantity of particulate material is conveyed through the device.

It is a further advantage of some embodiments of this invention that it can permit a temporal gradation in concentration of one or more of the  
30 particulate material components by varying the feed rate of one or more of the components relative to the other material components.

### BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a cross-sectional view of a prior-art vaporization device;

FIG. 2 is a three-dimensional view of one embodiment of an  
5 apparatus according to the present invention for vaporizing particulate materials  
and condensing them onto a surface to form a layer;

FIG. 3 is a cross-sectional view of one embodiment of a portion of  
the above apparatus for feeding particulate material according to the present  
invention, including one embodiment of an agitating device useful in the present  
10 invention;

FIG. 4 is a cross-sectional view of one embodiment of a portion of  
the above apparatus for feeding and vaporizing particulate material according to  
the present invention;

FIG. 5 shows a graphical representation of vapor pressure vs.  
15 temperature for two organic particulate materials;

FIG. 6a is a cross-sectional view showing one embodiment of an  
auger structure useful in this invention;

FIG. 6b is a cross-sectional view of the terminal end of the auger  
structure in FIG. 6a;

FIG. 6c is a relief view showing another embodiment of an auger  
20 structure useful in this invention;

FIG. 6d is a cross-sectional view showing another embodiment of  
an auger structure useful in this invention;

FIG. 7 is a cutaway view of another embodiment of an agitating  
25 device useful in the present invention;

FIG. 8 is a cutaway view of another embodiment of an agitating  
device useful in the present invention;

FIG. 9 is a cross-sectional view of a portion of another embodiment  
of an apparatus according to the present invention for vaporizing particulate  
30 materials and condensing them onto a surface to form a layer;

FIG. 10 is a cross-sectional view of a device according to the  
present invention including a deposition chamber enclosing a substrate; and

FIG. 11 is a cross-sectional view of an OLED device structure that can be prepared with the present invention.

### DETAILED DESCRIPTION OF THE INVENTION

Turning now to FIG. 2, there is shown a three-dimensional view of one embodiment of an apparatus according to the present invention for vaporizing particulate materials and condensing them onto a surface to form a layer. Vaporization apparatus 10 includes manifold 20, attached feeding apparatus 40, and attached feeding apparatus 45. Feeding apparatus 40 includes at least first container 50 and feeding path 60. Feeding apparatus 45 includes at least second container 55 spaced from first container 50 and feeding path 65. First container 50 is provided with a quantity of a first particulate material, such as a powder in one embodiment. Second container 55 is provided with a quantity of second particulate material. Feeding apparatus 40 can also include third container 70, and feeding apparatus 45 can also include fourth container 75. Third container 70 is associated with first container 50 and the first particulate material. Fourth container 75 is associated with second container 55 and the second particulate material. Third container 70 and fourth container 75 can receive first and second particulate materials, respectively, and transfer them to first container 50 and second container 55, respectively, as will become apparent. Manifold 20 includes one or more apertures 30 through which vaporized particulate material can exit to a substrate surface. Manifold 20 is shown in an orientation whereby it can form a layer on a horizontally-oriented substrate, but it is not limited to this orientation. Manifold 20 can be oriented vertically and can form a layer on a vertical substrate. Manifold 20 had been described in detail by Long et al. in commonly-assigned, above-cited U.S. Patent Application No. 10/784,585. Feeding apparatus 40 and feeding apparatus 45 are shown attached to opposite sides of manifold 20, but they can also be attached to the same side of manifold 20, or to the bottom of manifold 20 if the manifold is in a vertical orientation. The nature of the attachment of feeding apparatus 40 to manifold 20 will become clear.

Turning now to FIG. 3, there is shown a cross-sectional view of one embodiment of a portion of the above vaporization apparatus for feeding particulate material according to the present invention whereby particulate



material is effectively fluidized and transferred to the auger structure. First container 50 holds first particulate material 160, which can be in the form of a finely divided powder and is desirably of a uniform size, and which feeds into auger structure 80 in feeding path 60. Auger structure 80 passes through the interior of first container 50 and feeds into the manifold described above (not shown for clarity). At least a portion of auger structure 80 is rotated by motor 90 so as to transfer the first particulate material at a controlled volumetric rate or pressure along feeding path 60 to a vaporization zone where the component material is vaporized and subsequently delivered to a substrate to form a layer. Feeding path 60, and therefore first particulate material in feeding path 60, can be maintained at a temperature below the desired vaporization temperature of the component material. To facilitate the movement of first particulate material 160 to auger structure 80, first particulate material 160 is fluidized by agitating first particulate material 160 by using an agitating device, e.g. piezoelectric structure 130 or an electromechanical vibrator. Such fluidized material is more readily transferred to auger structure 80 by gravity feed.

The addition of optional third container 70 to hold additional first particulate material 100 provides several additional advantages. A large quantity of first particulate material 100 can be charged in the apparatus, allowing continuous operation of the device for extended periods of time. By sensing the quantity of particulate material in first container 50, e.g. by measuring the height of the column of first particulate material 160, one can selectively meter the amount of first particulate material transferred from second container 70 to first container 50 and provide a substantially constant volume of first particulate material 160 in first container 50, e.g.  $\pm 5 \text{ cm}^3$ . In practice,  $10 \text{ cm}^3$  of particulate material is loaded in first container 50. Some embodiments described herein have great process latitude with respect to reliable particulate material feeding over a wide range of particulate material height in the container and can be run nearly to exhaustion without failing to feed particulate material. However, it is believed that multi-component mixing homogeneity is fostered if an optimum powder height is established and maintained in first container 50 to within  $\pm 10\%$ . This minimizes variations in the feeding rate of first particulate material 160 to feeding

path 60. Also, third container 70 can be arranged to be refillable without affecting the operation of first container 50, allowing the device to be continuously operated for even longer periods of time. First particulate material 100 is maintained in third container 70 by e.g. screens 110 and 120, whose mesh size is chosen to prevent the free flow of particulate material. Screens 110 and 120 can also be the mechanism for providing measured quantities of first particulate material 100 to move from third container 70 to first container 50. Screens 110 and 120 may be contacted by agitating devices (not shown) that can be actuated to cause a quantity of particulate material 100 to pass through the screen mesh. Such devices include those to vibrate the screen, or a movable arm immediately above or below the screen to allow selective agitation of screens 110 and 120. A commercial flour sifter is one such device well adapted for use in this application. In these sifters, three screens are used and the top surface of each screen is contacted by rotatable arms that extend radially from the center of the sifter. The arms have a V shaped cross section so as to force the powdered particulate material 100 into a converging space between the arm and the screen as the arm rotates to thereby force a controlled volume of powder through the screen. A sensing system based on the height of first particulate material 160 in first container 50 (or on an integrated signal derived from the deposition rate and time of operation) can serve to actuate the devices agitating screens 110 and 120 so as to maintain a nearly constant volume of particulate material 160 in first container 50. Agitating devices such as piezoelectric structures 140 prevent the buildup of particulate material 100 in the feed path to first container 50. Piezoelectric structures can be vibrated with multiple frequencies, e.g. a siren effect, to prevent the buildup of particulate material 100 at vibrational nodes. Feeding apparatus 45 of FIG. 2 operates in an analogous manner with second container 55, fourth container 75, feeding path 65, and motor 35. Feeding apparatus 45 may also include an analogous separate agitating device for fluidizing the second particulate material by agitating it and thereby transferring it to an auger structure, an analogous means for measuring the height of the column of second particulate material in second container 55 and for metering the amount of second particulate material transferred from fourth container 75 to second container 55, and an analogous

separate second auger structure that passes through the interior of second container 55 to move the second particulate material down feeding path 65 and transfer it to manifold 20.

For proper operation of feeding apparatus 40 and 45, it is important to maintain a uniform feed rate of particulate material 160. Particulate material 160 is generally provided in a powdered form. One important strategy for providing a free flow of particulate material 160 is to prevent bridging, a characteristic behavior of particulate materials such as powders that can occur when the powder particles self-assemble into a load-bearing structure about an opening or aperture and thereby obstruct the flow of powder through the opening. Bridging effects can occur, for example, when the dimensions of an aperture are too small to overcome a tendency of a particulate material to resist flow. Factors that may cause bridging can include particulate size relative to the aperture dimensions, humidity, electrostatic attraction between particles, vacuum levels, and friction. To alleviate this problem, the dimensions of an opening 230 at the interface of first container 50 and feeding path 60, as shown in FIG. 3 for example, must be sufficiently sized to overcome the bridging characteristics of the powdered material. This sizing requirement is best determined empirically, taking into account worst-case conditions for the particular particulate material 160 that must be supplied, in a free flowing manner, to feeding path 60. Maintaining a nearly constant volume of particulate material 160 in first container 50 also helps to promote a constant feed rate of particulate material 160 to auger structure 80. By properly sizing opening 230 and maintaining a sufficient volume of particulate material 160 in first container 50, a uniform feed rate can be achieved for many types of powdered particulate material 160, providing a fluidized flow without requiring any supplemental form of agitation.

Where the opening 230 must be narrow, feed rate uniformity can be assured when the particulate material 160 in proximity to the infeed portion of the screw auger is maintained in a fluidized state by an agitating device. This can be accomplished by slowly agitating particulate material 160 immediately above the auger screw or by inducing vibration, e.g. by piezoelectric structure 130, into particulate material 160 that is tuned to induce liquid-like behavior of the

powdered particulate material 160 but is not so energetic as to cause gas-like behavior.

Turning now to FIG. 4, there is shown in further detail a cross-sectional view of one embodiment of a portion of the above apparatus for feeding and vaporizing particulate material 160 according to the present invention. Auger structure 80 transfers first particulate material 160 along feeding path 60 into manifold 20 and heating element 170. Heating element 170 can be e.g. a heated screen and has been previously described in detail by Long et al. Manifold 20 includes a vaporization zone which is defined as the region of feeding path 60 immediately adjacent to heating element 170. A thin cross-section of particulate material 160 is heated to the desired rate-dependent temperature, which is the temperature of heating element 170, by virtue of contact and thermal conduction, whereby the thin cross-section of first particulate material 160 vaporizes. Similarly, feeding apparatus 45 of FIG. 2 transfers the second particulate material 160 into manifold 20 at a second heating element, where the second particulate material 160 vaporizes in manifold 20. The first and second vaporized particulate materials 160 are mixed in manifold 20, and subsequently delivered to a substrate surface to form a layer. The auger structure 80 and its rotation rate control the rate at which particulate material 160 is fed to heating element 170. This linearly controls the rate of vaporization and therefore the rate at which particulate material 160 leaves the manifold in the vapor state. Thus the feed rate of particulate material 160 to the auger structure and to the vaporization zone controls the deposition rate of the vaporized component material onto the desired surface. With two such structures, as in FIG.1, the relative feed rates of the first and second particulate materials 160 to the respective auger structures and the respective vaporization zones controls the relative partial pressures of the first and second particulate materials 160 in the manifold, and hence their relative deposition rates and concentrations in the deposited layer. One useful example is wherein one of particulate materials 160 is a host material and the other is a dopant.

Additionally, base 180 can be included. Base 180 is a heat-dissipating structure to prevent much of the heat from heating element 170 from

traversing the length of feeding path 60, and thus keeps the bulk of particulate material 160 significantly cooler than the conditions it experiences in the vaporization zone immediately adjacent to heating element 170. Means of heat dissipation for base 180 have been described by Long et al. in commonly-  
5 assigned, above-cited U.S. Patent Application 10/784,585. A steep thermal gradient thereby created protects all but the immediately vaporizing material from the high temperatures. The vaporized component vapors rapidly pass through heating element 170 and can enter into the heated manifold 20. The residence time of particulate material 160 at the desired vaporization temperature is very  
10 short and as a result, thermal degradation is greatly reduced. The residence time of particulate material 160 at elevated temperature, that is, at the rate-dependent vaporization temperature, is orders of magnitude less than prior art devices and methods (seconds vs. hours or days in the prior art), which permits heating organic particulate materials 160 to higher temperatures than in the prior art.  
15 Thus, the current device and method can achieve substantially higher vaporization rates, without causing appreciable degradation of organic components of particulate material 160.

Particulate material 160 can include a single component, or can include two or more different vaporizable components, such as organic material  
20 components, each one having a different vaporization temperature. The vaporization temperature can be determined by various means. For example, FIG. 5 shows a graphical representation of vapor pressure versus temperature for two organic materials commonly used in OLED devices. The vaporization rate is proportional to the vapor pressure, so for a desired vaporization rate, the data in  
25 FIG. 5 can be used to define the required heating temperature corresponding to the desired vaporization rate. In the case where particulate material 160 includes two or more organic components, the temperature of heating element 170 is chosen such that the vaporization is feed-rate limited, that is, the vapor pressure at the heating element temperature is substantially above the desired partial pressure of  
30 that component in the manifold, so that each of the organic material components simultaneously vaporizes.

Pressure develops in manifold 20 as vaporization proceeds, and streams of vapor exit manifold 20 through the series of apertures 30 shown in FIG. 2. Because only a small portion of particulate material 160—the portion resident in the vaporization zone—is heated to the rate-dependent vaporization temperature, while the bulk of the material is kept well below the vaporization temperature, it is possible to interrupt the vaporization by a means for interrupting heating at heating element 170, e.g. stopping the movement of auger structure 80. This can be done when a substrate surface is not being coated so as to conserve particulate material 160 and minimize contamination of any associated apparatus, such as the walls of a deposition chamber, which will be described below.

Because heating element 170 can be a fine mesh screen that prevents powder or compacted material from passing freely through it, the manifold can be used in any orientation. For example, manifold 20 of FIG. 2 can be oriented down so as to coat a substrate placed below it. This is an advantage not found in the heating boats of the prior art.

Turning now to FIG. 6a, there is shown a cross-sectional view of one embodiment of an auger structure useful in this invention. The auger structure 80 includes an auger screw 85 that is turned by motor 90. The distance between the threads of the screw helix and the thread height are chosen to be sufficiently large that powder tends not to pack into and rotate with the helix, but rather to remain at the bottom of a horizontally oriented auger tube and be transported linearly by virtue of the relative motion between the screw and the auger tube. For example, an auger screw with a 2.5 mm pitch screw lead and a 0.8 mm thread height has been found to be an effective combination in transporting and consolidating organic material powders in a horizontal orientation.

The inventors have found that auger dimensions have an affect on maintaining a uniform flow rate. Similar to the bridging effects noted above with respect to the size of opening 230, proper auger sizing and screw thread pitch is best determined empirically, considering worst-case conditions for the particular composition of particulate material 160.

The inventors have also found that the angle of auger screw threads can be optimized to facilitate free flow of particulate material 160 along feeding path 60. While optimal screw thread angle may vary somewhat depending on the particular component materials of powdered particulate material 160, it has been  
5 determined that screw thread angles ranging from not less than about 4 degrees to no more than about 15 degrees relative to the rotational axis of auger structure 85 provide optimal flow conditions for particulate materials 160 that are conventionally used.

Various materials and surface treatments of the auger shaft have  
10 been found to facilitate auger operation, allowing increased feed rates. While stainless steel may provide acceptable performance, additional benefit may be obtained by surface treatments such as electropolishing or by coatings, such as a coating of titanium nitride.

While continuous auger rotation at a sustained rate may provide an  
15 acceptable level of performance, added benefits may be obtained by pulsing the auger, providing rotation of the auger shaft in a repeated incremental fashion. A pulsing action reduces the tendency for powdered particulate material 160 to rotate with the auger screw by reducing the effective coefficient of friction between the auger screw and the particulate material. The powder feeding  
20 efficiency of auger structure 85 is thereby improved. Pulsing behavior may also be advantageous where it becomes useful to vary the feed rate over an interval, for example.

In the horizontal orientation, particulate material 160 travels along the bottom of auger screw 85 in a tumbling and dispersed form. At the terminal  
25 end of auger screw 85, a powder pressure of 1 Mpa can be developed that increases the bulk density of particulate material 160 to the point where it serves as a vapor seal, preventing vaporized material in the manifold having a pressure greater than the ambient vacuum level from flowing back along the auger screw to the powder source container. As shown in FIG. 6b, the terminal end of auger  
30 screw 85 is configured to have a thread-free portion 135 having a constant circular cross section over a small length to constrain the consolidated powdered

particulate material 160 to form a narrow annular or tubular shape. This narrow annular shape substantially improves the thermal contact and temperature uniformity through particulate material 160, between the temperature-controlled auger screw 85 and the temperature-controlled feeding path 60. This configuration additionally assures good temperature uniformity of particulate material 160 at a given transverse cross section relative to a circular cross section and substantially increases the attainable temperature gradient in particulate material 160 between the auger structure and the heating element. The powdered particulate material 160 is extruded from the auger structure in a tubular shape and is sufficiently consolidated that it can maintain the tubular extruded form for at least several millimeters upon exiting the support of the auger tube. This solid form prevents pressurized vapor, resulting from organic material vaporization, from flowing back into the auger structure and enables the powdered particulate material 160 to bridge the short gap between the end of the temperature-controlled auger structure and the heating element.

Thermal modeling of a powder dispensing system having this annular configuration where the heating element is spaced 130  $\mu\text{m}$  from the end of the auger structure 85 indicates that an average axial thermal gradient of  $0.5^\circ\text{C}/\mu\text{m}$  can be achieved through that portion of particulate material 160 spanning heating element 170 and the terminal end of the auger structure when the temperature differential between the two is  $270^\circ\text{C}$ . There can therefore be a  $100^\circ\text{C}$  temperature drop through the first 200  $\mu\text{m}$  of consolidated powdered particulate material 160. This gradient prevents the usual leaching of more volatile constituents from bulk volumes of mixed-component organic materials and enables a single source to co-deposit multiple organic materials. This large gradient is further instrumental in maintaining particulate material 160 in a consolidated powder form at the exit of the auger tube even when organic component materials that liquefy before vaporizing are employed.

The auger structure 80 shown in FIG. 6a is effective at transporting particulate material 160 powders horizontally, but is not as effective in transporting particulate material 160 vertically, since the particulate tends to simply rotate with the screw and not advance along the length of the structure.



Turning now to FIG. 6c, there is a relief view of another embodiment of an auger structure 95 useful in this invention. In this embodiment, auger structure 95 includes two or more auger screws, e.g. auger screws 85a, 85b, and 85c, with identical interlaced helical threads. All of the auger screws 85a, 85b, and 85c rotate in the same direction. Particulate material 160 that is packed between the threads of one auger screw, e.g. 85a, will be removed as the material rotates into contact with the interlaced thread of the second rotating auger screw, e.g. 85b, because the facing portions of adjacent screws move in opposite directions. Auger structure 95 thus overcomes the orientation restrictions of the single-screw auger structure of FIG. 6a while retaining the ability to consolidate powdered particulate material 160 into a solid shape and form a vapor seal. The discharge portion of auger structure 95 would have an elongated cross-section that can extend across the entire length of the manifold so as to inject material substantially uniformly along its length.

Turning now to FIG. 6d, there is a cross-sectional view of another embodiment of an auger structure 105 useful in this invention. Auger structure 105 includes a rotating helical thread 115, a stationary center portion 125, and a stationary outer tube, which in this case is feeding path 60. In this embodiment, only a portion of auger structure 105—the portion comprising helical threads 115—rotates and is turned by motor 90. Powdered particulate material 160 feeding with circular cross section helical threads has been demonstrated. The thread consisted of a steel wire 0.7 mm diameter formed into a helix of 5 mm outside diameter and 2.5 mm pitch. Smooth wires of other materials such as titanium and stainless steel are also suitable. The wire can also have a non-circular cross section, with a rectangular cross section being particularly advantageous as it provides additional rigidity to prevent the helical thread from changing dimensions as it encounters torsional resistance while pushing the powdered particulate material 160. Stationary center portion 125, in cooperation with feeding path 60, substantially prevents all but a thin film of powdered particulate material 160 from rotating with the auger. Auger structure 105 does not rely on gravity to accumulate powdered particulate material 160 and will operate in any orientation. Auger structure 105 also consolidates the powdered

particulate material 160 into a thin annular shape that substantially improves the thermal contact between particulate material 160 and temperature-controlled feeding path 60 and stationary center portion 125. These characteristics are significant in enabling the controlled vaporization of mixed component organic materials, and organic materials that liquefy before vaporizing. Thus this embodiment overcomes the orientation restrictions of the first auger structure while retaining the ability to consolidate the powdered particulate material 160 into a solid shape and form a vapor seal.

The above embodiments of this invention, based primarily on vaporization apparatus 10 of FIG. 2, are useful at atmospheric pressure and pressures down to about one-half atmosphere. Experimentally, it has been observed that fine powder is considerably more difficult to meter in a partial vacuum below half an atmosphere. The powdered particulate material 160 agglomerates as residual air molecules are removed, and undergoes a reduction of the elastic coupling between particles that is effective in communicating vibrational energy through powdered particulate material 160 under atmospheric conditions. This effect negatively influences the powder-feeding uniformity of the auger structure. Therefore, a different agitating device can be necessary. Turning now to FIG. 7, there is shown a cutaway view of another embodiment of an agitating device useful in the present invention for overcoming the limitations in low-pressure conditions. This embodiment employs three piezoelectric structures as the agitating device. Piezoelectric structures 150 and 155 are inclined at a steep angle and form opposite walls of a funnel at the bottom of first container 50. The bottom portion 190 of these two piezoelectric structures is not supported and leads directly to the infeed portion of auger structure 80. The unsupported portions of the piezoelectric structures have high vibration amplitude and are effective in fluidizing particulate material 160 in proximity to their surfaces. The third piezoelectric structure 130 is mounted below auger structure 80 and imparts vibration whose amplitude is essentially perpendicular to the vibration of the other two piezoelectric structures. The piezoelectric structures are driven by a frequency sweeping circuit. The changing frequency is instrumental in preventing the formation of nodes and improves the powder feeding efficiency

considerably. Auger structure 80 can be any of the above-described auger structures.

FIG. 8 is a cutaway view of another embodiment of an agitating device useful in the present invention for overcoming the limitations in low-  
5 pressure conditions. Opening 230 represents the lower end of the above-described first container 50. Rotating thread type device 210 includes left- and right-hand helically wound wires on a common shaft. Rotating thread type device 210 is positioned above the infeed portion of the auger structure such that the wires are substantially tangent to the threads of auger structure 80. The rotating thread  
10 should not interfere with the auger screw threads, but it will continue to operate effectively with as much as 1 mm clearance. Rotating thread type device 210 is slowly rotated via gear drive 220, by motor 90, which also turns auger structure 80. In practice, the rotational speed of the rotating thread type device 210 can vary depending on the particle size and properties of the particular particulate  
15 material 160, but a practical guide is to have the axial slew rate of the rotating thread match the axial slew rate of the threads of the auger screw. The wires of rotating thread type device 210 tend to push particulate material 160 toward the center of opening 230 and prevent powder bridging over auger structure 80. Auger structure 80 can be any of the above-described auger structures. This  
20 agitating device is well adapted to feeding mixed-component organic materials as it imparts very little energy to particulate material 160 and is therefore not likely to cause particle separation by size or density.

FIG. 9 is a cross-sectional view of another embodiment of an apparatus according to the present invention for vaporizing particulate materials  
25 160 and condensing them onto a surface to form a layer. In this embodiment, first container 50 and second container 55 are spaced apart, but positioned in such a way that the feeding paths (represented by first auger structure 250 and second auger structure 255, respectively) terminate in close proximity. First particulate material 240 in first container 50 is fluidized and then transferred by first auger  
30 structure 250 into mixing chamber 260 in manifold 20. Second particulate material 245 in second container 55 is fluidized and then transferred by second auger structure 255 into mixing chamber 260 in manifold 20, where it mixes with

first particulate material 240. The mixed first and second particulate materials 240 and 245 are vaporized by heating element 170, and can be delivered to a substrate surface by manifold 20. The relative feed rates of the first and second particulate material 240 and 245 to the respective auger structures and the  
5 respective vaporization zones controls the relative concentrations of materials in the deposited layer, as well as the deposition rate. Such an apparatus can enable a gradient in dopant concentration through the thickness of a deposited layer, or can create a smooth transition from one layer to the next by adjusting the concentration of the first particulate material 240 from 100 % to 0 % while the  
10 second host particulate material 245 concentration is simultaneously adjusted from 0 % to 100 %. Multiple auger screw systems can be repeated along the length of the source to independently feed different particulate materials so as to deposit a series of layers in this way.

In practice, the apparatus described herein is operated as follows.  
15 A first organic particulate material 160, which is useful in forming a layer on an OLED device, is provided into third container 70, and a second organic particulate material 160 is provided into fourth container 75. The first particulate material 160 is transferred in a controlled manner to first container 50 and the second particulate material 160 to second container 55 in such a way as to maintain a  
20 substantially constant volume of particulate materials in the first and second containers. Each particulate material 160 can be fluidized by means described herein and thereby transferred to a respective auger structure, which transfers the particulate materials 160 to one or more vaporization zones as described herein. At least one component of the particulate material 160 is vaporized in the  
25 vaporization zone(s) into a manifold 20, which delivers the vaporized material to the surface of an OLED substrate to form a layer, as will be described below.

As has been noted hereinabove, vacuum levels may tend to complicate the problem of metering out uniform amounts of finely powdered organic materials 160. Referring back to FIG. 2, it can be observed that a  
30 continuous column of particulate material 160 is maintained in feeding path 60. In one embodiment, this column of particulate material 160, if suitably compacted, can be utilized as a type of vacuum seal, where particulate

characteristics of particulate material 160 allow. With this arrangement, a high vacuum level can be present for particulate material 160 at heating element 170 and in the manifold 20. A lower vacuum level can then be maintained at first container 50, which may even be at atmospheric pressure. Even a partial seal  
5 could be advantageous. This sealing effect could also be used to isolate ambient gases used for storage of organic particulate material 160 in first container 50 and/or for organic particulate material 100 in second container 70. With some materials, for example, it is beneficial to store materials under an inert gas such as argon or helium.

10 Turning now to FIG. 10, there is shown an embodiment of a device of this disclosure including a deposition chamber enclosing a substrate. Deposition chamber 280 is an enclosed apparatus that permits an OLED substrate 285 to be coated with organic material transferred from manifold 20. Manifold 20 is supplied with organic material via feeding path 60 as described above. For  
15 clarity of illustration, only a single feeding path is shown. Deposition chamber 280 is held under controlled conditions, e.g. a pressure of 1 torr or less provided by vacuum source 300. Deposition chamber 280 includes load lock 275 which can be used to load uncoated OLED substrates 285, and unload coated OLED substrates. OLED substrate 285 can be moved by translational apparatus 295 to  
20 provide even coating of vaporized organic material over the entire surface of OLED substrate 285. Although vaporization apparatus is shown as partially enclosed by deposition chamber 280, it will be understood that other arrangements are possible, including arrangements wherein the entire vaporization apparatus, including any container or containers for holding powdered particulate material  
25 160, is enclosed by deposition chamber 280.

In practice, an OLED substrate 285 is placed in deposition chamber 280 via load lock 275 and held by translational apparatus 295 or associated apparatus. The vaporization apparatus is operated as described above, and translational apparatus 295 moves OLED substrate 285 perpendicular to the  
30 direction of emission of organic material vapors from manifold 20, thus delivering mixed vaporized organic material to the surface of OLED substrate 285 to condense and form a layer of organic material on the surface.

Turning now to FIG. 11, there is shown a cross-sectional view of a pixel of a light-emitting OLED device 310 that can be prepared in part according to the present invention. The OLED device 310 includes at a minimum a substrate 320, a cathode 390, an anode 330 spaced from cathode 390, and a light-emitting layer 350. The OLED device can also include a hole-injecting layer 335, a hole-transporting layer 340, an electron-transporting layer 355, and an electron-injecting layer 360. Hole-injecting layer 335, hole-transporting layer 340, light-emitting layer 350, electron-transporting layer 355, and electron-injecting layer 360 include a series of organic layers 370 disposed between anode 330 and cathode 390. Organic layers 370 are the organic material layers most desirably deposited by the device and method of this invention. These components will be described in more detail.

Substrate 320 can be an organic solid, an inorganic solid, or a combination of organic and inorganic solids. Substrate 320 can be rigid or flexible and can be processed as separate individual pieces, such as sheets or wafers, or as a continuous roll. Typical substrate materials include glass, plastic, metal, ceramic, semiconductor, metal oxide, semiconductor oxide, semiconductor nitride, or combinations thereof. Substrate 320 can be a homogeneous mixture of materials, a composite of materials, or multiple layers of materials. Substrate 320 can be an OLED substrate, that is a substrate commonly used for preparing OLED devices, e.g. active-matrix low-temperature polysilicon or amorphous-silicon TFT substrate. The substrate 320 can either be light transmissive or opaque, depending on the intended direction of light emission. The light transmissive property is desirable for viewing the EL emission through the substrate. Transparent glass or plastic are commonly employed in such cases. For applications where the EL emission is viewed through the top electrode, the transmissive characteristic of the bottom support is immaterial, and therefore can be light transmissive, light absorbing or light reflective. Substrates for use in this case include, but are not limited to, glass, plastic, semiconductor materials, ceramics, and circuit board materials, or any others commonly used in the formation of OLED devices, which can be either passive-matrix devices or active-matrix devices.

An electrode is formed over substrate 320 and is most commonly configured as an anode 330. When EL emission is viewed through the substrate 320, anode 330 should be transparent or substantially transparent to the emission of interest. Common transparent anode materials useful in this invention are  
5 indium-tin oxide and tin oxide, but other metal oxides can work including, but not limited to, aluminum- or indium-doped zinc oxide, magnesium-indium oxide, and nickel-tungsten oxide. In addition to these oxides, metal nitrides such as gallium nitride, metal selenides such as zinc selenide, and metal sulfides such as zinc sulfide, can be used as an anode material. For applications where EL emission is  
10 viewed through the top electrode, the transmissive characteristics of the anode material are immaterial and any conductive material can be used, transparent, opaque or reflective. Example conductors for this application include, but are not limited to, gold, iridium, molybdenum, palladium, and platinum. The preferred anode materials, transmissive or otherwise, have a work function of 4.1 eV or  
15 greater. Desired anode materials can be deposited by any suitable means such as evaporation, sputtering, chemical vapor deposition, or electrochemical means. Anode materials can be patterned using well known photolithographic processes.

While not always necessary, it is often useful that a hole-injecting layer 335 be formed over anode 330 in an organic light-emitting display. The  
20 hole-injecting material can serve to improve the film formation property of subsequent organic layers and to facilitate injection of holes into the hole-transporting layer. Suitable materials for use in hole-injecting layer 335 include, but are not limited to, porphyrinic compounds as described in U.S. Patent No. 4,720,432, plasma-deposited fluorocarbon polymers as described in U.S. Patent  
25 No. 6,208,075, and inorganic oxides including vanadium oxide (VOx), molybdenum oxide (MoOx), nickel oxide (NiOx), etc. Alternative hole-injecting materials reportedly useful in organic EL devices are described in EP 0 891 121 A1 and EP 1 029 909 A1.

While not always necessary, it is often useful that a hole-  
30 transporting layer 340 be formed and disposed over anode 330. Desired hole-transporting materials can be deposited by any suitable means such as evaporation, sputtering, chemical vapor deposition, electrochemical means,

thermal transfer, or laser thermal transfer from a donor material, and can be deposited by the device and method described herein. Hole-transporting materials useful in hole-transporting layer 340 are well known to include compounds such as an aromatic tertiary amine, where the latter is understood to be a compound  
 5 containing at least one trivalent nitrogen atom that is bonded only to carbon atoms, at least one of which is a member of an aromatic ring. In one form the aromatic tertiary amine can be an arylamine, such as a monoarylamine, diarylamine, triarylamine, or a polymeric arylamine. Exemplary monomeric triarylaminines are illustrated by Klupfel et al. in U.S. Patent No. 3,180,730. Other  
 10 suitable triarylaminines substituted with one or more vinyl radicals and/or comprising at least one active hydrogen-containing group are disclosed by Brantley et al. in U.S. Patent Nos. 3,567,450 and 3,658,520.

A more preferred class of aromatic tertiary amines are those which include at least two aromatic tertiary amine moieties as described in U.S. Patent  
 15 Nos. 4,720,432 and 5,061,569. Such compounds include those represented by structural Formula A.

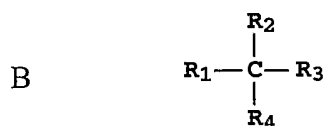


wherein:

Q<sub>1</sub> and Q<sub>2</sub> are independently selected aromatic tertiary amine moieties; and  
 20 G is a linking group such as an arylene, cycloalkylene, or alkylene group of a carbon to carbon bond.

In one embodiment, at least one of Q<sub>1</sub> or Q<sub>2</sub> contains a polycyclic fused ring structure, e.g., a naphthalene. When G is an aryl group, it is conveniently a phenylene, biphenylene, or naphthalene moiety.

25 A useful class of triarylaminines satisfying structural Formula A and containing two triarylamine moieties is represented by structural Formula B.

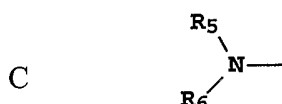




where:

$R_1$  and  $R_2$  each independently represent a hydrogen atom, an aryl group, or an alkyl group or  $R_1$  and  $R_2$  together represent the atoms completing a cycloalkyl group; and

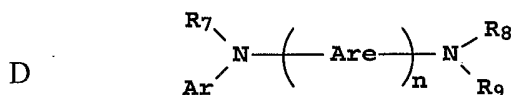
- 5  $R_3$  and  $R_4$  each independently represent an aryl group, which is in turn substituted with a diaryl substituted amino group, as indicated by structural Formula C.



wherein  $R_5$  and  $R_6$  are independently selected aryl groups. In one embodiment, at least one of  $R_5$  or  $R_6$  contains a polycyclic fused ring structure, e.g., a

- 10 naphthalene.

Another class of aromatic tertiary amines are the tetraaryldiamines. Desirable tetraaryldiamines include two diarylamino groups, such as indicated by Formula C, linked through an arylene group. Useful tetraaryldiamines include those represented by Formula D.



- 15 wherein:

each Are is an independently selected arylene group, such as a phenylene or anthracene moiety;

$n$  is an integer of from 1 to 4; and

Ar,  $R_7$ ,  $R_8$ , and  $R_9$  are independently selected aryl groups.

- 20 In a typical embodiment, at least one of Ar,  $R_7$ ,  $R_8$ , and  $R_9$  is a polycyclic fused ring structure, e.g., a naphthalene.

The various alkyl, alkylene, aryl, and arylene moieties of the foregoing structural Formulae A, B, C, D, can each in turn be substituted. Typical substituents include alkyl groups, alkoxy groups, aryl groups, aryloxy groups, and

25 halogens such as fluoride, chloride, and bromide. The various alkyl and alkylene moieties typically contain from 1 to about 6 carbon atoms. The cycloalkyl moieties can contain from 3 to about 10 carbon atoms, but typically contain five,

six, or seven carbon atoms--e.g., cyclopentyl, cyclohexyl, and cycloheptyl ring structures. The aryl and arylene moieties are usually phenyl and phenylene moieties.

The hole-transporting layer in an OLED device can be formed of a single or a mixture of aromatic tertiary amine compounds. Specifically, one can employ a triarylamine, such as a triarylamine satisfying the Formula B, in combination with a tetraaryldiamine, such as indicated by Formula D. When a triarylamine is employed in combination with a tetraaryldiamine, the latter is positioned as a layer interposed between the triarylamine and the electron-injecting and transporting layer. The device and method described herein can be used to deposit single- or multi-component layers, and can be used to sequentially deposit multiple layers.

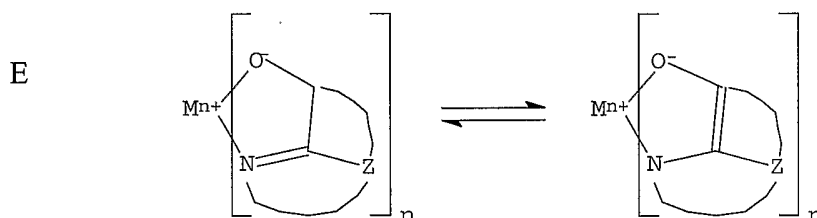
Another class of useful hole-transporting materials includes polycyclic aromatic compounds as described in EP 1 009 041. In addition, polymeric hole-transporting materials can be used such as poly(N-vinylcarbazole) (PVK), polythiophenes, polypyrrole, polyaniline, and copolymers such as poly(3,4-ethylenedioxythiophene)/poly(4-styrenesulfonate) also called PEDOT/PSS.

Light-emitting layer 350 produces light in response to hole-electron recombination. Light-emitting layer 350 is commonly disposed over hole-transporting layer 340. Desired organic light-emitting materials can be deposited by any suitable means such as evaporation, sputtering, chemical vapor deposition, electrochemical means, or radiation thermal transfer from a donor material, and can be deposited by the device and method described herein. Useful organic light-emitting materials are well known. As more fully described in U.S. Patent Nos. 4,769,292 and 5,935,721, the light-emitting layers of the organic EL element include a luminescent or fluorescent material where electroluminescence is produced as a result of electron-hole pair recombination in this region. The light-emitting layers can include a single material, but more commonly include a host material doped with a guest compound or dopant where light emission comes primarily from the dopant. The dopant is selected to produce color light having a particular spectrum. The host materials in the light-emitting layers can be an

electron-transporting material, as defined below, a hole-transporting material, as defined above, or another material that supports hole-electron recombination. The dopant is usually chosen from highly fluorescent dyes, but phosphorescent compounds, e.g., transition metal complexes as described in WO 98/55561, WO 00/18851, WO 00/57676, and WO 00/70655 are also useful. Dopants are typically coated as 0.01 to 10 % by weight into the host material. The device and method described herein can be used to coat multi-component guest/host layers without the need for multiple vaporization sources.

Host and emitting molecules known to be of use include, but are not limited to, those disclosed in U.S. Patent Nos. 4,768,292; 5,141,671; 5,150,006; 5,151,629; 5,294,870; 5,405,709; 5,484,922; 5,593,788; 5,645,948; 5,683,823; 5,755,999; 5,928,802; 5,935,720; 5,935,721; and 6,020,078.

Metal complexes of 8-hydroxyquinoline and similar derivatives (Formula E) constitute one class of useful host materials capable of supporting electroluminescence, and are particularly suitable for light emission of wavelengths longer than 500 nm, e.g., green, yellow, orange, and red.



wherein:

M represents a metal;

n is an integer of from 1 to 3; and

Z independently in each occurrence represents the atoms completing a nucleus having at least two fused aromatic rings.

From the foregoing it is apparent that the metal can be a monovalent, divalent, or trivalent metal. The metal can, for example, be an alkali metal, such as lithium, sodium, or potassium; an alkaline earth metal, such as magnesium or calcium; or an earth metal, such as boron or aluminum. Generally any monovalent, divalent, or trivalent metal known to be a useful chelating metal can be employed.

Z completes a heterocyclic nucleus containing at least two fused aromatic rings, at least one of which is an azole or azine ring. Additional rings, including both aliphatic and aromatic rings, can be fused with the two required rings, if required. To avoid adding molecular bulk without improving on function  
5 the number of ring atoms is usually maintained at 18 or less.

The host material in light-emitting layer 350 can be an anthracene derivative having hydrocarbon or substituted hydrocarbon substituents at the 9 and 10 positions. For example, derivatives of 9,10-di-(2-naphthyl)anthracene constitute one class of useful host materials capable of supporting  
10 electroluminescence, and are particularly suitable for light emission of wavelengths longer than 400 nm, e.g., blue, green, yellow, orange or red.

Benzazole derivatives constitute another class of useful host materials capable of supporting electroluminescence, and are particularly suitable for light emission of wavelengths longer than 400 nm, e.g., blue, green, yellow,  
15 orange or red. An example of a useful benzazole is 2, 2', 2''-(1,3,5-phenylene)tris[1-phenyl-1H-benzimidazole].

Desirable fluorescent dopants include perylene or derivatives of perylene, derivatives of anthracene, tetracene, xanthene, rubrene, coumarin, rhodamine, quinacridone, dicyanomethylenepyrans compounds, thiopyran  
20 compounds, polymethine compounds, pyrilium and thiapyrilium compounds, derivatives of distyrylbenzene or distyrylbiphenyl, bis(azinyl)methane boron complex compounds, and carbostyryl compounds.

Other organic emissive materials can be polymeric substances, e.g. polyphenylenevinylene derivatives, dialkoxy-polyphenylenevinylenes, poly-para-  
25 phenylene derivatives, and polyfluorene derivatives, as taught by Wolk et al. in commonly assigned U.S. Patent No. 6,194,119 B1 and references cited therein.

While not always necessary, it is often useful that OLED device 310 includes an electron-transporting layer 355 disposed over light-emitting layer 350. Desired electron-transporting materials can be deposited by any suitable  
30 means such as evaporation, sputtering, chemical vapor deposition, electrochemical means, thermal transfer, or laser thermal transfer from a donor material, and can be deposited by the device and method described herein. Preferred electron-

transporting materials for use in electron-transporting layer 355 are metal chelated oxinoid compounds, including chelates of oxine itself (also commonly referred to as 8-quinolinol or 8-hydroxyquinoline). Such compounds help to inject and transport electrons and exhibit both high levels of performance and are readily  
5 fabricated in the form of thin films. Exemplary of contemplated oxinoid compounds are those satisfying structural Formula E, previously described.

Other electron-transporting materials include various butadiene derivatives as disclosed in U.S. Patent No. 4,356,429 and various heterocyclic optical brighteners as described in U.S. Patent No. 4,539,507. Benzazoles  
10 satisfying structural Formula G are also useful electron-transporting materials.

Other electron-transporting materials can be polymeric substances, e.g. polyphenylenevinylene derivatives, poly-para-phenylene derivatives, polyfluorene derivatives, polythiophenes, polyacetylenes, and other conductive polymeric organic materials such as those listed in *Handbook of Conductive*  
15 *Molecules and Polymers*, Vols. 1-4, H.S. Nalwa, ed., John Wiley and Sons, Chichester (1997).

An electron-injecting layer 360 can also be present between the cathode and the electron-transporting layer. Examples of electron-injecting materials include alkaline or alkaline earth metals, alkali halide salts, such as LiF  
20 mentioned above, or alkaline or alkaline earth metal doped organic layers.

Cathode 390 is formed over the electron-transporting layer 355 or over light-emitting layer 350 if an electron-transporting layer is not used. When light emission is through the anode 330, the cathode material can include nearly any conductive material. Desirable materials have good film-forming properties  
25 to ensure good contact with the underlying organic layer, promote electron injection at low voltage, and have good stability. Useful cathode materials often contain a low work function metal ( $< 3.0$  eV) or metal alloy. One preferred cathode material is include of a Mg:Ag alloy wherein the percentage of silver is in the range of 1 to 20 %, as described in U.S. Patent No. 4,885,221. Another  
30 suitable class of cathode materials includes bilayers includes a thin layer of a low work function metal or metal salt capped with a thicker layer of conductive metal. One such cathode includes a thin layer of LiF followed by a thicker layer of Al as

described in U.S. Patent No. 5,677,572. Other useful cathode materials include, but are not limited to, those disclosed in U.S. Patent Nos. 5,059,861; 5,059,862; and 6,140,763.

When light emission is viewed through cathode 390, it must be  
5 transparent or nearly transparent. For such applications, metals must be thin or one must use transparent conductive oxides, or a combination of these materials. Optically transparent cathodes have been described in more detail in U.S. Patent No. 5,776,623. Cathode materials can be deposited by evaporation, sputtering, or chemical vapor deposition. When needed, patterning can be achieved through  
10 many well known methods including, but not limited to, through-mask deposition, integral shadow masking as described in U.S. Patent No. 5,276,380 and EP 0 732 868, laser ablation, and selective chemical vapor deposition.

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The invention has been described in detail with particular reference to certain preferred embodiments thereof, namely for delivery of organic  
20 particulate materials to a vaporization zone. However, it can be appreciated that the present invention applies more broadly to particulate materials, including organic and other types of particulate materials. The term "particulate materials" can include a broad range of substances in particulate form, including, but not limited to, crystals, nanotubes, powders, needles, flakes, and other solid materials  
25 that can be classified as discontinuous, for example. Moreover, the particulate materials may be provided in a mixture containing a quantity of inert material or materials acting as a carrier for the component material. Inert carriers could include other types of solid materials as well as pastes and liquids, particularly liquid materials having higher viscosities. Any inert material selected must be  
30 compatible with the vaporization process, such that the inert carrier is appropriately discarded before or during the vaporization of the component particulate material. For example, the inert carrier can be selected from materials

having a much higher vaporization temperature than the desired particulate component material. As just one example, particulate material 100 (FIG. 3) could be a mixture containing sand and the particulate component material that is to be vaporized. The utilization of such an inert carrier, with suitable mixing

5 techniques, would allow the metering of minute quantities of a component particulate material, such as an organic particulate material, for vaporization.

**PARTS LIST**

5	vaporization device
6	source
7	source
8	source
9	vapor plume
10	vaporization apparatus
15	substrate
20	manifold
30	aperture
35	motor
40	feeding apparatus
45	feeding apparatus
50	first container
55	second container
60	feeding path
65	feeding path
70	third container
75	fourth container
80	auger structure
85	auger screw
85a	auger screw
85b	auger screw
85c	auger screw
90	motor
95	auger structure
100	particulate material
105	auger structure
110	screen
115	helical thread



**Parts List cont'd**

120	screen
125	center portion
130	piezoelectric structure
135	thread-free portion
140	piezoelectric structure
150	piezoelectric structure
155	piezoelectric structure
160	particulate material
170	heating element
180	base
190	bottom portion
210	rotating thread type device
220	gear driver
230	opening
240	first particulate material
245	second particulate material
250	first auger structure
255	second auger structure
260	mixing chamber
275	load lock
280	deposition chamber
285	OLED substrate
295	translational apparatus
300	vacuum source
310	OLED device
320	substrate
330	anode
335	hole-injecting layer
340	hole-transporting layer

**Parts List cont'd**

350	light-emitting layer
355	electron-transporting layer
360	electron-injecting layer
370	organic layers
390	cathode

**CLAIMS:**

1. A method for vaporizing particulate material and condensing it onto a surface to form a layer, comprising:
  - (a) providing a quantity of first particulate material in a first  
5 container and a quantity of second particulate material in a second container spaced apart from the first container, the first and second containers respectively having first and second openings;
  - (b) transferring the first particulate material through the first  
10 opening in the first container into a manifold and vaporizing the first particulate material in the manifold;
  - (c) transferring the second particulate material through the  
second opening in the second container into the manifold and vaporizing the second particulate material in the manifold, whereby the first and second  
vaporized particulate materials are mixed; and
  - 15 (d) delivering the mixed vaporized materials from the manifold to the surface to form the layer.
2. The method of claim 1 further including fluidizing particulate material delivered from at least the first or the second container.
3. The method of claim 1 further providing a third and a  
20 fourth container respectively associated with the first and second containers, each for transferring particulate material to its corresponding first or second container.
4. The method of claim 3 further including metering the  
amount of particulate material transferred from the third and fourth containers to the first and second containers, respectively, to provide a substantially constant  
25 volume of particulate material in the first and second containers.
5. The method of claim 1 wherein the step of transferring the first particulate material into a manifold comprises:
  - (a) transferring the first particulate material through the first  
opening to a first auger; and
  - 30 (b) rotating at least a portion of the first auger to transfer the first particulate material from the first container along a feeding path to a first vaporization zone wherein the first particulate material is vaporized.

6. The method of claim 5 wherein the step of transferring the second particulate material into a manifold comprises:

(a) transferring the second particulate material through the second opening to a second auger; and

5 (b) rotating at least a portion of the second auger to transfer the second particulate material from the second container along a feeding path to a second vaporization zone wherein the second particulate material is vaporized.

7. The method of claim 1 wherein at least one of the first or second particulate materials comprises an organic particulate material.

10 8. The method of claim 5 wherein the first auger comprises a helical thread having an angle between 4 and 15 degrees relative to the axis of the auger.

9. The method of claim 5 wherein the surface of the auger is treated.

15 10. The method of claim 9 wherein the surface of the auger is coated with titanium nitride.

11. The method of claim 9 wherein the surface of the auger is electropolished.

12. The method of claim 1 wherein at least the first container is  
20 held at a pressure below atmospheric pressure.

13. The method of claim 5 further comprising agitating the first organic material by using an agitating device.

14. The method of claim 13 wherein the agitating device includes a piezoelectric structure or a rotating thread type device.

25 15. The method of claim 6 wherein the first auger structure passes through the interior of the first container and the second auger structure passes through the interior of the second container.

16. The method of claim 6 wherein the feed rate of the first and second particulate materials to the auger structures controls the deposition rate of  
30 the vaporized particulate material and the concentration of materials in the layer.

17. The method of claim 1 wherein the manifold includes a separate vaporization zone in the manifold.

18. The method of claim 1 wherein the first particulate material includes two or more different organic material components.

5 19. The method of claim 1 wherein the second particulate material includes two or more different organic material components.

20. The method of claim 5 wherein rotating at least a portion of the first auger comprises rotating the first auger in a repeated incremental fashion.

10 21. The method of claim 17 wherein the vaporization zone includes a heating element.

22. The method of claim 5 wherein the temperature of the first particulate material in the feeding path is maintained below the desired vaporization temperature.

15 23. A method for vaporizing particulate material and condensing it onto a surface to form a film, comprising:

(a) providing a quantity of first particulate material in a first container and a quantity of second particulate material in a second container spaced apart from the first container, the first and second containers respectively having first and second openings;

20 (b) transferring the first particulate material through the first opening in the first container into a manifold;

(c) transferring the second particulate material through the second opening in the second container into a manifold

wherein the first and second particulate materials are mixed; and

25 (d) vaporizing the mixed first and second particulate materials in the manifold and delivering such vaporized first and second particulate materials to the substrate surface to form the layer.

30 24. The method of claim 23 further providing a third and fourth containers respectively associated with the first and second containers each for transferring particulate material to its corresponding first or second container.

25. The method of claim 24 further including metering the amount of particulate materials transferred from the third and fourth containers to the first and second containers, respectively, to provide a substantially constant volume of material in the first and second containers.

5                   26. The method of claim 23 further including separately fluidizing the particulate material in the first and second containers, respectively, and transferring such fluidized materials.

27. The method of claim 26 wherein each fluidizing step includes agitating the particulate material by using an agitating device.

10                   28. The method of claim 27 wherein the agitating device includes a piezoelectric structure or a rotating thread type device.

29. The method of claim 23 wherein the feed rate of the first and second particulate materials to the manifold controls the deposition rate of the vaporized particulate material and the concentration of materials in the layer.

15                   30. The method of claim 23 wherein a first auger structure passes through the interior of the first container and a second auger structure passes through the interior of the second container.

31. The method of claim 23 wherein the first particulate material includes two or more different organic material components.

20                   32. The method of claim 23 wherein the second particulate material includes two or more different organic material components.

33. The method of claim 1 wherein at least the first particulate material comprises an inert carrier.

25                   34. The method of claim 23 wherein the first particulate material comprises an organic particulate material.

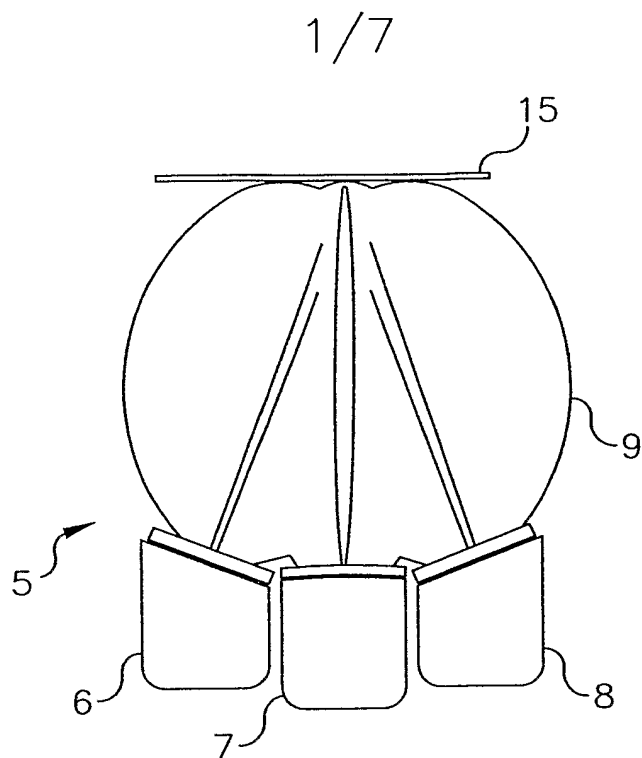


FIG. 1  
(PRIOR ART)

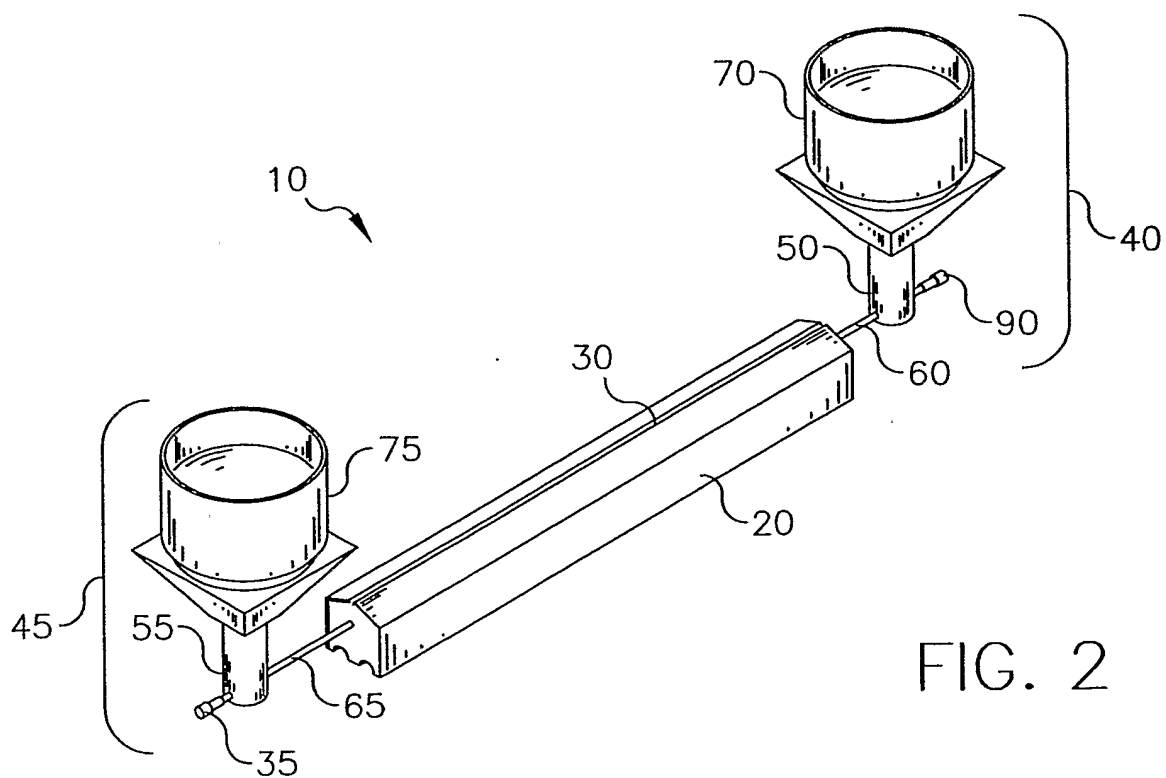
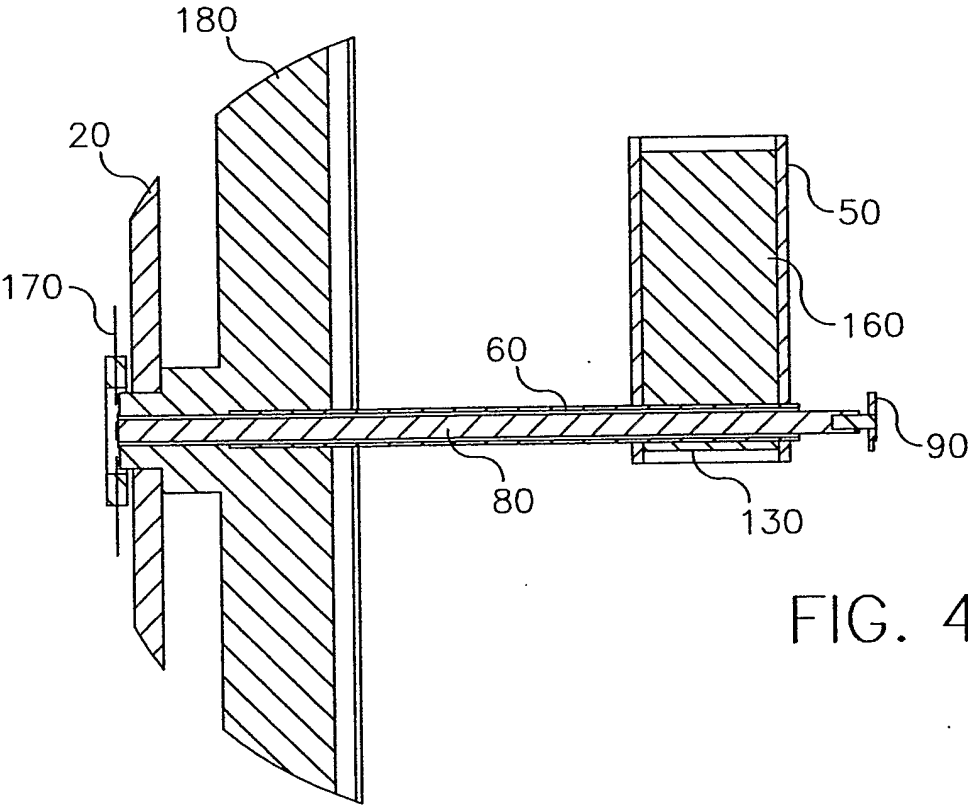
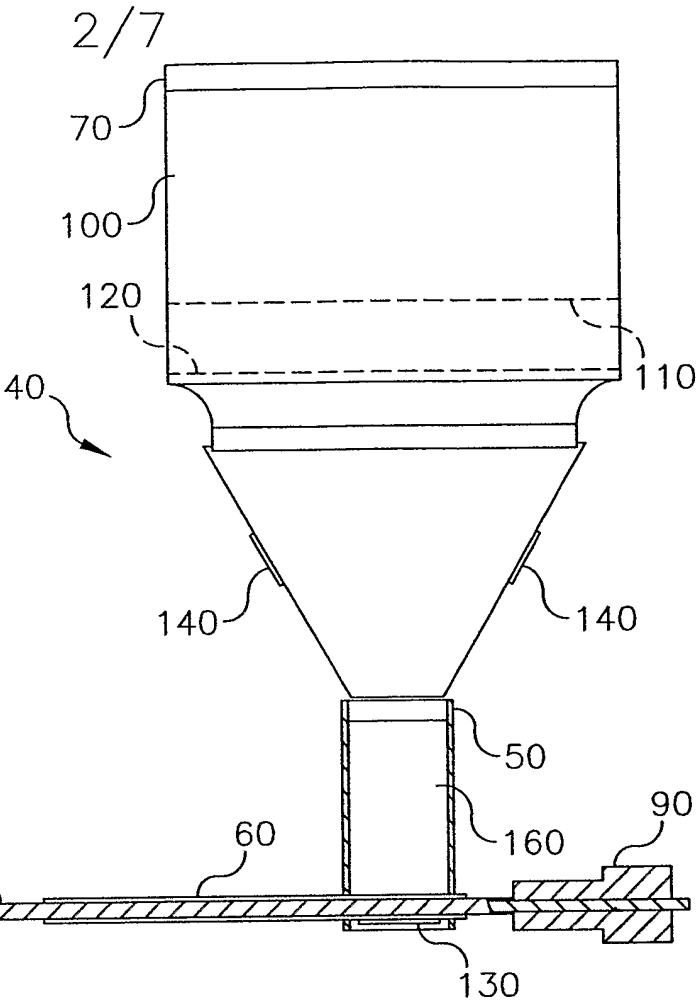


FIG. 2





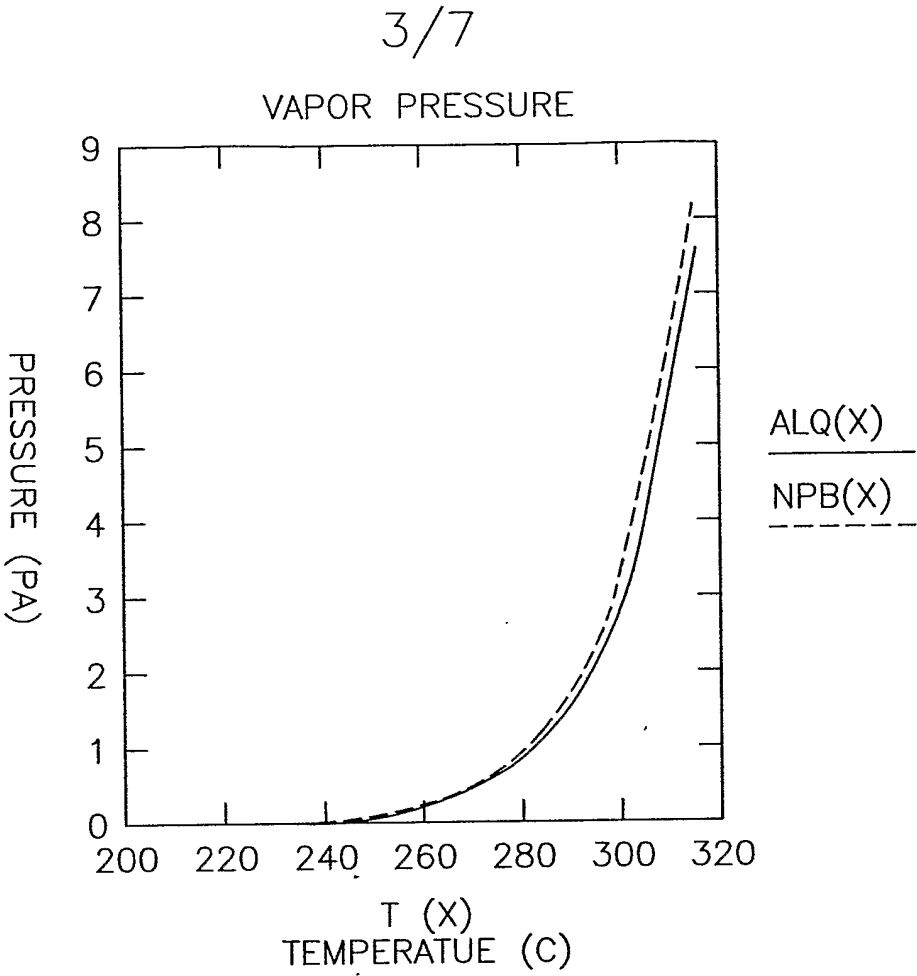


FIG. 5

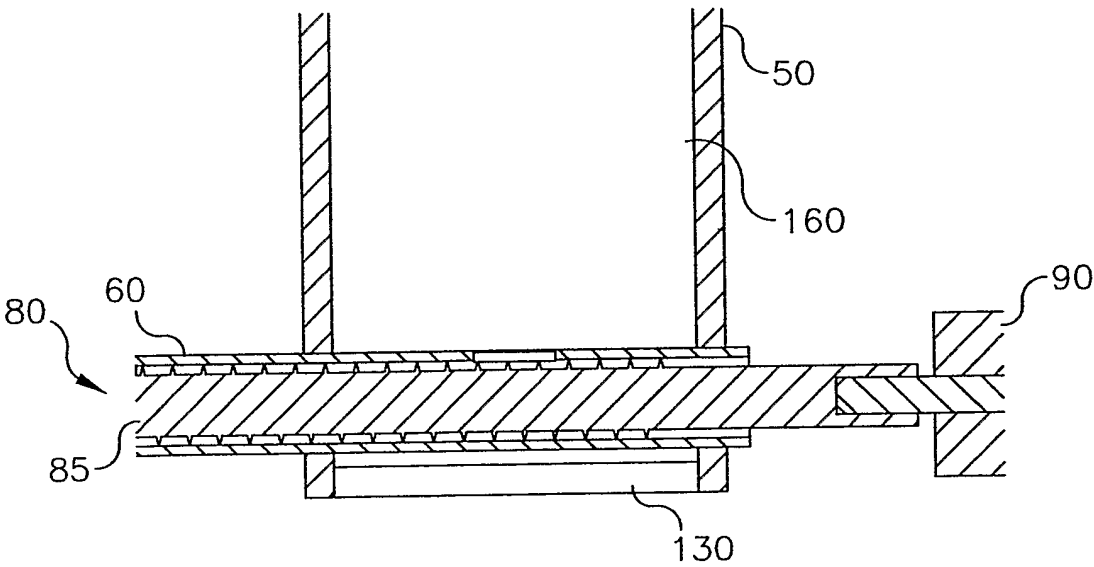


FIG. 6A

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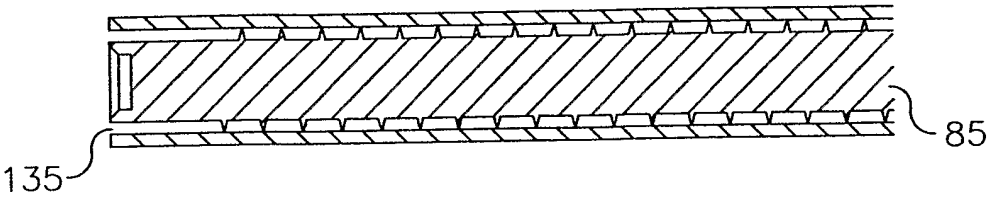


FIG. 6B

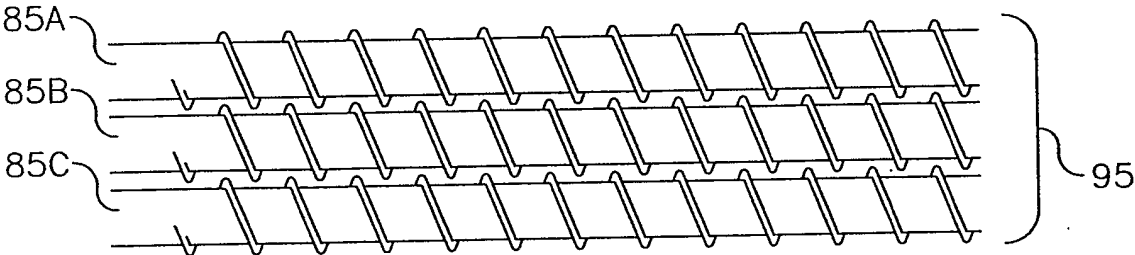


FIG. 6C

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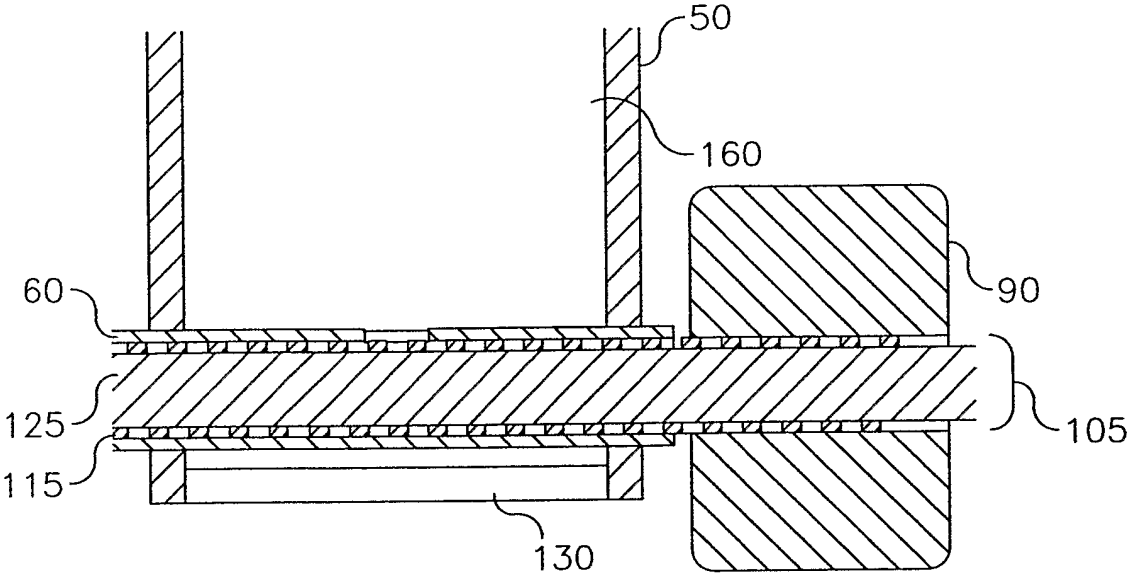


FIG. 6D

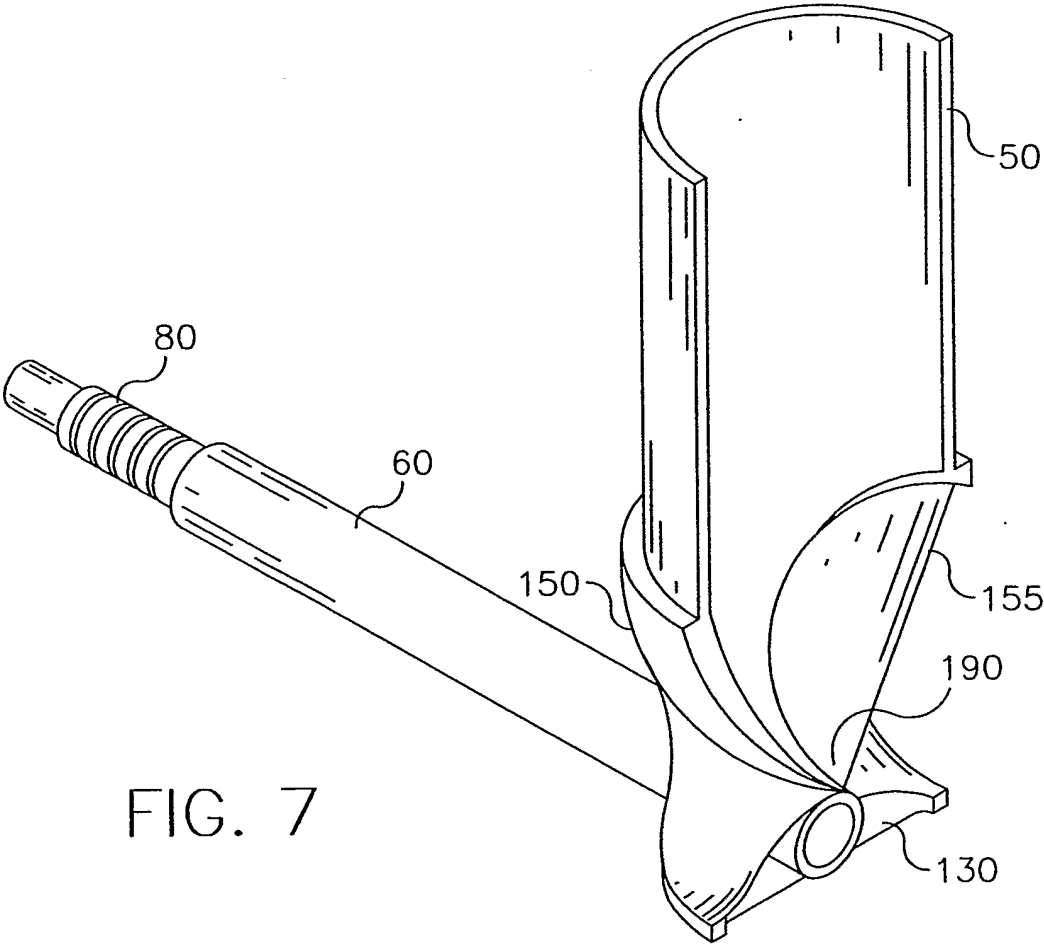


FIG. 7

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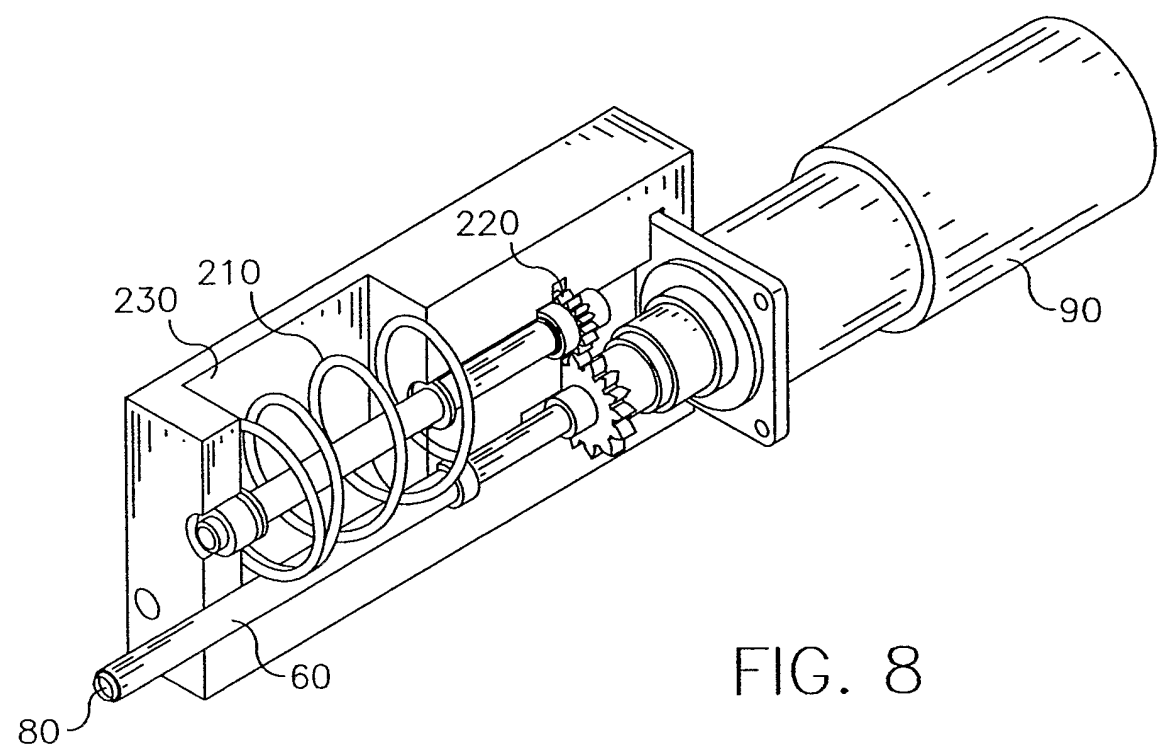


FIG. 8

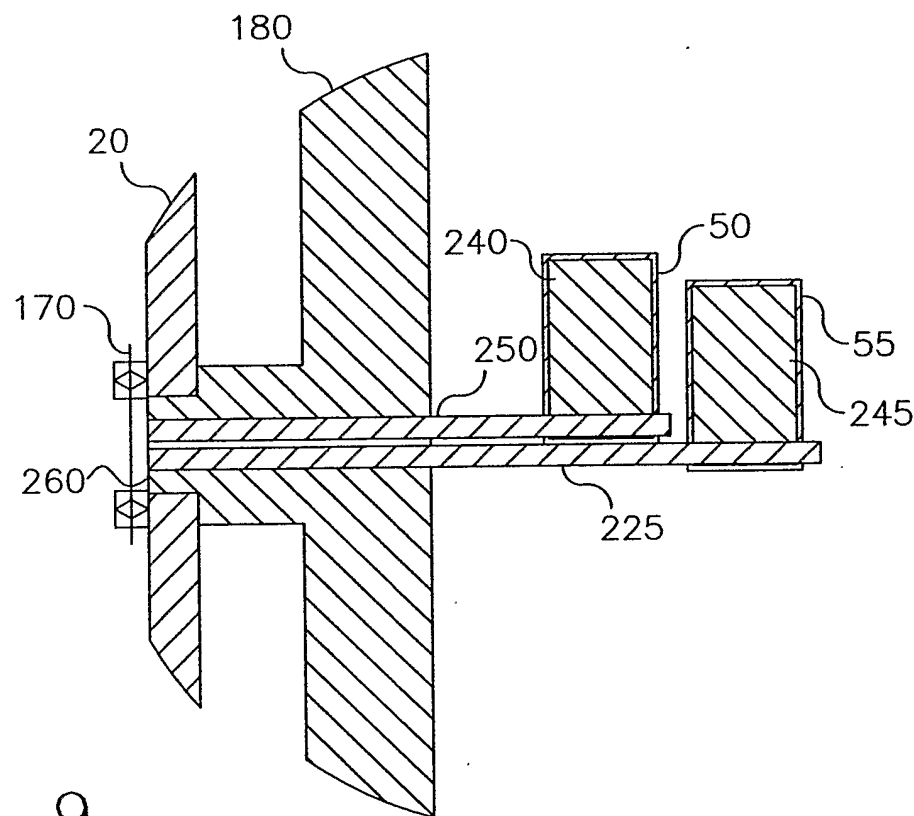


FIG. 9

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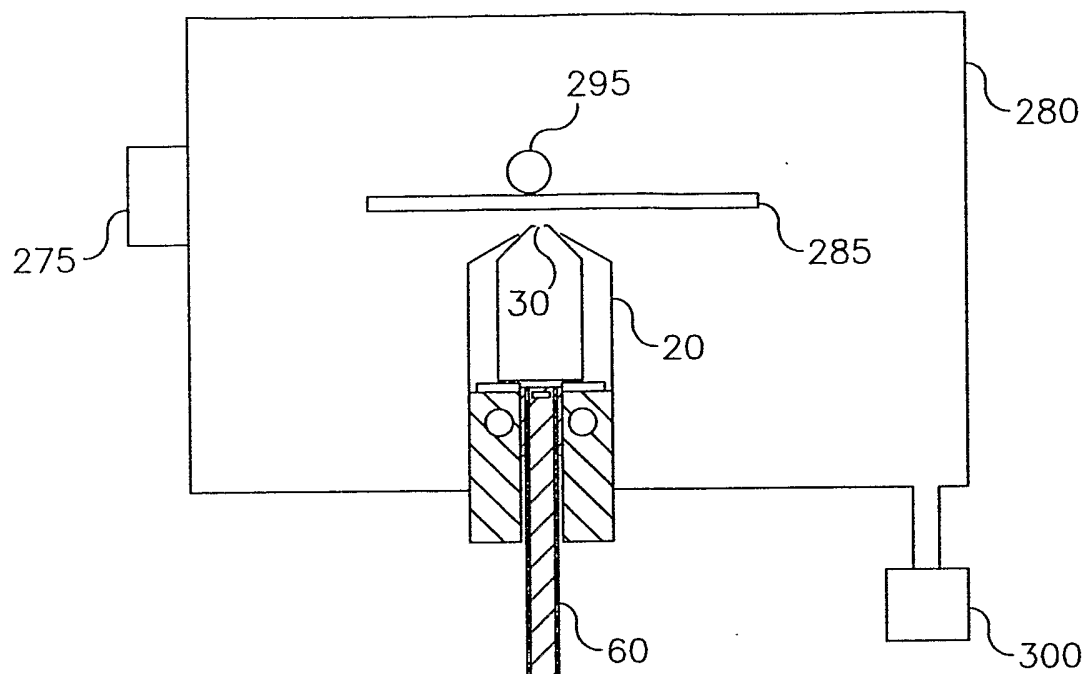


FIG. 10

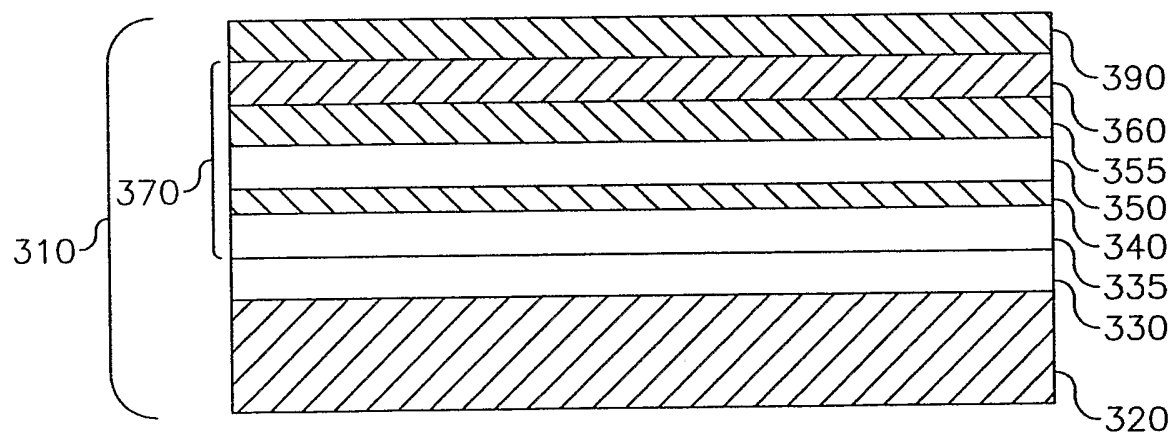


FIG. 11