

Flexible Organic Light-Emitting Diodes: Structure and Fabrication

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Abstract: Flexible organic light-emitting diodes (OLED) technology with thin, light in weight, and free in design have been developed and perfected rapidly. The versatility of flexible OLED technology has been effectively proved in the sectors of displays, lighting, and medicine. In this article, the structure and fabrication of flexible OLED were introduced. Normal flexible OLED device construction includes flexible OLED substrate, barrier layers, electrode and organic layer. The flexible OLED is made on the substrate. The barrier layer is used to shield the OLED material from airborne oxygen and humidity. The electrodes are used for electricity, and the organic layer is used for light. The production processes of flexible OLED include coat-debond with advantages such as good stability and thermal conductivity, bond-debond with the advantage of simple and convenient and roll-to-roll [R2R] method with advantages of low cost, and mass production. Additionally, the application fields of these methods have been highlighted.

Keywords: Flexible OLED, Structure, Fabrication.

1. Introduction

Organic light-emitting diodes (OLED), are mainly used for flat panel display. As we all know, the most important thing to make TV is the upstream liquid crystal panel. However, with the development of science and technology, advanced panel technology is gradually replacing the LCD panel. Since 2017, OLED has shown a trend of rapid development. With the development and improvement of OLED materials, flexible OLED is of great significance in various aspects. Flexible OLEDs are OLEDs built on non-rigid substrates such as plastic, Polyimide, or metal foil. Flexible OLED displays can be used in various electronic fields such as bendable, bendable, roll-up, and commercial displays due to their thin, lightweight, and free design. Currently, the versatility of flexible OLED technology has been effectively proved in the sectors of displays, lighting, and medicine. Early OLED displays are not as impressive as now. While features such as ultra-thin, automatic lighting, high contrast, and power saving were known, the technology was still immature and had many limitations. Even so, some global display manufacturers are still full of confidence in OLED display panels, continue to invest in technology research and development, and finally break through technical barriers and usher in new development opportunities. With flexible OLED in the stage of development, the characteristics of collapsible lags far behind the LCD panel. Today, the market has largely shifted toward OLED displays, which are now standard on high-end TVs and mobile phones. Flexible OLED materials will also be the development direction of the display industry in the future.

A fully flexible OLED device is capable of withstanding mechanical stress, primarily caused by bending, stretching, and repeated folding. Additionally, it benefits from strong light transmission, electrical conductivity, surface uniformity and adherence, chemical stability, low-cost mass production, and other properties. However, there are several technical obstacles to the development of flexible OLED displays, which call for improvements in production methods and materials. Furthermore, flexible OLED devices now experience difficult manufacturing issues such as lower yield and high production costs as a result of lengthy manufacturing processes. Using the positive of low-cost equipment manufacturing cost reduction strategy is desirable. Earlier in 2006, the Park found that creating effective permeation barriers on polymer substrates might be challenging [1]. Currently, a plastic substrate is the best option. Utilizes polyimide substrate frequently. Outstanding thermal stability, strong radiation resistance, chemical stability, and superior mechanical properties are all

characteristics of polyimide as a material [2]. Polyimide has been widely used due to its amazing qualities such as high-temperature solder resistance, high strength, high modulus, and flame retardant. Additionally, in 2016, issues with transparent thin film barriers in the fabrication of flexible OLED devices were highlighted [3]. Currently, the flexible OLED display had good gas barrier properties thanks to the multi-layer thin film encapsulation (TFE). Future progress in OLED technology is anticipated to come from flexible goods that are higher-performing, thinner, lighter, indestructible, and require less electricity [4]. In this article, the structure and fabrication of flexible OLED were introduced.

2. General structure of flexible OLED

As shown in Figure 1, the figure below shows a normal flexible OLED device construction. Flexible OLED is a solid-state light-emitting device with a simple structure. The organic layer is sandwiched between two electrodes, and barrier layers are wrapped to block the corrosion of moisture and air. On the substrate, the barrier layer at the bottom, the bottom anode, multiple organic layers are in the middle and the cathode is on the top and then wrapped with the barrier layer, just like the figure. The basic structure of flexible OLED is the same as that of rigid OLED, but to achieve flexibility, the substrate and packaging layers need to be changed. Figure 1 illustrates how gases can enter flexible OLED devices from all sides. As a result, barrier technologies must be used to prevent such gas penetrations.

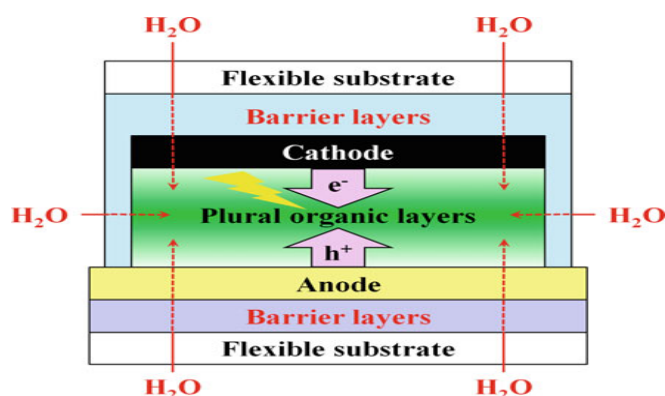


Figure 1. Normal flexible OLED device construction [5].

2.1 Flexible OLED substrate

The biggest difference between flexible and rigid OLED devices is not the functional material, but the substrate material. Rigid OLEDs usually use glass as the substrate material, while flexible OLEDs use plastic substrates as the flexible substrate. At present, the factors to be considered in the selection of substrate materials include heat resistance, water and oxygen penetration resistance, and expansion characteristics. Materials that can be used to make flexible substrates are polyimide, ultra-thin glasses, stainless steel foils, flexible films (barrier films), and so on. At present, Polyimide is a significant flexible substrate for flexible OLED devices. Polyimide (PI) refers to the main chain containing the imide ring (-Co-NH-CO-) of a class of polymers, and it has high-temperature resistance and excellent insulation performance.

2.1.1 Heat resistance

The flexible substrate's temperature resistance is typically linked to the OLED assembly procedure. In the OLED device preparation process, the semiconductor layer and organic functional layer are mostly prepared by the thermal evaporation process, and the process temperature is higher than 400 °C. Ordinary plastic substrates are not stable at this temperature. Polyimide (PI) is widely used as a flexible display substrate for OLEDs because it can achieve better heat resistance and stability. However, common polyimide materials are transparent yellow, which limits the application of

bottom-emission OLED. Therefore, researchers are developing transparent π -substrates (CPI), which would degrade the original properties of PI. To address this problem, cellulose nanocrystals CNC/CPI hybrid substrates were developed, which have excellent mechanical and thermal properties and can be improved by the interaction between CNC and CPI. The thermal decomposition temperature of the composite is 555 °C and the thermal expansion coefficient is 31.62 PPM K⁻¹ [6].

2.1.2 Water and oxygen penetration resistance

However, Polyimides have one issue that all polymer materials have, and that is the high water vapor transport rate (WVTR). Due to the increased water transport rate, TFT characteristics and even OLED performance would be destroyed as water passes through the polymer layer. In general, inorganic materials have low transmission rates, but rigid structures are not suitable for flexible OLED devices. Recent studies have shown that the water vapor transport characteristics of pure polymer materials can be greatly improved by fabricating polymer/nano-inorganic multilayer structures, which can remain flexible and flexible. Recently, researchers have fabricated laminated substrates based on PI/inorganic materials, and the results show that the laminated structures exhibit lower WVTR coefficients than the single-layer PI substrates. In the actual test, the OLED samples with laminated structure substrate still maintain good working stability for 384 hours, without obvious dark and bad spots. On the contrary, under the same test conditions, the performance of OLED devices with single layer PI substrate deteriorates significantly, with many dark spots and bad spots [3].

2.1.3 Expansion characteristics

Another advantage of laminated substrate materials is expansion stability. The results in Figure 2 show the size changes of different substrate materials at different temperatures. It is obvious from Figure 2 that the laminated structure exhibits lower size variation than the single-layer substrate at high temperature, and its expansion characteristics are close to those of glass, which fully indicates that this structure can achieve good process matching.

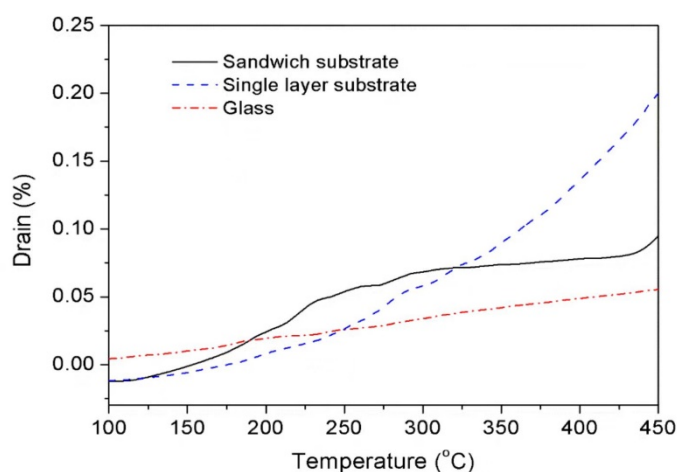


Figure 2. The dimensional stability of flexible substrate [6].

2.2 Barrier layers

Protecting OLED materials against oxidation and ambient moisture damage is one of the biggest hurdles in the progress of flexible OLED systems. These devices require encapsulants and protective coatings with incredibly low oxygen and moisture permeabilities. By WVTR with the unit of g/m²/day, gas barrier properties are frequently assessed. Most OLED devices need moisture permeability of less than 10⁻²⁶ g/m²/day as the bare minimum to provide appropriate lifetime stability. To ensure low Moisture and oxygen Permeability, OLED displays ultimately require a complex stacked layer as a permeable barrier to shield them from the environment's permeability with oxygen and water. At present, the technology of making barrier layers includes multi-layer barriers using wet and dry layers, dry gas barrier layers deposited by roll-to-roll (R2R)PE-CVD and multi-layer barriers

using sputtering. For the first method, a dry SiO_x barrier and a wet polysilazane barrier were created by a team from Yamagata University. The barrier layer's WVTR was 1.6×10^5 g/m²/day [10]. For the second method, WVTR can reach 5.1×10^5 g/m²/day when SiO_x barrier layers with an IZO electrode are applied to regular PEN films. But due to expensive equipment and slow deposition rates, PE-CVD processes frequently have significant prices. The third method is composed of Si₃N₄, Al₂O₃, and other layers, it can achieve good gas barrier qualities with a WVTR of at least 10^5 g/m²/day [4].

2.3 Electrode

2.3.1 Anode

One side of the electrode must be transparent in flexible OLED systems, hence transparent ITO (indium tin oxide) conductive glass or plastic with a high work function is typically chosen as the anode. The high work function is also convenient to improve the efficiency of hole injection. ITO glass has been commercial and can be directly on the preparation of the flexible OLED, but to improve the performance of OLED should be carried out on the surface of the ITO film processing, making it adapt to the organic thin film. Common treatment methods are thermal treated, acid or alkali treatment, argon plasma treatment, UV-ozone treatment and inert gas sputtering, Surface oxidation, and so on. For example, the researchers found that when ITO was heat-treated, its current strength also increased, meaning that its conductivity increased and therefore reduced OLED reaction time when applied to OLED [8]. For argon plasma treatment, researchers used 0.3 Torr, 80 W power argon plasma to treat the ITO surface, reducing surface roughness. From the light emission results of the device, the procedure lowers the gadget's starting voltage and increases the output of light from the device [9].

2.3.2 Cathode

Since injecting electrons is more challenging than injecting holes, the cathode should select a metal with the lowest work function possible to up the effectiveness of the electron infusion. The luminescence efficiency and service life of OLED devices are significantly impacted by the size of the metal work function. Higher luminescence efficiency and easier electron injection result from a lower metal work function. Additionally, a lower work function will result in less Joule heat being produced during operation, which will significantly extend the life of the device, as will a reduced interface barrier between organic and metal. Cathode materials generally include a single-layer metal cathode, alloy cathode, layered cathode, and doped composite electrode. The single-layer metal cathode, such as Ag, Al, Li, Mg, Ca, and In, however, are easily oxidized and unstable in air. For alloy cathodes, such as Mg:Ag, When a single metal cathode film is evaporated, a large number of defects will be formed, resulting in poor oxidation resistance. When the cathode is evaporated, a small amount of metal will preferentially diffuse into the defect, making the whole organic layer very stable. The advantage is to improve the quantum efficiency and stability of the device. It can form a stable and strong metal film on the organic film. On the other hand, The researchers Lau et al. found that Sm:Ag can significantly improve the contrast of OLED by simple thermal evaporation [6]. For a layered cathode, in this type of cathode, a barrier layer such as LiF is added between the luminous layer and the metal electrode. They form a double electrode barrier layer with Al, which can significantly enhance the device's performance by a layer of extremely thin insulating material, such as LiF, Li₂O, MgO, and Al₂O₃, and a thicker layer of Al. It is found that the LiF layer effectively blocks hole injection, enhances the balance of carrier injection, and improves the brightness and efficiency of the device. 1 nm thick LiF hole buffer layer has the best performance, and the efficiency is nearly 1.5 times higher than that without a buffer layer [11]. For doped composite electrodes (typical device structures include ITO, NPD and ALQ), the low work function organic layer metal is sandwiched between the organic luminescence layer and cathode, which can significantly enhance the luminance and performance of the device.

2.4 Organic layer

The organic layers in the majority of OLED devices typically consist of multiple layers that each serves a specific purpose. These layers are known as Hole Transport Layer (HTL), Electron Injection Layer (EIL), Emitting Layer (EML), Hole Blocking Layer (HBL), Hole Injection Layer (HIL), Electron Transport Layer (ETL), and so on. Each organic layer is typically between a few and 100 nanometers thick. As a result, the organic layer is only a few tens or hundreds of nanometers thick overall [5]. For EML, because both electrons and holes need to be transported in the luminescent layer, good electron-hole transport ability is needed to ensure that it is possible to combine enough electrons and holes to produce light. Commonly used EML materials are Alq₃, Almq₃, and blue TBADN. Fadavieslam et al. has found that the thickness of the EML will affect the performance of OLED, when the thickness is reduced from 75 nm to 35 nm, open from 4 V voltage reduced to 1 V, but the current density largely increased, therefore, OLED resistance and power reduction [12]. With EML, the luminescence condition is theoretically available. However, due to the low carrier mobility of organic materials, in practical flexible OLED devices, to ensure sufficient luminescence efficiency, EIL ensures the injection of carriers, ETL ensures the transmission of carriers, and HBL ensures the blocking of carriers from flowing out of the luminescence layer.

3. Fabrication of flexible OLED

Flexible OLED uses a different manufacturing technique from rigid OLED devices. A few standard methods of producing flexible OLED devices are displayed, they are the coat-debond, bond-debond, and roll-to-roll (R2R) methods.

3.1 Coat-debond method

At present, the “coat-debond” method is a common method in the mass production of flexible OLED displays. To do this, a polymer solution (usually polyimide) is coated on a virtual glass substrate and then baked to form a polymer film on the glass substrate on top of the substrate, making the bottom gas barrier layer, OLED devices (usually including active matrix arrays) and packaging structure. After the fabrication of the device, the fake glass substrate is removed by laser stripping (LLO), mechanical layering and other methods. Zhu et al. found through experiments that the mechanical debonding process between PI substrate and glass template is the key to realizing flexible OLED [13]. Additionally, the peeling layer (DBL) can be created to provide a weak, organic link between PI and glass, and then mechanical peeling, to solve the laser lift (LLO) glass surface quality is high, no defects, ensure uniform laser dose, to prevent the sticking caused by deformation and debond failure of the flexible panel. This approach produces a material with the benefits of low debond force, elimination of static charge, quick debond speed, pitch stability, and thermal conductivity.

3.2 Bond-debond method

Another method is called “bond-debond”. This approach involves adhering the flexible film to the substrate that looks like glass, whether it has a barrier layer or not. When the flexible film lacks a barrier layer, the barrier layer is generated on the flexible film on the substrate by combining it with the virtual substrate. A package structure and active matrix array are constructed for an OLED device. The ability to directly build high-performance electrical devices on flexible substrates is an advantage of this strategy. Through automated standard tool sets for flat panel displays and semiconductors, this technology makes it easier to produce flexible plastic and foil substrates. It is compatible with traditional automated tools, and it is not necessary to develop completely new machines capable of handling flexible substrates used in the manufacturing of flexible OLED light sources.

3.3 Roll-to-roll (R2R) method

Additionally, the R2R method can use in the manufacturing process of flexible OLED. Two typical R2R manufacturing techniques, albeit the process, can be altered in many ways. These methods make use of rolled flexible substrates, including ultra-thin glass stainless steel foil flexible films, etc. The

bottom electrode is prepared on the flexible roller substrate using the R2R process. In one case, the OLED device is fabricated using the R2R process and then cut into a single flexible OLED device in the other case, after the barrier layer and bottom electrode are fabricated, the roller substrate is cut into a single substrate and the cut substrate is attached to the virtual substrate and then the OLED device is fabricated. High throughput and continuous production are benefits of R2R technology. These features lead to a decrease in expected fabrication costs and capital investment. This method is suitable for manufacturing large area lighting OLED and OLED displays.

4. Conclusion

Flexible OLEDs with superior light transmission, electrical conductivity, surface uniformity and adherence, chemical stability, and low-cost mass production have been widely used in displays, lighting, and healthcare. This paper mainly introduces the structural composition and manufacturing technique of flexible OLED. A flexible OLED includes flexible OLED substrates, barrier layers, electrodes, and the organic layer. Flexible OLED substrates have to consider heat resistance, water and oxygen penetration resistance, and expansion characteristics. Barriers are used to shield the OLED components from oxidation and moisture damage in the environment. Electrode contains both positive and negative terminals and this article introduces the materials needed to make them, organic layer contains a hole transport layer, an electron injection layer, emitting layer, a hole blocking layer, a hole injection layer, an electron transport layer, and so on. The coat-debond, bond-debond, and roll-to-roll (R2R) methods for the preparation of OLED were also highlighted. In the future, the cost of manufacturing flexible OLEDs should be as low as possible, and it should be more widely used in civil equipment to make mobile phones with curved screens and folding screens, to make life more convenient. The factory can also understand the nature and requirements of each component in preparation.

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