From Structure to Performance - Enhancing the Efficiency and Longevity of OLED Devices

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Abstract. Organic light-emitting diodes (OLEDs) have emerged as the most advanced and popular display technology, offering exceptional color quality, efficiency, and design flexibility. Despite their widespread adoption in consumer electronics, OLEDs, particularly Blue OLEDs, face significant challenges such as limited lifespan, efficiency constraints, and high production costs. This paper comprehensively illustrates the operational principle, device structures, and evaluation standards of OLED technology and devices. It explains the sandwich and multi-layer structures and the physical process of carrier injection, recombination, and exciton formation. The study also delves into the challenges in depth, focusing on rapid degradation and lower energy efficiency, and discusses potential solutions, like advancements in thermally activated delayed fluorescence (TADF) materials. It underscores the need for continuous innovation in material development and device engineering to maintain OLED's competitive edge in the display technology market. Moreover, the paper underscores the promising future of OLED, especially in applications demanding superior display quality and flexibility, and highlights the technology's potential to lead the next generation of electronic displays.

Keywords: OLED, Display, Excitons, TADF.

1. Introduction

With the development of human society about to enter the 21st century, the capture, processing, storage, and display of information have been closely related to the acquisition of knowledge and the improvement of the quality of life. Since more than 70% of the information humans obtain comes from vision, the display technology closely related to vision has become an indispensable part of information technology. After decades of hard work, information display devices have gone from simple LEDs to traditional black and white, color, Cathode Ray Tube (CRT) displays, then colorful Liquid Crystal Display (LCD) and Plasma Display Panel (PDP) displays. Since 2000, the organic light-emitting diode (OLED) display technology, known as the third-generation display technology, has received significant attention from the industry because of its excellent performance, which is incomparable to other display technologies. In the rapidly evolving world of display technologies, OLED can precisely meet the needs and offer unparalleled advantages in terms of high-quality images (high resolution, extremely high contrast, high responsiveness, wide viewing angle), portability (ultra-thin, ultra-light), power consumption and efficiency, and design flexibility.

OLEDs have greatly improved the quality of life and user experience, from smartphones to large-screen televisions. Despite their excellent performance and aesthetic benefits, they also expose long-standing bottleneck problems like lifetime, manufacturing costs, and color stability problems [1, 2]. The color stability of white OLEDs also depends on the consistency of the life of organic semiconductor materials with different luminous colors [3]. In addition to designing and synthesizing organic luminescent materials with high efficiency and stability, it is also necessary to develop packaging materials with high water-oxygen barrier capacity and practical packaging methods. In recent years, new display technologies such as Mini-LED and Micro-LED have challenged the dominance of OLED in fast-changing civilian areas [4].

This paper thoroughly examines OLED technology, including its operational principles, device structures, and the role of multi-layer design in optimizing light emission and carrier balance. It explores physical processes like carrier injection, recombination, and exciton formation. It evaluates

key performance parameters such as luminance efficiency, stability, lifespan, and color accuracy, especially in the context of challenges. The research also discusses advancements in material science, particularly the development of thermally activated delayed fluorescence (TADF) materials, offering potential solutions to issues such as degradation and energy inefficiency.

2. Operation Principle of OLED

2.1. Device Structure

OLED technology has greatly developed in the past decades, and its structure has also transformed many times [5]. The efficiency and life of OLED are closely related to the device structure, and the structure widely used at present belongs to the "sandwich " structure; that is, the luminous layer is sandwiched between the cathode and the anode (one side is a transparent electrode to obtain the surface luminous effect). Due to the low film temperature of OLED, the Indium Tin Oxide (ITO) glass electrode is usually used as the anode [6]. A single or multi-layer organic semiconductor film was prepared by vacuum evaporation or spin coating on the ITO electrode, and a metal cathode was prepared on the organic film.

The materials of most organic luminescent devices are unipolar; there are very few bipolar organic semiconductor materials possessing the same transporting characteristics of holes and electrons at the same time; in most cases, only electrons or only holes can be transferred. If a unipolar organic material is used as the light-emitting material of the single-layer device, there will be an imbalance between the injection and transmission of electrons and those of holes. In the luminous region, the carrier with small mobility is prone to be injected to the electrode. If the electrode is a metal electrode, it is easy to lead to luminescence quenching, and this quenching will reduce the utilization rate of excitons, resulting in the reduction of the luminous efficiency of the device.

Due to the disadvantage of a single-layer structure, most OLED devices currently adopt a multi-layer structure. This milestone double-layer structure was first proposed by Kodak in 1987 [5]. The structure model is shown in Fig.1 [7].

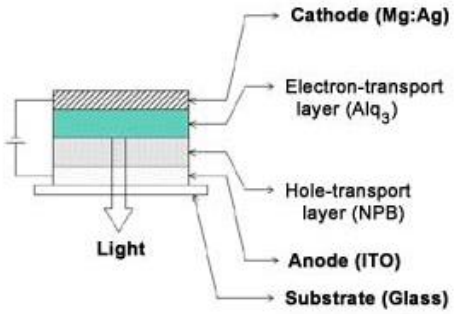

Fig. 1 Structure of double layer OLED [7]

The structure can effectively adjust the composite area of electrons and holes away from the electrode and balance the carrier injection rate, which greatly improves the luminous efficiency of the device. The main feature of this structure is that the light-emitting layer (EML) material transports electron (hole). It is necessary to add a layer of hole (electron) transport material to adjust the rate and number of holes and electrons injected into the light-emitting layer. This layer of hole (electron) transport material also plays a role in blocking the electron (hole) layer, so that the composite of injected electrons and holes occurs near the light-emitting layer.

A three-layer OLED device consists of an Electron Transport Layer (ETL), a Hole Transport Layer (HTL), and an EML, as shown in Fig.2 [8].

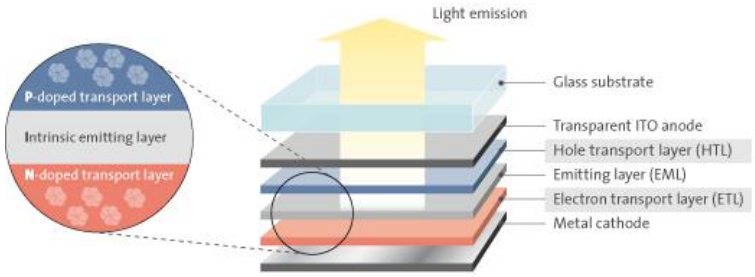

Fig. 2 Structure of triple layer OLED[8]

The structure was first proposed by Adachi research group in Japan [9]. The advantage of this device structure is that the three functional layers each perform their duties, which is very convenient for selecting functional materials and optimizing the device structure performance. In the actual OLED device structure design, in order to optimize the performance of OLED devices, give full play to the role of each functional layer, and further improve the luminous brightness and luminous efficiency of OLED, multi-layer device structure is used based on three-layer structure, excess carriers are therefore restricted and allocated. This device structure not only ensures good adhesion between the functional layers and the substrate but also makes it easier for carriers from the anode and metal cathode to be injected into the organic semiconductor functional film.

To improve the performance of devices, a variety of more complex device structures continue to appear. However, because most organic materials have the characteristics of insulation, only at a very high electric field strength (about 10 V/cm) can the carrier be transferred from one molecule to another, so the total thickness of the organic semiconductor film does usually not exceed 100 nanometers, otherwise the driving voltage of the device will be even higher.

2.2. Operation principle

Organic Electro-Luminescence (organic EL) utilizes the photoelectric functional properties of organic semiconductor materials to convert electrical energy directly into light energy. OLEDs belong to carrier injection luminescence, which forms excitons through the combination of holes injected from the anode and electrons injected from the cathode in the emission layer and releases energy in the form of light energy. It involves a series of physical processes such as carrier injection, migration, exciton formation and diffusion, which can be roughly divided into five stages, as shown in Fig.3 [10].

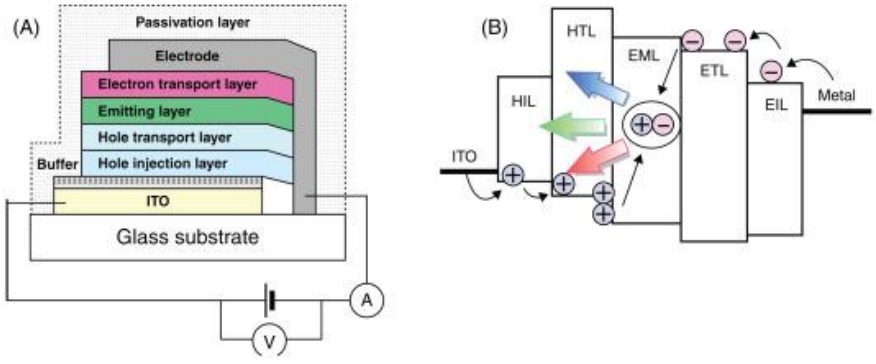

Fig. 3 (A) Structure of multilayer OLED. (B) Light emitting mechanism from an OLED device [10]

2.2.1 Carrier injection

Under the effect of the electric field, electrons and holes enter the organic functional film layer adjacent to the electrode from the cathode and anode respectively. There are 2 types of contacts between the electrode/organic layer interface: Ohmic Contacts and Schottky Contacts. Under ohmic contact, charge carriers are easily injected into the organic layer, and this is mostly the case at the anode-hole transport layer interface. Under Schottky contact, carrier injection is difficult, and most of the carriers gather at the electrode/organic layer interface, especially at the cathode/electron

transport layer interface. Richardson-Schottky (R-S) Thermionic emission theory and Fowler Nordheim(F-N) tunneling are commonly used to describe carrier injection in the basic theory and mechanism of OLED [11].

The thermal emission injection current depends on the interface barrier and the electric field and is related to temperature [12]. Carriers must have enough energy to overcome the organic/metal interface barrier to enter the organic layer, which is described mathematically:

$$J_{RS} = A^{*}T^{2}\exp\left(-\frac{E_{\Phi}}{KT}\right) \tag{1}$$

$$E_{\Phi} = E_{v} - E_{F} \tag{2}$$

Where J is the emission current density, A* is the Richardson constant, E is the escaped work, K is the Boltzmann constant, and T is the temperature.

Under a high electric field, the tunneling current of the barrier generally follows F-N rule [13]. When an electric field is applied between the electrode and the organic layer, the band will tilt and bend, and the injection of charge carriers is like passing through a wall. The current density described by F-N can be expressed as:

$$J_{FN} = BE^{2}\exp\left(\frac{-b}{E}\right) \tag{3}$$

$$b = \frac{4\sqrt{2m^{*}}}{3\hbar}x^{3/2} \tag{4}$$

Where m* is the effective mass of the electron, e is the unit charge, E is the electric field, x is the barrier height, and B is the preexponential factor.

2.2.2 Carrier mobility

Carrier mobility is one of the important parameters measuring the carrier transport ability of organic films. The molecular orbitals in organic semiconductor materials rarely overlap, and the band structure of organic semiconductor materials is mostly localized and discontinuous. The carrier mobility mainly adopts the jumping mechanism, so its mobility is much lower than that of inorganic semiconductor materials with a continuous band structure. In general, organic semiconductor materials are low-conductivity materials.

2.2.3 Carrier recombination and exciton formation

In the emission layer, the holes and electrons meet and recombine to form high-energy state excitons. In inorganic semiconductor materials, excitons are spatially confined because of the high delocalization of carriers, leading to low exciton binding energy, and thus can exist at low temperatures [14].

In contrast, in organic semiconductor materials, due to higher binding energy, excitons can stably exist at room temperature. When excitons return to the ground state in a radiative transition, EL occurs, and the emission color is determined by the bandwidth of the luminous material itself.

After the carrier recombination, two kinds of excited states will be formed due to the different spin configurations of electrons. One is the singlet excited state formed by non-spin-pairing ground state electrons, which can release energy back to the ground state in the form of fluorescence. The other is a triplet excited state formed by spin-paired ground state electrons, which usually cannot release energy back to the ground state in the form of fluorescence. Both singlet and triplet excitons are produced at the same time. However, according to theoretical estimates and experimental verification, the ratio of singlet to triplet excitons is about 1:3, which means only about 25% of the excitons can emit fluorescence, while the remaining 75% can only be non-radiative and transited back to the ground state [15]. Therefore, making full use of triplet excitons and reducing non-radiative transitions are important directions of OLED research and development.

2.2.4 Drift and diffusion of excitons

Exciton is in an unstable excited state once it forms, and will diffuse and drift in the emission layer. The distance of exciton diffusion and migration is about 20~30 nm, the lifetime of singlet excitons is about 1~10ns, and the lifetime of triplet excitons is on the order of micro- or even milliseconds, that is, triplet excitons are more easily captured by defect levels and deactivated [16].

In many cases, exciton diffusion and drift are unfavorable to device performance. For example, when there is a quenching center or electrode quenching, it will lead to exciton deactivation. Therefore, it is ideal that the exciton recombination region is located in the center of the emission layer.

2.2.5 Luminescence of excitons by radiative transition

If the exciton is not captured by defect levels or quenching centers during its lifetime, it returns back to the ground state by consuming its energy through radiative transition, resulting in EL, and the luminous color is determined by the energy level difference from the excited state to ground state [17].

As shown in Fig.4, the total spin of singlet excitons is zero, and the radiative transition process of the electron from the singlet S1 to the ground state S0 is called fluorescence radiation (labeled F); the total spin of triplet excitons is not equal to zero, and the radiative transition process from triplet state T1 to S0 is called phosphorescent radiation (labeled P) [18]. Although the ratio of triplet excitons to singlet excitons is generally 3:1, under normal circumstances, the triplet excitons of most luminescent materials have very low luminescence efficiency, because the transition from the triplet state to the ground state is spin-forbidden.

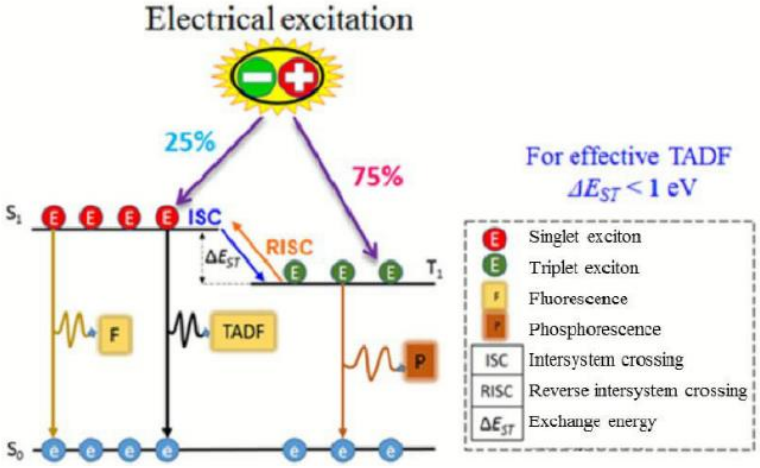

Fig. 4 Physical process of excitation [18]

When the exciton transitions to a lower state without emitting a photon, the process is called non-radiative transition. Non-radiative transition includes two types: internal conversion and intersystem crossing (ISC). Internal conversion refers to the process where the exciton returns to a lower potential surface of the same spin multiplicity by losing energy; ISC refers to the process where an exciton transitions to lower potential surface of a different spin multiplicity by losing energy.

By special methods in specific material, S1 and T1 can be strongly coupled, allowing ISC between the two levels, and the energy difference between the two states is made to be much smaller than typical organic molecules. This smaller energy gap enables reverse intersystem crossing (RISC) to occur, where excitons in the T1 are converted to S1 in a thermally activated process. The excitons in S1 state can therefore decay back to S0 and release fluorescence. Since the RISC process is slow, the fluorescence from the initial triplet excitons occurs later than the fluorescence from the excitons initially created in S1. The whole process is called Thermally Activated Delayed Fluorescence (TADF). By the mechanism of TADF, the luminescence efficiency is increased compared to that of phosphorescence.

3. Performance Evaluation

The basic performance parameters of OLEDs mainly include luminous color and chroma, luminous brightness and efficiency, stability and life and others.

3.1. Spectrum and color coordinates

The valence electron structure of organic EL material determines the absorption spectrum and fluorescence spectrum of the material, and also determines the EL spectrum of the device. Luminous spectrum is the dynamic distribution of luminous energy according to wavelength, frequency or wave number, usually expressed by the relative value of light energy according to the distribution of wavelength Φ (λ).

In organic EL research, there are usually two kinds of luminous spectra of organic materials and OLED devices: photo-luminescence (PL) spectrum and electro-luminescence (EL) spectrum. PL spectrum is excited by an external monochromatic light source with constant excitation wavelength and intensity, which is used to characterize the energy transfer process or other characteristics of the material [19]. The EL spectrum is mainly excited by electric energy, and the EL spectrum of devices with different current densities can be obtained. By comparing the EL spectrum and PL spectrum of the devices, the location of the composite region and the relevant information of the actual luminous substance of the OLED can be obtained. EL spectrum can be used to characterize chroma of the device. Chroma is quantified in International Commission on illumination (CIE) 1931 Color Coordinates, as shown in Fig.5 [20].

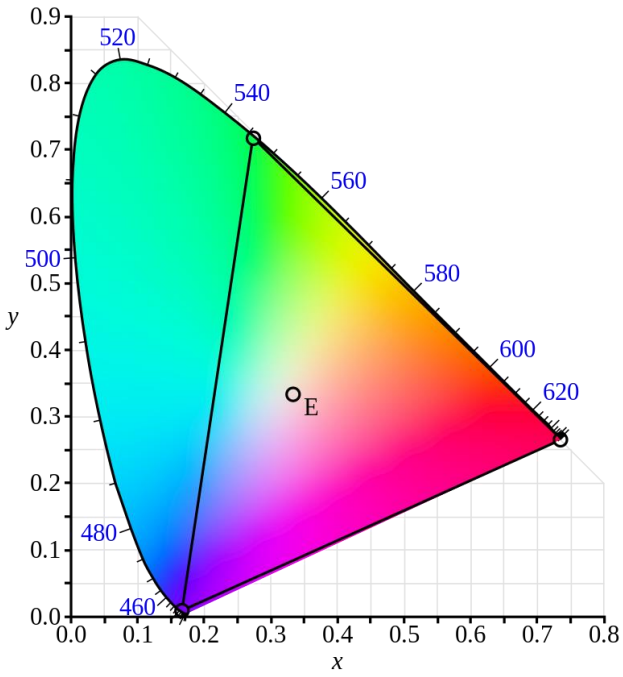

Fig. 5 CIE 1931 chromaticity coordinates of spectrum [20]

3.2. Luminance brightness and threshold voltage

Luminance is a physiological physical quantity, which is not only related to the radiation energy of the device when it emits light, but also related to human vision. Luminous brightness is the luminous intensity per unit area perpendicular to the beam propagation direction in the unit of nit. The luminescence distribution of OLED follows a cosine distribution, that is, brightness is a constant independent of direction.

Threshold voltage is defined as the driving voltage applied to a device of 1-nit brightness, also called Turn-on voltage. It is often considered an indicator for the energy performance of a luminous device.

3.3. Stability and life time of device

The stability and device life of OLED are the key parameters that restrict the commercial practical application. OLED device life time includes two parts: working life and storage life. For OLED devices put into the market, the working life is required to reach more than 10000h under continuous use conditions (uninterrupted use for more than one year), and the storage life is greater than 5 years [21].

The working life of the OLED device refers to the continuous working time of the device when the luminance of the device drops to half of its initial brightness under constant current density. The accepted starting brightness of the measurement service life is 100cd/m. Generally, the initial brightness of the device under test is several times the brightness value, and the life value when it is reduced to half brightness is measured, and then multiplied by the corresponding multiple, that is, the working life of the device.

3.4. Efficiency

The luminance efficiency represents the ability of converting electric energy to luminous energy [22]. Quantum Efficiency (QE) precisely describes the degree of excellence of the LE mechanism. QE is divided into internal quantum efficiency (IQE, η_{int}) and external quantum efficiency (EQE, η_{ext}). IQE is defined as the ratio of photons produced to the number of electrons injected, while EQE is the ratio of photons emitted from the device surface in the observation direction to the number of electrons injected.

$$\eta_{int} = \frac{N_{int}}{N_c} \tag{5}$$

$$\eta_{ext} = \frac{N_{ext}}{N_c} \tag{6}$$

$$\eta_{ext} = \eta_{int}(1 - \sqrt{1 - \frac{1}{n^2}}) \approx \eta_{int}\frac{1}{2n^2} \tag{7}$$

In the formula, n is the refractive index of luminescent material, typically ranging from 1.5 to 2. In other words, IQE is 4 to 8 times EQE. Evidently, the majority of light produced by the device is not emitted externally. Since the OLED have a multilayer structure, the light energy emitted from the EML is lost due to waveguide effects or reabsorption due significant difference in the refractive indices of luminescent materials, ITO anode and substrate. During light emission, most of the light energy is reflected back and absorbed by the device materials. Only a portion of light transmitted through the edges of the device. Therefore, it is crucial to enhance optical coupling for improvement of luminous efficiency of the device.

Apart from the characteristic of material transportations of carriers in transport layer and emission layer also affects the luminous efficiency: (1) limitations of quantum mechanics principles; (2) non-radiative decay of singlet excitons; (3) imbalance of electrons and holes during the injection process.

Considering these factors, a high-performance OLED device must meet four conditions: (1) a high fluorescence efficiency; (2) It should enhance and balance carrier injection efficiency to achieve a higher exciton formation rate; (3) Exciton recombination zone is away from the organic/metal electrode interface to reduce exciton quenching caused by the electrodes; (4) a high optical coupling output efficiency.

4. Challenges, progress and solutions

Fluorescent and phosphorescent OLEDs are two main types of OLEDs. In real applications, fluorescent OLEDs refers to the first-generation OLED technology, while the phosphorescent OLED (PhOLED) is considered to be the second generation. However, both fluorescent OLED and PhOLED have to face a series of challenges, especially for blue OLED [23].

4.1. Life time and degradation

The biggest problem for OLEDs is the limited lifetime. In real application and test, dark spots would eventually form on the screen over time, which means the cells are malfunctional.

This phenomenon is the degradation of organic luminescent material [24]. It occurs due to the accumulation of non-radiative recombination centers and luminescence quenchers in the emissive zone. Degradations can be reduced fundamentally by material improvements, improved electroluminescent mechanism, and device structure optimization.

The degradation is not only a challenge for material, but also packing techniques. OLED luminescent materials are extremely sensitive to oxygen and moisture, when being exposed to oxygen or moisture, the organic materials quickly degrade as they oxidize, generating black spots pixel by pixel or even lead to delamination of the electrode layer.

Currently, the most popular packing techniques is Thin-Film Encapsulation (TFE), especially organic/inorganic TFE [25]. The major advantage of this approach, particularly the integration of both organic and inorganic layers, is that it combines the flexibility and barrier properties of organic materials with the robust moisture and oxygen barrier capabilities of inorganic materials. This hybrid structure significantly enhances the durability and operational lifetime of OLED devices, making them more suitable for flexible and wearable applications.

4.2. Color imbalance (color shift)

The cause of color shift is the different degrees of degradation of different color cell. Blue light has a higher photon energy compared to red or green light. To emit blue light, the organic materials must handle higher energy transitions. These high energy transitions can lead to more rapid degradation of the organic materials, as the molecules are subjected to greater stress and break down more quickly. Blue light output will decrease relative to the other colors of light, so the screen usually has a red shift or green shift over time.

Most commonly, manufactures optimize drive technology and control algorithm, such as AMOLED, and optimize the size and arrangement of regular RGB subpixels to reduce the current density through a single cell to equalize lifetime at full luminance. Fig.6 shows RGBG pentile pixel of Samsung S20 AMOLED screen [26].

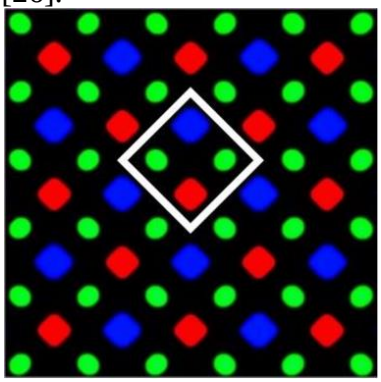

Fig. 6 RGBG pentile pixel of the Samsung S20 AMOLED screen [26]

4.3. Energy efficiency

As detailed above, the theoretical IQE limit of fluorescent is about 25%, while that of PhOLED can be 100%. In fact, red and green phosphorescent material can achieve close to 100% IQE, and they have been used in mass production applications on OLED display. However, blue phosphorescent materials have always exposed weaknesses in terms of color purity and longevity. In the commercial OLED display, fluorescent instead of phosphorescent materials are commonly used as compromise.

Thermally Activated Delayed Fluorescence (TADF) was adopted to solve the issue of low efficiency and instability of blue phosphorescent material. The advantage of TADF materials is that

the IQE is close to 100%, which is much higher than 25% of the first generation of fluorescent materials, and there is no problem that the second generation of phosphorescent materials need to use heavy metals and have high costs, so TADF materials are known as the third generation of OLED materials. TADF materials generally use donor-acceptor structures with high torsion angles to achieve a small ΔE_{ST}, but this also leads to strong Intramolecular Charge Transfer characteristics, causing the emission full-width at half-maximum (FWHM) to widen.

To overcome this issue, Professor Takuji Hatakeyama's team at Kwansei Gakuin University in Japan proposed a blue TADF material containing B and N atoms called DABNA, which has a small ΔEST, high efficiency, but no donor-acceptor structure, resulting in a narrower FWHM. The multiple resonance effects of boron and nitrogen atoms not only minimize the vibrational coupling between the S0 state and the S1 state but also minimize the energy gap between the S1 state and the T1 state. The electroluminescent wavelength of this material is 469nm, with an FWHM of 18nm, a maximum EQE of 34.4%, and an EQE of 26.0% at 1,000 nits, which was remarkably higher than blue OLED ever reached [27].

5. Conclusion

This paper has provided an in-depth exploration of OLED technology, explaining its remarkable advantages in display applications, such as superior color quality, energy efficiency, and design flexibility. However, it has also underscored the significant challenges that persist, particularly with Blue OLEDs. The issues with lifetime and efficiency continue to hinder widespread adoption. Through detailed research of these challenges, it becomes clear that OLED technology has made substantial development, the path forward is fraught with technical obstacles and requires innovative solutions.

Future prospects for OLED technology are bright. Ongoing research focuses on enhancing material stability, improving manufacturing techniques, and expanding the scope of applications. Particularly for Blue OLEDs, the development of TADF points out the direction to address primary limitations. As the industry continues to develop, the adaptations and innovations of OLED will be significant in maintaining its leading position in the whole display technology area. The exploration of OLED's potential and solutions to its challenges will not only enhance its performance but also expand its applicability in the continuously growing market of electronic displays.

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