Smart phones and televisions that use organic electroluminescence display are coming into use and “organic EL or OLED” has become a familiar term. OLED can reproduce high-quality images and thus hold promise for next-generation displays. Japan has produced much of the leading-edge technology, and Japanese companies introduced the world’s first OLED displays to the market in 1997. Now, 20 years later, we have reached the point where mass consumer electronic merchandisers are marketing high-end OLED televisions. For OLED TVs to enter true widespread use, however, a number of challenges must be overcome. Here, we describe the most recent OLED technology and explain the challenges that stand in the way of further market penetration.

1. Introduction

The term organic electroluminescence (EL) originally referred to the phenomenon in which an organic material emits light when an electric current passes through it, but now it is most often used to mean an electronic device that exhibits this phenomenon. Light emission by organic EL was first reported in the middle of the 20th century, and the basic device structure was established early in the 1980s. After about 30 years of technical progress, practical 50-inch or larger models of large-screen displays became available. Organic EL devices or OLED (organic light-emitting diode) emit light themselves, so they are thinner, lighter, have higher contrast and response rates, and other superior qualities compared with LCDs, which require backlighting, so they hold promise as next-generation large-screen displays. As TV displays, 11-inch models of practical organic EL TVs became available in 2007, followed by 15-inch models in Europe and elsewhere in 2010, and 55-inch models of organic EL Hi-Vision TVs appeared in 2013. Now, in 2017, 65-inch or larger models of OLED TVs that offer 4K resolution are being sold by various companies. This progress in the development of practical OLED TVs in recent years has given users the opportunity to compare their quality with conventional displays, and although the excellent image quality of OLED TVs is gradually increasing their popularity, cost is still hindering large-scale adoption.

New markets for OLEDs are also being explored, including applications that use unique features of these devices that differentiate them from other light-emitting devices. For example, the feature of surface light emission with easy dimming makes them suitable for museum lighting applications, the flexibility of shape is useful for high-end interior lighting, and the particular softness of the light from surface-emission light sources is an advantage in illumination for nighttime rounds in hospitals.

Another application is as flexible displays. The structure of OLED consists of layers of organic thin films, enables the construction of ultrathin displays, and is optimum for realizing a flexible display that is highly convenient in use because of features such as high portability and ease of storage and installation. In our R&D, we are targeting a next-generation broadcast service that provides a sense of presence with a higher quality by using these characteristics of OLED displays. One objective is to realize a thin and lightweight sheet-type display for a large TV screen for home viewing of 8K Super Hi-Vision broadcasting (which we refer to as simply 8K). Test satellite broadcasting of 8K began in 2016 and the regular service will begin soon in December 2018. We are also studying a display for a hand-held television that would enable viewing anywhere at any time, would not break even if dropped, and is easily storable.

Considering these latest trends, the following sections describe the principle of OLED, which has seen remarkable technical progress, and the problems it faces. Technical trends concerning materials and devices are also explained.

2. The Principle of OLED and Issues for OLED TV

2.1 OLED principle

The structure of OLED involves organic thin films sandwiched between two electrodes. When a DC current is applied to the electrodes, holes (+) and electrons (−) are injected from the electrodes and recombine in the light-emitting layer, producing light emission. The basic device structure is a transparent positive electrode (ITO),...
etc.) formed on a glass substrate, a hole injection layer\(^*2\), a hole transport layer\(^*3\), a light emission layer, an electron transport layer, an electron injection layer, and a negative electrode formed on top [Fig. 1 (a)]. The injected charges are efficiently moved to the light emission layer, where they recombine to emit light [Fig. 1 (b)]. To improve efficiency, the device performance can be optimized by selecting appropriate materials for the layers surrounding the emission layer and by adjusting properties such as the layer thickness. The color of the emitted light depends largely on the color of the emission layer material. The organic layers are from 30 to 50 nm thick and the total thickness, including the electrodes, is several hundred nanometers.

A device with the structure shown in Fig. 1 (a) emits light towards the substrate at the bottom, so it is called a bottom emission structure. A top emission structure that emits light from above the substrate is also used. In the top emission structure, the bottom electrode is formed as a reflective layer made of a silver alloy or a similar material and the top electrode is formed of a thin silver alloy or transparent ITO.

There are two types of light emission by OLED: fluorescence and phosphorescence. Fluorescent organic materials have been used since the beginning of development, but phosphorescent organic materials appeared in the late
1990s, and devices that use nearly 100% of the injected charges have been implemented. In recent years, phosphorescent materials have been used for OLED with red and green emission.

The differences between fluorescence and phosphorescence are illustrated in Fig. 2. The charges (holes and electrons) move to the light-emitting material, where they recombine and form excitation states of two energies [Fig. 2 (a)]. Molecules in these energy states are called excitons. Of the two types of excitons, singlet excitons are more readily emit light by fluorescence, and triplet excitons more readily lose energy as heat (molecular vibration, etc.) without emitting light, a process referred to as nonradiative deactivation. The two types of excitons are produced in a proportion of 3 to 1, with singlet excitons accounting for 25% of the total and triplet excitons accounting for 75%. Phosphorescent emitters can use triplet excitons to emit light, and there have been many reports of triplet exciton phosphorescence in organometallic compounds that include iridium, platinum, and osmium. Such phosphorescence is a result of the heavy-atom effect in which there is rapid light emission caused by iridium or another heavy metal. In phosphorescent materials, intersystem crossing (ISC) is promoted by the heavy-atom effect and singlet excitons are easily converted to triplet excitons. This enables 100% of the applied charge to be used in phosphorescence. In practical application, however, the development of fluorescent emitters that have a long device driving lifetime is moving forward, but phosphorescent devices that have a practical lifetime, and blue devices in particular, have not yet been developed.

2.2 Challenges for OLED TVs

The characteristics of OLED TVs and LCD TVs are presented in Fig. 3, where the relative values for each item are indicated by the position from the center to the outside, with the lowest value at the center and highest value on the outer edge. For image quality and degree of freedom of the shape on the right side of Fig. 3, OLED devices are advantageous. Although the performance of OLED as display devices is high, LCD devices are superior with respect to manufacturability and other cost factors shown on the left side of the figure.

Concerning image quality, the contrast ratio of LCD TVs has been improved to 3000:1 by adopting backlighting control technology. However, the black level of TVs that use OLEDs, which themselves emit light, can be infinitely high, depending on the manufacturer, so the contrast ratio as well as the response and viewing angle of OLED TVs are superior. Although the black level of LCD TVs has been improved by distributing many LEDs over the panel to provide

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4 A molecule whose excited-state electron is in the singlet state (one type of spin state, which is a magnetic property of the electron).

5 A molecule whose excited-state electron is in the triplet state (one type of spin state, which is a magnetic property of the electron).

6 The phenomenon in which intersystem crossing (see note 7) occurs more easily for heavy atoms that have high atomic numbers.

7 The phenomenon in which there is a change between the singlet state and the triplet state in the spin of electrons in molecules that are in an excited state.

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Figure 3: Characteristics of OLED TVs and LCD TVs
backlighting and controlling them, there cannot be as many LEDs as there are pixels, so unevenness in backlighting and leakage of light into the periphery at low brightness are fundamental problems. Accordingly, OLEDs can be considered to be effective for images that include many dark scenes, such as movies, because of their ability to produce natural color tones even at low brightness.

Concerning color reproduction, both types of displays have achieved a color reproduction range sufficient for Hi-Vision TV, although problems remain for the wide color gamut of 8K.

The response (the time required for a change in luminance) of an OLED is no more than a few microseconds, whereas the response of a liquid crystal device is determined by the movement speed of liquid crystal molecules and ranges from several milliseconds to several tens of milliseconds. While the LCD response is sufficient for Hi-Vision and other video where the frame rate is 60 Hz and the frame interval is 16.6 ms or less, the response of OLEDs is at least three orders of magnitude faster than that of LCDs.

Next, we consider the structure of displays. The OLED device itself is extremely thin (approximately 0.5 µm including electrodes), so the display thickness depends mainly on the thickness of the substrate (glass or film, etc.), and the thickness increases if a surface protective film or a color filter is used. The 11-inch models of OLED TVs sold in 2007 had a thickness of 3 mm and the models currently being sold are 5 mm thick. The display can be made even thinner by using plastic film for the substrate instead of glass, and a bendable TV or a roll-up TV could be implemented. LCD TVs, on the other hand, require many other components, such as a glass substrate, backlighting, a polarizing plate, and color filters, so it is difficult to make them thinner. Current commercial LCD TVs are much thinner than previous models, but they still have an average thickness of about 10 mm. Also, because the performance of LCDs is sensitive to the distribution of the thickness of the space in which the liquid crystal material is sealed (several micrometers), the device characteristics change when the display is bent or pressed forcefully. OLEDs, on the other hand, are robust against deformation and impact and thus suitable for flexible display applications. LCDs are advantageous for enabling a large screen size, but 77-inch widescreen OLED TVs are also available on the market.

While OLED TVs were very expensive a few years ago, the production yield has greatly improved and prices have fallen sharply. For example, 55-inch OLED Hi-Vision TVs are now available for 200,000 yen or less. Another factor is that there is currently only one major manufacturer of OLED panels, and a future increase in the number of manufacturers should lead to lower prices. On the other hand, OLED power consumption is currently about 1.5 times higher than that for LCDs, although OLEDs have the potential to achieve a power consumption of about half that of LCDs. For example, a 55-inch commercial OLED TV consumes 400 W or more, which is a major obstacle to widespread adoption. One reason for the high power consumption is that some fluorescent materials that contribute only 25% of the injected charge to light emission are used. Another reason is that two-thirds of the emitted light is wasted when RGB (red, blue, green) three-color filters are used with white-emitting OLEDs. Ideally, low power consumption can also be achieved for large-screen TVs by using phosphorescent materials or other materials that have nearly 100% light emission efficiency and are individually colored red, green, and blue. The next chapter explains the most recent trends in OLED development, including increased material efficiency.

3. Trends in OLED Devices
3.1 Development of materials for higher performance and lower cost

In section 2.1, we described OLED devices that use fluorescence and phosphorescence as the most basic types of OLED devices. Fluorescent devices use fluorescent materials to emit light and are referred to as first-generation OLEDs; their R&D has progressed since the late 1980s. The facts that only singlet excitons contribute to light emission in fluorescent emitters and that the singlet exciton production rate is 25% establish an upper limit on light emission efficiency (internal quantum efficiency)

\[ \text{\textsuperscript{8}} \text{ The ratio of the number of photons generated within the emitting device to the number of carriers injected into the emission layer.} \]
many years, and these materials have a long life and relatively low cost. Nevertheless, light emission efficiency remains low because only one-quarter of the applied charge is used.

The features of each generation of OLED materials and devices are shown in Fig. 4. To improve the efficiency of fluorescence, highly efficient phosphorescent emitters have been developed (upper right of Fig. 4). Because phosphorescent emitters can use triplet excitons to reach an emission efficiency of 100%, as explained in section 2.1, the energy-saving effect is large. Devices including phosphorescent emitters are referred to as second-generation OLEDs. However, these materials are expensive because they use iridium or other noble metals.

Consequently, in recent years, attention has turned to the third generation of OLEDs (upper left of Fig. 4). The third-generation devices feature the use of thermally activated delayed fluorescence (TADF) emitters instead of noble metals for light emission. TADF emitters are fluorescent materials in which the energy levels of the singlet (S1)*9 and triplet (T1) excitons are very close together (a difference of 0.2 electron volts or less)*10, as shown in Fig. 5. This opens up the possibility of obtaining fluorescent light emission from both singlet excitons and triplet excitons by upconverting the energy of triplet excitons to singlet excitons, thus converting nearly 100% of the electrical energy to light emission. The conversion of triplet excitons to singlet excitons creates a delay in fluorescent light emission of several microseconds to several tens of microseconds, which is why the process is referred to as “thermally activated delayed fluorescence”. The development of the most recent high-performance TADF materials is being accelerated by using computational chemistry. There are also reports of materials whose light extraction efficiencies are improved through material designs that incorporate control of the light emission direction by adjusting the molecular orientation*11 or other such means*9. The result is a clearly higher device performance in terms of external quantum efficiency*12 relative to materials that have no orientation control.

However, difficult problems in attaining color purity and a longer device lifetime remain in the application of TADF materials.

*9 Level refers to the energy of the electron orbit.
*10 electron volt: a unit of energy.
*11 Regular arrangement of organic molecules.
*12 The ratio of the number of photons radiating to the outside of the emitting device to the number of carriers injected into the emission layer.
emitters to display devices. To address these problems, research to establish a new method of extending the device lifetime using TADF materials has been progressing in recent years (Fig. 6). Such a method achieves higher fluorescent device efficiency by mixing a TADF material with a fluorescent material used in conventional devices that features both a long lifetime and good color purity (Fig. 6). As shown in the upper right of Fig. 6, energy is passed on to the TADF material from the host material, which carries electrical energy, and all of the energy within the TADF material is converted to singlet excitons. Then, all of the energy is passed on to the conventional fluorescent emitter (the material on the far right of Fig. 6) and 100% light emission is obtained. This device is also referred to as a fourth-generation OLED (upper center of Fig. 4). Because the TADF material compensates for the low efficiency, which is the weak point of fluorescent devices, the TADF material is referred to as an “assistant dopant” (a material added to assist a function). However, the difficulty of controlling the mixing ratio of the materials to achieve efficient energy delivery remains a problem.

Concerning long-lifetime devices with TADF, our labora-

![Figure 6: Principle of fourth-generation devices with TADF materials](image)

![Figure 7: Principle of a phosphorescent device with TADF material (generation 2.5)](image)
tory developed a highly efficient device whose lifetime approaches a practical level by using a TADF material rather than a luminescent emitter as the host material and a phosphorescent material for light emission (Fig. 7)⁸. Using the TADF material as the energy (charge)-carrying host material results in a simpler device structure. We clarified longer lifetime can be achieved using TADF materials that have smaller molecular sizes⁹. Because this phosphorescent device combines third-generation TADF materials that are expected to become practical in the future and second-generation phosphorescent materials that are already practical, it can be referred to as a 2.5-generation devices. Because this method can reduce the amount of expensive phosphorescent material used in existing practical phosphorescent devices by 80 to 90%, the cost of these devices can be greatly reduced. Accordingly, this technology is expected to become practical in the near future and is promising for obtaining future third-generation and fourth-generation devices that do not use noble metals.

### 3.2 Improving device stability in air

In ordinary OLEDs, alkali metals or other materials that are easily degraded by water or oxygen are used for the electron injection layer, so the devices must be sealed to prevent degradation. In particular, when a plastic film substrate is used, the substrate may be permeable to oxygen or water, so an expensive multilayer structure of organic and inorganic materials must be formed as a barrier to such intrusion. However, forming such a sophisticated barrier involves problems such as low productivity and the difficulty of fabricating large sealing layers without defects. It is therefore desirable to make the device itself stable in air to produce OLED that use a plastic film substrate.

To achieve the above goal, we are reviewing conventional electron injection materials that are susceptible to degradation by air and water, and moving forward with R&D on OLEDs that have high emission and are also stable in air¹⁰⁻¹³. For example, an OLED that has an inverted structure and uses air-stable zinc oxide or a similar material for the injection layer has attracted attention and its R&D is in progress. In the inverted OLED, the cathode and anode are reversed from the usual top and bottom locations [Fig. 8 (a)]. The inverted structure makes it possible to use zinc oxide or a similar material that is stable in air for the electron injection layer. However, zinc oxide and other metal oxide layers are formed by sputtering¹³, and the light-emitting layer and other organic material layers in the usual structure are damaged by the plasma used in the sputtering process. With the inverted structure, however, the zinc oxide electron injection layer can be formed before the organic layers, thus averting damage to the organic layers.

*¹³ A film-forming method in which a material is bombarded with accelerated ions. Particles of the material are scattered and adhere to the substrate to form the film.

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<th>Emission area during continuous operation in air (unsealed(exposed in air))</th>
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<td>Just after fabrication</td>
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<tr>
<td><strong>Inverted OLED</strong></td>
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**Figure 8:** Inverted OLED and its stability in air
The high resistance to oxygen and water of OLEDs that have the inverted structure has been verified by NHK and Nippon Shokubai. In 2014, it was shown that there was no degradation in an inverted OLED within 250 days of storage, whereas the emission area of an ordinary OLED with simple sealing decreased by about half after 100 days of storage. Furthermore, a recent comparison of continuous operation in air for an OLED that has the conventional structure and an OLED that has the inverted structure shows good stability during operation in air and improvement of the device lifetime for the inverted structure (Fig. 8 (b)).

A major problem with the inverted structure, however, is the need to improve the electron injection; therefore, metal oxides other than zinc oxide are being developed as materials. For example, improved electron injection has been obtained by using an electrode material composed of calcium and aluminum oxides and having a small work function.

There have also been attempts to improve the stability in air of ordinary OLEDs, and improvement achieved by using a gold and magnesium alloy for the cathode has been reported.

As described above, the improvement of stability in air is being well researched and good results for the lifetime as well as the emission efficiency are beginning to appear. These results are obtained using innovative technologies that will facilitate the management of moisture and oxygen in manufacturing processes and greatly reduce the cost of barrier films and other such components, hence we will continue to develop them.

3.3 Development of OLEDs for wide-color-gamut displays

A wide-gamut color system that features high color reproducibility has been adopted as an 8K standard that can provide high definition and a high sense of presence and has been standardized as Recommendation ITU-R BT.2020. High efficiency and a long service life have long been the subject of R&D toward solving the main problems for OLEDs, and we are finally at the stage where materials that exhibit a practical level of emission performance have begun to appear on the market. Because such development involved material systems based on the Hi-Vision color system (Recommendation ITU-R BT. 709), the development of materials that satisfy BT.2020 is now required.

R&D on OLEDs with a wide color gamut recently began, and success was finally achieved by improving color purity through the use of color filters and technology for extracting only particular wavelengths from a device with a top emission structure (referred to as a cavity structure).

In addition to improving color via the utilization of such device structures, R&D on improving the original color purity of the most recent highly efficient materials such as phosphorescent materials and TADF materials has begun. For example, materials that have a structure in which the bonds are spread out in a planar mesh (Figs. 9 (a) and (b)) easily provide color purity with narrow bands in the emission spectrum compared with the conventional iridium complex. A blue TADF emission material that contains boron is shown in Fig. 9 (a) and a green emission material that contains platinum is shown in Fig. 9 (b). In particular, a device using the green phosphorescent emitter produces a green color that is close to the specification in BT.2020. In addition, there have been many reports on high-color-purity devices that use quantum dots, perovskite crystals, or other such materials as luminescent dyes.

4. Conclusion

OLED display devices have a fast response, high contrast, and other excellent characteristics for video. Although a short device lifetime and low production yield were major problems a few years ago, the lifetime is now at a practical level and manufacturability has improved, so the production of next generation of light-emitting devices is well researched.

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*14 The minimum energy required to remove one electron from the surface of a substance to an infinite distance.

*15 A complex is a metal atom that is surrounded by molecules that are arranged at fixed positions.

*16 A substance composed of semiconductor nanoparticles that have diameters of several nanometers and which exhibits characteristic light absorption or emission. The light wavelength depends on the particle diameter, so pure color emission can be reproduced by reducing the dispersion in the particle size.

*17 A material that has a crystal structure that is typified by a perovskite and has attracted attention as a material for high-temperature oxide superconductors and solar cells.
tion of large-screen OLED displays is possible and even 60-inch or larger OLED TVs are available in consumer electronics stores.

Having reached this stage in OLED development, even more innovative technology for materials and devices that will enable a larger screen size, longer device lifetime, lower cost, and flexible display devices is being developed.

As a result of future development, we can expect technology for lower power consumption and a wider color gamut in addition to flexibility. At our laboratory, we are researching fundamental technologies toward the realization of a sheet-type display for a new form of TV that will be essential for achieving widespread adoption of 8K home viewing.

References
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