Improvement of Current-Voltage Characteristics in Organic Light Emitting Diodes by Application of Reversed-Bias Voltage

To cite this article: Dechun Zou *et al* 1998 *Jpn. J. Appl. Phys.* **37** L1406

Improvement of Current-Voltage Characteristics in Organic Light Emitting Diodes by Application of Reversed-Bias Voltage

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(Received September 2, 1998; accepted for publication September 25, 1998)

The effects of reversed-bias application on current-voltage and luminance-voltage characteristics of standard-type double-layer organic light emitting diodes [OLEDs], ITO/TPD(50 nm)/Alq_3(50 nm)/Mg:Ag, were investigated. Both the magnitude of reversed-bias and the duration of reversed-bias application were systematically changed. Evident voltage shifts towards the lower voltage side in current-voltage and luminance-voltage characteristics were observed in the diodes which were treated under various reversed-bias condition. The longer the duration of reversed-bias application, the larger the voltage shift was. A maximum voltage shift of 0.7 V was observed in the diode treated under a -10 V bias for 3 h. Little change in luminance-current density relationships was observed for diodes which were treated under various reversed-bias conditions. The results were interpreted in terms of the movement of ionic impurities and orientational rearrangements of permanent dipoles in organic layers.

KEYWORDS: organic electroluminescence, reverse bias, voltage shift, internal field, external field, enhancement

One of the most important subjects related to the practical applications of organic light emitting diodes (OLEDs) is the improvement of device durability. Durability of OLEDs is generally dependent on many factors, such as device structures, organic materials, electrode materials, processing conditions, driving methods and so on. Extensive research has been performed on these various aspects.[1–8] Among them, a driving scheme has recently been considered to be one of the most important factors for the improvement in the performances of OLEDs. This aspect was not seriously considered earlier, because DC driving was considered to be one of the major advantages of OLEDs. Indeed, DC driving is one of the essential features of OLEDs which discriminates them from inorganic AC electroluminescent devices.

Two important issues, which cannot be disregarded concerning the driving modes of OLEDs, should be pointed out: one is the coexistence of a high electric resistance and a high charge mobility in organic materials used in OLEDs.[9,10] This class of organic materials are basically dielectric insulators. Even in such insulating materials, small amounts of ionic impurities, which may migrate within insulating materials, exist no matter how carefully they are synthesized and purified. The other is that the electric field applied to OLEDs is extremely high, typically on the order of 10^6 V/cm. It is assumed, therefore, that ionic impurities move slowly in the presence of high applied electric field and hence form an internal electric field in the opposite direction to the applied field, despite the fact that their mobilities are extremely low compared with those of holes and electrons.

It is well known that a constant current driving mode can achieve a longer lifetime than a constant voltage driving mode. Especially, a pulsed driving mode combined with a reversed-bias component improves the lifetime of OLEDs.[11] Little is known, however, about why the driving mode influences the durability of OLEDs. We recently reported the reversed-bias induced recovery phenomenon of degradation in multilayer OLEDs and proposed that the movement of ionic impurities was a key factor which causes the degradation of OLEDs.[12,13] In particular, the initial degradation mechanism occurring within the first few minutes after a bias existence of ionic impurities. Ionic impurities are believed to be a dominant factor which causes recoverable degradation in OLEDs through the formation of an internal electric field in the opposite direction to the applied external field. The strength of the internal electric field may become comparable to that of the external electric field. The formation of an internal field leads to a decrease in the effective electric field for charge injection and transport.

In this letter, we will report our new experimental findings about the effects of the reversed-bias application on as-fabricated OLEDs. We will demonstrate the fact that the reversed-bias application not only accelerates the degradation recovery as reported by us earlier[14] but also significantly improves the performances of as-fabricated OLEDs. We will also show that the observed effects of reversed-bias can be explained by the same model which we previously proposed for the interpretation of recoverable degradation in OLEDs.[15]

Standard double-layer OLEDs of ITO/TPD/Alq_3/Mg:Ag were fabricated by a conventional vacuum-vapor deposition process. The size of the emitting area was $2 \times 2\,mm^2$ and 8 devices were formed on the same glass substrate at the same time. The OLEDs formed on the substrate were transferred into a vacuum cryostat and measurements of all diodes on the same substrate were continuously performed without breaking the vacuum. This ensured reproducibility of our luminance-current density-voltage (L-J-V) data. A source-measure unit (Keithley 238) and luminance meter (Topcon BMW-5A) were used for L-J-V measurements. For rapid-mode measurements, relative luminance was directly detected using a photomultiplier. All measurements were controlled by a computer, and each L-J-V curve could be obtained in 40 ms only.

First, L-J-V curves were repeatedly measured for several different diodes on a substrate by using the rapid-mode measurements. No detectable difference in L-J-V curves was observed. This observation ensured that no degradation process occurred during our rapid-mode 40 ms measurements, even though the voltage was scanned from 0 V to 10 V. Hence it was concluded that we can safely conduct our examination of the effect of reversed-bias application on device performance

To each diode, the reversed-bias voltage of −10 V was applied and maintained for a fixed duration. The time interval under the reversed-bias treatment was varied from 0 to 4 h on a logarithmic scale. Figures 1(a) and 1(b) show the J-V and L-V data obtained from the diodes with different reversed-bias time intervals. Due to a sensitivity saturation of the photomultiplier, relative luminance curves in the figure show saturation at the current density of about 6.0 mA/cm^2. The luminance values at the saturation points correspond to a brightness of 270 Cd/m^2. All the J-V curves for the diodes with different treatments fall on the curve for the virgin diode if the curves are shifted horizontally along the increasing voltage axis. Moreover, all the L-V curves also fall on the original L-V curve with exactly the same amount of voltage axis shifting. In other words, the reversed-bias treatment caused a decrease in applied voltage for attaining fixed current density and luminance values. A longer time interval under the reversed-bias treatment gives rise to a lower voltage to achieve the same current density and luminance. This observation suggests that the charge injection barrier decreases or effective charge injection/transport voltage increases by the reversed-bias treatment. The amount of voltage shift was as large as 0.7 V after a −10 V bias treatment for 3 h. It should be noted that this value is comparable to the reported values of charge injection barriers for conventional OLEDs. It should be emphasized that under the reversed-bias treatment better J-V characteristics can be achieved than those of an as-fabricated fresh diode.

The L-J-V curves for a series of diodes treated with various magnitudes of the reversed-bias were examined and are shown in Fig. 2. When the reversed-bias voltage was increased, the amount of drive voltage shift was increased. This observation exactly corresponds to the case where the time interval of reversed-bias application is increased.

Figure 3 displays the plots of relative luminance against current density for all the diodes with different reversed-bias treatments. The open marks indicate data from diodes treated under constant reversed-bias voltage for different time intervals and closed marks indicate the data obtained from diodes treated with different bias voltages for a constant time interval. All data points fall on the same line, even though they are from different diodes which have suffered different bias conditions. This result is consistent with our previous report[14] and indicates that the reversed-bias application does not cause a change in the quantum efficiency of electroluminescence in our observed luminance range (<270 Cd/m^2), even though it significantly contributes to the improvement in luminance-voltage characteristics. In other words, the reversed-bias application produces no effect on charge injection and transport balance, but gives rise to a large influence on the drive voltage necessary for sufficient current flow through diodes.

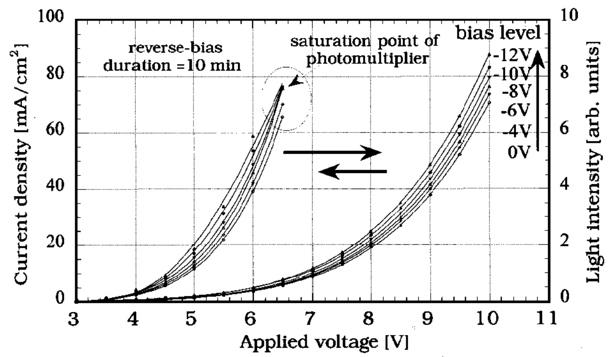
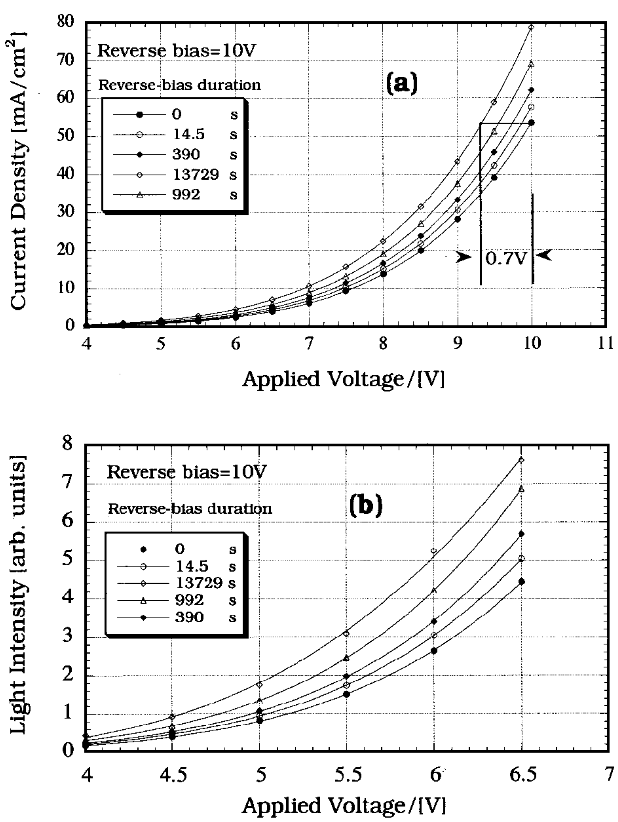


Fig. 2. Current-voltage-luminance characteristics of ITO/TPD/Alq$_3$/Mg:Ag devices under various reverse bias levels.



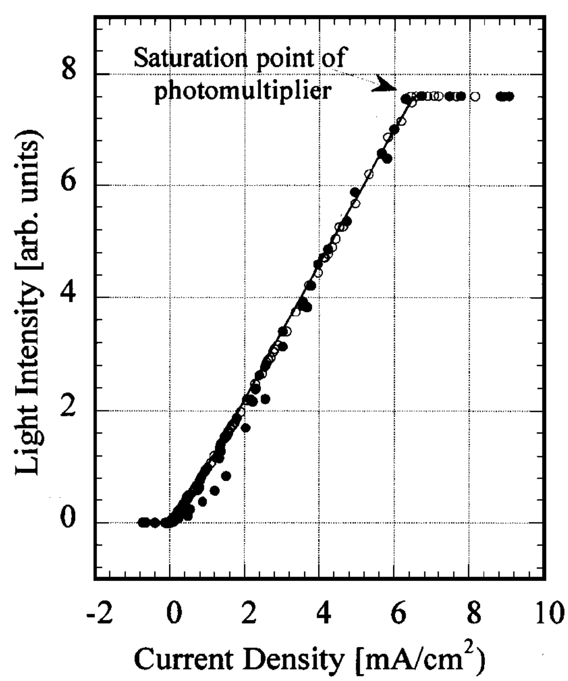


Fig. 3. Plots of light intensity against current density for a low current

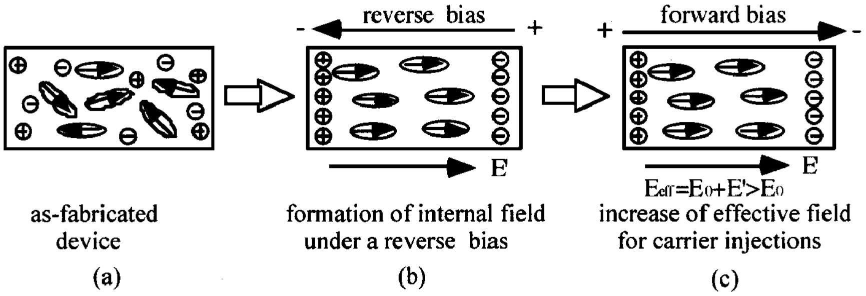


Fig. 4. A model for the formation of internal electric field based on the orientation of dipoles and movements of ions.

The origin of voltage shift can be attributed to two factors, the decrease in carrier injection barrier (interfacial effect) and increase in effective electric field for charge transport (effect in bulk region). We have no experimental evidence at this stage for discriminating between these two factors or to determine which one is dominant. The point we emphasize here is that the internal field model, which we proposed in previous publications for explaining the degradation mechanism, especially the phenomena related to the spontaneous and reversed-bias induced recoveries in OLEDs,[13–17] is also applicable for the explanation of the improvement in J-V characteristics by the reversed-bias application. In contrast to the interpretation for the degradation due to a forward-bias application, the internal field formed under the reverse bias treatment has the same direction as the forward-bias driving. Thus it causes an increase in the effective applied voltage, and this gives rise to the improvement in J-V and J-L characteristics of the diodes. Of course, in our devices, the movement of ions and orientation of permanent dipoles could cause additional current just like the charge and discharge current in a condenser which does not contribute to the emission of the device. Fortunately, this fraction of current is far less than the injection current and its influence on emission efficiency can be ignored.

Figure 4 shows a simplified model which explains the increase of effective electric field by the reversed-bias application. The model assumes that some kinds of ionic impurities and permanent dipoles are present in as-fabricated OLEDs. Ionic impurities and permanent dipoles are assumed to be randomly distributed in the organic layers (Fig. 3(a)). When a reversed bias is applied to an OLED, an internal field (E') in the forward direction is produced (Fig. 3(b)). On the other hand, when a forward external bias (E_0) is applied, the effective electric field ($E_{\text{eff}} = E_0 + E' > E_0$) is expected to be larger than the applied external field (Fig. 3(c)). Consequently, a larger current density and luminance can be obtained, and an enhancement of EL intensity is observable even under the same driving voltage.

In summary, marked voltage shifts towards the lower voltage side were observed both in L-V and I-V characteristics in the OLEDs treated under a reversed-bias. Decrease in the charge injection barrier or increase in effective electric field are possible origins. Reversed-bias treatment leads to improvement in J-V characteristics and brings about an increase in energy efficiency but produces no change in quantum efficiency. Our finding is directly related to the fact that device performances are closely related to the driving mode. Constant current driving and pulse driving modes with a proper reversed-bias component can achieve better device performance than a constant voltage driving mode. An internal field model is proposed and is shown to be useful for qualitative interpretation of the observed phenomena. A quantitative analysis of voltage shifts due to reversed-bias application in OLEDs will be published elsewhere.

Acknowledgment

This work has been partly supported by the Core Research for Evolutional Science and Technology, Japan Science and Technology Cooperation (CREST/JST).

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