A Novel Analytical Method of Charge Injection and Emission Behaviors in Tandem Organic Light Emitting Diode

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Abstract
We propose a novel analytical method of charge injection and emission in tandem organic light emitting diode (OLED) using displacement current and current density-voltage-luminance (DC-JVL) measurement. We found that the current and emission in the low-voltage regions of the tandem OLEDs are good indicators of the reliability of the tandem OLED.

Author Keywords
DC-JVL; displacement current; tandem OLED

1. Introduction
Top-emission tandem organic light-emitting diodes (OLEDs) have advantages such as high efficiency and long lifetime [1]-[4]. Optimizing the materials of each layer and the device structure of the tandem OLED is essential to realize stable display characteristics. To optimize the tandem OLED characteristics, current density-voltage-luminance (JVL) measurement has commonly been used in the OLED industry [5][6]. However, these measurements are only reliable at high current densities, due to the limited sensitivity of the source meter and a luminance meter, so it is difficult to see the charge injection behavior of tandem OLEDs at low current densities. To investigate the charge injection behavior in a low current density region, we proposed the displacement current and current density-voltage-luminance (DC-JVL) measurement and found that there is a gap between a charge injection voltage and an emission voltage due to the emission loss in the OLEDs [7]. In this study, we measured the DC-JVL characteristics of tandem OLEDs and investigated small current changes and dark emissions at low current density and compared them with the conventional JVL measurement. We also investigated small current changes, dark emissions, and the lifetime by changing the CGL (p-type) thickness. We found that the charge recombination in EML1 and EML2 depends on the thickness of the CGL (p-type), and the simultaneous charge recombination in EML1 and EML2 is indispensable to achieve a reliable tandem OLED.

2. Experiments
2.1 Sample preparation
Figure 1 shows the structure of the blue top emission tandem OLED examined in this study. There are two emission layers (EML) in the tandem OLED to improve the current efficiency compared to a single layer OLED. We prepared two tandem OLEDs, sample A and sample B, with different thicknesses of the CGL (p-type).

2.2 JVL and DC-JVL measurement
We measured the conventional current density-voltage-luminance (JVL) characteristics of the OLED by using the source meter (Keithley 2400) and the luminance meter (Topcon BM-9). The measurement conditions were 0 - +10 V (voltage range), 0.05 V (voltage step), and 0.5 sec (measurement delay of each step).

Figure 2 (a) shows the equivalent circuit of a displacement current measurement (DCM) for a capacitance and a resistance (CR) sample which are connected in parallel [8]. A triangular waveform voltage is applied to a CR sample, and the current vs. voltage characteristics are obtained as shown in Figure 2 (b). The total current (I) flowing through a capacitance (I_C) and a resistance (I_R) is defined by

$$I = I_C + I_R = C\frac{dV}{dt} + \frac{V}{R} \tag{1}$$

In case of $dV/dt > 0$ and $dV/dt < 0$, the currents of a positive slope (Ip) and a negative slope (In) become

$$I_p = C\frac{dV}{dt} + \frac{V}{R} \tag{2}$$

$$I_n = -C\frac{dV}{dt} + \frac{V}{R} \tag{3}$$

Thus, the difference between I_p and I_n is proportional to the capacitance as shown in equation (4).

$$\Delta I = I_p - I_n = 2C\frac{dV}{dt} \tag{4}$$

By using this theory, we calculated the capacitance and the resistance of the tandem OLEDs before charge injections.

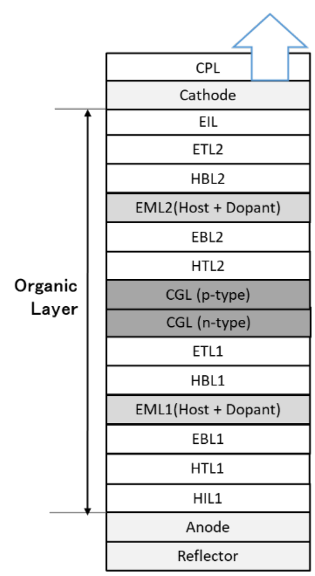

Figure 1. Structure of the tandem OLED.

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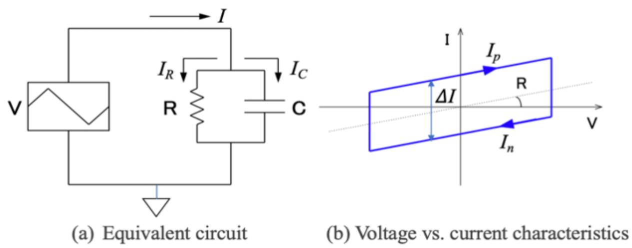

Figure 2. DCM: (a) Equivalent circuit of CR sample measurement by applying triangle waveform voltage and (b) voltage vs. current characteristics of CR sample.

We employed the DCM1000 DC-JVL measurement system (TOYOTech) to obtain the DC-JVL characteristics of the OLED. Figure 3 shows the equivalent measurement circuit of the DCM1000. A triangle waveform voltage was applied to an anode on a glass substrate of an OLED sample, and current was measured from a cathode by using a current-to-voltage (I/V) converter to obtain a displacement current and current density as a function of voltage (DC-JV). An EL intensity was measured by using a photodiode (Hamamatsu photonics S2386-8K) and an I/V converter. Each I/V converter had multiple feedback resistances, so the current measurement range was automatically optimized to the measurement current value. Measurement frequency and voltage were 0.01 Hz and 0 to +10 V (Ramp rate: 0.2 V/sec), respectively. Since the OLED showed uneven emission due to dark spot formation, we didn't calculate the current density and the luminance by using an area of the electrode. Thus, the results are a current (A), an EL intensity (a.u.), and a relative luminance (a.u.).

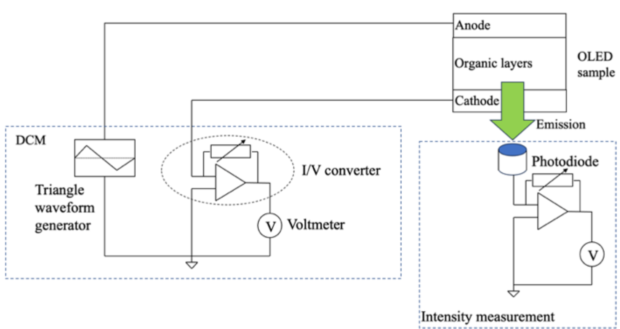

Figure 3. Equivalent circuit of DC-JVL measurement.

2.3 Lifetime measurement

We measured the lifetime of the tandem OLEDs. We applied the constant current of 50 mA/cm^2 to the samples and measured changes in the luminance over time. We also normalized the luminance values by the data of time=0 for comparisons.

3. Results and discussions

3.1 Comparison between JV and DC-JV characteristics

Figure 4 shows the JV (source meter) and the DC-JV (DCM1000) characteristics of the OLED from 0 V to 10 V. We observed a small current change around 3.0 V in the positive voltage sweep of the DC-JV. On the other hand, the JV measurement was hard to confirm the existence of current change due to the limited current measurement sensitivity of the source meter.

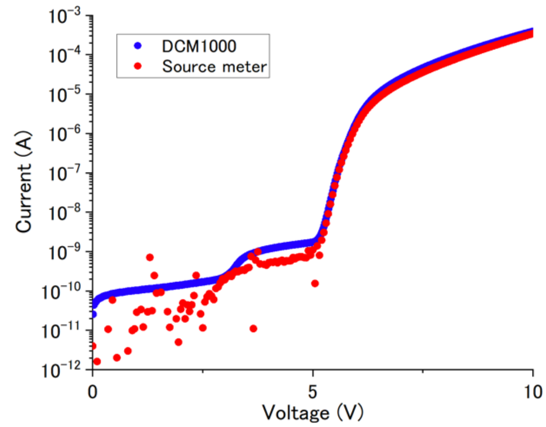

Figure 4. DC-JV and JV characteristics.

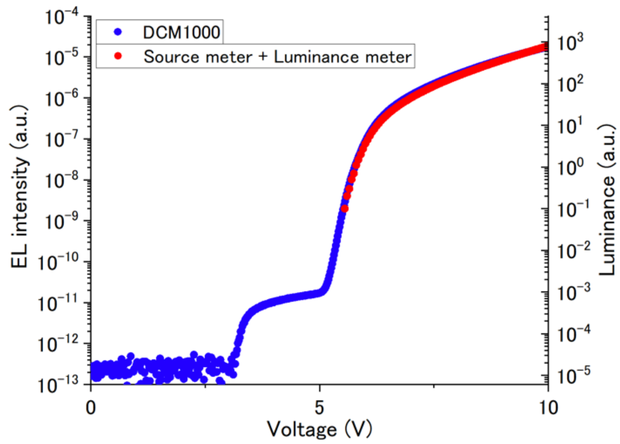

Figure 5. EL intensity and luminance-voltage characteristics.

3.2 Comparison between Luminance-voltage and EL intensity-voltage characteristics

Figure 5 shows the luminance (conventional luminance meter) and the EL intensity-voltage (DCM1000) characteristic of the OLED. We could observe the extreme dark emission from 3.0 V in the DCM1000, but the conventional luminance meter could not observe dark emissions due to the low sensitivity of the luminance meter.

By comparing the DC-JV and the EL intensity-voltage characteristics, we found that the current change at 3.0 V was not only a charge injection current but also a current due to the recombination of holes and electrons because the emission was observed at the same voltage. We have previously measured a single-layer OLED with DC-JVL measurement, where we observed similar current changes without any emission, suggesting that the hole accumulation at the HTL interface occurs without charge recombination [9]. Thus, by performing DC-JVL measurements with high sensitivity for detecting current and luminance, it is possible to distinguish between charge accumulation and charge recombination in the current range that was difficult to detect with conventional JVL measurements.

3.3 DC-JVL characteristics by changing the CGL (p-type) thickness

Figure 6 shows the DC-JV characteristics of the tandem OLEDs having a different thickness of CGL (p-type). The CGL (p-type) thickness of sample B was thicker than that of sample A. We observed that the current at 3.0V significantly decreased by increasing the thickness of CGL (p-type). Accordingly, the EL intensity of sample B turns out to be negligible (Fig.7).

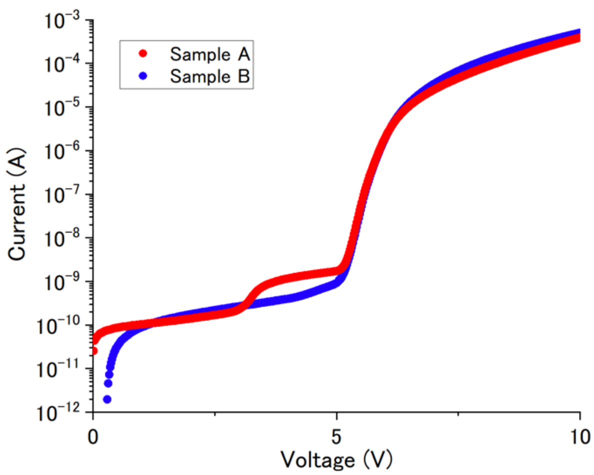

Figure 6. DC-JV characteristics of the tandem OLEDs.

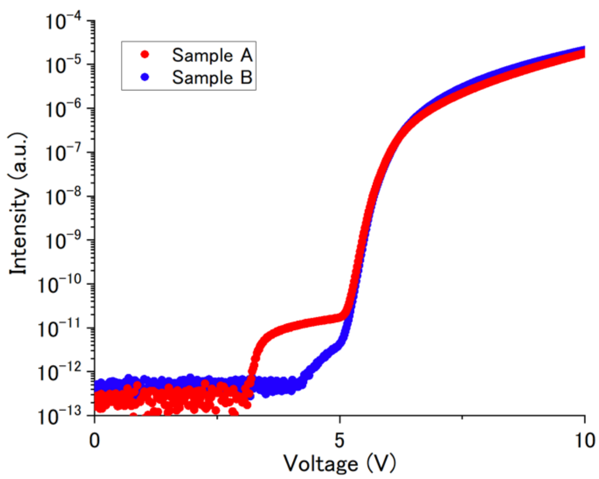

Figure 7. EL intensity-voltage characteristics.

3.4 Capacitance and resistance by DCM

A portion of the cyclic DCM characteristics of the OLEDs are plotted on a linear scale in Figure 8. Table 1 shows the calculation result of the resistance and the capacitance of the tandem OLEDs in the range of 0 V and 1.0 V, where no charge injection occurs from the electrodes. As the CGL (p-type) thickness increased, the resistance and capacitance of sample B became smaller and larger, respectively, than those of sample A. These changes could indicate an increase in intrinsic charges in sample B.

Previously, we observed a symmetric small current change in the positive and negative voltage sweep of the single α-NPD/Alq_3 OLED due to the charge accumulation without charge recombination [8]. On the contrary, we observed an asymmetric current change in the range of 3.0 V and 5.0 V in the negative voltage sweep of sample A. Based on the result, we suggest that the current change is not a change of capacitance of the tandem OLED, but a charge injection into an EML followed by a charge recombination.

Indeed, we have measured the EL spectra of sample A operated at the voltage between 3.0 V and 5.0 V and confirmed that the emission peak is located at 452 nm, which is consistent with the EL spectra measured at higher than 5 V. The threshold voltage of the emission from sample A is 3.0 V, which is slightly higher than the photon energy of the blue EL emission at 452 nm (2.74 eV). Therefore, the observed emission in this voltage range should be considered the result of charge recombination in one of the emitting layers (EML1 or EML2) in sample A followed by the charge recombination in another emitting layer. Conversely, simultaneous charge recombination in EML1 and EML2 may occur in sample B.

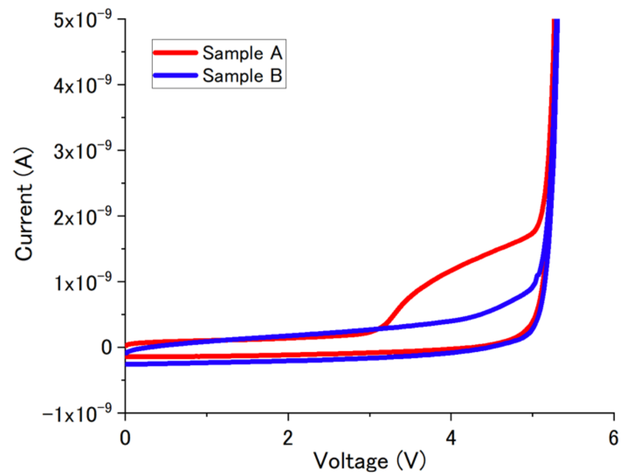

Figure 8. DCM characteristics of the tandem OLEDs with a linear scale.

Table 1. Calculation results of the resistance and the capacitance.

Sample	Resistance (GΩ)	Capacitance (pF)
A	26.7	567
B	12.0	667

3.5 Lifetime of the tandem OLEDs

Figure 9 shows the lifetime of the tandem OLEDs operated at the current density of 50 mA/cm^2. We found that sample B, which has thicker CGL (p-type), exhibits a longer lifetime than sample A. This result suggests that the simultaneous charge recombination in EML1 and EML2 is indispensable to achieve a reliable tandem OLED.

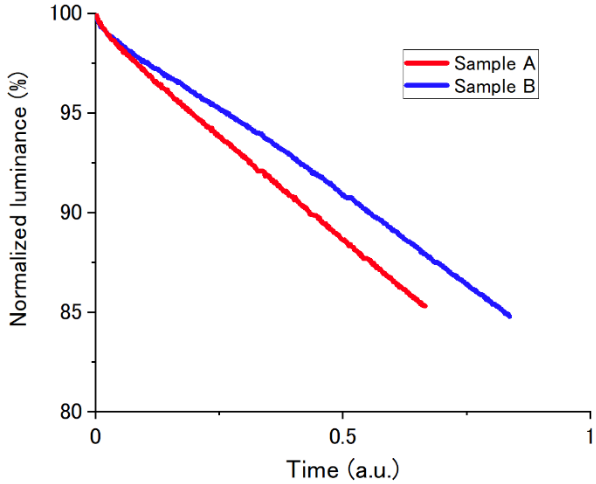

Figure 9. Normalized luminance vs. time of the tandem OLEDs operated at the current density of 50 mA/cm^2.

4. Conclusions

We measured the displacement current and current density-voltage-luminance (DC-JVL) of the tandem OLEDs and compared the results of the conventional JVL measurement.

Distinct behaviors of charge injections, accumulations, and recombination in a tandem OLED can be discerned through the simultaneous measurement of displacement current and current-voltage-luminance in the low current and dark EL emission levels. We found that the current and emission in the low-voltage regions are indicative of the reliability of the tandem OLED.

5. References

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