Impedance spectroscopy for lifetime analysis of OLED

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Abstract

The frequency response analysis of complex impedance spectra using small perturbation ac impedance spectroscopy is an informative method of OLED performance characterization and lifetime analysis. Using simple RC equivalent circuit mode,l macroscopic nonliniear transport properties of semi-conductive emission/transport layers can be analyzed and parameterized. We present the bias voltage dependence and aging effect in impedance spectra measured from an ITO/CuPC/TPD/Alq_3/LiF/Al OLED device, and discuss possible failure mechanism based on impedance model parameters.

1. Introduction

Ability of fast responsive self-emission of the organic light-emitting diodes (OLEDs) has attracted a lot of interests for their applications to a full-color mobile display as the next generation to the liquid crystal displays (LCDs). Among many of device characteristics required for commercial development of display devices, lifetime is one of the key issues for OLEDs [2]. In order to reach a targeted level of device quality, understanding of failure mechanism in terms of a few key measurement parameters from the simple and straightforward testing methods is important

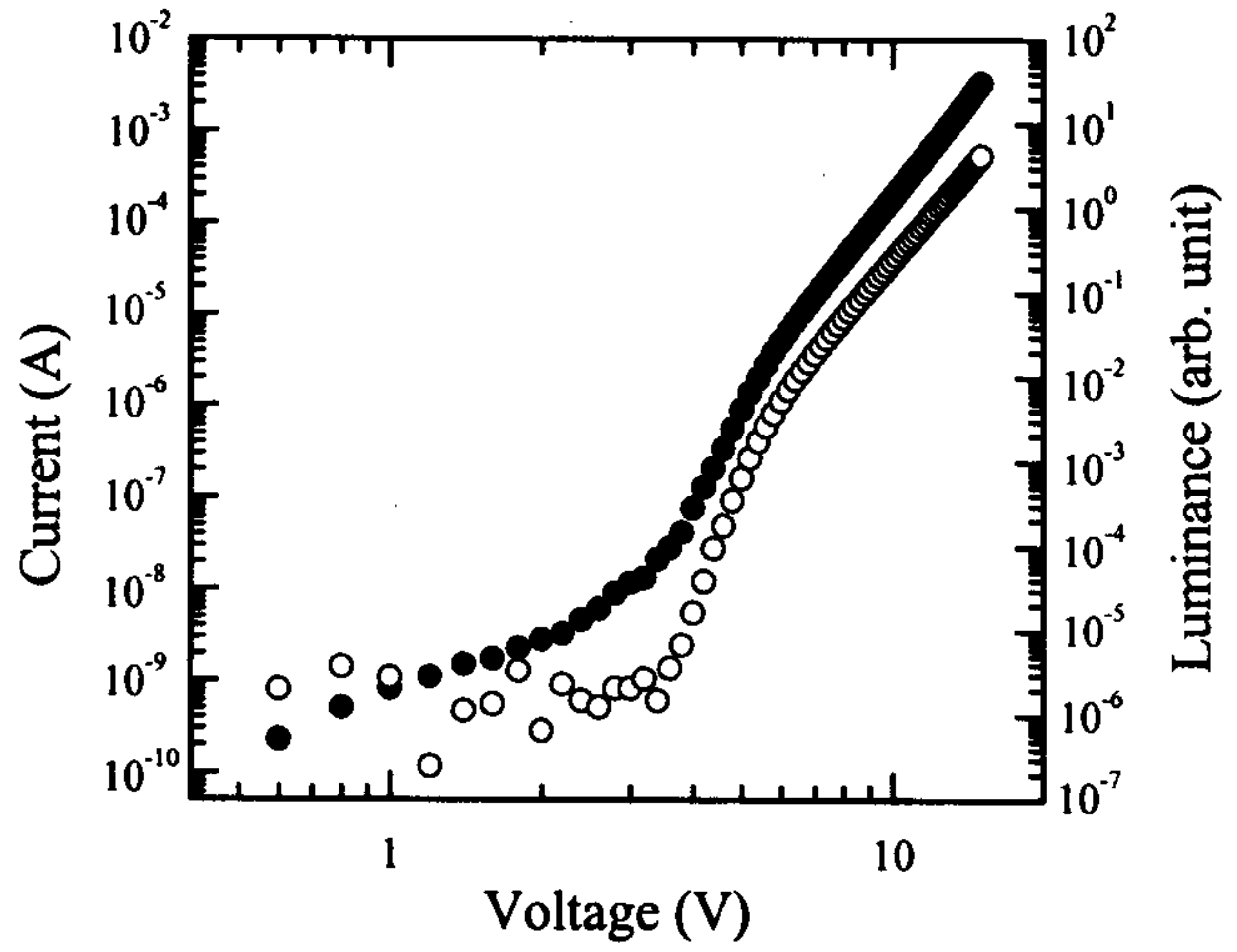
Since 2001, we have been dedicated to development of the OLED reliability and lifetime testing equipment supported by Technology Development Service Program of the Ministry of Science and Technology (MOST) of Korea. The *Polaronix IV* is a new instrument designed especially for OLED device characterization with a variety of functions for I-V-L measurement, dc and pulse lifetime measurement under various acceleration conditions such as high temperature, humidity as well as various atmospheric conditions such as controlled level of vacuum, N_2 or O_2. In the course of development of power driving electronic module for the high frequency pulse operation of OLED, we examined applicability of frequency response analysis [1] to OLED device characterization and failure analysis, based on inherent capability of ac impedance measurement using *Polaronix IV* pulse power driving electronics.

In this work, we present and discuss the bias voltage dependence and aging effect in frequency dependent complex impedance spectra measured from an ITO/CuPC/TPD/Alq_3/LiF/Al device. Macroscopic nonlinear transport properties of semi-conductive layers in the device are analyzed and parameterized by using simple RC equivalent circuit model. The variation of impedance characteristics upon self-degradation of the device after exposure to air provides useful information on phenomenological failure mechanism of OLED devices.

2. Experimental

The devices are fabricated by using successive vacuum-depositions of CuPC (1500 Å), TPD (500 Å), Alq_3 (700 Å), LiF (5 Å), and Al electrode (1000 Å), without breaking vacuum, onto etched and cleaned indium tin oxide (ITO) substrates. The ITO substrates with a sheet resistance of about 10 Ω/□ are supplied by Samsung Corning Inc. The overlap area of the Al and ITO electrodes is about 4 mm^2. The evaporation rates are about 2 Å/s, measured by a quartz crystal oscillator, under a base pressure of about 1×10^{-6} Torr.

The current-voltage-light (I-V-L) characteristics were measured under vacuum simultaneously with a Keithley 236 source-measure unit and a Keithley 2000



multimeter equipped with a PMT through an ARC 275 monochromator.

Figure 1 The voltage dependence of current (●) and luminance (○) of ITO/CuPC(1500Å)/TPD (700Å)/Alq_3(700Å)/LiF(5Å)/Al.

The complex impedance spectrum measurements for samples spontaneously aged under air without special packaging against for device degradation were carried out at ambient conditions by using Autolab PGSTAT 30 with the FRA2 impedance electronic interface module. The voltage-controlled sinusoidal perturbation signals of 50 mV amplitude and variable frequencies from 1 Hz to 1 MHz were applied with various dc bias in the range of 0 ~ 5 V for device operation.

3. Results and discussion

Fig. 1 shows the current-voltage-luminescence (I-V-L) characteristics of a freshly prepared ITO/CuPC /TPD/Alq_3/LiF/Al device measured under vacuum. The onset voltage of light emission occurs at 3.6 V. At high voltages the I-V-L characteristics exhibit a power-law dependence, indicating the trap-limited current [2]. Near the onset voltage, current-voltage data show nonlinear dependence. The device shows an external QE of about 1.9 % and a luminous efficiency of 3.8 lm/W at luminance 100 cd/m^2 (at bias voltage 8 V and current density 1 mA/cm^2).

After 42 hour's exposure to air without special packaging, the device performance was significantly reduced. The current density at 8 V bias voltage decreased by 48% to 0.65 mA/cm^2. Table 1 summarizes the change of device performance through self-aging after exposure to air. After 82 hours no more light emission was observed up to 20 V.

Table 1 Device performance after exposure to air.

Time (hrs)	Current* (mA)	Performance Ratio** (%)	Note
0	0.411	100	under vacuum
42	0.260	63.2	
55	0.215	51.9	
62	0.189	45.7	
66	0.168	40.6	
72	0.065	15.7	
82	0.031	7.8	
90	0.002	0.5	no emission

* measured at bias voltage 8.0 V
** current ratio to initial current value 0.411 mA

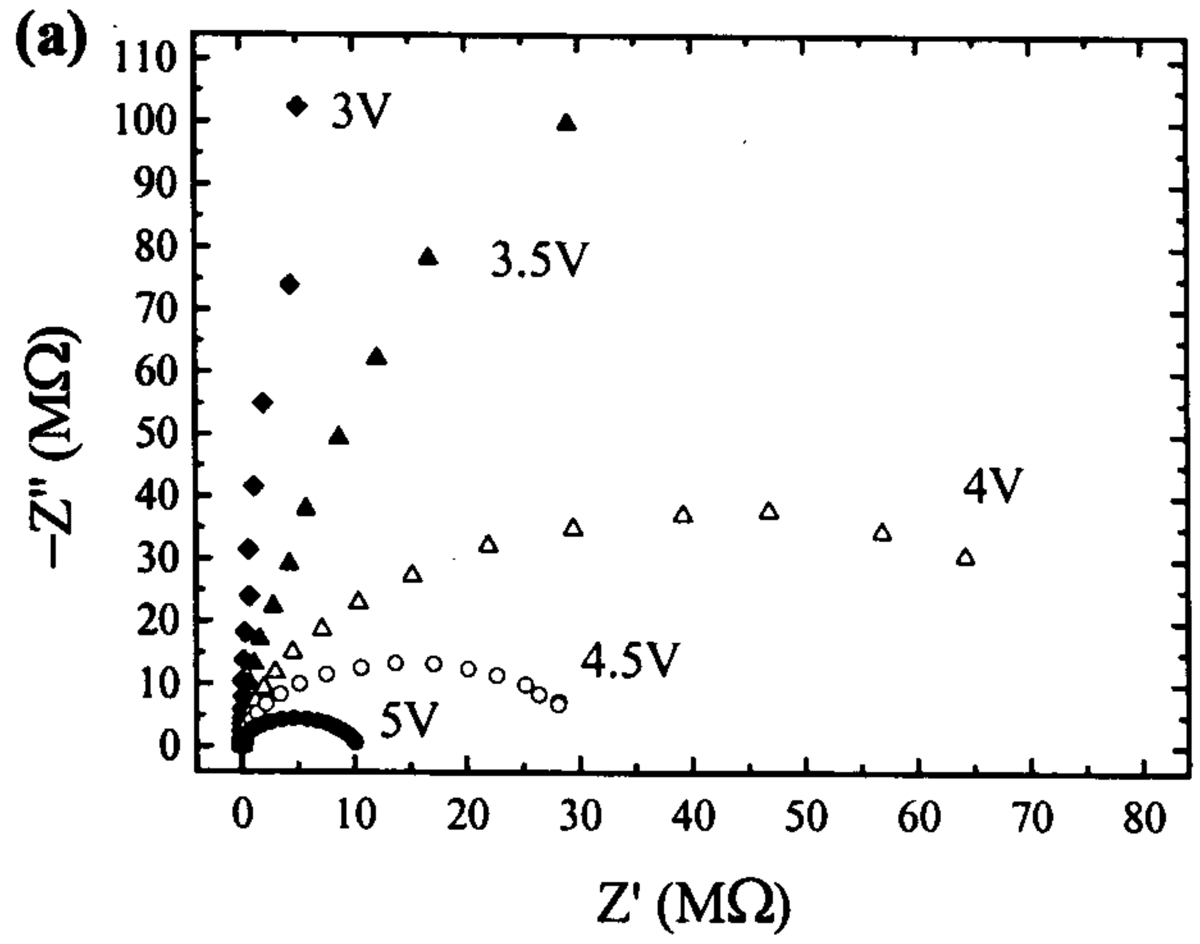
The impedance spectra of ITO/CuPC/TPD/Alq_3/LiF /Al device show generally a semi-circle in the Nyquist plot, as shown in Fig. 2. This indicates that, in measured frequency (f) range of 1 Hz ~ 1 MHz, the impedance characteristics can be interpreted by the simple equivalent circuit model consisting of a RC pair connected in parallel (R_p and C_p) with a serial resistance (R_s), as shown in Fig. 3. In this case, the complex impedance can be expressed as,

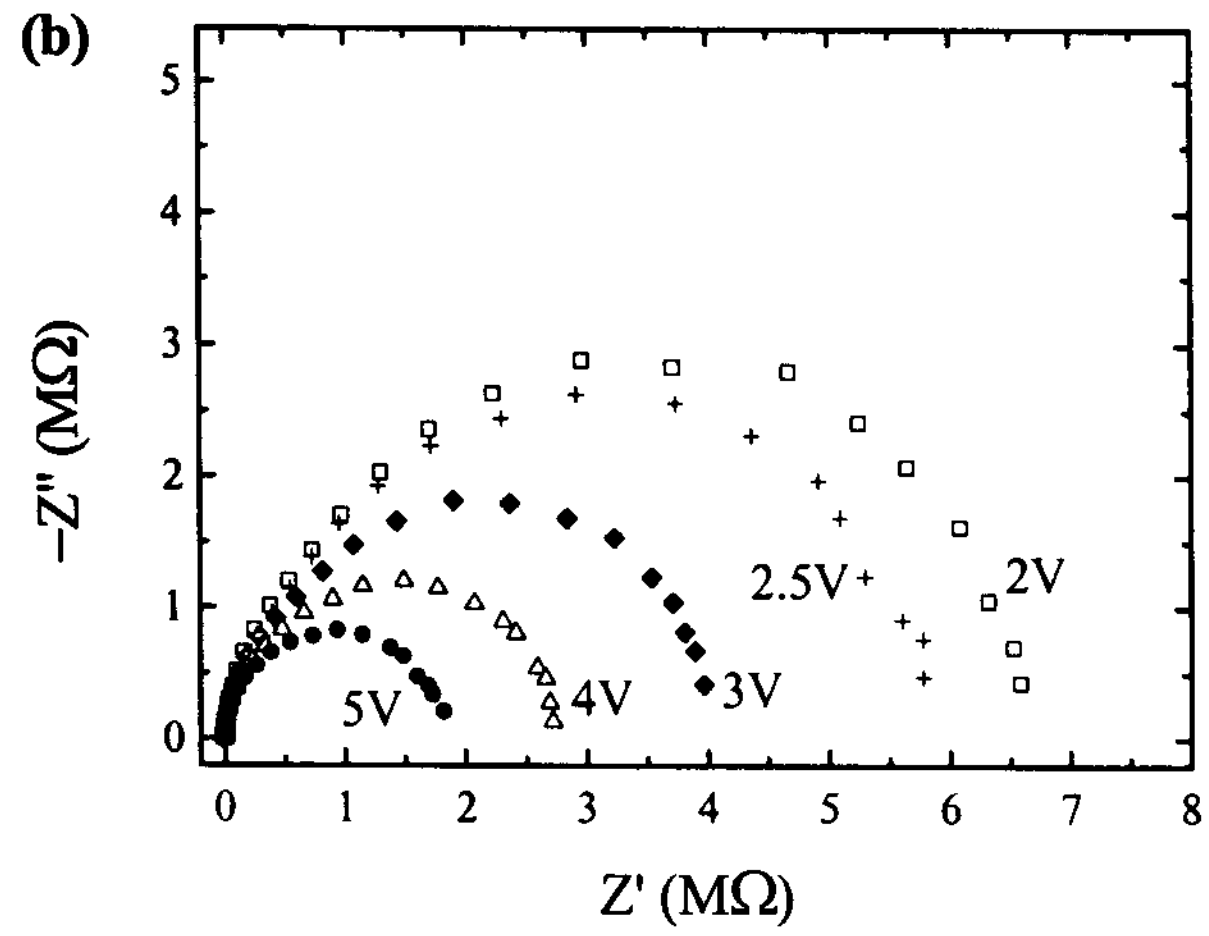
$$Z = R_s + \frac{R_p}{1+\omega^2\tau^2} - j\omega\tau\frac{R_p}{1+\omega^2\tau^2} \quad (1),$$

where = 2π f, and = R_pC_p is the time constant.

At high perturbation frequency the real part of complex impedance reaches below 40 Ω, which is several orders of magnitude smaller than that of low frequency, and shows little variation upon bias voltage and air exposure time, indicating that contribution of serial resistance to complex impedance is very small and originates from electrical conductors in the device such as ITO and Al electrode [3].

The parallel components of the equivalent circuit R_p and C_p, which contribute to the complex impedance as a semicircle with radius $R_p/2$ in the Nyquist plot, can be interpreted as nonlinear transport properties of semi-conductive emission and transporting layers in the device. These parameters are nonlinear element, and dependent upon bias voltage and air exposure time, as shown in Fig. 2 (a), (b) and (c). The conductivity through the layer increases due to high carrier injection at higher electric field, consistent with the decrease of R_p with increasing bias voltage.





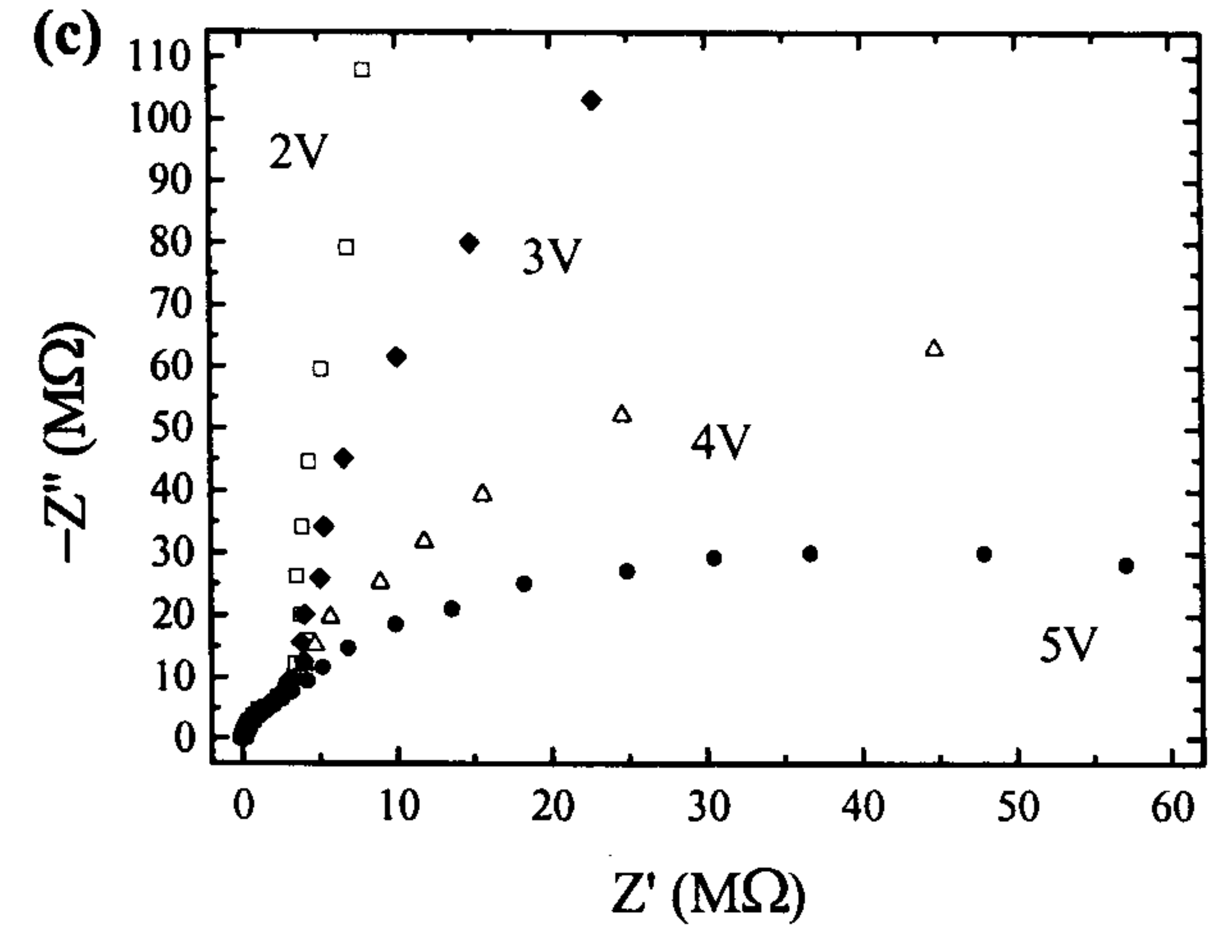


Figure 2 Variation of complex impedance spectra measured upon exposure to air after (a) 62 hrs, (b) 72 hrs and (c) 90 hrs at various bias voltage: (●) 5 V, (0) 4.5 V, (△) 4 V, (▲) 3.5 V, (◆) 3 V, (+) 2.5 V, and (□) 2 V.

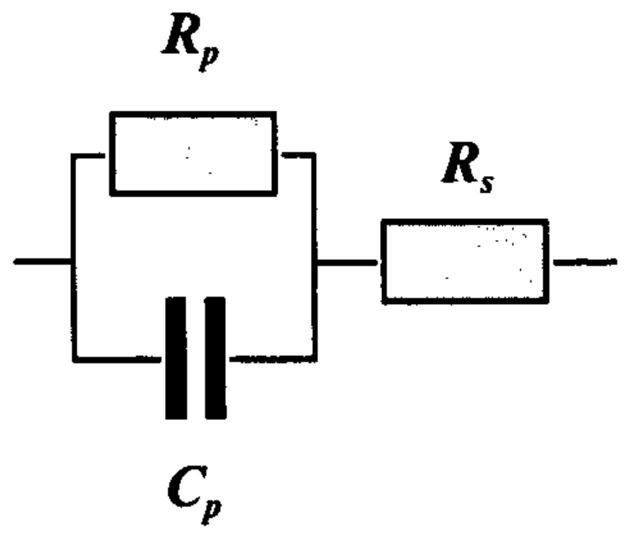


Figure 3 A simple equivalent circuit model for the complex impedance spectrum analysis of OLED (R_p: parallel resistance, C_p: parallel resistance, and R_s: serial resistance).

Fig. 4 shows nonlinear characteristics of the imaginary part of the complex impedance. The peak frequency of dielectric absorption in the spectra corresponds to the time constant in the equivalent circuit model. Our calculation shows that variation of C_p is relatively small in the range from 0.8 nF to 1.1 nF, and up-shift of absorption frequency with increasing bias voltage arises mainly from the decrease of R_p.

The variation of impedance spectra upon exposure to air can be briefly summarized in plots of impedance spectra at constant bias voltage, as shown in Fig. 5. The radius of semicircle in the Nyquist plot generally increases with exposure time. After 72 hours of exposure, however, an abrupt decrease of the radius was observed. The impedance parameter calculation shows that both R_p and C_p are decreased, while relative change of R_p is three times larger than that of C_p. One possible explanation for this abrupt change of parameters can be the formation of inhomogeneous local conductive path in microscopic scale due to degradation of active layers [4]. The voltage dependence of impedance spectra before the change in Fig. 2 (a) shows significant variation of R_p near the onset voltage, which was initially 3.6 V. The small R_p with relatively weak voltage dependence after 72 hours of exposure in Fig. 2 (b) suggest that onset voltage has shifted to higher voltage than 5 V, and the impedance spectroscopy in measured range of frequency sees only the non-emissive transport through conductive areas. A complete degradation of the device after 90 hours of exposure introduces a secondary semicircle in impedance plots in Fig. 2 (c), suggesting the formation of a depletion region type Schottky barrier [5]. The calculated value of C_p, which is similar to that before degradation, and large increase of R_p indicates that the device is losing diode characteristics and became more capacitor-like.

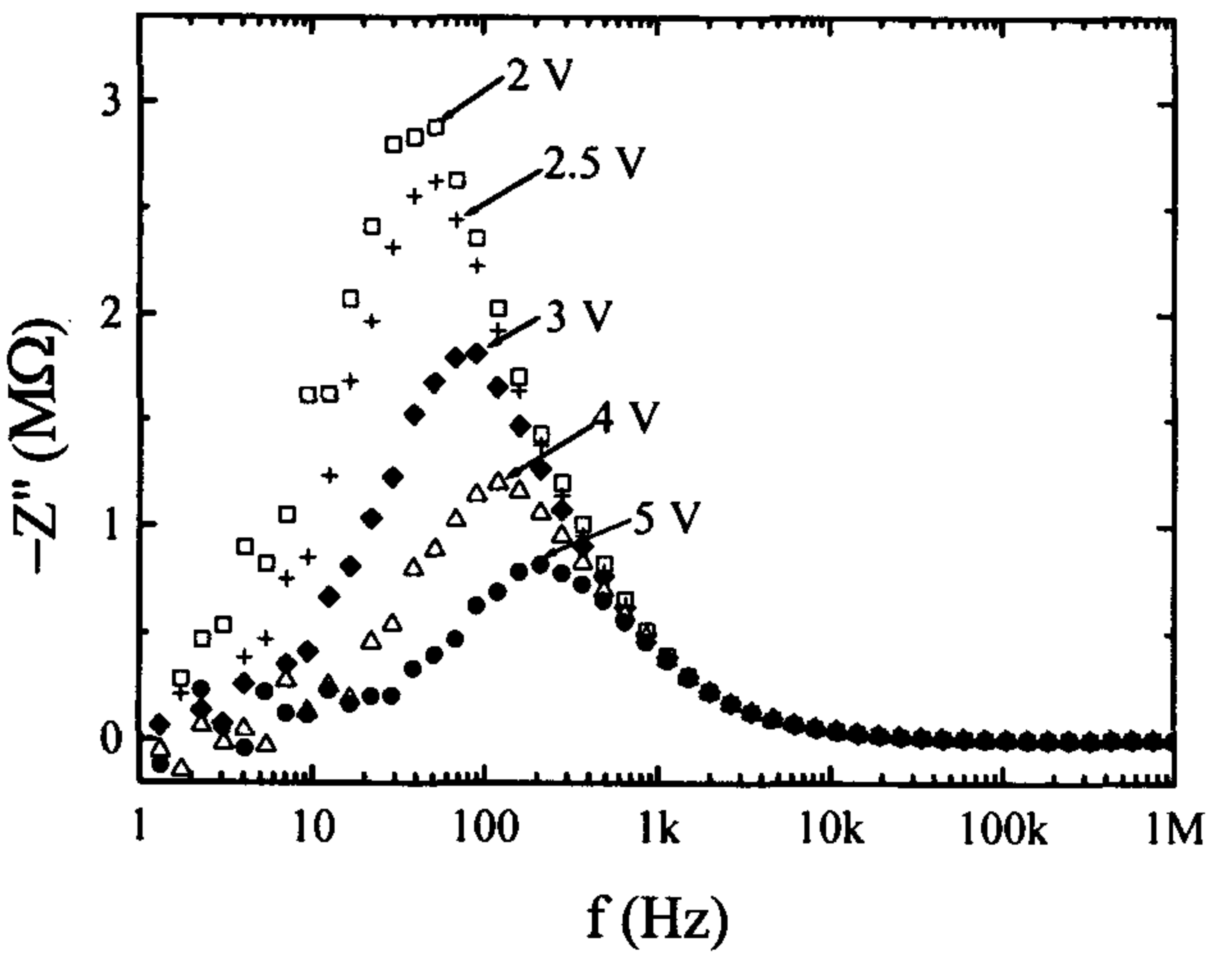
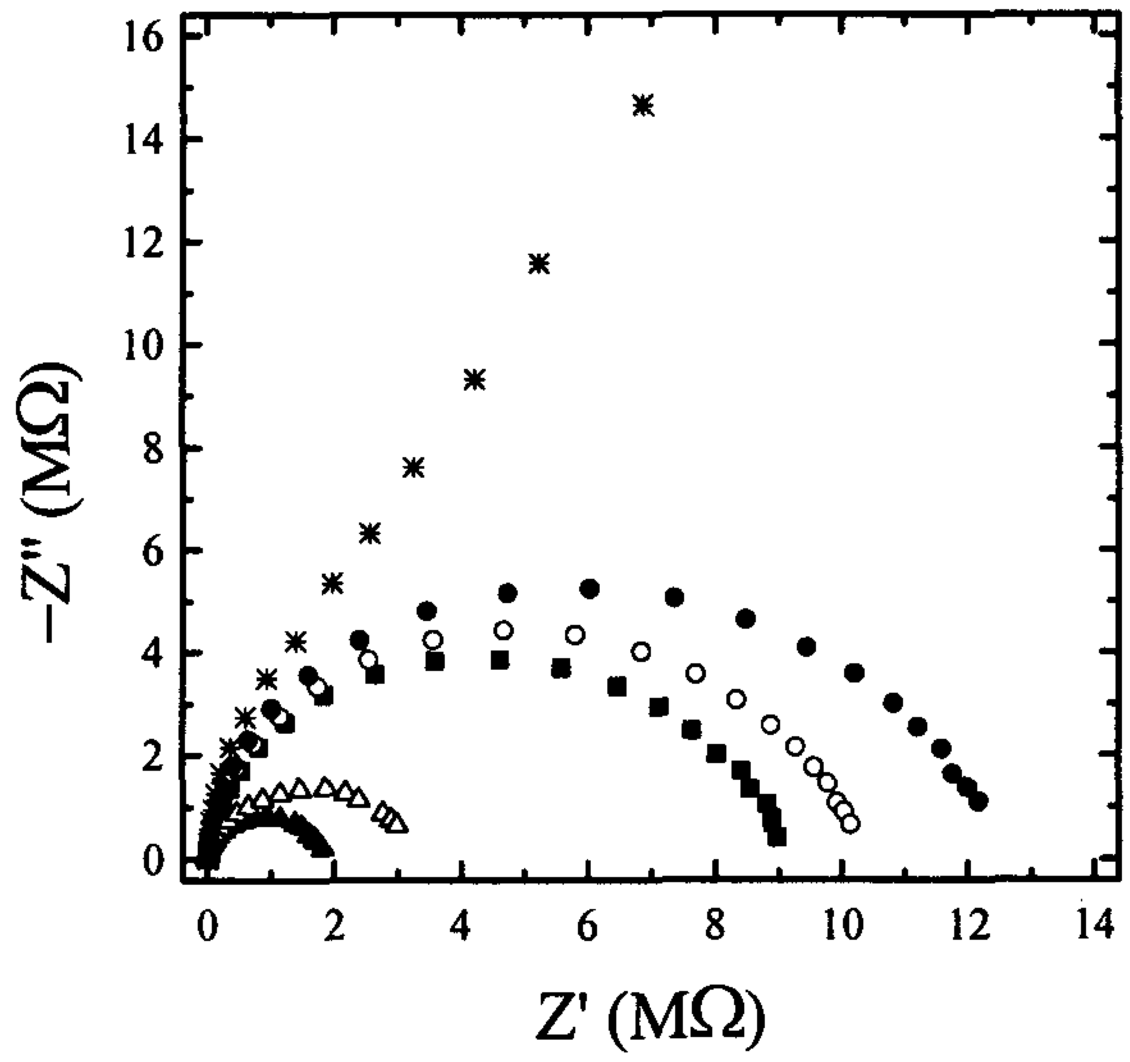


Figure 4 Variation of the imaginary part of the impedance spectra measured after 72 hours exposure at various bias voltages: (●) 5 V, (△) 4 V, (◆) 3 V, (+) 2.5 V, and (□) 2 V.

Figure 5 Variation of complex impedance spectra measured at dc bias 5.0 V with the exposure time to air: (■) 55 hrs, (0) 62 hrs, (●) 66 hrs, (▲) 72 hrs, (△) 82 hrs, and (✱) 90 hrs.

4. Conclusion

We conclude that frequency response analysis of complex impedance spectra of OLED device using simple RC equivalent circuit model can be developed as a powerful tool for the study of nonlinear transport properties and failure mechanism. We could observe drastic variation of impedance spectra upon change of bias voltage as well as upon device degradation through the exposure to air. Quantitative lifetime analysis using impedance parameters seems to be possible based on detailed understanding of failure mechanism. Further experiments on measurement and



analysis of impedance spectra for various OLED devices, and particularly at bias voltages beyond onset voltage of light emission, are necessary for the generalization of the observed phenomena.

5. Acknowledgements

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