Abstract

We have studied the degradation mechanisms of green fluorescent organic light emitting diodes (OLEDs) with tris(8-hydroxyquinoline) aluminum as an emitter with transient luminescent spectroscopies. While transient photo-luminescent (PL) spectroscopy with optical pulses allowed us to investigate the dynamics of singlet exciton of the emitter, transient electro-luminescent (EL) spectroscopy with electrical pulses provided us the information about the dynamics of the triplet excitons of the emitter. The transient PL showed a reduction in singlet lifetime of the emitter of degraded devices. This indicates that the device degradation is partly ascribed to the reduction in PL quantum yield, which is due to singlet quenching by neutral quenchers. Capacitance-voltage analysis revealed that the neutral quenchers may work as traps for positive charges. Singlet exciton demonstrated no change in lifetime with and without EL operation, thus the device degradation is not caused by quenching of singlet exciton with positive charges. The transient EL of OLEDs showed delayed EL decay within 16 μs, which was smaller than lifetime of triplet exciton (25−170 μs). This indicates that the delay EL of OLEDs is generated via triplet-triplet annihilation (TTA). The transient EL showed a reduction in intensity of the delayed EL, which was ascribed to the triplet quenching with positive charges.

As for degradation mechanisms of highly efficient blue OLEDs utilizing the TTA process, the blue devices demonstrated maximum external quantum efficiency of 10.47% at 4 mA/cm^2 which exceeds the theoretical limit (5%) of the fluorescence OLEDs that make use of only singlet exciton generated by the charge recombination. Similar EL decay time was observed in OLEDs where an emitting layer was composed of a host material with and without an emitting dopant. The delayed EL is thus due to the TTA process occurring in the host material. The blue devices with the emitting dopant showed less than 3% luminance loss of the initial luminance of 1,000 cd/m^2 after 100 h operation. For device degradation analysis, we accelerated the device degradation until 13% luminance loss of the initial luminance of 1,650 cd/m^2. The transient PL of degraded OLEDs demonstrated no change in the singlet lifetime of the dopant material, which indicates that the device degradation is not due to the quenching with neutral quenchers of singlet exciton of the dopant material. The lifetime of singlet of the dopant showed a decrease with the application of positive current, which suggests that degradation is caused by the quenching with positive quenchers of singlet exciton of dopant. The reduction in the intensity of the delayed EL of degraded devices indicates that the degradation is also associated with the reduction in the generation of additional singlet exciton via the TTA in the host layer.

Keywords: Organic light-emitting diodes, transient luminescent spectroscopies, triplet-triplet annihilation, quenching of singlet exciton, quenching of triplet exciton.