Fast-response organic-inorganic hybrid light-emitting diode

Takeshi Fukuda*1, Bin Wei2, Eiichi Suto3, Musubu Ichikawa3 and Yoshio Taniguchi3

1 Faculty of Engineering, Saitama University, 255 Shimo-okubo, Sakura-ku, Saitama-shi, Saitama 338-8570, Japan
2 Key Laboratory of Advanced Display and System Applications, Ministry of Education, Shanghai University, P.O.B. 143, 149 Yanchang Road, Shanghai 200072, P.R.China
3 Faculty of Textile Science and Technology, Shinshu University, 3-15-1 Tokida, Ueda, Nagano 386-8567, Japan

Received zzz, revised zzz, accepted zzz
Published online zzz

PACS 42.70.Jk, 42.72.Bj, 61.72.Uj, 78.55.Hx

* Corresponding author: e-mail fukuda@fms.saitama-u.ac.jp, Phone: +81 48 858 3526, Fax: +81 48 858 3526

We demonstrated important changes produced on the modulation speed of hybrid organic-inorganic light-emitting diodes to examine applicability as a light source for visible optical communications. Fabricated device structure was 4,4’-bis[N-(1-naphthyl)-N-phenyl-amino]biphenyl/4,4’-(bis(9-ethyl-3-carbazovinylene)-1,1’-biphenyl)/4,4’-bis[9-dicarbazolyl]-2,2’-biphenyl/ZnS/LiF/MgAg. This device showed the improvement in the modulation speed using ZnS instead of an organic material, tris(8-hydroxyquinoline)aluminum. A maximum cutoff frequency of 20.6 MHz was achieved.

Introduction Since a first report of efficient electroluminescence (EL) emission from triphenylamine derivative, namely tris(8-hydroxyquinoline)aluminum (Alq3), organic light-emitting diodes (OLEDs) have attracted much attention for flat-panel displays and lightings. To date, several breakthroughs have led to significant enhancements of performance in OLEDs, such as the improvement of a charge-carrier balance into an emitting layer (EML) [1] and the efficiency of injecting electrons into an organic layer from a metal cathode [2], the usage of high-carrier mobility electron/hole transport materials [3], and high-efficiency fluorescence- and phosphorescence-organic emitting materials [4].

In addition, OLEDs are also expected as light sources for optical communication systems because of their fast electro-optical conversion speed [5]. There have been several studies on the influence of device parameters on transient properties of OLEDs, such as the fluorescence lifetime (FL) of organic light-emitting materials [5, 6], the capacitance determined by an emitting area [7], the electron injection barrier at metal/organic and organic/organic interfaces [8, 9], and particularly the carrier mobility of electron/hole transport materials [3]. Here, the modulation speed of OLEDs has been estimated as a cutoff frequency of EL intensity, and a maximum cutoff frequency of up to 25 MHz has been achieved for the OLED with a small area of 300 µm circle [10]. However, a high-voltage of more than 10 V is necessary to realize the fast-modulation speed. In order to apply OLEDs to optical communications, further improvement in both a lower drive voltage and a faster modulation speed are desired.

Previous papers demonstrated that a drive voltage of OLEDs could be reduced using high-carrier mobility organic materials because of the efficient carrier transport into organic layers [3]. The reported carrier mobility of organic materials is sufficiently high for OLED-displays, however, it is considered not to be high enough for light sources for optical communications. One approach to reduce the drive voltage is the usage of an organic-inorganic multilayered structure [11]. This is because that the carrier
mobility of inorganic materials is much higher than that of organic materials [12, 13]. It is surmised that the efficient carrier transport using inorganic materials is also considered to be important to improve the modulation speed of OLEDs [3]. However, very little is known about the effect of the inorganic carrier transport material for transient properties of OLEDs.

In this paper, we investigated transient properties of hybrid organic-inorganic light-emitting diodes (HOLEDs) with an inorganic electron transport material, ZnS. The frequency dependence of EL intensity and the current density-voltage-luminance (J-V-L) characteristics of OLEDs with ZnS and Alq3 as electron transport layers (ETLs) are measured to investigate the effect of electron transport materials on transient properties.

**Experimental**

HOLEDs were fabricated on a glass substrate covered with a patterned indium tin oxide (ITO) anode. The ITO layer was deposited in a sputtering deposition system, and the thickness was 150 nm. The prepared glass substrate was cleaned in deionized water, detergent, and isopropl alcohol sequentially under ultrasonic waves, and then treated with oxygen plasma for 5 minutes. Then, organic/inorganic/metal layers were deposited successively using a vacuum deposition system at a base pressure of below 5.0 x 10⁻⁶ Torr.

Each layer consisted of 4,4’-bis[N-(1-naphthyl)-N-phenyl-amino]-biphenyl (α-NPD) as a hole transport layer (HTL), 4,4’-bis[9-dicarbazolyl]-2,2’-biphenyl (CBP) doped with 0.5wt%, 4,4’-(bis(9-ethyl-3-carbazolylene)-1,1’-biphenyl (BCzVBi) as an EML. We employed two species materials as electron transport layers (ETLs), namely, ZnS and Alq3 for devices A and B, respectively. The electron injection layer (EIL) of a LiF layer and a cathode of MgAg (9:1 w/w) and Ag were evaporated on the top of ETL layers. The device structures were α-NPD [40 nm]/BCzVBi:CBP [20 nm]/ZnS (device A) or Alq3 (device B) [20 nm]/LiF [0.4 nm]/MgAg [100 nm]/Ag [50 nm].

The deposition rates were maintained at 1.0 Å/s for α-NPD, ZnS and Alq3, 5.0 Å/s for BCzVBi doped CBP, MgAg and Ag, and 0.1 Å/s for a LiF as determined using a quartz crystal monitor. The emitting areas of all the devices were fixed at 1 mm². The FL of the 0.5wt% BCzVBi doped CBP neat film was 1.5 ns, and it was short enough to realize the fast modulation speed [8].

We measured J-V-L characteristics using an electrometer (HP4140B, Hewlett Packard) and a luminance color meter (BM-7, Topcon). The current efficiency was determined from J-V-L characteristics. In addition, we also measured the EL intensity as a function of the frequency of an applied sine wave voltage to evaluate the modulation speed of HOLEDs. The sine wave and bias voltages were applied to the HOLEDs using a programmable FM/AM standard signal generator (SG-7200, KENWOOD) and a DC power supply (IPS-3610D, ISO-TECH), respectively. The output EL intensity was observed using an avalanche photodiode (S5343, Hamamatsu Photonics) and an oscilloscope (DL-1740, Yokogawa Elec.). The frequency dependence of EL intensity was measured by changing the modulation frequency of the sine wave voltage ranged from 0.1 to 20 MHz. The cutoff frequency of the measurement setup was about 900 MHz. Therefore, there was no influence of measurement results of the frequency response.

**Results and discussion**

Figure 1 shows J-V-L characteristics of devices A and B, which consisted of ZnS and Alq3 as ETLs, respectively. The light turn-on voltage of the device A was lower than that of the device B. In our experiment, the exact mechanism of electron transport from a metal cathode to an EML. However, this experimental result in Fig. 1 indicates that the electron injection efficiency from a metal cathode of the device A is higher than that of the device B. The difference in the electron transport is most likely due to the electron mobility of ZnS and Alq3 [12, 13]. Furthermore, the luminance of the device A was higher than that of the device B. It achieved 500 cd/m² at 7 V, a lower voltage than that of the device B.

We show in Fig. 2, the current efficiency-current density characteristics of devices A and B. The current efficiency of the device A was 0.5 cd/A at a current density of 100 mA/cm², while that of the device B was 1.5 cd/A. The decrease in the current efficiency of the device A may be due to the unbalanced carrier in the EML because of the
of a large emitting area and a low-drive voltage. We attribute that the modulation speed was limited by other
limiting factors, such as the FL of organic light-emitting materials, the capacitance of emitting areas, and the energy
gap at metal/organic and organic/organic interfaces.

On the contrary, the cutoff frequency of the device B decreased with the decrease in the applied sine wave
voltage. Previous papers demonstrated that the electron mobility of Alq3 decreases with the decrease in the applied
voltage [13]. Therefore, the decrease in the electron mobility at a lower applied voltage causes the long electron
traveling time from a metal cathode to an EML, which led to the poor cutoff frequency of the device B, as shown in
Fig. 4.

In summary, we demonstrated fast-response HOLEDs with a ZnS electron transport layer for the application of
light sources to optical communications. The modulation speed and J-V-L characteristics have been evaluated. By
utilizing a ZnS as an ETL, we found that the modulation speed can be improved compared with the result obtained
using an organic electron transport material, Alq3. This result is mainly attributed to the high electron mobility of a
ZnS electron transport layer. A maximum cutoff frequency of 20.6 MHz has been achieved, and the cutoff frequency
is independent of the applied voltage.

Acknowledgements The part of this work was supported by Fujikura Ltd. and CLUSTER (the second stage) of Ministry of
Education, Culture, Sports, Science and Technology, Japan.

References

Copyright line will be provided by the publisher