Elastomeric composition for intrinsically stretchable organic light emitting diodes

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1 ABSTRACT

In order to manufacture intrinsically stretchable organic light emitting diodes, the surface energy of the mixed composition containing elastomers was analyzed as a highly efficient stretchable emitting layer (EML), and the performances of the devices were compared when elastomers and EML materials were mixed.

2 INTRODUCTION

In the past decades, research on stretchable electronics has been actively conducted and has made significant progress. Many researchers have been working on making devices with excellent mechanical properties that are bendable, stretchable, and twistable. To impart tension to electronic devices, we have applied special architectures such as out-of-plane wrinkles, rigid islands with deformable interconnects, and topographic buckles. However, we found that implementing a sophisticated architecture is very difficult.. Therefore, we studied the devices that can be intrinsically stretched to avoid the process of fabricating devices with such structures as aforementioned.

Thus, we are developing intrinsically stretchable organic light emitting materials to prepare such an intrinsically stretchable organic light emitting diode (OLEDs). Basically, in order to make the light emitting layer elastic, the elastomer and the emitting materials were mixed or blended. Especially, we found the best conditions to provide the ideal compatibility to maintain the luminescent properties by analyzing the difference in surface energy between the two materials. Then, we have manufactured devices using various elastomers, and confirmed that the better the compatibility, the better the device performance. This means that we could find the best composition as an intrinsically stretchable luminescent materials without increasing hopping distance between emitting species inside elastomeric matrix.

However, poor compatibility can lead to a longer distance between the luminescent material and the elastomer, which can interfere with hopping and reduce efficiency. As a result, it was concluded that the use of an elastic material with a surface energy similar to that of the luminescent material can improve the performance of stretchable OLED devices.

EXPERIMENT

The indium tin oxide (ITO) substrate with a 9 mm² emission area was used to make a solution processed OLED device. The substrates were cleaned with acetone and isopropyl alcohol for 20 min each by sonication and then treated with UV-ozone for 10 min. PEDOT: PSS (Heraeus Clevios ™ P VP CH 8000) as a hole injection layer (HIL) was spin coated under ambient conditions and immediately transferred to an N₂ filled glove box and annealed at 160 °C for 20 min. Then it was irradiated by UV-O3 treatment equipment to eliminate all the remained organic impurities and to improve the wettability of substrate. The materials as a hole transport layer (HTL) was spin casted and annealed. In the case of hole transport materials (HTM) requiring thermal crosslinking, the pre-annealing and crosslinking were performed at 100 ° C for 10 minutes (pre-annealing) and then at 220 ° C for 60 minutes (crosslinking) to give TX-HTM (thermally crosslinkable -HTM). The emitting layer (EML) was then prepared by spin-coating by using EML solution with red, green host (RH, GH) doped with 5% of red, green dopants (RD, GD) blending with 35% KH-E1. The annealing of EML was performed at 80 ° C for 10 min inside glove box. The electron transport layer (ETL) was thermally deposited at a rate of 0.5 Å / s under typical vacuum conditions, and LiF and AI were also thermally deposited at 0.3 Å / s and 3 Å / s under typical vacuum conditions.

The following device structures are used for this study **Device A**: ITO / PEDOT:PSS (40nm) / TX-HTM : TFB(20%, 20nm)/ RH : RD (5%, 30nm) / TPBi (50nm) / LiF (1nm) / AI (100nm)

Device B: ITO / PEDOT:PSS (40nm) / TX-HTM : TFB(20%, 20nm) / RH : RD : KH-E1 (5%, 35%, 30nm) / TPBi (50nm) / LiF (1nm) / Al (100nm)

Device C: ITO / PEDOT:PSS (40nm) / TX-HTM : TFB(20%, 20nm)/ GH : GD (5%, 30nm) / TPBi (50nm) / LiF (1nm) / AI (100nm)

Device D: ITO / PEDOT:PSS (40nm) / TX-HTM : TFB(20%, 20nm) / GH : GD : KH-E1 (5%, 35%, 30nm) / TPBi (50nm) / LiF (1nm) / Al (100nm)

3 RESULTS

Many research groups studying stretchable OLEDs reported results of blending elastomers with super yellow (SY) polymers.[UCLA] So, we also did a feasibility study with that direction as well, but for SY, the driving voltage increased sharply as the blending ratio of the elastomer increased. Besides, it was concluded that the method of mixing polymer-based emitting materials with elastomers was inefficient due to a significant decrease in current efficiency. We tentatively conclude that the reason why the combination of these materials causes inefficient charge transport behavior is that the hopping distance between fibrils is too far apart than expected. Figure 1 shows the contact angle of elastomers and emitting materials used in this study. We observed the contact angles of 83.269°, 20.376°, and 34.574° for Elastomers KH-E1, KH-E2, and KH-E3, respectively. Meanwhile, the emitting materials, SY, Host doped with RD, Host doped with GD showed contact angles 82.911°, 82,495°, and 80.120°, respectively.

Based on these results, we came to the conclusion that KH-E1 is the elastomer that has the best compatibility with the EML materials we used.

Fig. 2 shows the information about the devices prepared in this study. The energy band diagram and the device structure were also shown in this figure.

Fig. 3(a) shows the current density - voltage (J-V) and luminance - voltage (L-V) characteristics of the EML layer depending on the presence or absence of the elastomer in red OLEDs. The driving voltage reaching 1000 cd/m² was 7.5V for Device A and 9.5V for Device B. The blending ratio of KH-E1 in EML was about 35 wt. %, which is a minimum ratio needed for stretching of the EML layer. Fig. 3(b) shows current efficiency and power efficiency curves. The maximum current efficiency achieved was 18.8 cd/A for Device A and 18.3 cd/A for Device B, showing only ignorable difference when elastomers were mixed, while power efficiency was 16.7 lm/W for device A and 11.7 device Β. 3(c) shows the lm/w for Fig. electroluminescence spectra of the red devices which shows no serious difference after mixing an elastomer inside FMI

Likewise, Fig. 4(a) shows the J-V and L-V characteristics of the EML layer depending on the presence or absence of the elastomer in green OLEDs. The driving voltage reaching up to 1000 cd/m² was 6V for Device C and 7.5V for Device D. The blending ratio of KH-E1 was also 35 wt. % to stretch the EML layer. The maximum current efficiency was 38.7 cd/A for Device C and 33.2 cd/A for Device D. Very interestingly, we couldn't see a serious decrease in efficiency even after we mixed 35 wt. % of elastomers in EML. Meanwhile, power efficiency was 33.5 lm/W for Device C and 21.5 lm/w for Device D. Fig. 4(c) shows the electroluminescence spectra of the green devices and also shows no difference

upon addition of elastomer.

Fig 5(a) is a diagram of the strain-stress relationship obtained in the process of stretching a specimen manufactured using Instron and returning it to its original state. As shown in the figure, when tensile stress is applied to our EML, it is increased up to 20%, but it is difficult to restore to the original dimension and permanent deformation of about 6% occurs. In the future, we plan to continue to conduct compositional research to increase the reproducibility of these characteristics.

Fig. 5(b) shows the original and stretched EML showing photoluminescence. Overall, it was found that the color of the EML remained the same at all times, regardless of whether the EML was pulled or contracted.

4 CONCLUSIONS

We have found an elastomer for a light emitting layer suitable for producing stretchable OLEDs as a solution process. In particular, we found a method of blending an elastomer whose surface energy is similar to that of a light-emitting material to enable stretching and without deteriorating EML properties.

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Fig. 3 (a) J-V-L (b) current efficiency – luminance (c) normalized EL (electroluminescent) spectra (at a brightness of 1000 cd/m2) of the solution processed red PHOLEDs of Device A and B.



Fig. 4 (a) J-V-L (b) current efficiency – luminance (c) normalized EL (electroluminescent) spectra (at a brightness of 1000 cd/m2) of the solution processed green PHOLEDs of Device C and D.



Fig. 5(a) Stress - Strain of thin polymer film substrates coated with EML (b) stretched EML showing photoluminescence