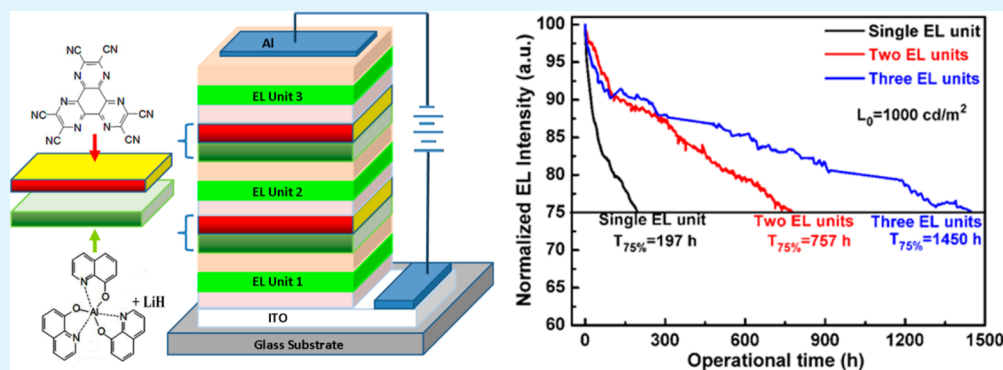


# Lithium Hydride Doped Intermediate Connector for High-Efficiency and Long-Term Stable Tandem Organic Light-Emitting Diodes

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## Supporting Information



**ABSTRACT:** Lithium hydride (LiH) is employed as a novel *n*-dopant in the intermediate connector for tandem organic light-emitting diodes (OLEDs) because of its easy coevaporation with other electron transporting materials. The tandem OLEDs with two and three electroluminescent (EL) units connected by a combination of LiH doped 8-hydroxyquinoline aluminum ( $\text{Alq}_3$ ) and 1,4,5,8,9,11-hexaazatriphenylene-hexacarbonitrile (HAT-CN) demonstrate approximately 2-fold and 3-fold enhancement in current efficiency, respectively. In addition, no extra voltage drop across the intermediate connector is observed. Particularly, the lifetime ( $T_{75\%}$ ) in the tandem OLED with two and three EL units is substantially improved by 3.8 times and 7.4 times, respectively. The doping effect of LiH into  $\text{Alq}_3$ , the charge injection, and transport characteristics of LiH-doped  $\text{Alq}_3$  are further investigated by ultraviolet photoelectron spectroscopy (UPS) and X-ray photoemission spectroscopy (XPS).

**KEYWORDS:** *n*-type dopant, LiH, tandem OLEDs, intermediate connector, long lifetime

## 1. INTRODUCTION

Organic light-emitting diodes (OLEDs) have been considered to be excellent candidates for next-generation flat-panel displays and solid-state-lighting sources owing to their advantages of high brightness, long lifetime, and low power consumption.<sup>1</sup> Nevertheless, development of both novel device structure and good organic materials are essential for the realization of high efficiency and long-term OLEDs. It is known that the OLED is actually a current-driven device. In order to achieve higher brightness, an OLED has to be operated at a relatively high current density, which will shorten the device lifetime. Fortunately, a tandem OLED, which includes at least two electroluminescent (EL) units connected via an intermediate connector can effectively resolve the aforementioned issue. The intermediate connector commonly functions as an internal electrode to produce charge carriers and to facilitate opposite carriers injection into the adjacent EL units. Therefore, a typical intermediate connector can be formed by conductive films,<sup>2,3</sup> organic heterojunction,<sup>4–7</sup> and *n*-doped/metal oxide junction<sup>8–13</sup> with good optical and electrical characteristic.<sup>14–16</sup> Particularly, an organic heterojunction based connector

generally consists of an *n*-type doped organic layer and a *p*-type doped organic layer to form a *p-n* junction. The *n*-type dopants in the doped organic layer are typically alkali metal, alkali metal compounds, alkaline earth metals, or alkaline earth metal compounds such as Li,<sup>17</sup> Cs,<sup>18</sup>  $\text{Cs}_2\text{CO}_3$ ,<sup>19</sup>  $\text{Li}_3\text{N}$ ,<sup>20,21</sup>  $\text{Li}_2\text{CO}_3$ ,<sup>22</sup> and  $\text{CsN}_3$ .<sup>23</sup> Recently,  $\text{KBH}_4$ <sup>24</sup> and  $\text{C}_{60}$ <sup>25</sup> were also used as the *n*-type dopant with strong electron donating properties. However, the aforementioned dopants are not atmospherically stable with the existence of moisture and oxygen. Moreover, the deposition rate of the alkali metals is difficult to control during evaporation. The alkaline carbonates keep changing the evaporation behaviors with reduced doping effects when the dopant sources are used multiple times. On the other hand, alkali metal nitride and the alkali metal azide may have lab safety issues if handled with carelessness. In fact, practically useful *n*-type dopants are much needed for real production. Therefore, it is necessary to search for air-stable

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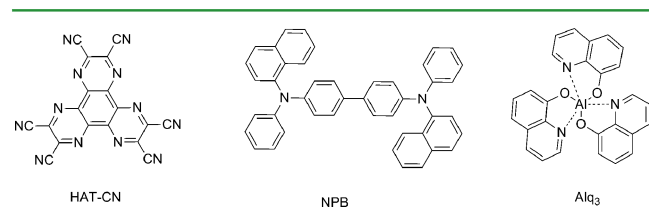
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and relatively easy evaporable *n*-type dopants in order to make good intermediate connectors of tandem OLEDs.

Herein, we demonstrate a simple but effective material, lithium hydride (LiH), as an *n*-dopant in the intermediate connector of tandem OLEDs. Although the decomposition temperature of LiH is about 850 °C, it is still lower than that of LiN<sub>3</sub> and Li<sub>2</sub>CO<sub>3</sub>. Therefore, LiH can be coevaporated relatively easily with common electron transporting materials due to its low melting temperatures. Particularly, LiH is decomposed into Li and H<sub>2</sub>, and only Li remains in the film during evaporation, which has a positive effect on the device stability because no other impurities will be introduced. LiH doped Alq<sub>3</sub> combined with HAT-CN are successfully used as intermediate connector for fabricating green and red fluorescent tandem OLEDs with two and three EL units. As a result, a more than 2-fold and 3-fold enhancement in current efficiency (12.3 cd/A and 9.6 cd/A vs 4.1 cd/A) can be obtained in two and three EL units OLED, respectively. More importantly, the tandem OLEDs present remarkably improved device stabilities owing to the stable and easy evaporable processed LiH as an *n*-dopant.

## 2. EXPERIMENTAL SECTION

**2.1. Materials.** LiH powder was purchased from Aldrich. The Alq<sub>3</sub>, *N,N'*-bis(1-naphyl)-*N,N'*-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB) were purchased from Nichem Fine Technology Co., Ltd. LiH doped Alq<sub>3</sub> was used as an *n*-doped organic layer, which was combined with HAT-CN acting as an intermediate connector in tandem OLEDs. NPB was used as a hole-transporting layer. Figure 1 shows the molecular structures of organic materials upon investigation.



**Figure 1.** Molecular structures of the organic materials used in this work.

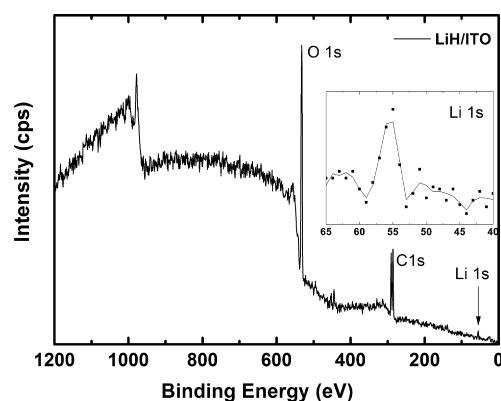
**2.2. Device Fabrication.** OLEDs were fabricated on cleaned glass substrates precoated with 110 nm ITO (15 Ω/sq) and treated by UV ozone for 15 min. All layers were deposited by thermal evaporation under a vacuum pressure around 10<sup>-6</sup> Torr. LiH was evaporated at the rate in a range of 0.22–0.44 Å/s. The other organic materials and metal were evaporated at the rates in a range of 3–5 Å/s and 8–10 Å/s, respectively. Green color OLEDs with single, two, and three EL units were fabricated, respectively. The layer structures and units for all devices are listed in Table 1. The doping ratio of LiH in Alq<sub>3</sub> was kept as an optimized ratio of 1.2 vol %.

**2.3. Measurement.** The layer thickness and deposition rate of each material were monitored in situ using an oscillating quartz thickness monitor. A photometer (Photo Research Spectra Scan PR 655) was used to measure the electroluminescent (EL) spectra and

Commission International de l'Eclairage (CIE) coordinates of all devices. Combined with a constant current source (Keithley 2400 Source Meter), the current–voltage characteristics were measured simultaneously. The active area of the devices is 0.09 cm<sup>2</sup>. X-ray photoelectron spectroscopy (XPS) was used to investigate the composition of evaporated LiH film. Ultraviolet photoelectron spectroscopy (UPS) was carried out to evaluate the energy levels of LiH doped- and nondoped Alq<sub>3</sub> films. UV/vis spectrophotometer (PerkinElmer Lambda 750) was used to measure the transmittance of charge generation layer. The devices were encapsulated by the cover glasses for lifetime evaluations.

## 3. RESULTS AND DISCUSSION

**3.1. LiH Decomposition during Evaporation.** The LiH decomposition has been reported to proceed based on the mechanism<sup>26</sup> of 2LiH → 2Li + H<sub>2</sub>. During evaporation, LiH is decomposed into Li and H<sub>2</sub>. Only Li is deposited onto the substrate because H<sub>2</sub> is extracted from the chamber by continuous pumping. For the confirmation of Li liberation, a composition analysis of evaporated LiH film was carried out by XPS. Figure 2 shows a full XPS scan for the evaporated LiH (10



**Figure 2.** A full XPS scan of 10 nm LiH evaporated on ITO-coated glass substrate; (inset) Li 1s core level of the as-prepared film.

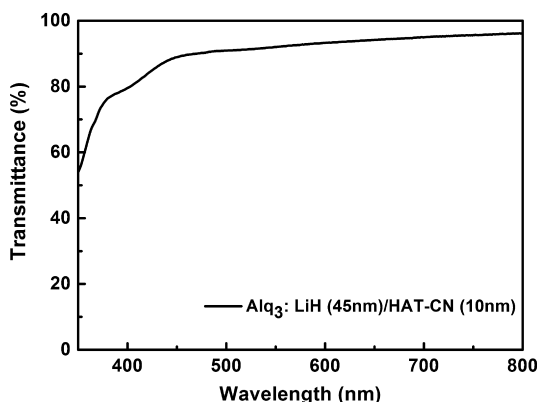
nm) on ITO substrate. The inset of Figure 2 is the magnification near 56 eV. The Li 1s core level was observed at 56 eV, which was suggested to have originated from Li and lithium oxide due to the liberation of Li from LiH during evaporation. Therefore, LiH is expected to have an *n*-type doping effect with other organic materials during coevaporation.

**3.2. LiH as an *n*-Dopant in the Intermediate Connector.** **3.2.1. Optical Transmittance.** One requirement for the tandem OLEDs to function efficiently is that the optical transparency of the layers, including the organic EL units and the intermediate connector, should be as high as possible to permit efficient light extraction from the device. According to a simple calculation, a tandem OLED will not work effectively if the optical transmission of the intermediate connector is below

**Table 1.** Layer Structures of the OLED Devices

device	layer structure
single EL unit	ITO/HAT-CN (10 nm)/NPB (75 nm)/Alq <sub>3</sub> (60 nm)/LiH (2 nm)/Al
two EL units	ITO/HAT-CN/EL-G/OC/EL-G/Alq <sub>3</sub> :LiH/Al
three EL units	ITO/HAT-CN/EL-G/OC/EL-G/OC/EL-G/Alq <sub>3</sub> :LiH/Al
EL-G	NPB (70 nm)/Alq <sub>3</sub> (30 nm)
OC	Alq <sub>3</sub> :LiH (1.2 vol %, 45 nm)/HAT-CN (10 nm)

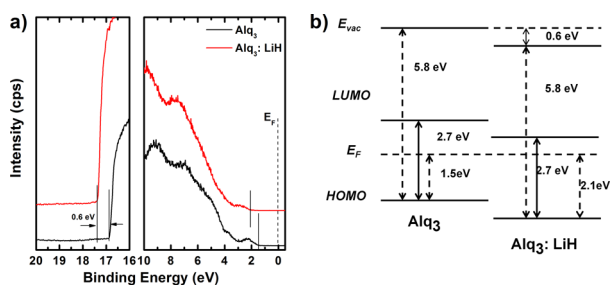
70% at the emitting peak wavelength. No matter how many EL units there are in the device, the EL efficiency can never be doubled compared to the reference OLED with single EL unit.<sup>27</sup> Figure 3 shows the optical transmittance of Alq<sub>3</sub>:LiH



**Figure 3.** Transmittance spectrum of Alq<sub>3</sub>:LiH (45 nm)/HAT-CN (10 nm).

(45 nm)/HAT-CN (10 nm) in the visible light range. The intermediate connector exhibits a high transmittance of 86–96% in the wavelength range of 430–780 nm, which is suitable as an intermediate connector in the tandem OLEDs.

**3.2.2. Doping Effects of LiH in Alq<sub>3</sub>.** Another important issue in tandem OLEDs is the effective charge injection and good transport for the intermediate connector. To investigate the role of LiH in the electron injection and transport characteristics in the intermediate connector, we further examined the UPS and XPS spectra of LiH doped Alq<sub>3</sub> films. Figure 4a shows the UPS spectra for the secondary electron

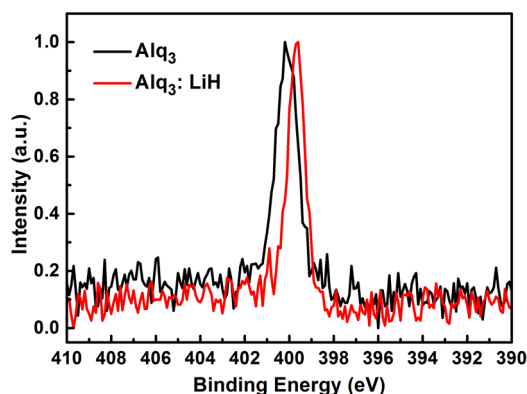


**Figure 4.** (a) UPS spectra of Alq<sub>3</sub> and Alq<sub>3</sub>:LiH. (b) Schematic energy-level diagrams of pristine Alq<sub>3</sub> and Alq<sub>3</sub>:LiH films.

cutoff region and the highest occupied molecular orbital (HOMO) region of the Alq<sub>3</sub> and Alq<sub>3</sub>:LiH (1.2 vol %) films. The edge of the HOMO for pristine Alq<sub>3</sub> is located 1.5 eV below the Fermi level. Meanwhile, the ionization energy is decided to be 5.8 eV. After a 1.2 vol % LiH doping into Alq<sub>3</sub>, the HOMO level relative to the Fermi level shifts toward a higher binding energy by about 0.6 eV compared to that of pristine Alq<sub>3</sub>, indicating that the Fermi level of Alq<sub>3</sub> was moved toward its lowest unoccupied molecular orbital (LUMO) by the LiH doping. The band gap of Alq<sub>3</sub> is about 2.7 eV,<sup>28</sup> and the LUMO of Alq<sub>3</sub>:LiH is just 0.6 eV above the Fermi level, which indicates an *n*-doped Alq<sub>3</sub> film by LiH doping. Therefore, the electron injection barrier expects to be reduced, owing to the strong *n*-type doping effect of LiH. In addition, the secondary cutoff position of Alq<sub>3</sub> shifted below about 0.6 eV after LiH-doped Alq<sub>3</sub>, as shown in Figure 4a, suggesting that the vacuum

level shifts down toward the Fermi level. This means that the LiH doped with Alq<sub>3</sub> forms charge exchanges, which will produce a dipole at the interface. Figure 4b shows a schematic diagram of energy levels of Alq<sub>3</sub> and Alq<sub>3</sub>:LiH (1.2 vol %) for comparison. Obviously, the electron barrier is decreased after doping because LUMO level of the Alq<sub>3</sub>:LiH shifts down toward the Fermi level.

Figure 5 shows the N 1s core levels in the XPS spectra of the pure Alq<sub>3</sub> and LiH-doped Alq<sub>3</sub> (1.2 vol %) films. In the case of

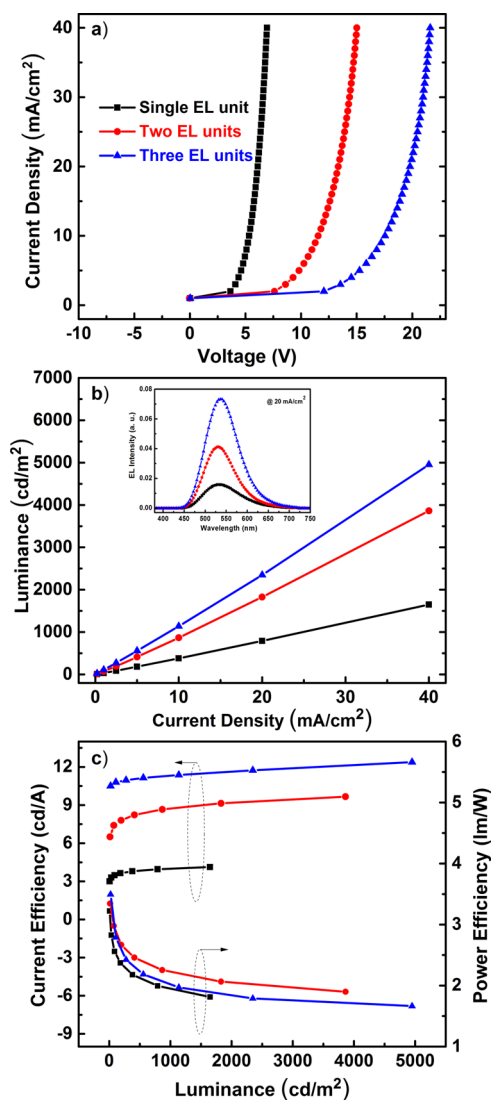


**Figure 5.** XPS spectra of N 1s core level of pristine Alq<sub>3</sub> and Alq<sub>3</sub>:LiH films.

LiH doping into Alq<sub>3</sub>, the core level of N 1s shifts toward lower binding energy by 0.6 eV. It means that the charge transfer from Li to Alq<sub>3</sub> occurs effectively, which facilitates electron transfer from HAT-CN to the LiH-doped Alq<sub>3</sub> layer.

**3.3. Tandem OLEDs.** Figure 6 compares the EL performance of Alq<sub>3</sub>-emission based fluorescent green OLEDs with single, two, and three EL units by using Alq<sub>3</sub>:LiH/HAT-CN as the intermediate connectors (ICs). The typical performance parameters including turn-on voltage ( $V_{on}$ ), current efficiency ( $\eta_c$ ), power efficiency ( $\eta_p$ ), and Commission International de l'Éclairage (CIE) coordinates ( $x, y$ ) are summarized in Table 2.

Under an operational current density of 20 mA/cm<sup>2</sup>, the OLEDs with single, two, and three units require a driving voltage of 6.2, 13.9, and 20.6 V corresponding to the luminance of 791, 1828, and 2347 cd/m<sup>2</sup>, and the current efficiency of 3.9, 9.1, and 11.7 cd/A, respectively. The driving voltage increases by about two or three times with the EL unit, whereas the tandem OLEDs exhibits more than two or three times improvement in luminance. The inset in Figure 6b shows the EL spectra of OLEDs with single, two, and three EL units measured at 20 mA/cm<sup>2</sup>. Obviously, the tandem OLEDs with two or three units connected by Alq<sub>3</sub>:LiH/HAT-CN layers demonstrate more than two times and/or three times enhancements in the luminance intensity while keeping the color basically unchanged. With a combined result, the tandem OLEDs present higher power efficiency than the reference device with only one EL unit. It suggests that the IC has excellent charge injection and transport characteristics. In addition, the extra voltage across the IC can be neglected due to the power efficiency of the OLEDs with two and three EL units are all higher than that of the single EL device. Similar results were also observed in 4-(dicyanomethylene)-2-*t*-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4*H*-pyran (DCJTb) based red tandem OLEDs using Alq<sub>3</sub>:LiH/HAT-CN as the IC (EL performance can be seen in Figures S2 and S3 in the Supporting Information).



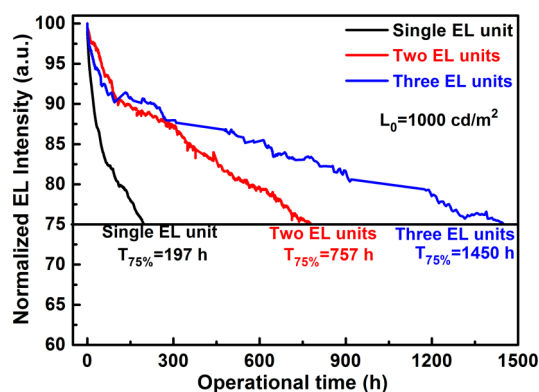
**Figure 6.** EL performances of green fluorescent OLEDs with single, two, and three EL units. (a) Current density–voltage characteristics. (b) Luminance–current density characteristics and (inset) EL spectra at 20 mA/cm<sup>2</sup> in the OLEDs with single, two, and three EL units. (c) Current and power efficiency–luminance characteristics.

**Table 2. Electroluminescence Characteristics of the Devices**

device <sup>a</sup>	V(V) <sup>b</sup>	$\eta_c$ (cd/A) <sup>c</sup>	$\eta_p$ (lm/W) <sup>c</sup>	CIE (x,y) <sup>d</sup>
single EL unit	2.8	4.1, 3.5, 4	3.2, 2.4, 1.9	(0.33, 0.56)
two EL units	5.8	9.6, 7.6, 8.8	3.3, 2.7, 2.1	(0.33, 0.58)
three EL units	9.9	12.3, 10.8, 11.3	3.5, 2.8, 1.9	(0.37, 0.55)

<sup>a</sup>The devices of Alq<sub>3</sub> emission based fluorescent green OLEDs with single, two, and three EL units. <sup>b</sup>Voltages at 0.2 mA/cm<sup>2</sup>. <sup>c</sup>Efficiencies in the order of maximum at 1000 and 10 000 cd/m<sup>2</sup>. <sup>d</sup>Commission International de l'Éclairage coordinates measured at 20 mA/cm<sup>2</sup>.

Figure 7 shows the operational lifetime ( $T_{75\%}$ , defined as the time when the actual luminance decays to 75% of the initial value) of green fluorescent OLEDs with single, two, and three EL units. The  $T_{75\%}$  lifetime of the device was measured at the same initial luminance of 1000 cd/m<sup>2</sup>. As mentioned above, compared to the single EL unit based device, the tandem OLEDs need only about one-half (in a two EL unit based device) or one-third (in a three EL unit based device) of the current density for obtaining the same luminance, which



**Figure 7.** Operational stability of devices with single, two, and three EL units with an initial luminance of 1000 cd/m<sup>2</sup>.

suggests that the device lifetime would be improved by using the tandem structure. From this figure, the lifetime ( $T_{75\%}$ ) of OLED with single, two, and three EL units is 197, 757, and 1450 h, respectively, corresponding to 3.8 and 7.4 times improvements in the device lifetime, which means that LiH can indeed function as a good *n*-type dopant in the IC for tandem OLEDs with improved device lifetime.

#### 4. CONCLUSIONS

In summary, we demonstrate that LiH can effectively function as an *n*-dopant for Alq<sub>3</sub>, which can be combined with HAT-CN as an intermediate connector for tandem OLEDs. The tandem OLEDs with two and three EL units were successfully fabricated with 2-fold and 3-fold enhancement in current efficiency, respectively, by using Alq<sub>3</sub>:LiH/HAT-CN as an intermediate connector. In addition, no obvious voltage drop resulted from the ICs in the tandem OLEDs, which is beneficial for lowering the device power consumption. The lifetime ( $T_{75\%}$ ) of OLED with two and three EL units was substantially improved by 3.8 times and 7.4 times, respectively. The doping effect of LiH into Alq<sub>3</sub>, the charge carrier behavior and energy levels of LiH-doped Alq<sub>3</sub> were investigated in detail by XPS and UPS analyses. Enhanced current efficiency in the tandem OLEDs was achieved by good charge carrier balance owing to improved the carrier injection and the reduced barrier at the intermediate interfaces. These results reveal the potential of LiH as a novel *n*-type dopant, which provides an alternative for robust intermediate connector materials in tandem OLEDs.

#### ■ ASSOCIATED CONTENT

##### Supporting Information

Layer structures of red fluorescent OLED devices; proposed energy level diagram of a red tandem OLED with three units; EL performances of green OLEDs with different thickness of Alq<sub>3</sub>:LiH (35, 45, 55, and 65 nm); current density–voltage curves; current density–luminance curves; current efficiency and power efficiency of the single-unit and tandem OLEDs; EL spectra at 20 mA/cm<sup>2</sup> in the OLEDs; EL performances of red fluorescent OLEDs with single, two, and three EL units; current density–voltage–luminance curves; current efficiency and power efficiency of the single-unit and tandem OLEDs; and EL spectra at 20 mA/cm<sup>2</sup> in the OLEDs with single, two, and three EL units. This material is available free of charge via the Internet at <http://pubs.acs.org>.



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## Notes

The authors declare no competing financial interest.

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