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Lifetime Improvement of Organic Light Emitting Diodes using LiF Thin Film and UV Glue Encapsulation

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This work demonstrates the use of lithium fluoride (LiF) as a passivation layer and a newly developed UV glue for encapsulation on the LiF passivation layer to enhance the stability of organic light-emitting devices (OLEDs). Devices with double protective layers showed a 25-fold increase in operational lifetime compared to those without any packaging layers. LiF has a low melting point and insulating characteristics and it can be adapted as both a protective layer and pre-encapsulation film. The newly developed UV glue has a fast curing time of only 6 s and can be directly spin-coated onto the surface of the LiF passivation layer. The LiF thin film plus spin-coated UV glue is a simple packaging method that reduces the fabrication costs of OLEDs. [DOI: 10.1143/JJAP.47.5676]

KEYWORDS: OLED, encapsulation, packaging, UV glue, LiF

1. Introduction

In recent years, organic light-emitting diodes (OLEDs) have seen increased application in portable displays, such as MPEG Audio Layer III (MP3) players, mobile phones, and mobile televisions due to the advantages of organic materials and an optimized vertical structure. Much research has been devoted to improving OLED performance by modifying the electrode structures[1,2] or incorporating buffer layers.[3–5] It is widely recognized that a common cause of degradation in OLEDs is the influence of moisture and oxygen.[6] However, longer device operating lifetime is a critical issue for OLEDs. The most popular encapsulation technique for protecting OLEDs from moisture and oxygen uses epoxy and a cover glass or a metal lid with large amounts of desiccant.[7] However, these thick and heavy lids limit OLED applications in mobile and flexible displays. The desiccant typically resides in the device, which affects the light passing through the cover glass of top emitting OLEDs.

Many encapsulation techniques have been proposed to avoid moisture and oxygen attacks. Dielectric layers such as silicon oxide,[8] silicon nitride,[9] and aluminum oxide[10] from plasma-enhanced chemical vapor deposition (PECVD) or plasma-enhanced atomic layer deposition (PEALD) are attractive and widely used as passivation layers. In addition, the dielectric/organic mixed multilayers[11] are also popular candidates. To date, some layers, such as silicon carbide,[12,13] carbon-coated oxide layer,[14] and the fluorocarbon film,[15] proved that carbon-incorporated thin films present an excellent barrier against moisture permeation due to their hydrophobic nature. However, the thermal expansion between the aluminum cathode top and such subsurface layers are different.

The use of thin films instead of glass or metal lids has been an interesting development in OLED encapsulation, such as in the SiN/2-methyl-9,10-di(2-naphthyl)-anthracene (MADN)/SiON multilayer encapsulation technique.[16] The motivation for us is to find a useful organic passivation layer to coat onto the aluminum cathode to balance the thermal stress and to produce a lightweight, thin, flexible device. The merits of LiF are described as follows: (1) The evaporation temperature for LiF is low enough that will not destroy the organic devices during the evaporating processes compared to the temperatures required for silicon oxide or silicon nitride in sputtering or PECVD. (2) The systems and processes for evaporating LiF are simple and low-cost. The same vacuum system as for evaporating cathode metals can be employed for LiF passivation. It is unnecessary to use sputtering or PECVD. (3) The LiF film has high electrical resistance due to its wide band gap. Using LiF film as a first passivation layer does not affect the OLED's electrical properties. The main purpose of using LiF thin film as passivation layers is to protect the organic layers and not to be destroyed by UV glue, which is spin-coated directly onto OLED cathode surfaces. LiF film is employed as the cathode passivation layer using a double protective layer encapsulated in a newly developed UV glue onto the LiF layer to improve OLED performance.

2. Experimental Methods

Patterned indium tin oxide (ITO) glasses with a sheet resistance of 5 ohm/sq were used as the anode and substrate throughout these experiments. The routine cleaning procedure included sonication in acetone and methanol, rinsing in de-ionized water for 5 min, drying in an oven, followed by a 90 s oxygen plasma treatment.[17] Forty-nanometer-thick *N*,*N*′-bis(naphthalen-1-yl)-*N*,*N*′-bis(phenyl)-benzidine (NPB) was used as the hole transport layer (HTL) and 60-nm-thick tris(8-hydroxyquinolinato)aluminum (Alq_3) as the electron

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Table I. Lifetime of devices encapsulated with LiF thin film plus ITRI's UV glue on cathode surfaces with different experimental parameters (but UV glue curing time is fixed at 6 s).

No.	Sub.	Anode	NPB (nm)	Alq_3 (nm)	Cathode	LiF thin film layer (nm)	UV glue	UV glue spin time (two steps)		Half-lifetime (h)
								1500 rpm (s)	3500 rpm (s)	
A	Glass	ITO	40	60	LiF/Al 0.2/70	×	×	×	×	4
B						80	×	×	×	18
C							○	20	30	63
D							○	30	30	63
E						100	×	×	×	33
F							○	20	30	70
G							○	30	30	70
H						120	×	×	×	44
I							○	20	30	100
J							○	30	30	65

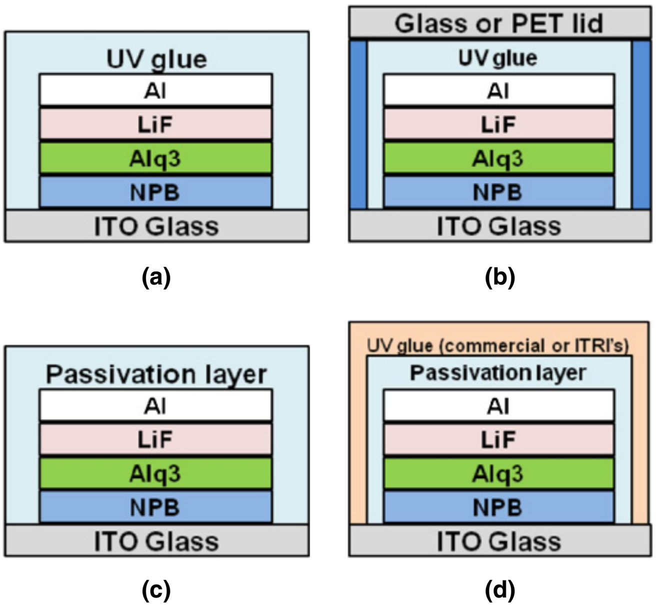

Fig. 1. (Color online) Packaging methods: (a) UV glue spin-coating only, (b) glass or PET lid for package, (c) LiF thin film passivation only, and (d) LiF plus UV glue spin-coating.

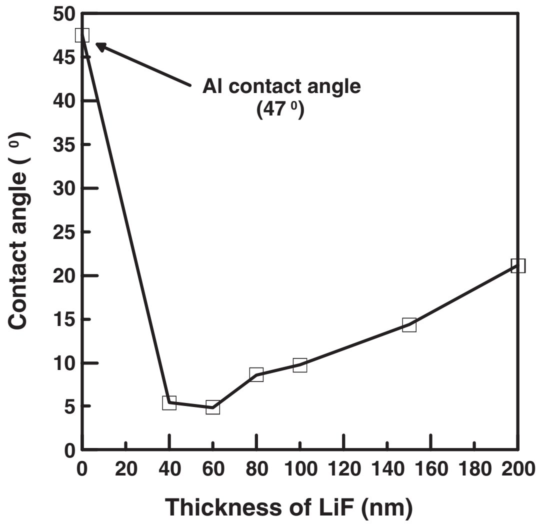

Fig. 2. Contact angles of water drops on the surface of LiF as a function of thickness.

transport and emissive layers (ETL). They were sequentially deposited on the ITO anode. A layer of 0.2-nm-thick LiF and 70-nm-thick Al was employed as the cathode. Deposition pressures were maintained at 10^{-6} mbar for the organic and metal layers. The deposition rates were monitored with a quartz oscillator and controlled at 0.5, 0.1, and 1 nm/s for LiF, organic, and Al, respectively. Shadow masks were used to define an active emitting area of 0.25 cm^2. After device fabrication, LiF film was directly deposited onto the device using vapor evaporation without breaking the vacuum. The OLED structure is glass/ITO/NPB/Alq_3/LiF/Al/LiF. To study OLED lifetime for various thicknesses of LiF passivation layers, the new UV glue developed by ITRI Material Research Laboratories was spin-coated onto the LiF film surface in a first step that was 1500 rpm for 20 s and a second step that was 3500 rpm for 30 s, as shown in Fig. 1(a). Keithley 2400 and Spectra Scan PR650 were applied to measure the current versus voltage (I–V), luminance versus voltage (L–V) characteristics and luminescence spectra of the completely manufactured components. A lifetime measuring system especially designed for OLEDs was applied to measure the OLED lifetime for 5 mA current, the measured lifetimes for different encapsulations are shown in Table I.

3. Results and Discussion

3.1 LiF passivation layer

Figure 1 shows the schematic structures of various OLED packaging methods. The structure in Fig. 1(c) was used for the protective ability test with and without the LiF passivation layer. Figure 2 shows the contact angles of water drops for OLEDs coated using 40–200-nm-thick LiF film on the cathode. As seen from Fig. 2, when the LiF thickness increases from 40 to 200 nm, the contact angle varies from 5 to 21°, all of which are much smaller than the contact angles of Al film. Second, the solubility of LiF film in water is 0.133 wt % at 25.4 °C.[18] The water solublility and small contact angles reveal the hydrophilic properties of LiF.

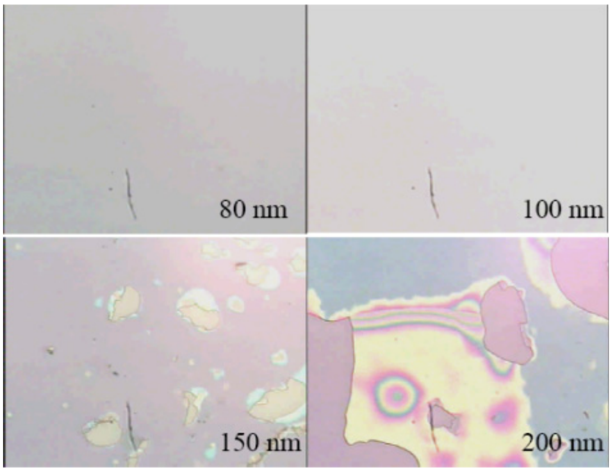

Fig. 3. (Color online) Surface of the LiF film for thicknesses of 80, 100, 150, and 200 nm.

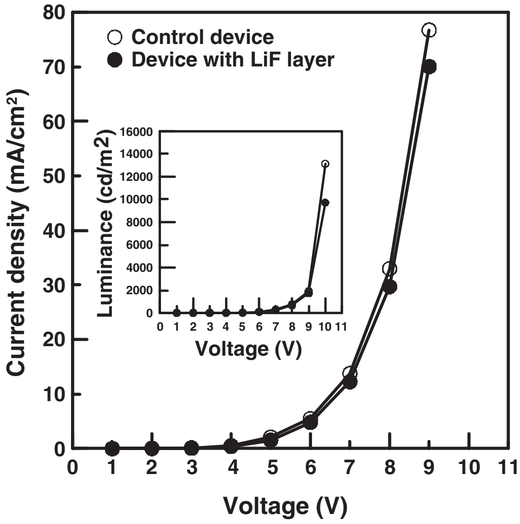

Fig. 4. *J–V* and *L–V* characteristics with and without LiF film.

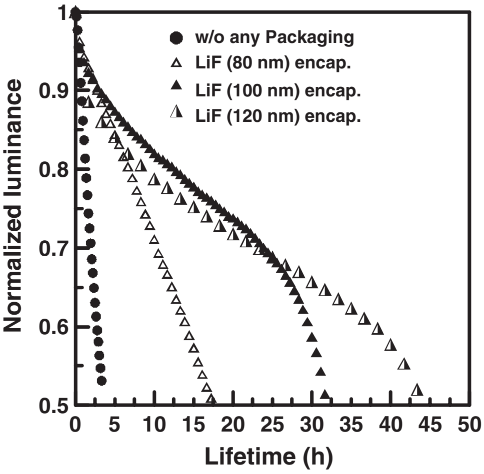

Fig. 5. Lifetime for different thicknesses of LiF passivation thin film.

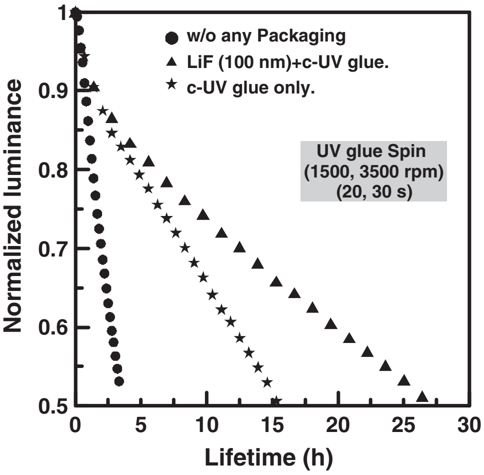

Fig. 6. c-UV glue tests.

This kind of passivation film will cause the OLEDs degrade easily. Therefore, these results suggest that little permeation took place when the LiF layer was deposited at a thickness over 80 nm. That benefit contributes to the OLED lifetime. The adhesive force between LiF and Al was observed as shown in Fig. 3. The LiF films deposited with 80- and 100-nm-thick Al were smooth. At a thickness of 150 nm the LiF film appears to peeled slightly. Furthermore, the 200-nm-thick LiF/Al film coating peeled off dramatically. Figure 4 shows the *J–V* and *L–V* characteristics of devices with and without LiF film on the cathode. Apparently, no observable difference was found between the two devices. This shows that the LiF deposition process did not cause any detectable damage to the underlying OLED. Figure 5 shows the normalized luminance versus operating time for different LiF passivation film thicknesses of 80, 100, and 120 nm. The half-lifetime was 4 h for the device without a LiF coating layer. The half-lifetime gradually increased to 18, 33, and 44 h when LiF layers were employed. For comparison with the uncoated device with 4-h half-lifetime, the optimum protective layer thickness of 120 nm produced an elevenfold longer lifetime than the device without any LiF film passivation layer. The increased half-lifetime can be attributed to a low rate of moisture permeation into the LiF passivation layer. The lifetime clearly shows that a 120 nm LiF passivation layer coating on a cathode can significantly enhance OLED lifetime.

3.2 *Comparisons of different UV glues*

Figure 6 shows the lifetime for spin-coating commercialized UV glue (c-UV glue) directly and with double protective layers. The packaging element directly spin-coated with c-UV glue produced shorter half-lifetime. The hydrosphere in the colloid affects the element and causes a shorter lifetime. Hence, the LiF passivation layer is desirable for improving device lifetime for low oxygen and moisture permeation rates through the c-UV glue coating. As shown in Fig. 7, the newly developed UV glue from Industrial Technology Research Institute of Taiwan (ITRI's UV glue) was employed for the encapsulation tests. Figure 8 shows the unit lifetime for devices without any packaging, LiF/c-UV glue, LiF encapsulation only, and LiF/ITRI's UV glue. The LiF encapsulated device exhibited superior lifetime compared with the LiF/c-UV glue device. The reduction in lifetime when the LiF passivation layer was added occurred because the c-UV glue has a longer curing time of 90 s. An

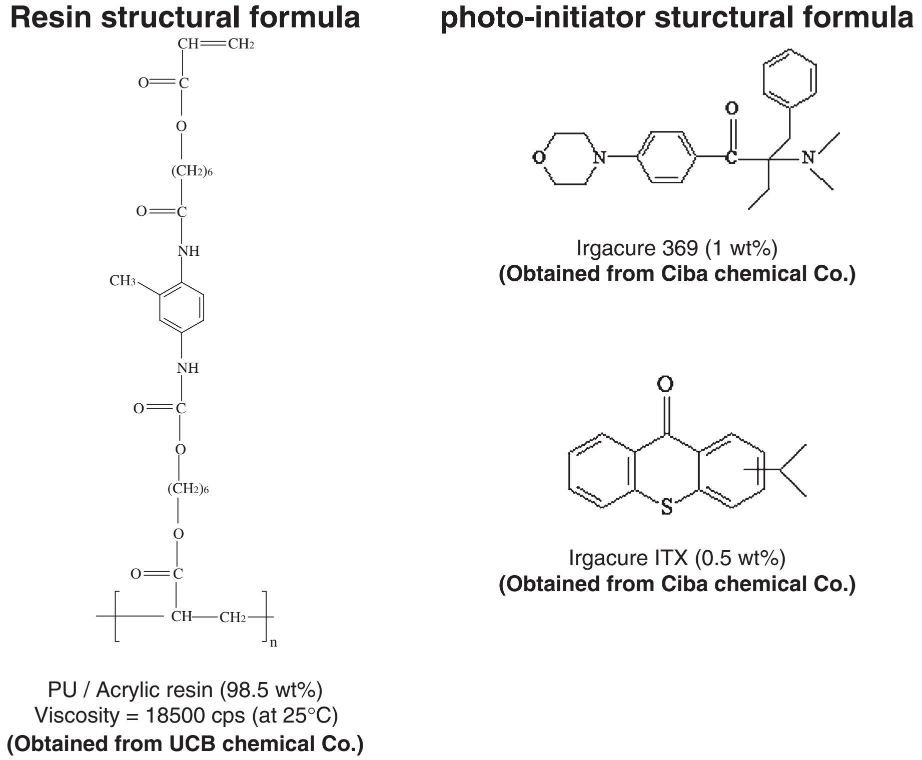
Resin structural formula

photo-initiator sturctural formula

Irgacure 369 (1 wt%)
(Obtained from Ciba chemical Co.)

Irgacure ITX (0.5 wt%)
(Obtained from Ciba chemical Co.)

PU / Acrylic resin (98.5 wt%)
Viscosity = 18500 cps (at 25°C)
(Obtained from UCB chemical Co.)

Fig. 7. The new-developed UV glue (ITRI's UV glue) is composed of a high-density resin and a special photo-initiator. The ITRI's UV glue has a shorter curing time to prevent damage to the OLED device during the UV light exposure period.

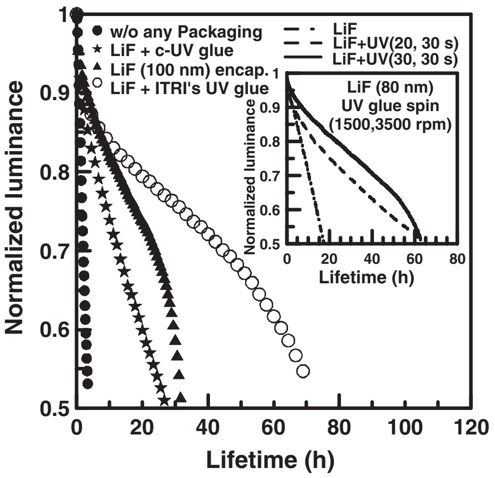

Fig. 8. Tests of ITRI's UV glue, where LiF is 100 nm thick. The inset in this figure shows the lifetime of UV glue on a LiF passivation layer (80 nm) for different spin times.

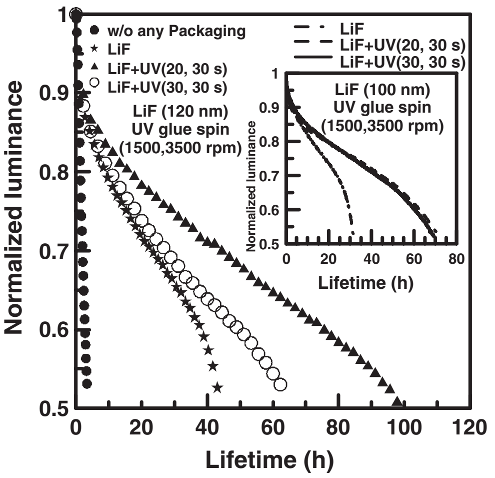

Fig. 9. Lifetime for UV glue on LiF passivation layers (120 nm) for different spin times. The inset in this figure shows lifetimes for UV glue onto LiF passivation layers (100 nm) for different spin times.

organic device will suffer degradation from longer exposure to UV irradiation. The newly developed ITRI UV glue has a fast encapsulation curing time of only 6 s. Using the LiF/IRIT's UV glue as a double protective layer produces a threefold lifetime increase compared with a LiF/c-UV glue encapsulated device. Based on these results, we consider the LiF film to be a passivation layer and the ITRI's UV glue as a coating layer for double protective packaging devices.

3.3 Double protective layer

To further block the influence of moisture and oxygen, the ITRI's UV glue was spin-coated as a second packaging layer upon a LiF passivation layer. The first step was spin-coating at 1500 rpm for 20 and 30 s for uniform deposition. The second step was spin-coating at 3500 rpm for 30 s for a thinner deposition. The insets in Figs. 8 and 9 show that when the subsurface passivation layer thickness was 80 or 100 nm, at 1500 rpm, the half-lifetime was similar for spin times of 20 and 30 s. As shown in Fig. 9, different from 120-nm-thick LiF layer thickness, the half-lifetime was increased to 100 h as the spins time in the first step was controlled to 20 s and the spin time in the second step was held to 30 s. However, when the first spin time was adjusted to 30 s, the half-lifetime was dramatically reduced to 65 h for the same

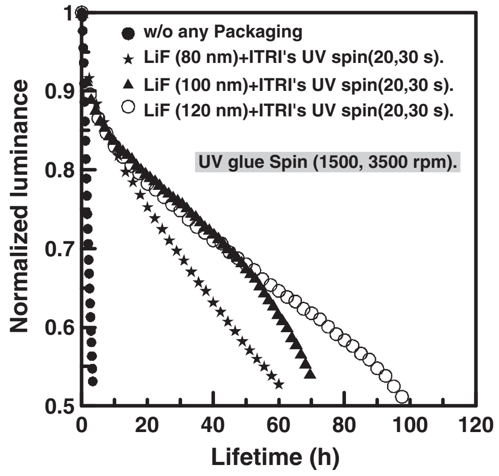

Fig. 10. Lifetime for UV glue on LiF passivation layers of different thickness as a function of spin time (20 or 30 s).

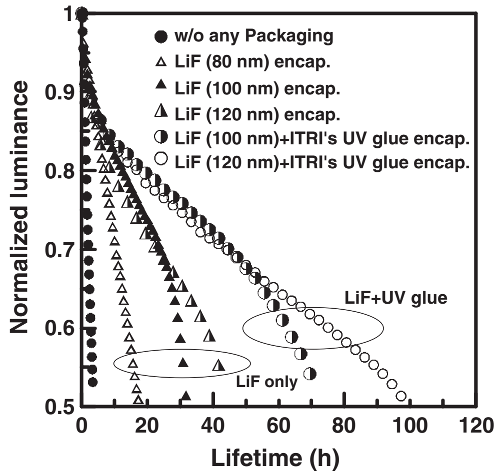

Fig. 11. Lifetime for ITRI's UV glue on LiF passivation layers of different thickness.

spin time in the second step. This result shows that a longer first step spin time causes a decline in device half-lifetime. This decline can be attributed to weak adhesion among the interfaces for a thicker film (120 nm) at a longer spin time. Obviously, the double protective layers may prolong the lifetime for an adapted structure. Figure 10 shows the normalized luminance versus operating time for different LiF film thicknesses with UV glue (20 or 30 s). It was found that the half-lifetime measured as a function of LiF film thickness for UV glue spin times of (20 or 30 s) and the half-lifetime gains with compared to a device with no packaging were 15.75, 17.5, and 25 at thicknesses of 80, 100, and 120 nm, respectively. Figure 11 shows the lifetime without any packaging, with different LiF thicknesses, and with double protective LiF/ITRI's UV glue encapsulation. Devices with LiF passivation layers have longer lifetimes than devices without any packaging, and devices with ITRI's UV glue encapsulated onto a LiF passivation layer exhibits superior lifetime. In this paper a relative comparison between LiF only, LiF plus commercialized UV glue, and LiF plus ITRI's UV glue as an encapsulation for OLEDs was studied. Because a home-made system was used for OLED fabrication and the organic materials and device structures were not optimized, the device characteristics may not be competitive with the standard ones used by other groups who may use conventional encapsulation technology. But the conclusion can be made in this report that our devices produced with ITRI's UV glue and encapsulated on 120-nm-thick LiF film showed a 25-fold increase in half-lifetime compared with devices without any packaging.

4. Conclusions

We proposed a simple method using LiF film as passivation to enhance OLED lifetime. The LiF layer significantly slows down moisture diffusion into the device but does not affect the luminescent characteristics. The newly developed UV glue (from ITRI) has a short curing time of 6 s, compared with commercial UV glue of 90 s. The proposed method produces a threefold lifetime improvement over c-UV glue. ITRI's UV glue can be applied by spin-coating onto a LiF passivation layer to reduce cost and realize a simple packaging method. Devices produced with ITRI'S UV glue encapsulated onto 120-nm-thick LiF films showed a 25-fold increase in the half-lifetime compared to devices without any packaging.

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