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Bulk-like Al/Ag bilayer film due to suppression of surface plasmon resonance for high transparent organic light emitting diodes

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ABSTRACT

We demonstrate the enhanced optical and electrical properties of an ultrathin silver (Ag) film by applying an aluminum (Al) seed layer between LiF and Ag as a transparent cathode for higher-transparency organic light-emitting diodes (OLEDs). Although the thickness ranges from 4 to 8 nm, the ultrathin Ag film is a continuous and uniform bulk-like film with an Al seed layer, which suppresses the surface plasmon absorption. Compared to an Ag-only cathode, the measured transmittance spectra were considerably increased, comparable with the theoretical calculations of a bulk Al/Ag bilayer film. The Al/Ag bilayer cathode has a transmittance of 87% at a 550 nm wavelength and a sheet resistance of 19.5 Ω/sq with a 4-nm-thick Ag layer. The transparent OLED devices that employed the Al/Ag cathode showed a transmittance of 72% at a 550 nm wavelength for an Ag thickness of 6 nm.

1. Introduction

Organic light emitting diodes (OLEDs) are superior to other types of displays for use as transparent displays [1]. Traditionally, OLEDs need to have only one transparent electrode, and it emits light in one direction, either top or bottom. However, transparent organic light emitting diodes (TOLEDs) must have transparent electrodes on both sides because TOLEDs are bidirectional devices [2]. Indium tin oxide (ITO) film is the most commonly used transparent electrode, but it has limitations. Specifically, it is difficult to apply to the top electrode (cathode) due to the damage to the organic layer [3]. One of the candidates, which is free from deposition damage, is silver (Ag) film. It has been commonly used as a transparent cathode because it can be thermally evaporated to minimize the damage. Furthermore, Ag has a high conductivity and a relatively low loss in the visible and near-infrared (NIR) ranges [4–9]. Hung et al. reported important results on the cathode structure of LiF/Al/Ag for excellent electron injection and optically low-loss top electrodes [10]. However, they used a relatively thick Ag film in the 20–50 nm range, which resulted in a relatively low transmittance of the cathode electrode. Bilayer cathode structures, such as a Ca–Ag layer [11], Ba/Ag bilayer [12], and Sm/Ag bilayer [13] have been investigated for the improvement of optical transparency and electrical conductivity. A much thinner Ag layer is necessary for higher transmittance, but it is well known that Ag grows in the Volmer-Weber (VW) mode on a dielectric layer [14,15]. Specifically, many studies have been reported that produced ultrathin, smooth Ag films using the following two techniques. First, impurity doping with Calcium (Ca) and Al in Ag has been employed to achieve smooth surfaces [16]. Al-doped Ag film has a significantly reduced RMS roughness of less than 1 nm [17,18]. The second technique involves surface energy modification using a seed layer, which influences the Ag layer wetting on substrates. Specifically, Ca, gold (Au) and Al seed layers of only 1 nm thickness significantly affected the morphology of the subsequently deposited Ag film [19,20]. Those studies demonstrated that Al is a good material both as an impurity and as a seed material for achieving a smooth Ag layer. Accordingly, the Al/Ag bilayer is a commonly used electrode in both top-emitting and bottom-emitting OLED devices. However, no study has been performed on the role of an Al seed layer for uniform bulk-like Ag films.

In this paper, we report the use of an ultrathin Al seed layer to improve the morphology of the Ag surface for higher transmittance. The transmittance spectra of a thermally evaporated thin Ag layer with and without a 1-nm-thick Al seed layer were analyzed using measurements and calculations with varied thickness of the Ag layer. In addition, we examined the surface morphology of the ultrathin Ag film using a field-emission scanning electron microscope

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(SEM). Finally, we investigated the systematic bi-directional light emission from TOLEDs as a function of the Ag cathode thickness.

2. Experimental setup

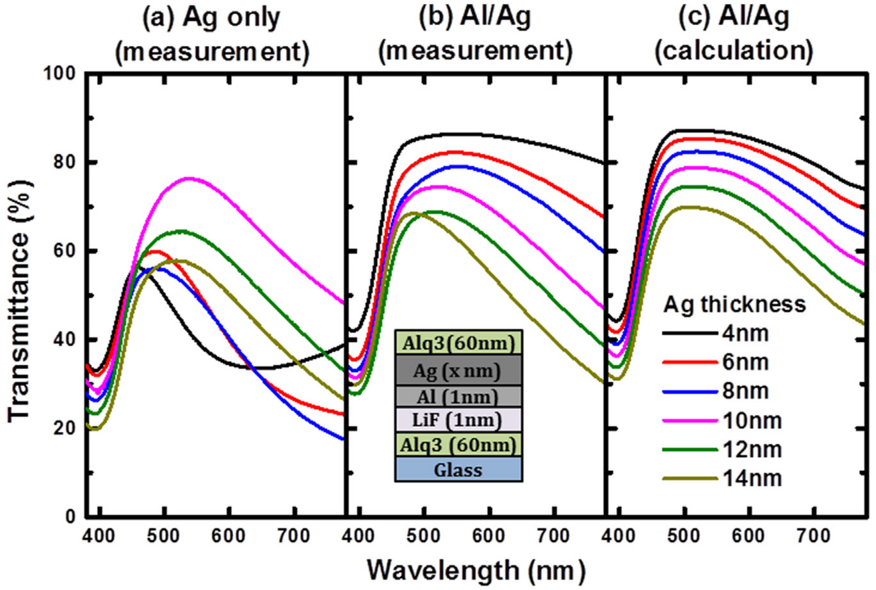
The cathode-only samples with the Alq3/LiF/Al (with or without)/Ag/Alq3 structure were prepared on glass as shown in the inset of Fig. 1 (b). The structure of the Ag-only cathode is Alq3/LiF/Ag/Alq3, and the structure of the Al/Ag bilayer cathode is Alq3/LiF/Al/Ag/Alq3. Additionally, the samples with Alq3/LiF/Al (with or without)/Ag that were used to examine the surface morphology and sheet resistance did not have a subsequent Alq3 layer deposition on the Ag layer. All layers were deposited using thermal evaporation after the samples were cleaned with deionized water, acetone and isopropyl alcohol in an ultrasonic bath. The deposition rates of the Alq3 and LiF, Al, Ag layers were 1, 0.3, 0.5, 0.5 Å/s, respectively. The rate and thickness of each layer were monitored using a quartz crystal. The tooling factors of each deposition source were estimated for a 100 nm thickness on ITO glass. The thicknesses of LiF and Al were fixed at 1 nm, and the Ag thickness was varied from 4 nm to 16 nm.

We investigated the systematic bi-directional light emission of TOLEDs with Ag thicknesses of 6 nm, 8 nm, and 12 nm. The TOLED devices consisted of a stack structure of ITO (150 nm)/poly(3,4-ethylenedioxythiophene) polystyrene sulfonate [PEDOT:PSS] (50 nm) as a hole injection layer/N,N′-Di(1-naphthyl)-N,N′-diphenyl-(1,1′-biphenyl)-4,4′-diamine [NPB] (60 nm) as a hole transport layer/tris-(8-hydroxyquinoline)aluminum [Alq3] (40 nm) as an emitting layer/lithium fluoride [LiF] (1 nm) as an electron injection layer/Al (1 nm) as a seed layer/Ag (x nm) as a cathode/Alq3 (60 nm) as a capping layer. PEDOT:PSS was spin-coated on ITO-patterned glass and heated to 120 °C for 30 min, and NPB was thermally evaporated at a rate of 1 Å/s. The fabrication methods of other layers are the same as for the cathode-only samples. The transmission spectra were measured using a UV-VIS spectrometer (T70+, PG instrument). Here, we measured the transmittance spectra of all of the samples with an air baseline condition.

The encapsulation process used a UV-cured resin and was performed using a dispenser (Shotmaster 300s, Musashi) in a N_2 atmosphere. The I-V-L characteristics of the TOLED devices were obtained using a source measure unit (Model 2400, Keithley) and a spectrophotometer (CS-2000, Konica Minolta).

3. Results and discussion

Fig. 1 shows the measured and calculated optical transmission spectra for the Ag-only and Al/Ag bilayer cathodes. At a thickness of 4–8 nm, the measured transmission of the Ag-only cathode shows a substantial difference in the shape and tendency of the spectra compared with the Al/Ag bilayer cathode. In addition, the sample with 4 nm thickness exhibits a strong optical absorption at approximately 600 nm, which is the typical feature of a localized surface plasmon resonance (LSPR) absorption [21,22]. However, the measured Al/Ag bilayer cathode transmission clearly exhibits an increased transmittance, and the shape of the spectrum is similar to



(d) Bulk refractive index, extinction coefficient

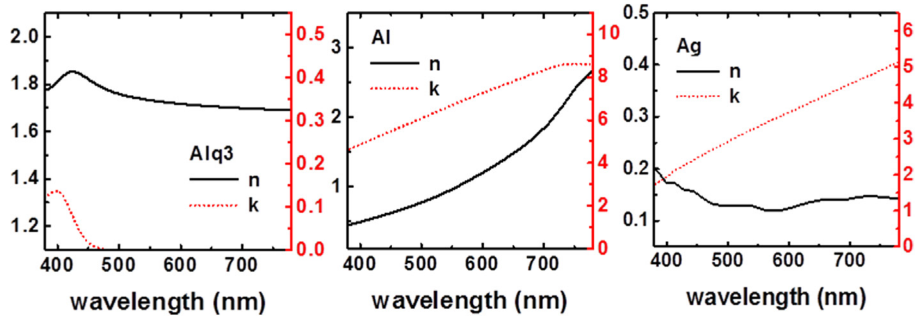


Fig. 1. Measured transmittance spectra of (a) Ag-only and (b) Al (1 nm)/Ag bilayer cathodes with different Ag thicknesses. (c) Calculated transmittance spectra of an Al (1 nm)/Ag bilayer cathode. (d) Bulk refractive index (n) and extinction coefficient (k) of Alq3, Al, and Ag, respectively. Inset in (b) is a schematic illustration of the cathode-only samples.