



## Solution-processed MoO<sub>x</sub> hole injection layer towards efficient organic light-emitting diode



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### ABSTRACT

Low cost, high throughput and large scale production techniques, such as roll-to-roll, printing and doctor blading, boost the favorite of electronic devices with all solution process. While MoO<sub>x</sub> are conventionally formed via high temperature and vacuum deposition, we develop a novel, lower-temperature, solution-processable MoO<sub>x</sub> hole injection layer (HIL) and cast successful application in organic light-emitting diodes (OLEDs). The characterization of MoO<sub>x</sub> is presented in detail using X-ray diffraction, atomic force microscopy, scanning electron microscopy, X-ray photoelectron spectroscopy, ultraviolet photoelectron spectroscopy and impedance spectroscopy measurements. The results show MoO<sub>x</sub> features amorphous phase structure, superior film morphology and exceptional electronic properties. With solution-processed MoO<sub>x</sub> as HIL, highly efficient OLED is demonstrated. The luminous efficiency has been enhanced by 56% in comparison with that of the counterpart using evaporated MoO<sub>x</sub>. The main reasons for the substantially improved performance are the tailored surface work function and appropriate hole injection capacity correspondingly result in optimizing carrier balance in OLED. Our results pave a way for advancing MoO<sub>x</sub>-based organic electronic devices with solution-processable techniques.

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### 1. Introduction

Organic light-emitting diodes (OLEDs) have been widely studied due to their potential applications to amazing full-color displays and environment-friendly lightings. Researchers' one of major targets is focused on enhancing device efficiency which predominantly depends on hole-electron balance in the emitting layer (EML). Multilayer-structure OLED incorporating electron injection layer and hole injection layer (HIL) is widely accepted for meeting such a requirement. Numerous HILs particularly the transition metal oxides such as MoO<sub>x</sub>, V<sub>2</sub>O<sub>5</sub>, NiO and WO<sub>3</sub> have recently been investigated for their favorable energy level alignments [1]. Among documented transition metal oxides, MoO<sub>x</sub> is favorite because of its non-toxicity, mild deposition temperature, wide range of thickness tolerance, good transparency and suitable work function [2–6]. The photoelectron spectroscopy studies show the work function of

MoO<sub>x</sub> is as large as 6.9 eV [7]. In-depth investigation clarifies that MoO<sub>x</sub> modification reducing hole injection barrier is ascribed to the interfacial dipole by charge transfer [7,8]. Further experiments indicate that the efficient hole injection at MoO<sub>x</sub>/organic interface is resulted from electron extraction from the highest occupied molecular orbital (HOMO) level of adjacent organic layer to the deep conduction band of MoO<sub>x</sub> [9,10]. It is also great interesting that MoO<sub>x</sub>-based device behaves extended life time and durability [8,11–13].

Rather than using cost-intensive vacuum deposition process, low-cost solution-processable techniques such as roll-to-roll, printing and doctor blading strongly boost the development of solution-processed HILs. A well acknowledged case is the application of poly(ethylene dioxythiophene):poly(styrene sulfonate) (PEDOT:PSS). However, the presence of moisture in aqueous PEDOT:PSS causes degradation [14]. The acidic nature of PEDOT:PSS also tends to etch indium-tin-oxide (ITO) electrode which reduces device durability [15,16]. Another alternative solution-processable HIL is graphene oxide (GO) [17,18]. However, GO behaves as a wide band-gap insulator with low conductivity and limited hole injection engineering [18]. Solution-processed MoO<sub>x</sub> as an ideal

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candidate attracts numerous attentions and have shown potential applications in organic photovoltaic cells [19,20], organic thin-film transistors [21] and OLEDs [1,9,13]. Reported methods of synthesizing solution-processed  $\text{MoO}_x$  are mainly focused on sol-gel/precursor solution [13,19,22–26], dissolving  $\text{MoO}_x$  powders in a water (ammonia water) or solvent [1,11,12,19,21,27,28], and deposition from  $\text{MoO}_x$  nanoparticles suspension [20,29]. However, high temperature annealing (at least 275 °C) or  $\text{O}_2$ /air plasma treatment is essentially required to form uniform layer in sol-gel method, which complicates device fabrication process [9,19,29]. It is well established that  $\text{MoO}_x$  behaves extremely low solubility and inferior dispersivity directly dissolved in water/solvent, resulting in poor coverage and rough morphology, although such a method possesses feasible accessibility [12,19,21,23,28]. It is also reported that aqueous  $\text{MoO}_x$  consists of small “islands” in the blend film [11]. To circumvent these problems, in this study we introduce a novel, lower-temperature, solution-processable  $\text{MoO}_x$  with superior dispersivity and film morphology. With the solution-processed  $\text{MoO}_x$  as HIL, highly efficient OLEDs are demonstrated.

## 2. Experiment details

### 2.1. $\text{MoO}_x$ synthesis

High-purity molybdenum (Mo) powders (10 g) were added into a beaker placed in the ice bath. Then gradually incorporating hydrogen peroxide ( $\text{H}_2\text{O}_2$ , 100 mL) and stirring for 60 min. Acetic acid (HAc, 100 mL) was added after the solution transforming from green to orange. The solution was further placed 8 h within the temperature range of 40–60 °C. A yellow dried gel was obtained by vacuum-distillation at 60 °C and then vacuum dried for 12 h. The dried gel was dissolved in ethanol with a ratio of 10 mg/mL and continuously stirred at 60 °C for 24 h till the solution exhibiting deep blue.  $\text{MoO}_x$  powders were obtained after vacuum drying at 60 °C for 12 h. Fig. 1 shows the schematic process of  $\text{MoO}_x$  powders synthesis. The  $\text{MoO}_x$  solutions for spin-coating with various concentrations were obtained by dissolving the synthesized  $\text{MoO}_x$  powders into ethanol.

### 2.2. Device fabrication

ITO coated glass was served as substrate and anode after routine chemical cleaning. The  $\text{MoO}_x$  HIL was obtained by spin-coating  $\text{MoO}_x$  ethanol solution at 3000 rpm for 30 s and subsequently heat treated at 200 °C for 30 min before loading into the multi-source deposition chamber. Organic stacks of N,N'-bis(naphthalen-1-yl)-N,N'-bis(phenyl)-benzidine (NPB, 50 nm), tris(8-hydroxy-quinolino)aluminium ( $\text{Alq}_3$ , 45 nm) and 4,7-diphenyl-1,10-phenanthroline (BPhen, 35 nm) served as hole transport layer, EML and electron transport layer, respectively, were sequentially piled by thermal deposition under a vacuum of  $4 \times 10^{-4}$  Pa. LiF (1 nm)/Al (200 nm) was served as conventional bilayer cathode. Fig. 2 shows the schematic device structure and molecular

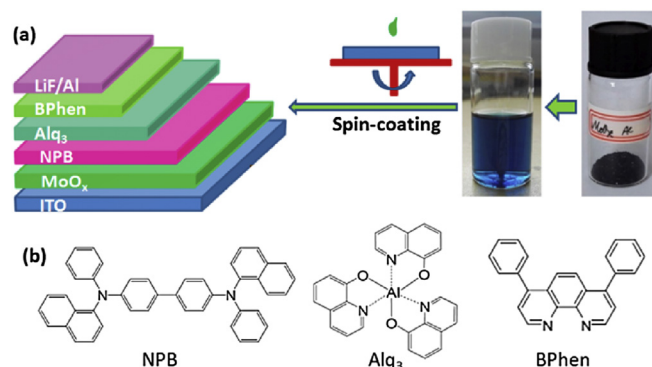


Fig. 2. (a) Device schematic structure of OLED with solution-processed  $\text{MoO}_x$  HIL. The images of  $\text{MoO}_x$  ethanol solution (5 mg/mL) and synthesized  $\text{MoO}_x$  powders are also incorporated. (b) The molecular structures of organic materials of  $\text{Alq}_3$ , NPB and BPhen.

structures of organic materials used. The layer thickness and deposition rate were monitored and controlled in situ by using an oscillating quartz thickness monitor. The typical deposition rate of organic layer was 1–2 Å/s while that of Al was 6–10 Å/s.

### 2.3. Measurement methods

The phase structure of synthesized  $\text{MoO}_x$  powders was characterized by X-ray diffraction (XRD, Bruker D8-ADVANCE).

The surface topography of solution-processed  $\text{MoO}_x$  was presented by using a field emission scanning electron microscopy (SEM, FEI Quanta 450 FEG).

The surface height image and surface potential data were mapped by atomic force microscopy (AFM, NT-MDT Solver P47) using a standard two-pass procedure [30]. A Kelvin probe force microscopy (KPFM) line was acquired in a lift mode after each topography line was acquired in tapping mode.

The X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS) were collected in an Escalab 250Xi Surface Analysis System (Thermo Scientific). A monochromatic aluminum  $K\alpha$  source (1486.6 eV) and UV lamp emitted He I (21.22 eV) radiation lines were used in XPS and UPS measurements, respectively. The signals were collected and processed by energy analyzer.

The current versus voltage ( $I$ - $V$ ) and electroluminescent characteristics of studied OLEDs were measured with a computer controlled Keithley 2400 Source Meter and an L88/OPT2000 Spectra/Luminance Meter which is calibrated with a Photo Research PR745 Spectra Scan.

The impedance spectroscopy of Hole- and Electron-only devices were measured using an Agilent 4294A Precision Impedance Analyzer in  $Z$ - $\theta$  or  $C_p$ - $D$  mode with amplitude of ac signals of 50 mV.

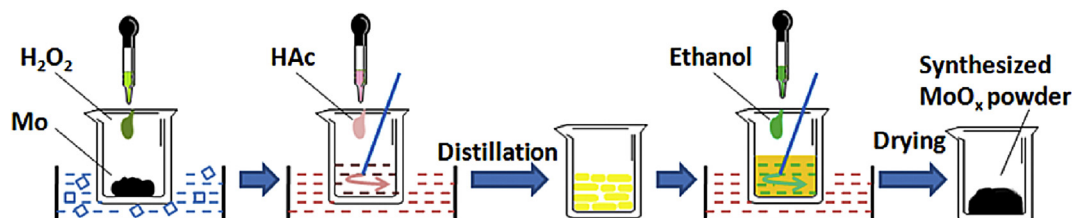


Fig. 1. Schematic process of  $\text{MoO}_x$  powders synthesis.

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