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Facilitation of transparent gas barrier using SiN_x/a-IZO lamination for organic light emitting diodes

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ABSTRACT

We investigate a full inorganic transparent barrier that alternately consist of a layer of SiN_x using plasma-enhanced chemical vapor deposition and a layer of amorphous indium-zinc-oxide (a-IZO) using plasma vapor deposition. Full inorganic thin film barriers showed a lower water vapor transmission rate (WVTR) of 5.21×10^{-4} g/m^2/day with three dyads of SiN_x/a-IZO stacking evaluated by Calcium (Ca) corrosion test. In consequence, the organic light-emitting diodes (OLEDs) encapsulated with laminated SiN_x/a-IZO barriers show longer continuous operation lifetime under environmental and driving conditions. This means that the fully inorganic encapsulated structure were quite suitable for the obstruction of water and oxygen gas permeation and their integrated OLEDs luminance decay time were improved by a considerable extent. These were attributed to the modification of barrier performance by the introduction of a-IZO film which has functions of structure decoupling, tortuous paths formation, and water or oxygen getter.

1. Introduction

Transparent gas-diffusion barriers fabricated at low temperature (lower than 100 °C) have received much attention for flexible organic electronic devices, such as organic light-emitting diodes (OLEDs), organic photovoltaics (OPV), and printed circuits [1–3]. Especially, OLEDs are allegedly considered to be highly sensitive to environment working conditions [4]. Hence, OLEDs urgently require encapsulated materials to segregate themselves from water or oxygen that with oxidizing capacity [5]. On this occasion, it is generally accepted that a water vapor transmission rate (WVTR) of less than 1×10^{-6} g/m^2/day and a oxygen transmission rate (OTR) of less than 10^{-3} cc/m^2/day are essential for the realization of OLEDs shelf-life time up to 10,000 h. Nowadays, the requirements can be met through the state-of-art glass to glass encapsulation technology [6,7]. But as for the encapsulation of flexible OLEDs, the thin film encapsulation (TFE) being endowed with immense prospects has been considered as an alternative scheme.

Researches about transparent gas-diffusion barriers that have been reported targeting at the lifetime enhancement for the OLEDs used a multilayered stack of inorganic layers separated by organic layers (known under the trade name of Barix™), which has a superior performance in WVTR approaching the target value of the 1×10^{-6} g/m^2/day [8,9]. As for the Barix™ structure, the inorganic layers playing as a major humidity and oxygen barrier are generally deposited by plasma-enhanced chemical vapor deposition (PECVD) or physical vapor deposition (PVD), while the introduction of organic film acts as an effective decoupling layer to circumvent the defect or pinhole in the inorganic layer to improve barrier performance [10]. Additionally, by applying multilayered barriers with appropriate materials and structures, a tortuous path for diffusion species can be created, which would lead to the increase of diffusion length, thereby lowering the overall permeability of the films [11]. However, organic thin film is mainly formed by means of printing or vacuum deposition and then cross-linked by some of the ultraviolet or thermal curing process. This implies that the additional methods are needed to weight, and the potential damages to the OLED devices would be introduced. Besides, organic film is apt to face many issues like reliability in extreme environments, particle contamination, and transparency. Most importantly, the complicated process reduces the production efficiency and yield in mass production. As a whole, those issues should be carefully considered during the design of inorganic/organic multilayer for encapsulation.

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Alternatively, an encapsulation solution that all composed of inorganic films has been proposed [12]. The atomic layer deposition (ALD) is inherently well-known for its thin, highly uniform, conformal coverage at monolayer thickness scale, and free pinhole with featureless microstructures, that promising the barrier performance of TFE [13–15]. So far, Duan's groups have reported of a single layer of ZrO_2 or Al_2O_3 deposited by ALD as the barrier materials, which shows excellent barrier properties with WVTR as 6.0×10^{-4} g/m^2/day and 8.7×10^{-6} g/m^2/day respectively [16,17]. Typically, Meyer's group has achieved a WVTR below < 10^{-6} g/m^2/day by adopting a nanolaminated structure that consisting of alternative layers of Al_2O_3 and ZrO_2 by ALD grown [18,19]. Singh's group has employed high-quality laminated structure of Al_2O_3 and TiO_2 as gas-diffusion barrier, and simultaneously they also optimize the technical process of ALD to reduce the deposition influence on OLEDs' performance [20]. But nowadays the development of TFE by traditional ALD technology is constrained by the low deposition rate and the realization of low temperature process, demanding with more investigation in the pipeline [21,22]. Although the emergence of some novel technologies, for instance, spatial ALD and plasma-assisted ALD [23,24], have been effectively solve these issues of low deposition rate and low temperature process, the ALD technology is still faced with the problem of industrialized production, such as the difficulty of masking pattern, by which the processing of the encapsulation is hard to be compatible with industrialized process of large-size OLEDs. Thus, the development and integration of high-barrier encapsulation films in the actual production of flexible OLEDs remains a challenging endeavor.

In this study, we focus on designs of multilayer structure with fully inorganic film as TFE technology, which adopts a system of SiN_x and amorphous oxide semiconductors (AOSs), deposited by PECVD and PVD respectively, to fabricate the laminated inorganic film. The material of SiN_x fabricated by PECVD with a low temperature process as a kind of industrial-scale barrier exhibits a good impermeability and high productivity. However, the main drawback associated with single material of SiN_x as encapsulated structure is the inherent defect or pinhole in the PECVD-deposited film, which causes a fast degradation rate of gas permeation characteristic. Therefore, preparation of decoupling layer arises at the right moment to block or fill the defect sites. For this work, an ultrathin amorphous indium-zinc-oxide (a-IZO) film deposited by PVD systems at room temperature is firstly adopted to be functioned as a decoupling layer with the multilayer structure as SiN_x/a-IZO stacking. The IZO film, as a kind of transparent conductive oxide (TCO) materials, can be fabricated by direct current sputtering with mature target technology, possessing the advantages in use of rapid growth and large-area deposition, and suggest to popularize use in Gen10 industrial process. But more importantly, IZO film has an excellent amorphous characteristic in large area within the substrate, and this property cannot be matched by other TCO materials, such as indium-tin-oxide (ITO), aluminum-zinc-oxide (AZO), and gallium-zinc-oxide (GZO), which tend to be crystallized. In addition, as compared to organic decoupling layer with a thickness of a few microns, the ultrathin a-IZO inorganic film has obvious competitiveness to fulfill lower WVTR properties as long as the thickness of decoupling layer determines the gas diffusion paths [25]. Based on candidate materials evaluation, we successfully fabricated a thinner multilayer structure of SiN_x/a-IZO film with lower WVTR as encapsulated layers for OLEDs and achieved an ideal lifetime for the real application in flat panel displays.

2. Experimental

2.1. Material and deposition method

The SiN_x layer with a thickness of 200 nm were firstly deposited using the PECVD method at 80 °C, and the growth rate was about 1.2 nm/s. The chamber pressure and deposition power was 100 Pa and 150 W, respectively. Then, the amorphous indium zinc oxide (In:Zn = 1:1, a-IZO) layer with a thickness of 30 nm was deposited at room temperature by DC magnetron sputtering in PVD system [26]. Therefore, a dyad of SiN_x/a-IZO laminated structure was fabricated. The a-IZO film with 30 nm thick, as a critical thickness in PVD manufacturing process, is to guarantee the uniformity on the large area substrate in the mass production. In this study, we presented gas permeability and OLEDs lifetime degradation with respect to the encapsulated barrier films. As for comparison, four types of encapsulated structure were fabricated to evaluate the barrier characteristic. The reference sample without encapsulation was named as Device A. The encapsulated structure with two layers of SiN_x was named as Device B. For multilayer structure, the Device C was fabricated with a-IZO layer inserting between two SiN_x layers, while Device D was completed with three dyads of SiN_x/a-IZO as repeated unit. In order to confirm the quality of encapsulated film, we also fabricate Device E with glass encapsulation as comparison. A schematic diagram of prepared TFE structures was shown in Fig. 1.

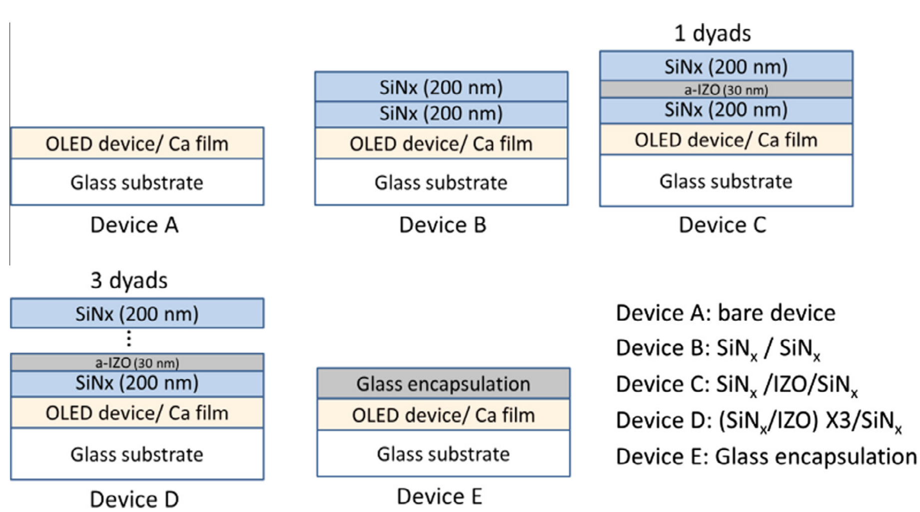

Fig. 1. Schematic diagram of the prepared TFE with variable structures.