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Diamine-cored tetrastilbene compounds as solution-processable hole transport materials for stable organic light emitting diodes

Min Jy Cho,^{‡,a} Kyu Min Sim,^{‡,b} Sa-Rang Bae,^a Hye Ok Choi,^a Soo Young Kim,^{*,a} Dae Sung Chung,^{*,b} Kwangyong Park^{*,a}

^a School of Chemical Engineering and Materials Science, Integrative research center for two-dimensional functional materials, Chung-Ang University, 84 Heukseok-ro, Dongjak-gu, Seoul, 06974, Republic of Korea

^b Department of Energy Systems Engineering, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 42988, Republic of Korea

ABSTRACT: A series of diamine-cored tetrastilbene (DTS) derivatives bearing various aliphatic and aromatic substituents was designed and synthesized for use as solution-processed hole transport layers (HTLs) in organic light emitting diodes (OLEDs). The chemical structures of the DTS derivatives were strategically designed to increase solubility in organic solvents as well as to avoid self-crystallization, and thus ensure a stable morphology under Joule heating while maintaining efficient hole transport capabilities. The five DTS derivatives, composed of different conjugation structures, yielded reasonably good hole transport behavior with a marginal charge carrier mobility of $\sim 10^{-5}$ cm²V⁻¹s⁻¹, which is similar to that of vacuum-deposited *N,N'*-bis(1-naphthyl)-*N,N'*-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB). Due to the high glass transition temperatures of the DTS derivatives, this satisfactory charge transport behavior and smooth surface morphology were conserved up to 180 °C. Green OLEDs were prepared using tris-(8-hydroxyquinoline) aluminum (Alq₃):C545T as the emission layer, and the OLED performances of the solution-processed DTS HTLs and the vacuum-deposited NPB HTL were compared. A high luminance efficiency of 11.5 cd A⁻¹ was obtained for one solution-processed DTS HTL, which exceeds that of the NPB HTL (10.01 cd A⁻¹). Furthermore, the DTS HTLs enabled a stable OLED operation, with double the accelerated half-life of the NPB-based device.

Key words: Diamine-cored tetrastilbene, hole transport layer, organic light emitting diodes, solution process

1. Introduction

Due to the rapid development of materials synthesis and lighting technologies, organic light emitting diodes (OLEDs) are considered as promising candidates for flat panel displays and solid-state lighting applications.[1-3] Recently, highly efficient emitters and out-coupling techniques yielded OLED current efficiencies exceeding 200 cd A⁻¹, thus meeting the requirements for commercial application.[4,5] However, these achievements are

based on a thermal evaporation method, which has drawbacks in terms of manufacturing costs and large area production.[6,7] Consequently, studies have focused on solution-based processes that are expected to allow low-cost plastic electronics by large area production and high-throughput methods.[8] However, in the case of solution-processed OLEDs, the efficiency is still very low compared to that of vacuum-processed OLEDs.[9-12]

Due to the structural complexity of multilayer (injecting, transporting, and emitting layers along with their interfacial layers) stacked OLEDs, determination of a decisive factor to enhance the efficiency of solution-processed OLEDs was desirable. Among the known layers and interfaces, the hole injection layer (HIL) and hole transport layer (HTL) lower the energy barrier between the anode and the emitting layer to allow efficient charge delivery and lighting.[13] In addition to favorable energy barrier characteristics, the HIL/HTL must also exhibit anode planarization, high transparency, sufficient conductivity, good electron blocking ability, and good exciton blocking ability. Thus, for developing highly efficient solution-processed OLEDs, optimization of the HIL/HTL is a priority. For the HIL, poly(3,4-ethylenedioxythiophene:polystyrene sulfonate) (PEDOT:PSS), metal oxides, and metal sulfides are the preferred materials due to their superior electrical/optical characteristics.[14-19] Among them, the majority of recent reports on solution-processed OLEDs have employed PEDOT:PSS as the HIL and reported their reasonably good performance. However, in the case of the solution processed HTL, there are very limited numbers of materials that have been known to allow a sufficient decrease of the energy barrier and satisfactory electrical and thermal stability.[20-22]

Ideally, in addition to the above-mentioned characteristics, HTLs must possess the following properties: i) The HTL must protect the emission layer (EML) from the acidity of PEDOT:PSS; ii) It must be deposited from a solvent orthogonal to that used for the HIL; and iii) It should be thermally stable to ensure the operational sta-

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