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Structure and Morphology of Organic LED Thin Films

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Organic light-emitting devices (OLED) are a fast growing research area because of their potential applications in low-power, flexible, cost-competitive and flat-panel displays. We report, for the first time, a TEM study of electro- (EL) and photo- (PL) luminescent composite films spin-cast from a combination of two dissimilar solvents. Fluorescent fluorene polymer poly(9,9-dioctylfluorenyl-2,7-diyl) (PFO) is the matrix and fac tris(2-phenylpyridine) iridium Ir(ppy)$_3$, a phosphorescent iridium (III) complex, is chosen as an emitting dopant. A high boiling-point (180°C) and more viscous orthodichloro-benzene (ODCB), and a low boiling-point (62°C) and less viscous chloroform ($CHCl_3$) are exploited as two dissimilar solvents. The liquid composite solutions are prepared using two solvent bases: a pure $CHCl_3$ (P) and a combined $CHCl_3$-2% ODCB (C). Four different film compositions were prepared: (a) P-based PFO; (b) C-based PFO; (c) C-based Ir(ppy)$_3$(0.1%)/PFO composite; and (d) C-based Ir(ppy)$_3$(4%)/PFO composite. Each film was spin-cast onto a glass substrate to ~80 nm thickness. TEM specimens were prepared by floating the thin films off the glass substrates and placing them onto holey carbon supports, and were examined in a Hitachi HF-2000 field emission TEM.

TEM images and representative diffraction patterns of the four films are shown in Fig. 1. An amorphous film forms when pure $CHCl_3$ is used as the solvent (Fig. 1a). When the combined $CHCl_3$-2% ODCB solvent is used, all films contain a crystalline so-called "β" phase, revealed by the faint ring in the insets of Figs. 1b-d. The pure PFO and composite films have slightly different crystalline structure: the interplanar spacing is 3.61±0.03Å in the pure PFO film, whereas that with 4 wt.% Ir(ppy)$_3$ has a larger spacing, 4.40±0.03Å. An interplanar spacing of 4.2 Å has previously been reported for the β phase formed in the pure PFO film prepared from an equilibrium thermodynamic process based on melt state [1] or friction transfer [2]. Therefore, the interplanar spacing is clearly a function of both material composition and film-formation conditions, similar to that of a highly strained structure. Diffraction from the Ir(ppy)$_3$ aggregation (Fig. 1d) indicates a single crystal with an interplanar spacing of 8.19±0.07Å.

The spin-cast films of the Ir(ppy)$_3$/PFO composite show significantly different exciton emission and energy transfer rates when prepared with the pure and combined solvents. TEM studies reveal that the combined solvents result in the coexistence of crystalline β and amorphous phases in the PFO. PL/EL measurements indicate that the coexisting crystalline/amorphous structures function as quantum wells and thus significantly influence both exciton emission and energy transfer characteristics in the Ir(ppy)$_3$/PFO composite films. As a result, such solvent-induced quantum-well like structures with the resultant morphology-dependent excitonic dynamic behavior presents a new pathway to further enhance EL/PL from the dye/polymer composites and EL from conjugated polymers based on rationally selected organic solvents [3].

[1] S.H. Chen, H.L. Chou, and A.C. Su, *Macromol*. 37 (2004) 6833.
[2] M. Misaki et al., *Macromol*. 37 (2004) 6926.
[3] Research at the Oak Ridge National Laboratory (ORNL) High Temperature Materials Laboratory (HTML) was sponsored by the Assistant Secretary for Energy-Efficiency and Renewable Energy, Office of FreedomCAR and Vehicle Technologies, U.S. Department of Energy under contract DE-AC05-00OR22725 with UT-Battelle, LLC.

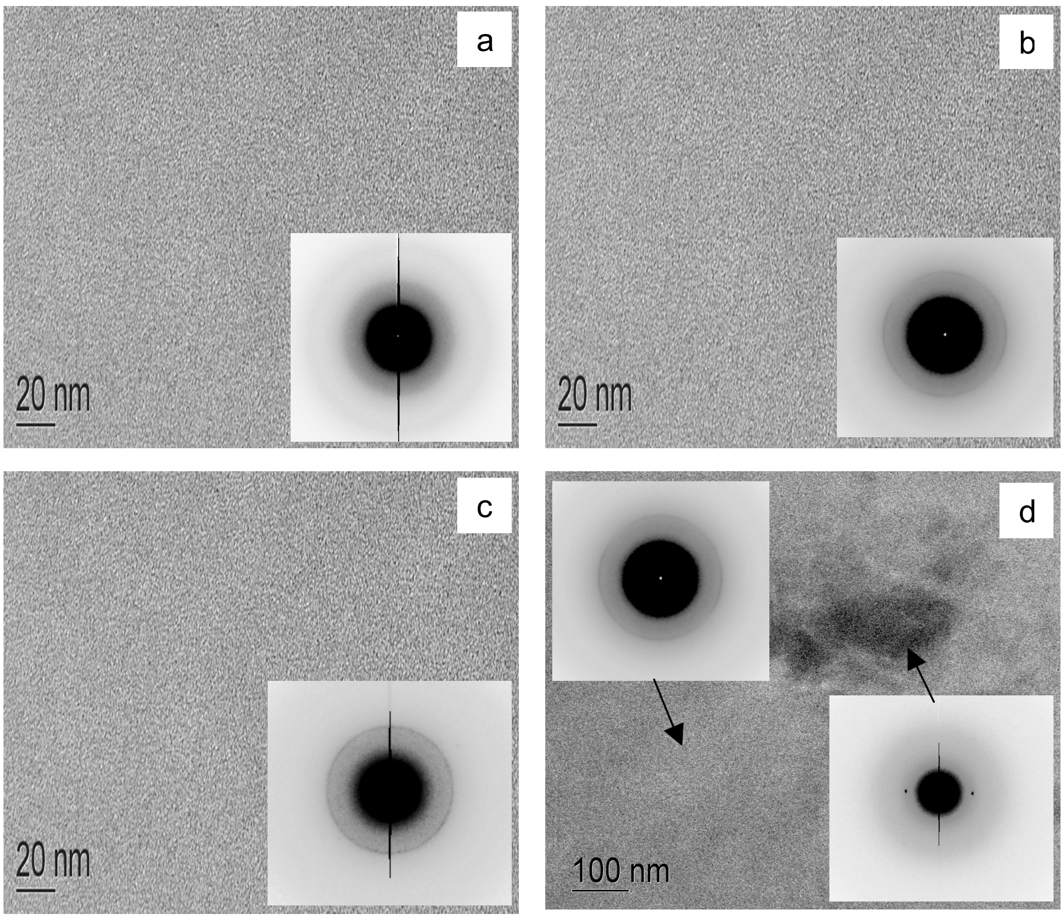

Fig. 1 – TEM images and representative electron diffraction patterns of four spin-cast films: (a) P-based PFO; (b) C-based PFO; (c) C-based $Ir(ppy)_3$(0.1%)/PFO composite; and (d) C-based $Ir(ppy)_3$(4%)/PFO composite. (P = $CHCl_3$; C = $CHCl_3$-2%ODCB.)