

Ambipolar N-doped PAHs. A unique strategy for the efficient TADF OLED emitters.

M. Lindner

Institute of Organic Chemistry, Polish Academy of Sciences, Kasprzaka 44/52, 01-224 Warsaw, Poland;

Although bowl-shaped N-pyrrolic polycyclic aromatic hydrocarbons (PAHs) can achieve excellent electron-donating ability, their application for optoelectronics is hampered by typically low photoluminescence quantum yields (PLQYs). [1] To address this issue, we report a efficient synthetic strategy towards nonplanar N-PAHs with intrinsic D-A electronic structures [2] which are, for the first time, applied as yellow to orange TADF/RTP OLED emitters. The entirely fused, and curved system leads remarkable PLQY (up to 86%) OLEDs fabricated with the developed N-PAHs as TADF/RTP emitters were unprecedented for this class of compounds showing EQE as high as 12% along with satisfactory operational stability. We next performed a facile post-functionalization of our core structure introducing a set of auxiliary donors at the *meta* position of protruding phenyl ring of quinoxaline.[3] This modification let us to obtain a set of highly emissive dyes (PLQY up to 96%) and successfully fabricate a series of TADF OLEDs devices with a yellow to orange-red electroluminescent emission color. Their analyses revealed significant EQE up to 21.9% and pronounced luminance (up to 31 000 cd/m²) which is the highest recognized for N-doped PAHs investigated in OLED domain.

In addition, we are focusing our efforts on the synthesis of curved nanographenes intended for optoelectronic applications. These structures contain 'dopants' (nitrogen atoms) and structural 'defects' (5, 6, and 7-membered rings), affecting the curvature of the entire system, enabling the improvement of all key photophysical parameters necessary for achieving a highly efficient emitter within one structural motif. The foundation of our work lies in a naphthalene monoimide (NMI) framework containing a 6-membered defect, specifically a six-ring ring generated as a result of an intermolecular tandem reaction between dibromo-NMI and carbazole, catalyzed by a palladium complex. Combining this system with its structural analogue which lacks a fully conjugated structure (no additional C-C bond), allowed us to pioneer a systematic strategy. This strategy combines both Förster resonance energy transfer (FRET) and the triplet-to-singlet electronic transition (TTS) as complementary mechanisms for efficient exciton transfer, marking a significant advancement for the fourth generation of high-efficiency (HF) OLED emitters. Through advanced photophysical research and characterization of OLED device demonstrators, we have demonstrated that this designed emitter system, with properly adjusted excited state energies, is the most effective emitter of yellow light to date [4]. This emitter boasts a photoluminescence quantum yield (PLQY) of 100% and achieves an external quantum efficiency (EQE) of 27%, showcasing a narrow emission band (40 nm), which holds particular significance for future applications.

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[2] J. Wagner, P. Zimmermann Crocomo, M.A. Kochman, A. Kubas, P. Data, M. Lindner, *Angew. Chem. Int Ed.* **2022**, *61*, e202202232. (**HOT PAPER**) Highlighted in Synfacts: 10.1055/s-0041-1738208, 18, 07, (0742), (2022)

[3] J. Wagner, D. Kumar, M.A. Kochman, T. Gryber, M. Grzelak, Kubas, P. Data, M. Lindner, *ACS Appl Mat. Interf.* **2023**, *15*, 31, 37728–37740

[4] K. Bartkowski, P. Zimmermann Crocomo, M.A. Kochman, D. Kumar, A. Kubas, P. Data, M. Lindner *Chem. Sci.* **2022**, *13*, 10119-10128