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Abstract in English

A New Class of Cyclazine-Based Curved Polycyclic Aromatic Hydrocarbons for Applications in Optoelectronic Devices

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Despite rapid development, modern organic electronics still faces fundamental structural limitations associated with classical aromatic systems. Traditional, planar polycyclic aromatic hydrocarbons (PAHs) exhibit a strong propensity for π-π stacking, which leads to a significant decrease in photoluminescence efficiency in the solid state. This doctoral dissertation presents a breakthrough approach to this problem, based on the precise molecular design and synthesis of a novel class of curved, bowl-shaped, N-doped PAH systems. Utilizing a unique 5-7-5 ring system, a N-doped molecular platform was established which adopts a concave core geometry, effectively suppressing π-stacking and paving the way for high efficiencies in devices.

A key element of the molecular engineering strategy involved utilizing a seven-membered ring as an antiaromatic spacer to decouple the HOMO and LUMO orbitals. This effectively minimizes the singlet-triplet energy gap (ΔE_{ST}), thereby promoting the thermally activated delayed fluorescence (TADF) mechanism. I developed a fully scalable, relatively straightforward synthetic route, yielding 14 chemically and thermally stable donor-acceptor (D-A) emitters, electronically diversified by varying the acceptor moiety.

Photophysical investigations revealed the high potential of the developed structures, enabling a tunable TADF/RTP (room-temperature phosphorescence) emission mechanism within appropriate host matrices. Prototype OLED devices achieved a maximum external quantum efficiency (EQE) of 12%, which represented one of the highest values reported in the literature for curved N-PAHs. By transitioning from D-A systems to donor-augmented D-A-D architectures, a substantial increase in EQE to 21.9% was achieved. Concurrently, record-high luminance values (up to 30,100 cd/m^2) were recorded, accompanied by a bathochromic shift in emission. In the D-A-D emitters, an O-alkyl chain was implemented to improve solubility; owing to its electron-donating character, it flattened the polycyclic system without disrupting the TADF emission.

The developed cyclizine-based core was further applied in perovskite solar cells (PSCs). Through peripheral functionalization, a series of T-shaped, dopant-free hole-transporting materials (HTMs) was synthesized. These novel N-PAH systems enabled a power conversion efficiency (PCE) of 20.39%, outperforming the commonly used reference material, featuring the desired hydrophobicity for perovskite protection.

These projects aimed to carefully analyze structural modifications and their impact on photophysical properties, and the results obtained can serve as a comprehensive guide for designing modern, versatile organic optoelectronic materials.