Stretchable OLEDs enabled by Mxene, exciplex and transfer printing

Xiangyu Shen,[1] Shaoyu Lin,[1] Qin Xue,[2] Igor F. Perepichka,[3,4,*] Yanqin Miao,[5,*] and Guohua Xie[1,6,7,*]

[1]Institute of Flexible Electronics (IFE, Future Technologies), Future Display Institute of Xiamen, Tan Kah Kee Innovation Laboratory, Xiamen University, Xiamen 361102, China
[2]Department of Physical Science and Technology, Central China Normal University, Wuhan 430079, China
[3]Department of Physical Chemistry and Technology of Polymers, Silesian University of Technology, Gliwice 44-100, Poland
[4]Centre for Organic and Nanohybrid Electronics (CONE), Silesian University of Technology, Gliwice 44-100, Poland
[5]College of Physics and Optoelectronics Engineering, Taiyuan University of Technology, Taiyuan 030600, China
[6]College of Physics and Information Engineering, Quanzhou Normal University, Quanzhou 362000, China
[7]State Key Laboratory of Organic Electronics & Information Displays & Institute of Advanced Materials, Nanjing University of Posts & Telecommunications, Nanjing 210023, China
*Correspondence: i.perepichka@bangor.ac.uk (I.F.P.); miaoyanqin@tyut.edu.cn (Y.M.); ifeghxie@xmu.edu.cn (G.X.)

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Organic light-emitting diodes (OLED)s, as a next-generation display technology, satisfy the demands of high brightness, light weight, flexibility, and even stretchability. Stretchable OLEDs overcome the limited deformability of traditional devices, realizing multi-mode deformations, such as bending, stretching, and torsion. They cater to the application requirements of various scenarios, which effectively broadens the applications of OLEDs beyond display, e.g., in electronic skin, smart textiles, biomedicine, and wearable devices. Fully stretchable OLEDs struggle with the inherent trade-off between high electroluminescence performance and excellent mechanical stretchability.

TRANSFER PRINTING FOR STRETCHABLE DEVICES

OLEDs maintain their dominant role in flat-panel displays. Nevertheless, the transition from conventional rigid architectures to stretchable OLEDs presents considerable challenges, notably in achieving a balance between stretchability and electrical conductivity.[1] Current fabrication strategies for stretchable OLEDs primarily follow two avenues: (1) the first one involves developing highly efficient and intrinsically stretchable emissive layers between the transport layers; (2) the second one is devoted to employing the stable electrode contacts and the interfaces between two adjacent layers. While polymers are frequently introduced to mitigate mechanical failure under strain, they are generally more insulating than small molecules used in the state-of-the-art OLEDs, thereby compromising device efficiency. Furthermore, polymers are limited by issues including interlayer miscibility, which hinders the fabrication of high-performance multilayer devices although they are compatible with spin coating and inkjet printing. As recently demonstrated, transfer printing offers a promising alternative fabrication approach for flexible devices by manipulating the interfacial work of adhesion.[2-3] This technique enables dry transfer of thin films, circumventing interlayer mixing issues and thus significantly reduces the complexity of molecular and device

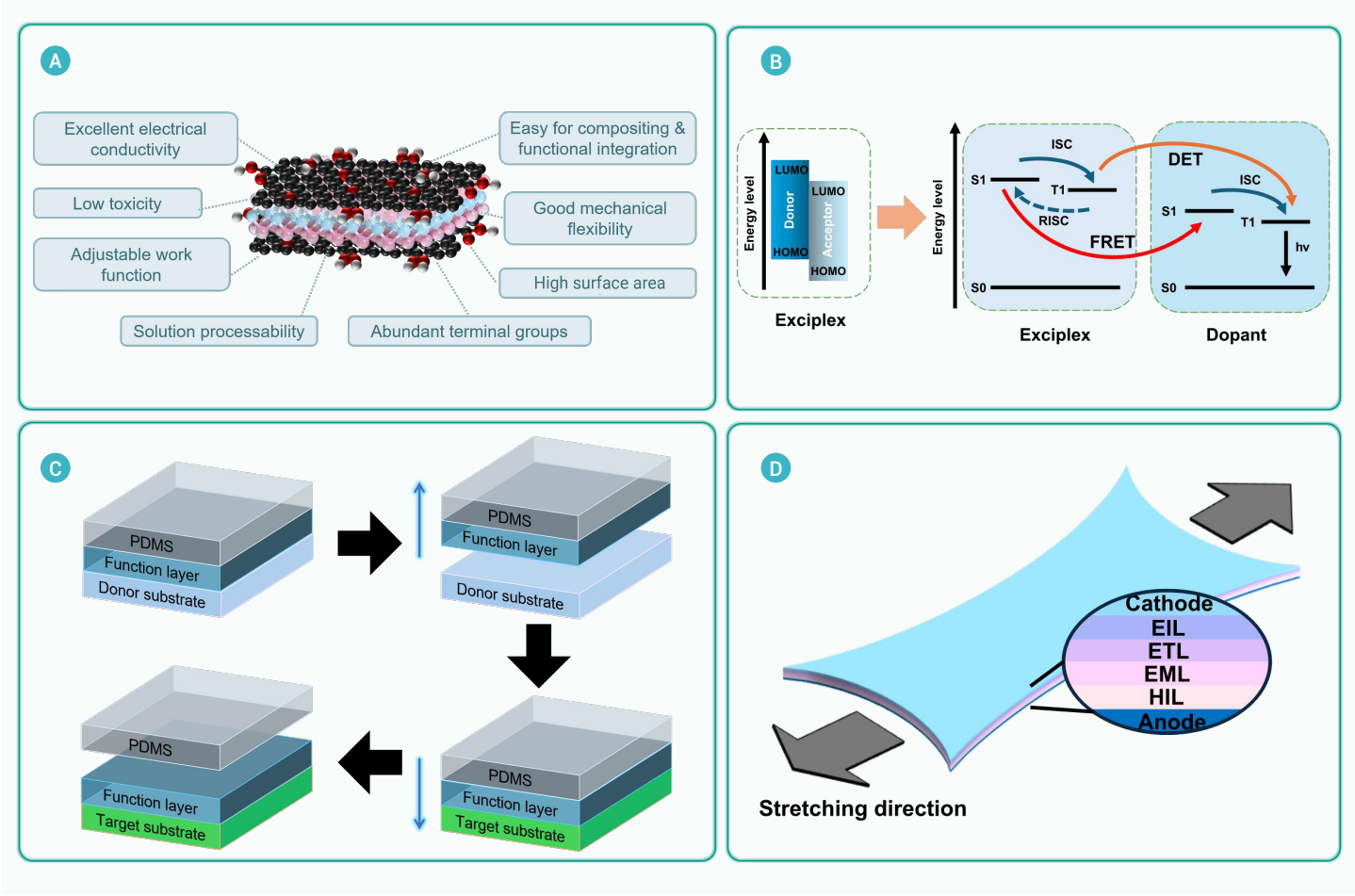

Figure 1. Schematic illustration of the fully stretchable OLED (A) Schematic diagram of MXene structure. (B) Energy transfer process of the exciplex host with phosphorescent complex. (C) Transfer printing process. (D) Schematic diagram of stretchable OLED.

designs.

MXENE FOR OPTOELECTRONICS

The typical thickness of organic functional layer in OLEDs is usually in the order of 10~200 nm. Therefore, they required the electrode with high conductivity and high transparency since rough surface can easily cause local leakage and even short circuit. In rigid OLEDs, indium tin oxide (ITO) is widely used as an anode due to its excellent electrical conductivity, high light transmittance and smooth surface morphology. However, traditional ITO film is difficult to meet the bending and stretching requirements for stretchable OLEDs, due to its intrinsic brittleness. Although AgNWs exhibit excellent electrical conductivity and high light transmittance, their excessively high surface roughness significantly impairs device performance. PEDOT:PSS ensures good film uniformity. Unfortunately, its intrinsic strong acidity drastically reduces device lifetime. Other metal-based electrodes such as ultrathin metal films and serpentine electrodes fail to balance high stretchability and high transmittance, and they show poor compatibility with solution processing.

MXenes (Figure 1A), a class of newly developed 2D layered transition metal carbide materials, have great potential to address above-mentioned issues. The chemical formula of MXenes is $M_{n+1}X_nT_x$ (n = 1 ~ 3), where M is an early transition metal, X is C or N (carbide, nitride, or carbonitride), and T represents the surface terminal functional groups of -O, -F, and –OH, respectively. They typically exhibit high electrical conductivity, good film-forming property, excellent optical transparency, tunable work function, and high chemical stability. These features make MXenes attractive for various optoelectronic devices.[4] In this work, MXene-based electrodes were surface-modified with perfluorosulfonic acid (PFSA) to raise the work function from 4.71 eV to 5.31 eV, enabled by the high ionization potential of fluorine atoms, to meet the anode work function requirement. For cathode applications, polyethyleneimine (PEI) and conjugated polyelectrolyte (CPE) were employed for surface modification, where the amino groups of the modifiers effectively reduced the work function. The modified MXene electrodes maintained excellent electrical conductivity and optical transmittance, showing promising application prospects in the optoelectronic field.

STATE-OF-THE-ART OF STRETCHABLE OLEDS

Recently, Lee and Gogotsi's groups brilliantly adopted MXene/AgNWs composite electrode for high-performance stretchable OLEDs with an exciplex host fabricated by transfer printing.[5] They found that MXene effectively welded the nanowire-nanowire junctions, thus enhancing long-range connection between AgNWs/MXene percolation networks. The presence of hydrogen-bonding interactions between AgNW ligands and MXene strengthened the mechanical and electrical stability of the electrode under stretching. The resulting MXene-contact stretchable electrodes (MCSE) showed a minimal resistance change under 500 cycles of 40% cyclic strain. Interestingly, after simple surface modifications, the work function of the MCSEs was tunable between 3.79 and 5.71 eV (starting from the initial 4.73 eV), which is suitable for the ideal cathode and anode of OLEDs, respectively.

TADF (thermally activated delayed fluorescence) materials possess the characteristic of reverse intersystem crossing, which enables efficient utilization of triplet excitons. When subjected to external tensile stress, the molecules in the thin films may undergo local changes in aggregation behavior and variations in intermolecular distance, which may further weaken the spin-orbit coupling and lead to a decrease in the reverse intersystem crossing rate. As for stretchable devices, the exciton utilization efficiency of TADF materials decreases with increasing tensile strain, which impairs the electroluminescence performance. In contrast, phosphorescent complexes have strong spin-orbit coupling due to the presence of heavy atoms, rendering their luminescent properties less susceptible to the tensile strain of the thin film.

Ingeniously, Lee's group constructed an emissive layer containing the exciplex cohost and phosphorescent dopant bis(2-phenylpyridine)(acetylacetonate)iridium(III) ($Ir(ppy)_2acac$) to improve stretchability by incorporating a thermoplastic polyurethane (PU) elastomer matrix, enabling the layer to withstand stretching of over 200% stretchability. Although the addition of insulating PU significantly increased the stretchability of the emissive layer, it separated the host and the dopant, due to the insulating property of PU. In addition, Dexter energy transfer between the host and the dopant, which relies on short-range electron exchange, is inhibited. To efficiently utilize the triplet excitons, 4,4 ',4 " -tris(*N*-carbazolyl)triphenylamine (TCTA) and 1,3,5-tris(1-phenyl-1*H*-benzimidazol-2-yl)benzene (TPBi) were employed to form the exciplex cohost. Singlet excitons of the cohost could be efficiently transferred to $Ir(ppy)_2acac$ through Förster resonance energy transfer (Figure 1B). Therefore, the exciton utilization efficiency was boosted from 12.6% (single host) to 57% (exciplex cohost). For fully stretchable devices, the emissive layer was initially grown on the octadecyltrimethoxysilane (OTMS) treated silicon wafer with the reduced the adhesion and subsequently transfer printed by polydimethylsiloxane (PDMS) stamp onto the target substrate with the MXene/AgNWs composite electrode (Figures 1C-D). The stretchable device exhibited a remarkable EQE of 17%, which still retained at approximately 80% even under a 60% tensile strain. After 100 cycles of 20% cyclic strain, the efficiency still retained 83% of the initial value. The insulating elastomer matrix will also reduce the charge mobility of the emissive layer, leading to unbalanced charge transport and further increasing the non-radiative loss of excitons. Meanwhile, the surface energy mismatch between the elastomer and conjugated small molecules may induce phase separation and aggregation-caused quenching (ACQ), which further lowers the triplet exciton capture efficiency. Therefore, achieving efficient utilization of triplet excitons within the insulating polymer elastomer matrix is a key bottleneck for highly efficient stretchable OLEDs.

CONCLUSION

Fully stretchable devices based on the exciplex-assisted phosphorescent emissive layer and MXene-contact stretchable electrodes have achieved breakthroughs in both electroluminescence performance and mechanical stretchability. However, such devices still face challenges including material compatibility, long-term stability, and scalable mass production. In the future, research on stretchable devices should further focus on the development of new material systems compatible with high stretchability, the optimization of device structures, and the expansion of application scenarios. Currently, some preliminary attempts to create full-color OLEDs via transfer printing technology has been demonstrated. This technology can effectively address interlayer miscibility issues in solvent processing. To achieve full-color stretchable OLEDs, the adoption of transfer printing technology would be essential. With technological breakthroughs and advances, fully stretchable OLEDs will be more affordable and thus unlock diverse practical application scenarios.

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DECLARATION OF INTERESTS

The authors declare no competing interests.