

变色有机电致发光器件的研究进展

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摘要: 碳基有机半导体光电器件易于从分子层级进行调控设计,近年来有机电致发光器件已逐步实现产业化。变色有机电致发光器件是指在单个或单种器件内实现多个色彩或色温变化的发光器件,使得单个像素点结构实现色彩或色温可调的功能,从而简化单个显示单元,是一种高分辨率的显示技术。依据变色原理的不同,变色有机电致发光器件分为器件结构变色、单发光层材料变色、外加驱动方式3个类别。对该领域最新研究进展、性能优势和发展现状进行了综述,进而探讨了变色有机电致发光器件存在的问题和发展趋势,拓展了研究思路,对推动变色有机电致发光器件的发展与应用具有指导作用。

关键词: 柔性电子;有机发光二极管;色彩和色温

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有机发光二极管(OLED)是新一代健康照明和高清晰度、高色域新型显示元器件,具有高效节能、大面积面光源、与柔性基底兼容等特点。相较传统硅基发光二极管(LED)光源,OLED有很大优势,其既可改善光谱从而避免蓝光危害,又可采用新颖分子材料优化色域,使得发光光谱色彩鲜亮逼真。目前,OLED已应用在健康照明和高色纯度显示领域中,在显示应用方面更可作为一种便携和可折叠拉伸的新型可穿戴电子产品,应用价值巨大。在最10年的时间里,OLED光电器件在固态照明和平板显示市场中始终有一席之地,并且有替代现有产品的趋势^[1-5]。自从第一个高效的OLED由邓青云院士^[6-8]在十九世纪八十年代发明以来,已经有了巨大的发展。例如:OLED器件的内量子效率通过磷光材料(由贵金属元素铱和铂等贵金属过渡金属配合物构成)和热活化延迟荧光(TADF)材料的应用已

经达到100%^[3, 9-10];白光OLED(WOLED)也可以用真空中物理气相沉积或者溶液法的制备,白光通过互补色双发光层或者RGB三基色三发光层得以实现^[11-13]。尽管如此,基于有机电致发光器件的产品价格依然非常昂贵,对于市场上的大部分消费者而言承担不起。因此,简化器件结构^[14-17]、制备工艺^[10, 18-20]及降低OLED器件产品的制造成本,以此扩大产品市场占有率势在必行^[21]。相对色温Correlated Color Temperature(CCT),在设计白光有机发光二极管(WOLED)时,是一个十分重要的概念,关乎人对色彩的心理感受。暖光是指低色温的光源,它的特征是红辐射相对较多、能量分布相对集中;冷光的色温及蓝光辐射比例较高,冷白光使人情绪上清醒和警觉,暖白光使人感到心情放松和舒适^[22-24]。新型变色有机发光二极管特点、分类及其应用如图1所示。

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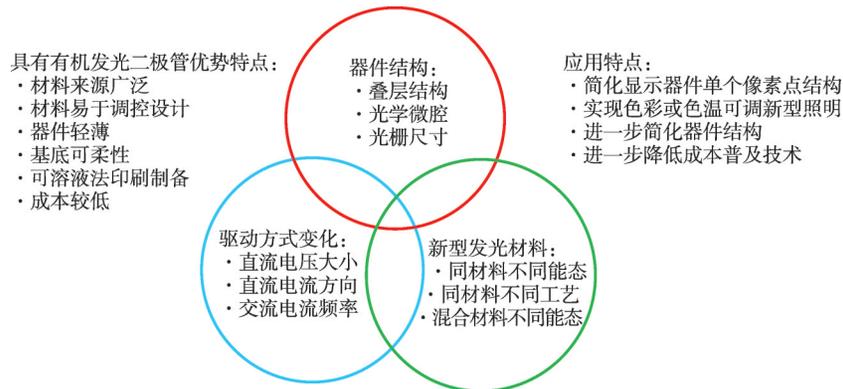


图1 新型变色有机发光二极管特点、分类及其应用

Figure 1 Schematic of characteristics, classification and application of novel color tunable organic light-emitting diodes

有机发光二极管是一种由多层功能薄膜构成的新型电致发光器件,而广义的变色有机发光二极管是在单个或单种有机发光二极管中实现色彩变化功能的新颖有机发光二极管。作为在单个或单种器件内实现多个色彩或色温变化的一种有机电致发光器件,变色有机发光二极管能简化器件结构和实现色彩或色温可调。变色有机发光二极管分为器件结构变色、材料变色、外加驱动方式变色,本文将详细综述3类器件结构的发展及最新研究进展。

1 器件结构变色

1996年,由美国普林斯顿大学的Forrest课题组^[25]提出OLED器件实现变色功能及颜色可调器

件的概念,即Color Tunable Organic Light Emitting Diodes(CT-OLEDs)。他们把两个互补色OLED器件,即在一个蓝光OLED和一个红光OLED中用一层镁银电极级联并接地,再由两个电压分别驱动两个互补色器件,利用驱动电压的变化实现了单个OLED器件的变色功能,该器件实现了单个器件从蓝光到红光的切换,展现出在照明和显示器件中的巨大潜力,开辟出CT-OLED器件的应用新领域。于1997年,Forrest组^[26]又将变色器件拓展至三基色OLED级联,实现了三基色和白色器件的切换和调控,这种基元光电器件如果用于显示领域中,将会使得原来的像素点尺寸最小化及分辨率最大化,在高清显示领域有巨大的应用前景。普林斯顿大学Forrest组发明的颜色可调有机电致发光器件如图2所示^[25-26]。

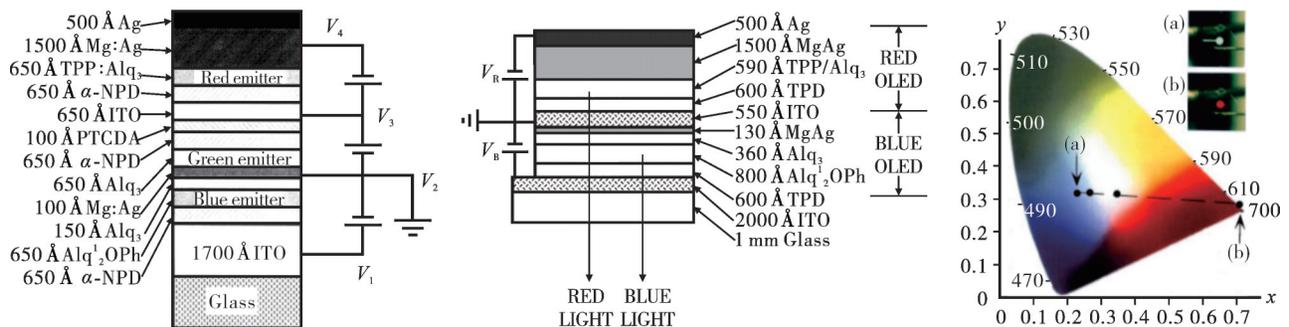


图2 普林斯顿大学Forrest组发明的颜色可调有机电致发光器件结构和CIE1931色坐标变化^[25-26]

Figure 2 Color tunable organic electroluminescent device structure developed by Princeton University Forrest Group

法布里-罗布(Fabry-Perot cavity theory)光学微腔是调控光电场的有效手段,其可以使得光谱变窄和在一定范围内可以调节,同时也利于提高OLED外量子效率,是进行光提取的一种重要手段。光学微腔的实现需要提高两个电极反射率,其中一个电极反射率需接近100%,另外一侧可观察光学微腔

效应,有窄化光谱使光强变强的效果。加拿大多伦多大学吕正红教授课题组^[27]采用法布里-罗布微腔效应,实现了同样材料结构的CT-OLED器件,器件结构如图3所示。该研究所用器件为易于实现微腔的顶发射,在ITO玻璃作为阳极的一侧蒸镀金属所用器件阴极采用复合电极LiF(0.5 nm)/Al(3 nm)/

Ag(30 nm)结构,该复合电极拥有良好的光电特性,在顶发射电极 Al 作为阴极的一侧采用新型含有富勒烯 TPD/Alq₃/C₆₀ 多个有机材料薄膜复合结构作为电子传输层,实现光电场的调控结构,使用绿光

材料 TPD/Alq₃ 作为发光层,通过器件膜的厚度调节实现发出蓝光、绿光、红光,器件亮度普遍超过 1000 cd·m⁻²,最高器件亮度接近 10 000 cd·m⁻²。

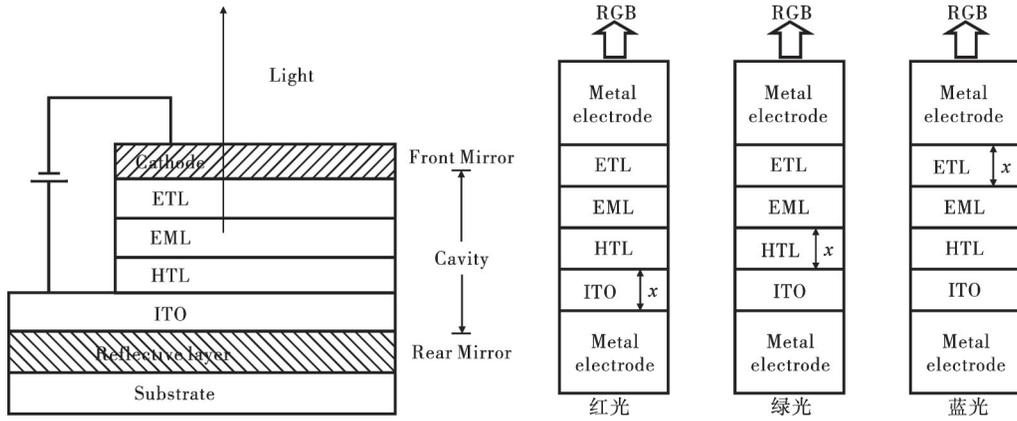


图 3 多伦多大学吕正红教授课题组采用法布里-罗布微腔效应实现同样材料和结构的器件红光,绿光,蓝光的连续调控^[27]

Figure 3 The Fabry-Perot microcavity effect was used to control the red, green and blue light of devices with the same material and structure by Lu Group

在法布里-罗布光学微腔的基础上,在全反射的电极一侧加入不同间距的光栅结构,可以对 OLED 器件内光电场实现进一步的光调控,器件结构如图 4 所示。韩国电子电信研究所研究者及其合作者德国德累斯顿工业大学 Karl Leo 教授^[28]设计出基于微

腔、金属光栅结构的色温可调白光 CT-OLED 器件结构和光栅结构,该结构可以很好的利用光学微腔和光栅间隔尺寸的变化实现冷暖白光的变化,白光可以从 3000 K 到 8000 K 色温的变化,实现如图 5 所示光谱和 CIE 变化的效果。

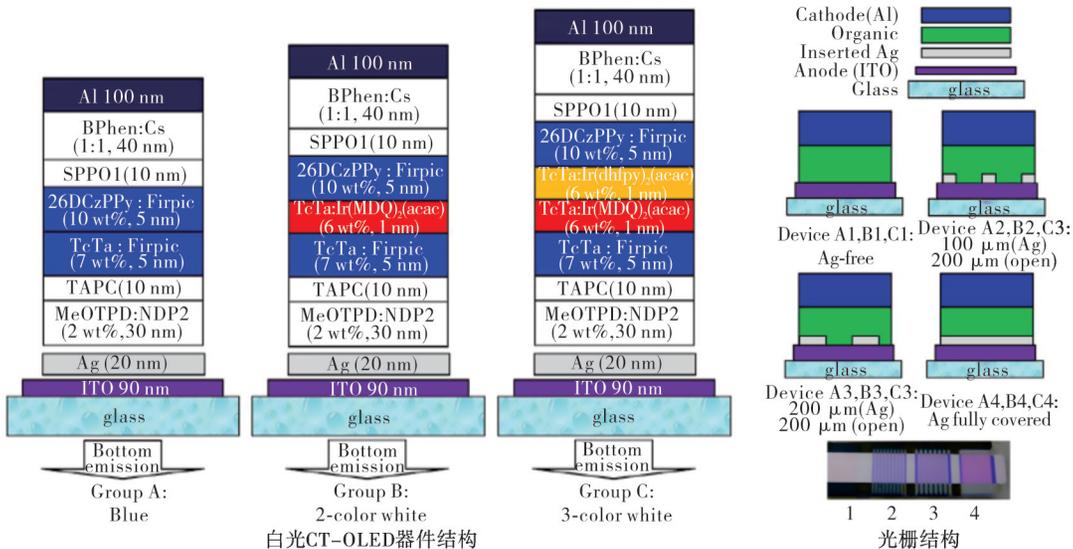


图 4 韩国电子电信研究所研究者及其合作者德国德累斯顿工业大学 Karl Leo 教授所设计基于微腔和金属光栅结构的色温可调白光 CT-OLED 器件结构和光栅结构^[28]

Figure 4 The CT-OLED device structure and grating structure based on micro-cavity and metal grating structure were designed by the researcher of electronics and telecommunications research institute of Korea and his collaborator professor Karl Leo of Technische Universitat Dresden in Germany

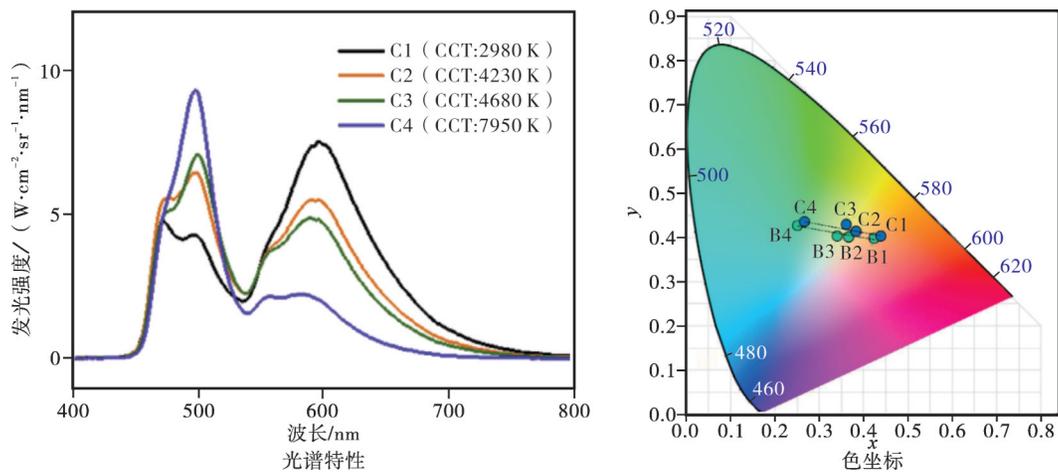


图5 基于微腔和金属光栅结构的色温可调白光CT-OLED器件的光谱特性和色坐标^[29]

Figure 5 Spectral characteristics and color coordinates of CT-OLED device based on microcavity and metal grating structure

从驱动电源性质的角度出发,采用交流电和直流电混合电源也可以实现颜色的变化。德国德累斯顿工业大学 Karl Leo 教授课题组^[29]基于交流电源驱动实现了CT-OLED器件(见图6),该器件结构由一个发蓝光的p-i-n结构的OLED器件和一个发黄光的p-i-n结构OLED器件通过银电极连接,并且在2个器件的3个电极处施加变流电源(可以实现直流和交流的切换),当采用正向电流时蓝光OLED器

件开启发出蓝光而对黄光而言是反方向电流器件不亮,当采用反向偏压时黄光OLED开启而对蓝光OLED而言是反向偏压器件不亮,当采用交流电流50 Hz电流驱动器件时两个器件都发亮且构成稳定的白光,并且随着频率变化白光实现了色温的调节,其中黄光OLED器件的功率效率高至 $75 \text{ lm}\cdot\text{W}^{-1}$,白光器件功率效率高至 $36.8 \text{ lm}\cdot\text{W}^{-1}$ 。

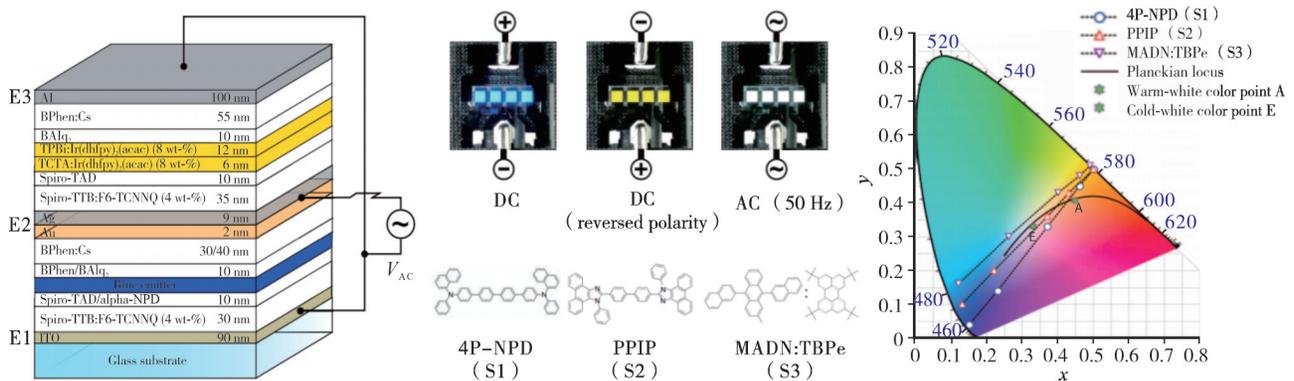


图6 德国德累斯顿工业大学 Karl Leo 教授课题组所设计基于交流电源驱动的变色 OLED^[29]

Figure 6 Prof. Karl Leo's research team at the Technical University of Dresden in Germany designed a CT-OLED driven by AC power

2016年,徐汀博士与上海交通大学何谷峰教授^[30]合作采用荧光有机发光材料作为超薄发光体,设计了一个新颖的黄光、蓝光双荧光超薄发光体变色CT-OLED器件结构,实现了颜色可调和色温可调的白光WOLED器件(见图7)。该器件变色色域广、高效率、低滚降,发光区域从天蓝光(0.22, 0.30)、冷白光(0.29, 0.33)、暖白光(0.43, 0.42)直到黄光(0.40, 0.45);中间调控层厚度对亮度、效率滚降

和色彩调控的机制有影响,在优化好的器件中载流子复合区域得到了扩大,两个发光体稳定的能量转移过程得到优化和激子的浓度淬灭得到抑制;所设计的CT-OLED开启电压2.82 V时最大亮度 $39810 \text{ cd}\cdot\text{m}^{-2}$ 、最大外量子效率达到6%,当所设计CT-OLED器件亮度从 $500 \text{ cd}\cdot\text{m}^{-2}$ 增加到 $5000 \text{ cd}\cdot\text{m}^{-2}$ 时器件的电流效率滚降低至11.1%;所设计的CT-OLED具有制备工艺简化和消耗有机发光材料少的优

势,从白光 WOLED 器件的角度看,器件可以实现色温 6932 K 到 3072 K 的调节,有助于实现健康照明光源。

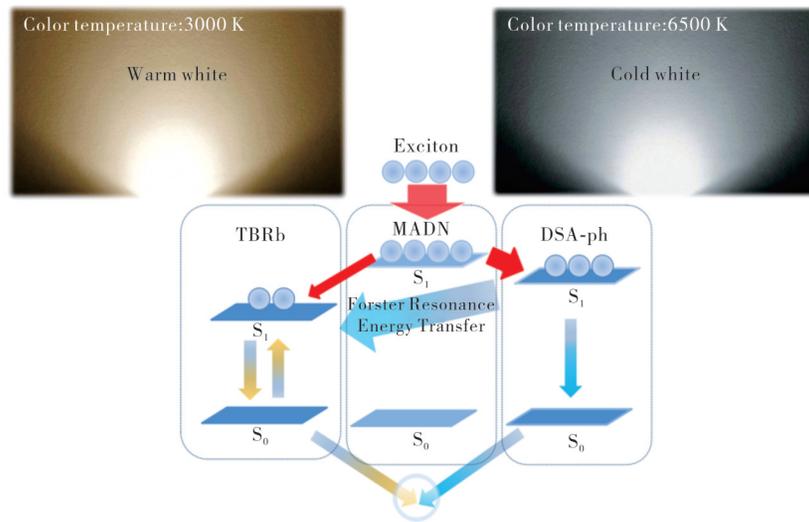


图 7 采用超薄发光体和长程能量转移机制实现冷暖白光调节白光 WOLED 效果^[30]

Figure 7 The mechanism of cold and warm white light by long range energy transfer of ultra-thin emitter in CT-OLED

WOLED 在节能环保绿色照明领域扮演重要角色,徐汀博士与苏州大学纳米科技学院研究者^[31]合作设计并制备了一种新型结构简化的高效 WOLED 器件(见图 8)。该器件是由互补色超薄发光体作为单个发光单元,采用叠层结构,通过高电子和空穴迁移率的界面激基复合物达到高效的能量传递,互补色发光单元分别是材料 PO-01 的黄光发光体和 FIrpic

材料的蓝光发光体。所设计的 WOLED 器件依据国际照明委员会标准 CIE 坐标为(0.36, 0.41),在不使用光萃取结构的情况下最高电流效率为 $41.5 \text{ cd} \cdot \text{A}^{-1}$ (EQE=18.59%),这个效率是目前使用全超薄结构达到的较好效率之一;所设计白光 OLED 的色温范围为 3686—4700 K,低色温的特点使得器件在健康白光照明方面有较大应用潜力。

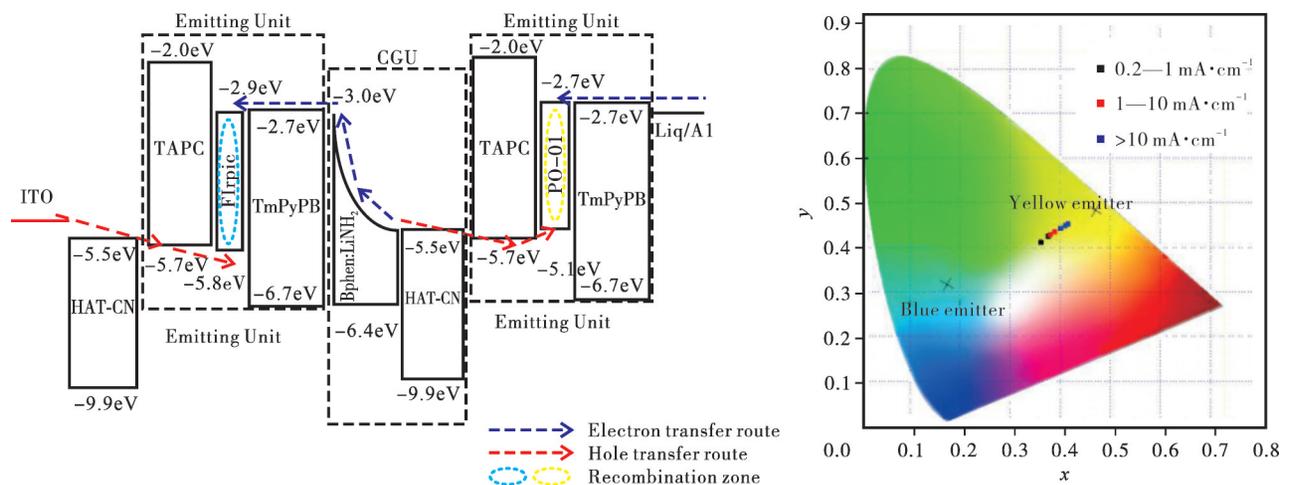


图 8 采用超薄发光体和叠层器件结构实现色温变化白光 WOLED 器件^[31]

Figure 8 The WOLED with color temperature variation is realized by ultrathin emitter and tandem device structure

2 材料变色

基于 CT-OLED 这一新概念,研究人员和科学

家不仅从器件结构来实现变色功能,而且从发光层组分子的角度出发,以稀土元素(如含有铕(Eu)元素 Eu^{3+} 离子)的有机物作为发光层,利用 Eu 发光峰窄的特性,与其他材料相混合,在不同电压驱动条件

下发光能量转移和载流子区域的变化,实现颜色可调的CT-OLED器件。

2006年,香港大学电子工程系Wallace教授课题组^[32]使用典型的空穴传输材料NPB及Eu³⁺作为空穴传输层和电子传输层,设计了结构为ITO/NPB

(40 nm)/Eu(DBM)₃bath(40 nm)/Alq₃(20 nm)/LiF/Al的器件,并且通过在NPB中掺杂perylene或者C545T,实现发光从红光到蓝光或者绿光的调控(见图9)。该器件结构简单,电流效率最高4 cd·A⁻¹,实现了多种颜色的调控发光。

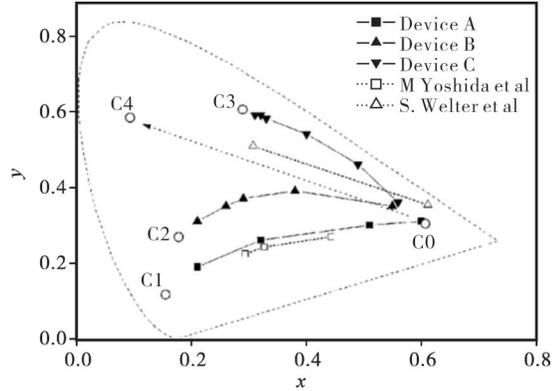
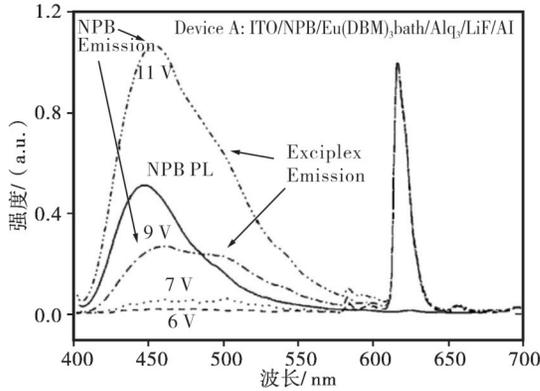


图9 香港大学Wallace教授所设计CT-OLED器件光谱特性和CIE色坐标的变化^[32]

Figure 9 Spectral characteristics and CIE coordinates of CT-OLED devices designed by Prof. Wallace, University of Hong Kong

2006年,华南理工大学曹镛院士团队^[33]提出高效单聚合物发光体光色可调白光OLED器件(见图10),实现在单个材料内单线态和三线态同时发光,

所发出白光达到CIE(0.33, 0.33)纯白光,开启电压5.9 V时电流效率达6.1 cd·A⁻¹。

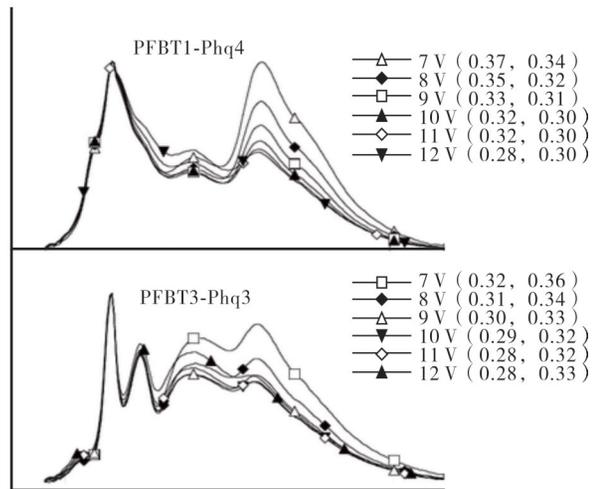
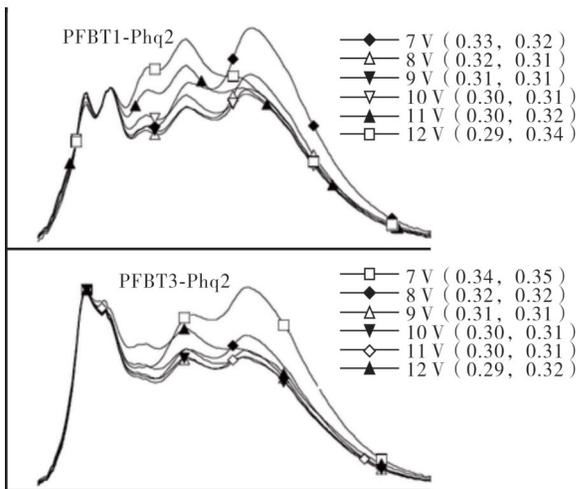


图10 华南理工大学曹镛院士团队提出高效单聚合物发光体光色可调白光OLED器件^[33]

Figure 10 Cao Yong, academician of South China University of Technology, and his team proposed an efficient single-polymer light-emitting white CT-OLED

2009年,意大利分子结构实验室^[32,34-35]采用全溶液法所制备的器件具有工艺简单的优势,基于新型含有铕(Eu)元素的有机分子材料和poly(9,9-dioctyl-fluorene)(PFO)电子传输材料,同样实现了CT-OLED的效果,其中外量子效率达到0.5%,结果如图11所示。

2016年,北京大学深圳研究生院孟鸿教授和何

谷峰研究员^[36]发现,当把2-芴蒽(FlAnt)和连二蒽(2A)以1:9的比例混合,得到掺杂体(Mixture1/9),基于Mixture1/9所制备的器件综合了FlAnt的蓝光和2A的黄绿光,在高驱动电压下发射出近白光,当驱动电压为10V时发射出的光几乎为纯白色,其中CIE(0.33,0.34),亮度为2560 cd·m⁻²(见图12)。

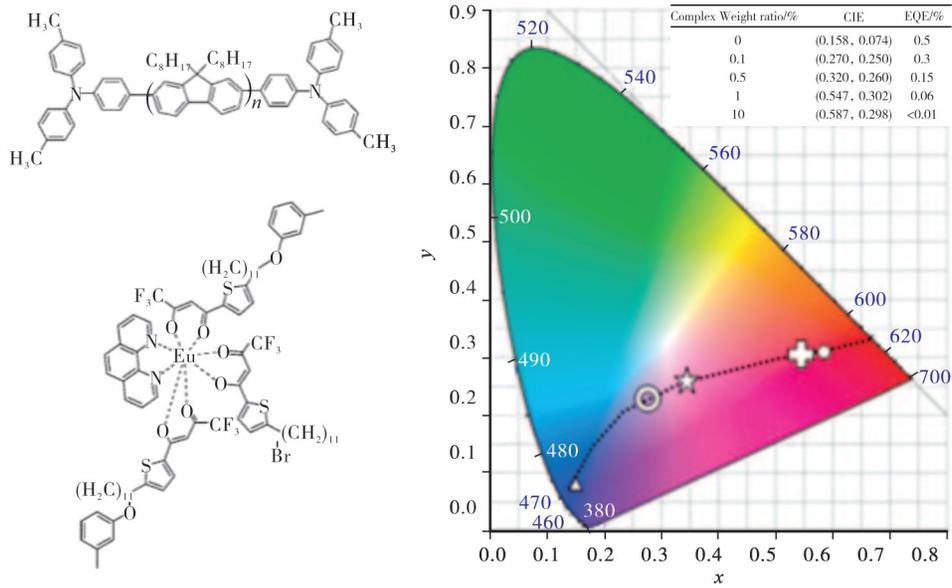


图 11 ITO/PEDOT:PSS/Eu(Phen)(L1-Br)(L1)2:PFO/Ca/Al 结构实现 CT-OLED 器件内量子效率和 CIE 1931 色坐标的变化^[34]

Figure 11 ITO/PEDOT:PSS/Eu(Phen)(L1-BR)(L1)2:PFO/Ca/Al structure to achieve the quantum efficiency and CIE 1931 color coordinate changes in CT-OLED devices

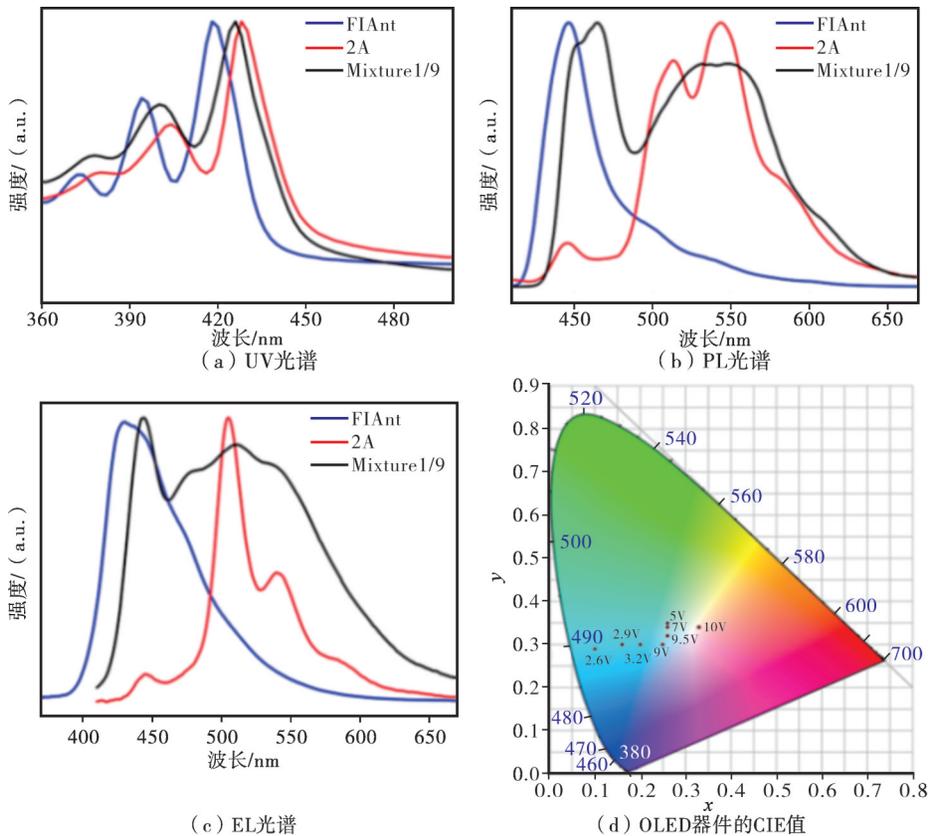


图 12 材料 FIAnt, 2A 和 Mixture1/9 的及基于 Mixture1/9 的 OLED 器件在不同驱动电压下的 CIE 值变化^[36]

Figure 12 UV, PL and EL spectra of materials FIAnt, 2A and Mixture1/9, and CIE changes of MIXture1/9 based OLED devices under different driving voltages

由于FIAnt和2A材料的单晶结构相似(见图13),少量FIAnt的掺杂没有造成体系内大量的陷阱。纯FIAnt和2A材料的OTFT器件迁移率分别是 0.22 和 $3.19 \text{ cm}^2 \cdot \text{Vs}^{-1}$,而基于Mixture1/9所制备的OTFT器件的迁移率却高达 $1.56 \text{ cm}^2 \cdot \text{Vs}^{-1}$,表明

少量FIAnt与2A材料的掺杂在实现白光的同时,也保证了足够好的电荷传输性质^[36]。这是第一次实现材料的高迁移率和纯白光双重性质,为自驱动的OLED照明和显示提供了材料基础,也为日后多功能材料的设计开辟了一条新的道路。

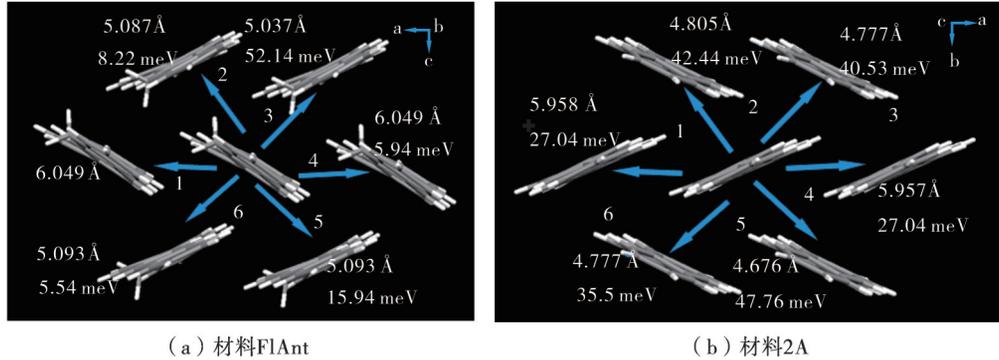


图13 材料FIAnt和2A的单晶结构^[36]

Figure 13 Single crystal structure of FIAnt and 2A

3 驱动变色

从交流电流载流子输运变化的角度出发,基于一种新型的电荷产生界面结构TCTA/MoO₃。新

加坡南洋理工大学半导体照明显示中心孙小卫教授课题组^[37]设计CT-OLED器件(见图14),器件把两个单载流子互补色超薄发光层器件用电荷产生层结构TCTA/MoO₃/TCTA级联,形成一个n-i-p-i-n的器件结构。

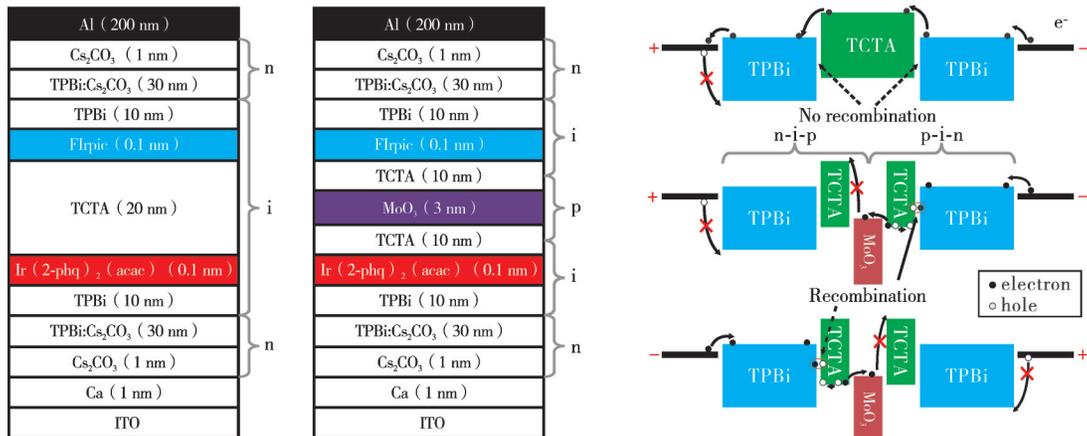


图14 南洋理工大学孙小卫教授课题组所设计CT-OLED器件^[37]

Figure 14 CT-OLED device designed by Professor Sun Xiaowei's research group in Nanyang Technological University

在交流电流源驱动下,当一侧电极为负极时电子注入到TPBi中,并且在外加电压驱动下电荷产生层产生对于电子空穴对并扩散至超薄发光层形成激子发光。在外加交流电驱动下,互补色橘红光和蓝光交替产生,随着偏压的变化复合区域也会有所偏移(见图15),从而实现了橘红光、冷暖白光到蓝光的切换变化,器件电流效率可以达到 $2 \text{ cd} \cdot \text{A}^{-1}$ ^[37]。

OLED发光效率通常蓝光器件低于黄光器件,为了形成良好的白光光谱,香港科技大学郭海诚教授课题组^[38]利用掩膜版工艺设计实现交流电CT-OLED器件结构并发现,正向偏压时两个蓝光OLED器件同时发光,反向偏压时器件发黄光,交流电时两个器件交替发光且可形成比较理想的白光,器件电流效率可以达到 $8 \text{ cd} \cdot \text{A}^{-1}$ 。

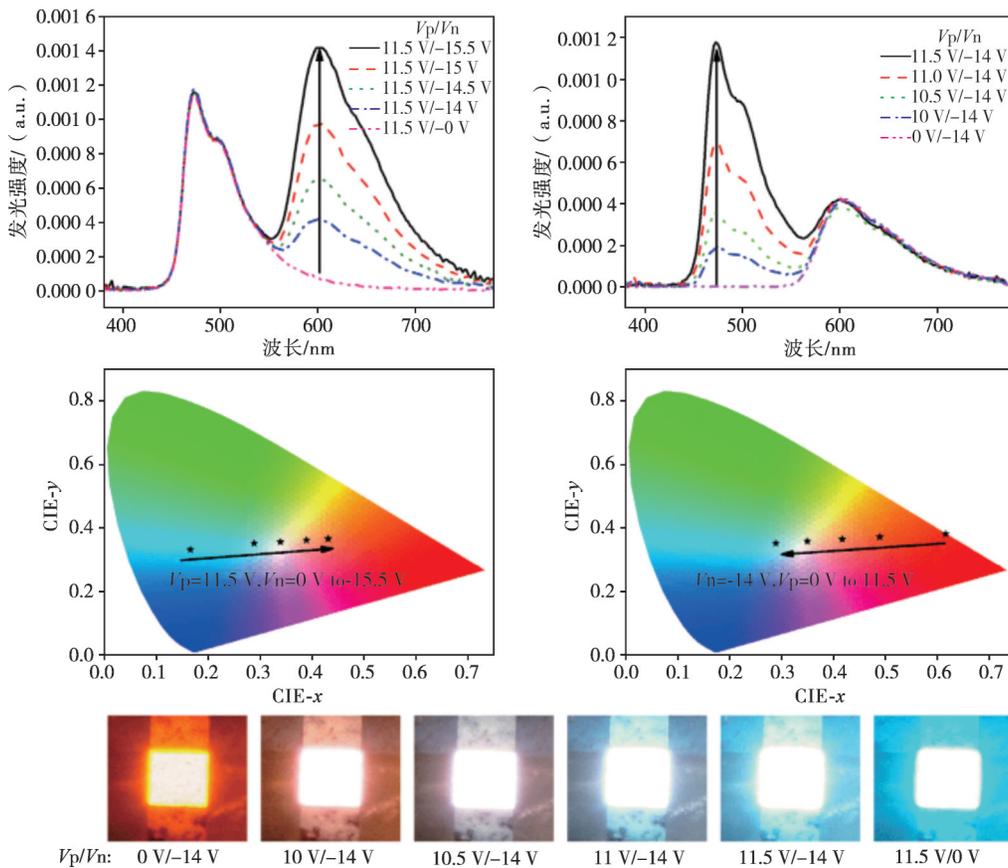


图 15 南洋理工大学半导体照明显示中心孙小卫教授课题组所设计 CT-OLED 器件性能^[37]

Figure 15 The device performance of CT-OLED device designed by Professor Sun Xiaowei's research group in Nanyang Technological University

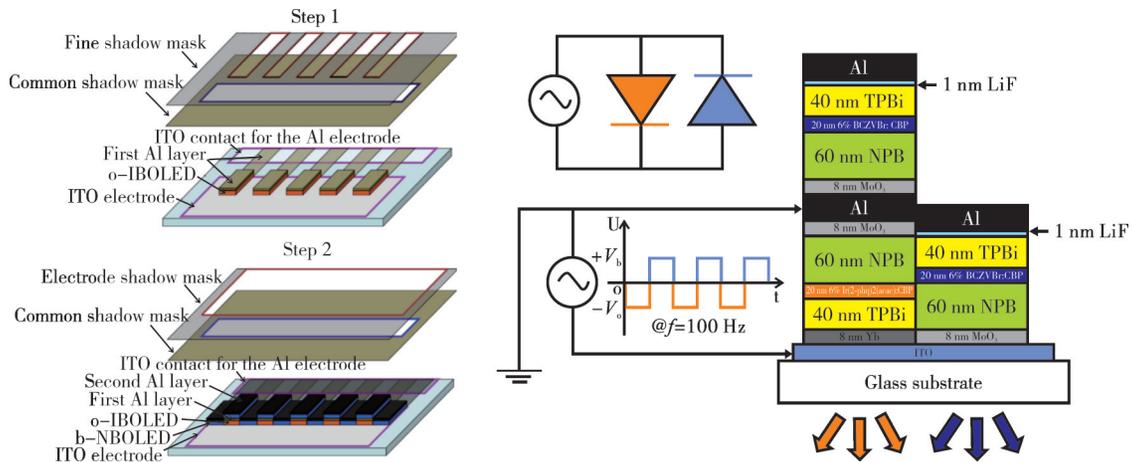


图 16 香港科技大学郭海诚教授利用掩膜版工艺设计实现新型交流电 CT-OLED 器件结构^[38]

Figure 16 Professor Hoi Sing Kwok of Hong Kong University of Science and Technology used mask technology to design an AC CT-OLED device structure

4 结语

变色有机电致发光器件可在单个发光器件中实

现色彩转换,在高分辨率显示方面有重要应用。变色有机电致发光器件分为器件结构变色、单发光层材料变色、外加驱动方式变色 3 个类别,目前变色有机电致发光器件依然存在效率低、工艺路线有待拓

展、所用材料有限的这些发展局限和需要解决的问题,可从如下3方面尝试新技术新方法突破目前技术瓶颈,丰富器件结构、提高发光色域和发光效率,助力变色有机电致发光器件产业化发展。

(1)器件结构和工艺方面,可在两个或三个发光体中插入高三线态的有机中间调控层,如TPBi、TCTA或者TAZ^[39-41],形成多个发光单元,与丰富的驱动方式配合,形成新的器件结构;工艺方面,一般采用溶液法工艺^[42],其也有进一步改善的空间,也可联合使用再蒸镀工艺,研制出更为丰富的器件

结构。

(2)发光材料方面,利用量子点碲化镉(CdSe)和硫化锌(ZnS),科学家设计和制备出变色量子点发光器件^[43]。随着材料科学进步,可将能发出高色纯度光的材料应用于可以拓展发光器件色域中,例如发光层采用硼-氮体系的热激活延迟荧光材料。

(3)高频交流驱动和电极界面修饰材料的应用,可结合新的器件结构和电极界面材料,进一步设计新型器件结构。表1总结了有机电致发光器件目前最新研究进展的问题及解决思路。

表1 有机电致发光器件目前最新研究进展的问题与解决思路

Table 1 The problems and solutions of the latest research progress of organic electroluminescence devices

变色有机电致发光器件分类	类别优势	类别商业化存在问题	类别解决思路
器件结构变色	技术路线丰富	单个发光单元结构复杂	结合新机理简化器件
材料变色	器件结构简单	不易稳定精准调控	探索新材料新机理
外加驱动方式	可精准调控频率	驱动外电路复杂	探索新器件结构

总之,在同一材料或器件中可实现变色有机电致发光器件,而且变色有机电致发光器件的功能和用途可以多样化,也可借鉴锂电池、超级电容等储能器件电极的先进技术,指导和制备色域更广、效率更高的变色有机电致发光器件,使其在高分辨率显示技术中发挥重要的作用。

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Progress in the Study of Color Tunable Organic Light Emitting Diode

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Abstract: Carbon-based organic semiconductor optoelectronic devices are easy to modulate from the molecular level design; organic electroluminescent devices have been gradually industrialized in recent years. Color tunable organic light emitting diodes (CT-OLEDs) are defined as the OLEDs realizing tuning color or color temperature in single device. CT-OLEDs effectively simplified structures of OLEDs with function of tuning color or color temperature applying in single pixel of display screen or lighting appliances. In this review article, we summarize that there are mainly three approaches to design and fabricate CT-OLEDs by novel device structures, novel luminescent materials and charging driving mode. Firstly, novel device structures of CT-OLEDs include complementary color connected in one OLED, Fabry-Perot cavity structure and optical grating structure. Secondly, the principle of CT-OLEDs using novel luminescent materials is that one layer of novel material emits light of different wavelengths from different energy states of one material, or mixed emitting materials as one layer emit light of different wavelengths from different energy states of different materials, or different treatment processes change the energy states of material in CT-OLEDs. Thirdly, CT-OLEDs realize tuning color or color temperature by charging driving electrical direction or frequency. Generally, CT-OLEDs show a great potential in applying in novel display and lighting in the future.

Keywords: flexible electronics; organic light-emitting diode; color and color temperature

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