



9,10-Bis(*N*-methylcarbazol-3-yl-vinyl-2)anthracene: High contrast piezofluoro-chromism and remarkably doping-improved electroluminescence performance



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ABSTRACT

This paper reported the synthesis, optical and electroluminescent properties of 9,10-bis(*N*-methyl-carbazol-3-yl-vinyl-2)anthracene (**MCA**). **MCA** could be facily prepared by the Wittig–Horner reaction of *N*-methyl-3-formylcarbazole and 9,10-bis(diethoxyphosphorylmethyl)anthracene and exhibited strong aggregation-induced and crystallization-enhanced emission (AIE/CEE) effect. Simple mechanical force could change **MCA** solid from initial green to final red emission, affording high contrast piezofluorochromism with large spectral shift of up to 90 nm. When **MCA** was used as the emissive material in light-emitting diodes, the doped devices showed remarkably improved electroluminescence (EL) performance whose EL efficiency was 8 times higher than that of the non-doped device. Moreover, the dopant concentrations did not affect significantly the fluorescence properties of doped films and EL performances of doped devices, demonstrating the potential advantage of AIE/CEE luminogens applicable for EL dopants.

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1. Introduction

Conjugated organic molecules exhibiting strong solid fluorescence and mechanically stable and meta-stable morphologies have been promising materials applicable in optical recording, active gain media for optically pumped lasers, and electroluminescence (EL) fields [1–15]. However, many conventional conjugated organic molecules suffer from intense intermolecular interactions and detrimental species such as excimers and exciplexes, which can induce notorious aggregation-caused quenching (ACQ) effect [16–19]. To mitigate these problems and obtain highly fluorescent solid materials, an optional way is to introduce bulky substituents

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into such chromophores [20–24]. The adjoint problem is that bulky groups usually dilute fluorophore concentration and hinder carrier transport. In 2001, Tang et al. discovered a novel anti-ACQ phenomenon: 1-methyl-1,2,3,4,5-pentaphenylsilole is non-emissive when dissolved molecularly in good solvents but become highly emissive upon aggregation in poor solvents and condensation in solid states, which was named “aggregation-induced emission (AIE)” [25]. Since then, a large number of AIE luminogens characterized by strongly twisted skeleton bearing rotatable aryl units have been developed and expected to be promising emitters for electroluminescence and piezofluorochromism (PFC) due to their strong solid-state fluorescence and diverse aggregation morphology [26–38]. Indeed, AIE luminogens have become the mainstay of PFC materials reported to date [3,39–41], and some EL devices with AIE luminogens as the emissive layer have been fabricated [42–46]. However, most present PFC materials based on AIE dyes show the PFC shifts within tens of nanometers under simple mechanical force [3,47–49], and those with very large PFC shifts (e.g., >80 nm) and well-marked colorimetric fluorescence are

still scarce [48,50]. On the other hand, when AIE luminogens are used as bulk emissive layers by vapor deposition, their EL performances seem to be not as good as expected [42–46]. This is probably due to the poor carrier transport caused by strong twisting skeleton and loose intermolecular stacking. Moreover, most AIE molecules are characterized by crystallization-enhanced emission and exhibit usually the decreased fluorescence efficiency in the amorphous aggregates. It is an innovative attempt to fabricate the highly fluorescent crystalline films of AIE molecules; however, this is quite difficult due to that vapor deposition affords amorphous films in principle. We have ever annealed and solvent-fumed the vapor-deposition films of AIE-active PFC dyes, but unlike the amorphized films produced by grinding or pressing, they can not be converted to high-fluorescent crystalline films. This is probably due to the lack of residual crystal seeds in vapor-deposition films [51]. To further improve EL performance of AIE dyes, an alternative way is to optimize device configuration. It is known that the doped devices could exhibit improved EL performance, but the conventional organic fluorophores usually show dopant concentration-dependent photoluminescence (PL) and EL properties [52–54]. We wonder how the EL performance is improved when AIE luminogens are used as fluorescent dopants. To our knowledge, there have been no such reports at present.

Here we have synthesized a simple AIE dye 9,10-bis(*N*-methylcarbazol-3-yl-vinyl-2)anthracene (**MCA**, Scheme 1), and its PFC and EL properties are investigated. We now present that **MCA** shows high contrast mechanochromic luminescence with very large PFC shift under simple mechanical force and remarkably doping-improved and dopant concentration-independent EL performance.

2. Experimental section

2.1. Materials

Starting materials and solvents were purchased from Aldrich Chemical Co., or Energy Chemical Co., China. Tetrahydrofuran (THF) was distilled over metallic sodium and dimethylformamide (DMF)

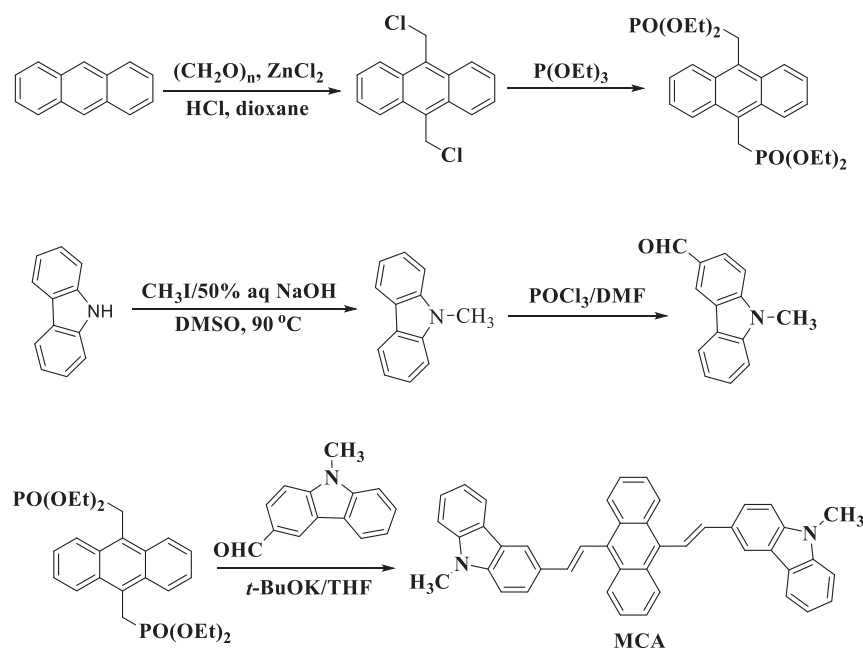
over calcium hydride before use. 4,4'-Dicarbazol-9-yl-1,1'-biphenyl (CBP), *N,N'*-di(1-naphthyl)-*N,N'*-diphenylbenzidine (NPB), 1,3,5-tris(1-phenyl-benzimidazol-2-yl)benzene (TPBi), and lithium fluoride (LiF) were purchased from Aldrich Chemical Co. and were utilized as host, hole-transporting, electron-transporting, hole-blocking, and interface modify-cation materials, respectively. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) (Baytron P4083, Bayer AG) was purchased from H. C. Starck Inc. They were used without further purification.

2.2. Preparation of aqueous dispersion and ground sample

The stock solution of **MCA** in THF with a concentration of 1.0×10^{-4} M was prepared and reserved. THF/water mixtures with different water fractions (aqueous dispersion) were prepared by slowly adding distilled water into the THF solution of **MCA** under ultrasound at room temperature, and the final concentration was kept at 1.0×10^{-5} M. Ground sample was prepared by grinding **MCA** powder on a glass plate with a metal spatula at room temperature.

2.3. Measurements

^1H and ^{13}C NMR spectra were recorded on a Bruker AC500 (500 MHz) spectrometer at 298 K by utilizing deuterated chloroform (CDCl_3) as the solvent and tetramethylsilane (TMS) as the internal standard. The MALDI-TOF-MS mass spectra were recorded using an AXIMA-CFRM plus instrument. Elemental analysis was performed on a PerkinElmer 2400. UV-vis absorption spectra were recorded on a Hitachi U-4100 spectrophotometer. Fluorescence measurements were carried out with a Hitachi F-4600 spectrophotometer. Solution fluorescence quantum yield (Φ) was determined at room temperature by the dilution method using fluorescein in water ($\text{pH} = 11$) as the reference. The quantum efficiencies of solid films were carried out with FLS980 Spectrometer. Thermal gravimetric analysis (TGA) was undertaken on a PerkinElmer thermal analysis system at a heating rate of $10^\circ\text{C min}^{-1}$ and a nitrogen flow rate of 80 mL min^{-1} . Differential scanning



Scheme 1. Synthetic route and structure of **MCA**.

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