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DNA as Functional Material in Organic-Based Electronics

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Abstract: Recently, biological materials such as DNA molecules, proteins, and albumen have been extensively investigated for various applications, as they are environmentally friendly and exhibit novel optical and electronic properties. Especially, over the last decades, DNA–lipid complex have been frequently reported as components of optical electronic devices. In this mini-review, the physicochemical performance of DNA–lipid complex is introduced, and then the related research progress in electronic devices such as organic thin film transistors and other optical-electrical devices are discussed. Finally, the challenges and prospects of other possible applications are also presented.

Keywords: DNA complex; OTFTs; optical electronic devices

1. Introduction

In recent years, with the development of electronic information technology and the massive use of plastic electronics, the materials waste problem has become increasingly serious, and plastic consumption and wastes have already been one of the major concerns in the world nowadays, especially in developing countries [1–3]. Therefore, it is essential to develop and investigate potential bio-degradable, bio-compatible, bioresorbable, or even metabolizable devices, which serve as the components of various electronic and photonic products. Environmentally-friendly electronics based on the natural materials such as polypeptide [4], albumen [5], and nucleic bases [6] are potential approaches that promise to meet these needs.

DNA (deoxyribonucleic acid), one of the most famous biopolymers, is a unique three-dimensional molecule with the sequence of four base pairs and carriers the genetic information for all living organisms. DNA exists in three different conformations including A, B, and Z forms. The most commonly known DNA structure is the B-form DNA and it possesses the right-handed helical sense, with diameter, rise per base pair of 2 nm, 0.34 nm, respectively [7]. DNA is negatively charged polyelectrolyte, and always are compensated by the positive charges inorganic cations, e.g., sodium ions [8]. Since purified DNA can usually only be soluble in water, which has been restricted in many fields for its further applications, so many efforts have been made to improve its solubility in organic solvents. For example, Okahata et al. [9] developed cationic surfactants such as cetyltrimethylammonium chloride (CTMA) in the preparation of DNA based complexes. Many other researchers have also utilized the DNA surfactant complex for the application in electronic devices.

Organic-soluble DNA–lipid complexes prepared with DNA and various cationic surfactants, including octadecyltrimethylammonium chloride (OTMA) and lauroylcholine (Lau), have gained considerable attention for their ease of fabrication process such as spin coating, dip-casting, or even printing techniques. Moreover, they show excellent physicochemical properties including high transparency, good thermal stability, and large bandgaps (4.7 eV). Among these cationic surfactants, CTMA is one of the most popular reported materials. Figure 1 illustrates the chemical structure of DNA, the surfactants, and the electrostatic interactions between them. The film of the DNA–CTMA complex has been employed in wave guides [10,11], lasers [12–14], and as a nonlinear optical material for second harmonic generation [15].

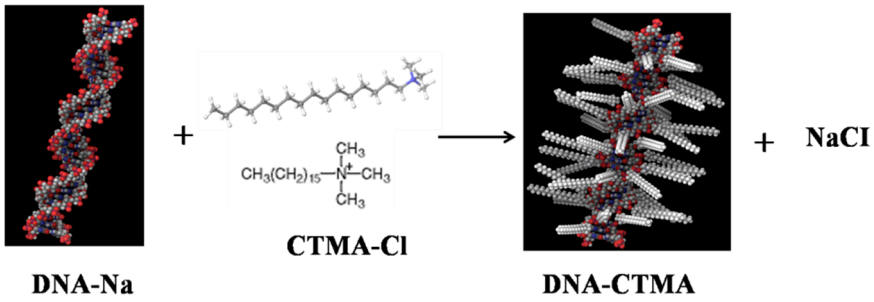

Figure 1. The ion exchange reaction between DNA and cationic surfactant.

In this mini-review, after a general introduction to the physicochemical performance of DNA–lipid complex, we summarized the recent research progress of DNA–lipid complexes in various electronic and photonic devices. In particular, we explain in details of the organic field effect transistors, in which the DNA–lipid complex functions as the gate dielectric or interlayer. At last, the future prospects of DNA in other electronic applications are presented, and the key issues for future development are discussed.

2. Physical Performance of DNA Thin Film

The special physicochemical properties enable DNA–lipid complex into various applications in optical-electronic devices. For example, the surface roughness of the DNA complex is very important when serving as the gate dielectric in OTFTs, because the charge transported at the interface between DNA complex and semiconductors [16]. In OLEDs, the excellent optical transparency in the visible range is necessary as it can be placed on either side of the diode. The characteristics of DNA and DNA complex have been investigated and briefly introduced as follows through various techniques including atomic force microscopy (AFM), current–voltage (I–V) performance, UV–vis and circular dichroism (CD) spectroscopy.

The surface profile of the DNA polymer film is scanned by AFM, as shown in Figure 2b (10×10 μm area). The DNA molecules (white dots) are distributed on the silicon substrate and are mutually aggregated with each other, i.e., forming the plurality of agglomerates. In comparison, the surface morphology of uniform film of DNA–CTMA complexes is shown in Figure 2c (2×2 μm). The RMS was estimated to be as low as 0.3 nm, indicating the DNA–CTMA film is smooth enough to be the gate dielectric layer in the OTFTs [16]. The optical properties of DNA complex film are characterized by the UV and CD spectroscopy, as shown in Figure 2d. From the UV spectroscopy, the DNA complex film shows a λ_{max} at 260 nm in the range between 220 to 300 nm, which could be attributed to the characteristic absorption of the nucleobases [17]. From the CD spectroscopy, one positive cotton effect and two negative cotton effects are found at about 280, 245, and 225 nm, respectively. Such features indicate that the DNA complex film existed in the configuration of B form [18].

The thermal stability of DNA and DNA–CTMA surfactant complex are given in Figure 2e, showing thermal degradation curves which indicate generally good thermal stability. It is very surprising that the DNA can be stable until 250 °C, suggesting that the DNA complex can tolerate various fabrications and may be the candidate for the application of temperature sensors. In addition, the resistivity of the DNA–CTMA and DNA alone were estimated, as shown in Figure 2h. And the current densities of DNA and DNA–CTMA were calculated to be 7.0 $\mu A/cm^2$ and 2.9 nA/cm^2 at 10 V, respectively. The good insulating properties of DNA-CTMA complex allow it to be the gate insulator layer in the OTFT devices [16].

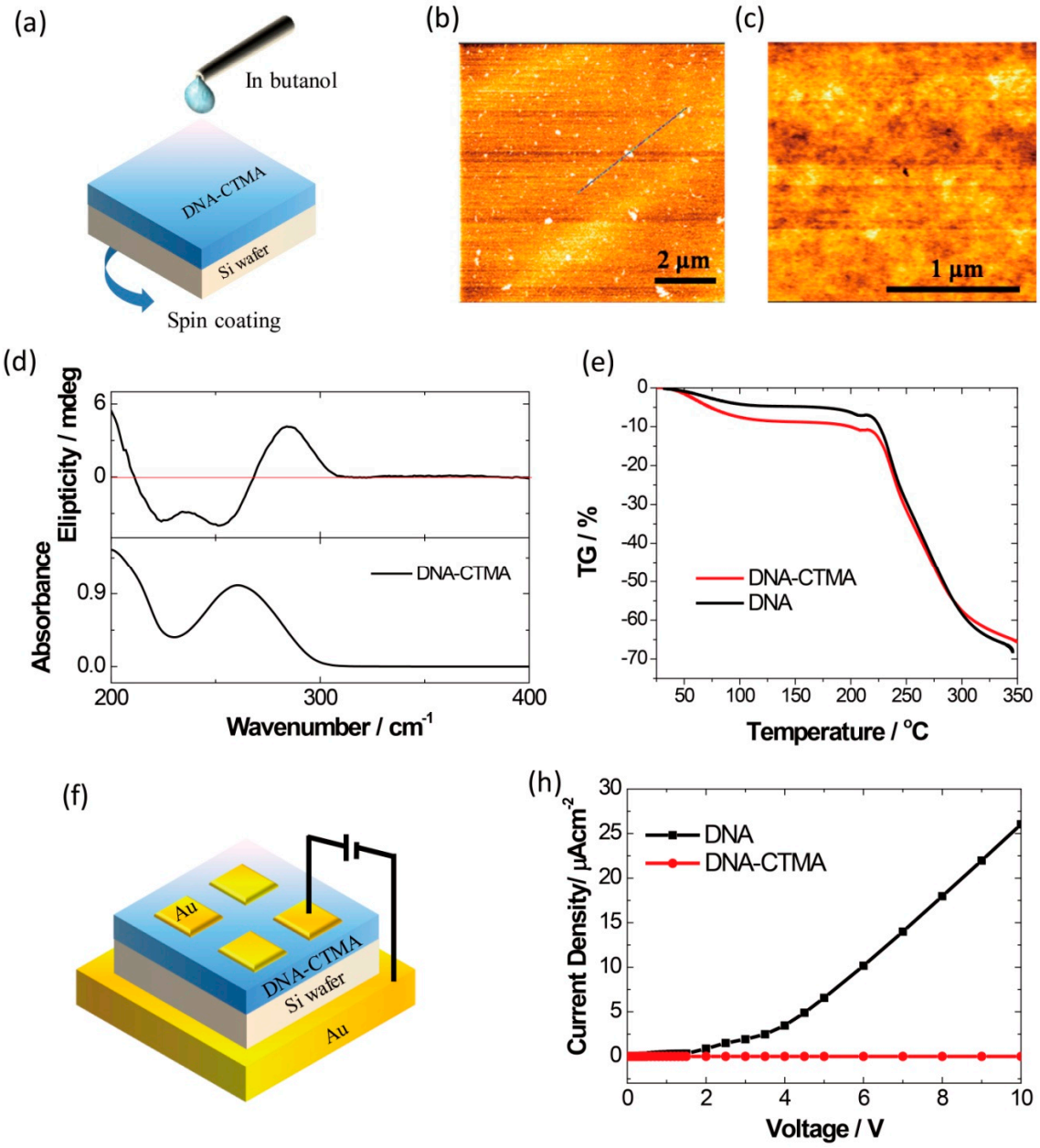

Figure 2. (**a**) The fabrication of DNA and DNA-complex thin films on a silica wafer by spin coating the butanol solution. AFM image of the deposited (**b**) DNA and (**c**) DNA–CTMA thin film characterizing the surface roughness; (**d**) UV–vis and CD spectral of DNA-complex film; (**e**) Thermogravimetric (TG) curves of DNA and DNA-complex; (**f**) Mental/insulator/mental (MIM) structure for I–V measurement; (**h**) I–V characteristics of DNA and DNA-complex thin film, respectively (Reproduced with permission from [19], Elsevier, 2016).

As discussed above, the generally good physicochemical characteristics and insulating properties of DNA and DNA–CTMA complexes would enable the applications in bio-organic electronics. In the following, we will next highlight the classical electronic devices based on DNA thin-films.

3. DNA as Functional Layers for OTFTs

The basic structure of OTFTs is composed of three functional layers: an insulating layer (gate dielectric), a semiconducting organic layer (*p*-type, *n*-type, or ambipolar), and three conducting electrodes (source, drain, and gate electrodes) [20]. The charge transport OTFTs is turned on when the gate field increases so that carriers accumulate at the interface between semiconductor and insulators. Hence, the performance of the OTFT critically relies on the physical and chemical performance of the dielectrics. When the dielectric layer has tunable polarity, the carriers can be depleted or accumulated more at the interface, allowing a memory effect and thus the OTFT memory device.

The dielectric materials in OTFT device can be classified into three categories: self–assembled monolayers (SAMs), inorganic, and organic materials [21]. Biocompatible and biodegradable dielectrics are applied to make the OTFTs sustainable and environmentally friendly. For instance, Uemura et al. developed the synthetic polypeptide poly (γ-methyl-L-glutamate) (PMLG), as the gate dielectric to prepare the organic field effect transistor memory as early as 2001 [22]. Jer-Wei Chang et al. [23] and So-Jung Kim et al. [24] demonstrated OTFTs based on chicken albumen as the gate dielectric. Recently, base pairs of DNA have also been applied as gate insulators in OTFTs as presented below.

The repeating units of DNA are nucleotides and each nucleotide is composed of nucleobases, including cytosine (referred by C), and thymine (T), adenine (A), and guanine (G). Among these base pairs, the adenine (A) and guanine (G) are found to have high dielectric constants, low dielectric losses, and high breakdown strengths, which are promising for being the dielectric of OTFTs. As shown in Figure 3, Mihai et al. [2] found that the base pair A and G based OTFTs exhibited the carrier mobility, operation voltage and the ON/OFF current ratio as 0.012 $cm^2 \cdot V^{-1} \cdot s^{-1}$, 15 V, 10^2, respectively, which needs to be the improved with a further optimized structure. Such a demonstration in applying the base pairs in OTFTs indicated a possibility to create 'all green' electronic devices.

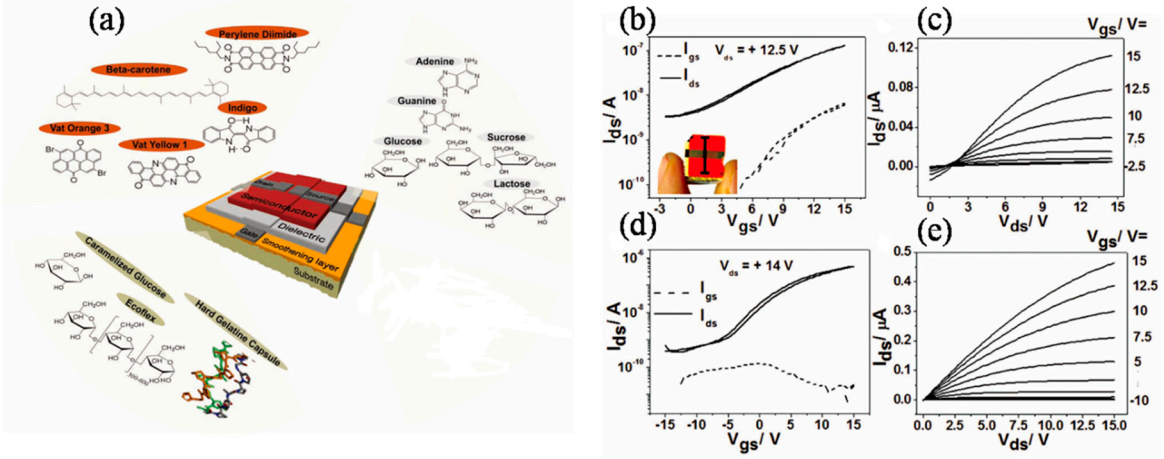

Figure 3. (**a**) Schematic configuration of the Bio-TFT device and chemical structures of natural materials; (**b**,**c**) Transfer and output characteristics of OTFTs based on four alternating layers of guanine and adenine as gate dielectric; (**d**,**e**) Transfer and output characteristics of OTFTs based on six inorganic layers of adenine and guanine as gate dielectric (Reproduced with permission from [1], Wiley-VCH, 2010).

Besides the application in basic OTFTs, bio-organic polymers derived from DNA have been widely applied to prepare photo-induced write-once read many-times memory devices [25], memristive memory devices [26] and OTFT memory devices [27,28]. The mechanism of the memorizing effect

caused by the DNA gate dielectrics remains poorly understood. Some of the researchers attribute the hysteresis behavior to the mobile ion impurities [29,30]. However, Liang et al. [19] demonstrated the memory window still exists even in a much lower temperature region, as shown in Figure 4. Since movement of ions may not explain such hysteresis, the memorizing effect could be attributed to the quasi-ferroelectric polarization originates from the orientation of the dipoles existed in the DNA complex.

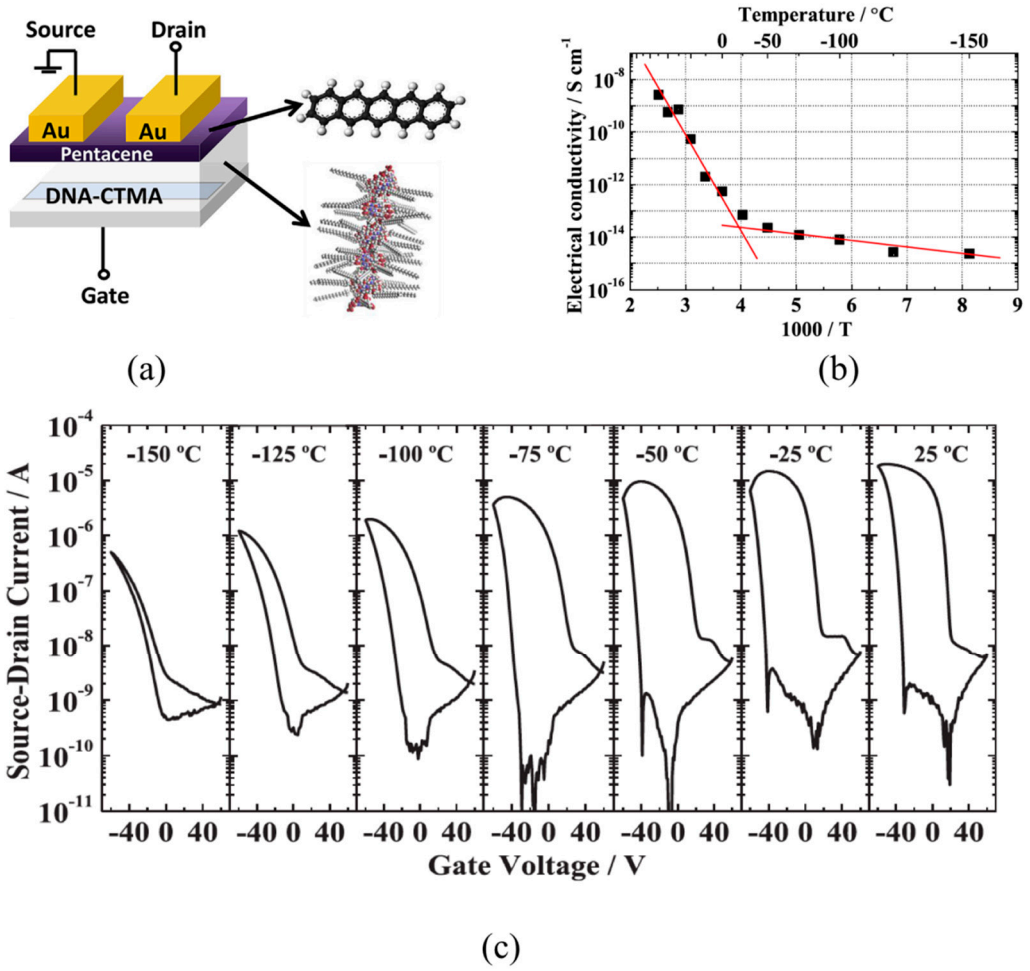

Figure 4. (**a**) Schematic configuration of the OFET memory device and chemical structure of DNA–CTMA and pentacene; (**b**) Arrhenius behavior of the metal/insulator/metal(MIM) device; (**c**) Transfer characteristics of DNA–CTMA based OTFT memory device at different temperature. (Reproduced with permission from [19], Elsevier, 2016).

Besides acting as the gate dielectric, DNA derivatives have also been employed as the insertion layer in the preparation of OTFTs. Such an insertion layer is developed to enhance the charge injection and the carrier mobility of OTFTs. In fabrications, the renewable, biodegradable polyelectrolyte DNA or DNA–CTMA complexes are easily deposited by solution processes using aqueous solvents or organic solvents, respectively [31,32]. For example, Wei et al. [32] integrated water-soluble DNA into OTFTs, in which an 8-nm thick DNA film was deposited as a buffer layer between electrode and pentacene film through spray-coating. Based on the characterization of surface morphology and microstructure of pentacene film, the authors suggested that the increase of carrier mobility from 0.035 to 0.153 cm^2/Vs can be ascribed to the decreasing of charge trapping at conducting channels as well as the effective inhibition of the bulk-like phase transition of pentacene.

Moreover, DNA was also used as the interlayer for the solution processed *n*-type or ambipolar OTFTs, as shown in the reports by Yuan Zhang et al. [33]. The schematic of OTFT structure and electric performance are shown in Figure 5a,b, respectively. The mobility and threshold voltage for *n*-type $PC_{70}BM$ and ambipolar BTDPP2 based device are summarized in Table 1. It demonstrated the DNA interlayer effectively enhanced charge carrier injection in OFETs. The improvement of the performance is attributed to the dipole layer originated from DNA and the enhanced injection from Au source electrode to the semiconducting layers.

Table 1. Device parameters comparison of OFETs fabricated with or without DNA interlayer [33].

	$PC_{70}BM$ (*p*-Type)			BTDPP2 (Ambipolar Type)		
	μ_{lin} [cm^2/Vs]	μ_{sat} [cm^2/Vs]	V_{th} [V]	μ_{lin} [cm^2/Vs]	μ_{sat} [cm^2/Vs]	V_{th} [V]
With DNA	1.7×10^{-2}	1.6×10^{-2}	10	2.3×10^{-3}	1.1×10^{-2}	6.5
Without DNA	7.5×10^{-3}	9×10^{-3}	16	4.8×10^{-4}	4.4×10^{-3}	12

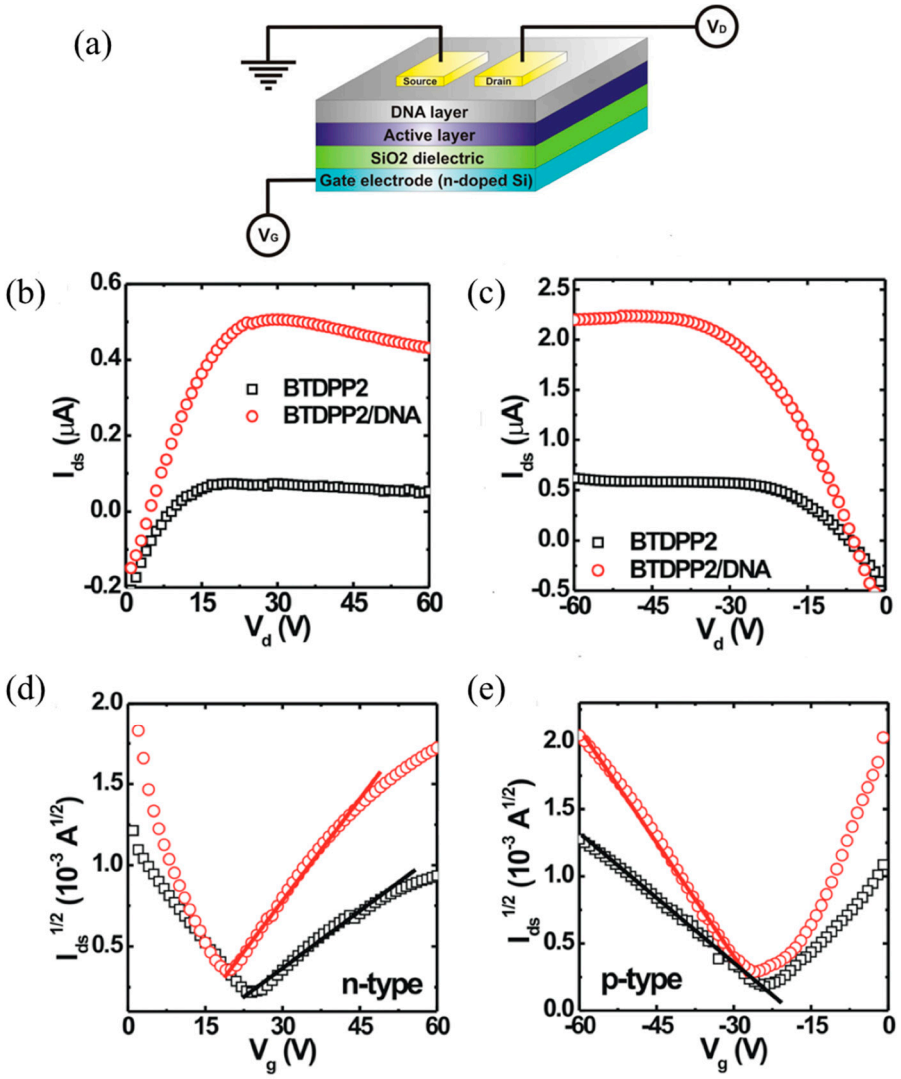

Figure 5. (**a**) Schematic configuration of the OFET device based on DNA as interlayer. Output and transfer characteristics of *n*-type (**b**,**d**) and *p*-type (**c**,**e**) modes with DNA and without DNA as the interlayer. (Reproduced with permission from [33], Wiley-VCH, 2012).

4. DNA for Other Optical Electronic Devices

Alongside OTFTs and OTFT-based memory, OLEDs have also received huge attention in the flexible printed electronics [34–39]. In the past decades, OLEDs have been actively explored for the low cost (through roll to roll processing), high contrast ratio, and wide color fullfillness. OLEDs have become a part of the heart in the display technology and have been used in wide applications like smart phones and music players. Since in 1987, Tang et al. improved the device performance through applying TPD, Alq_3, and Mg:Ag as the hole transport layer, luminous layer, cathode layer, respectively. The electromechanical luminescent devices exhibit the low driving voltage (~10 V), high luninence (>1000 cd/m^2), high efficiency (1.5 $lm \cdot W^{-1}$) [40]. In 1990, the Burroughes developed the electroluminescent device with polymers [41]. The research created the infinite possibility for the choice of materials and also opened new avenues for the preparation of large area electroluminescent devices.

Among the various polymers, the DNA and the nucleic acid bases were explored as hole blocking layers (HBL) or effective electron blocking layers (EBL) in Bio Organic light-emitting diodes (BioLEDs) [42–45], which enhanced the blue and green light emission effectively. As shown in Figure 6, Hagen et al. [46] demonstrated that the BioLEDs show a maximum luminous efficiency of about 8 and 1 cdA^{-1}, respectively. The operating voltages and turn-on voltages of these two devices were demonstrated as 10~25 V and 4~5 V, respectively. The green and blue BioLEDs exhibits the highest luminance of 21,000 cdm^{-2} and 1500 cdm^{-2}, respectively. These values indicated that the luminance improved about 10 times compared with the polymethyl methacrylate or polyvinyl carbazole based device.

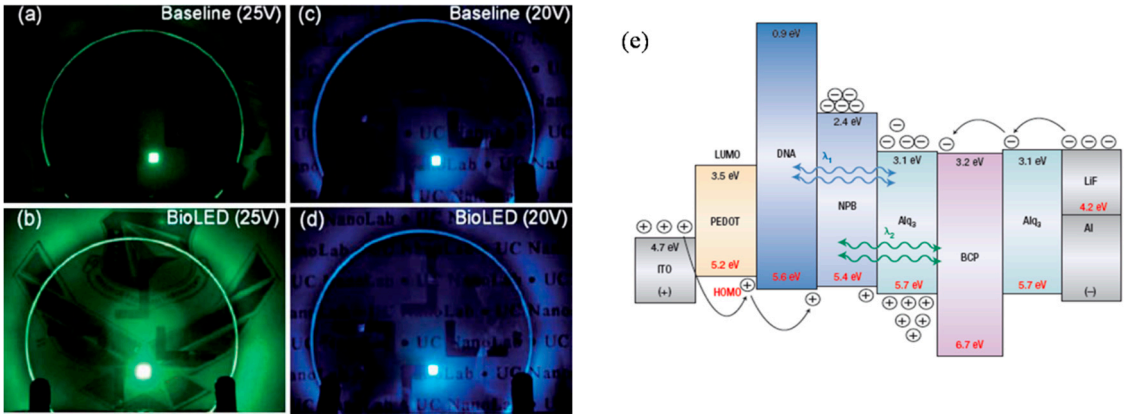

Figure 6. (**a**–**d**) Photographs of green-emitted (Alq3) and blue emitting (NPB) based BioLEDs devices in operation. (**e**) The energy-level diagrams for two color based BioLEDs (Reproduced with permission from [46], AIP Publishing, 2010).

In addition, significant advances have also been obtained in the phosphor based bio-hybrid light emitting diodes (Bio-HLEDs). Such devices exhibit high brightness and, most importantly, are ecofriendly and low cost. Park et al. [47] have demonstrated Bio-HLEDs through applying the pure DNA–curcumin-based phosphors as the chromophore material, as shown in Figure 7. The device shows a lower luminous efficiency drop rate and a bright luminescence of 62% and 0.0551 s^{-1}, respectively. Such results envision large-scale industrial applications of low-cost and ecofriendly illumination devices. Moreover, DNA can be even used in color-tunable OLEDs, which are of great importance for the good control of pixels in large-screen based display with enhanced resolution and color quality. Kobayashi et al. [48] developed the DNA/Pan/$Ru(bpy)_3^{2+}$ based color tunable OLEDs for the first time. The resulting OLEDs exhibited multicolor emission, namely, the color of the device change from green to yellow to red when applying the voltage of 10, 14, 18, 20 V, respectively. The device structure and characteristics are shown in Figure 8.

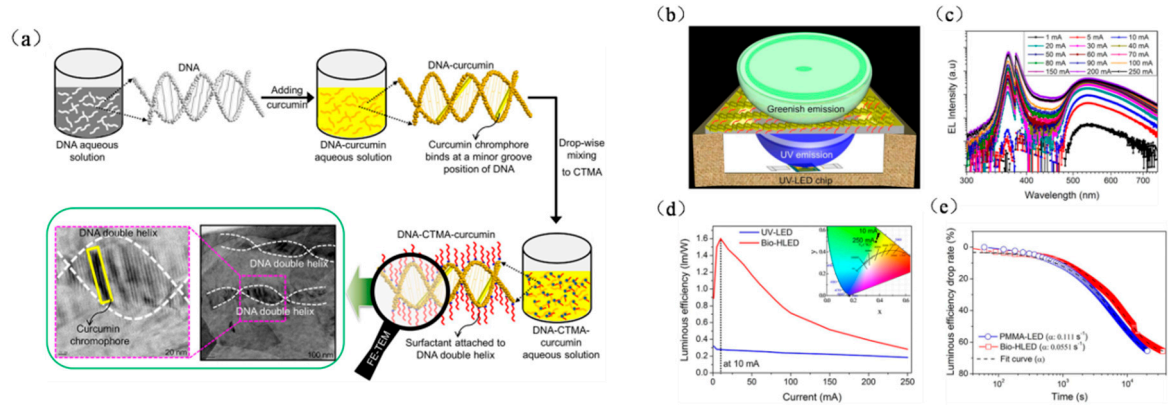

Figure 7. (**a**) Fabrication process of DNA–CTMA–curcumin aqueous solution; (**b**) Schematic configuration of DNA based Bio-HLED; (**c**) EL spectral; (**d**) Luminous efficiency spectral and the inset shows CIE colour; (**e**) Time-dependent characteristics of luminous efficiency drops of PMMA and DNA based OLEDs (Adapted with permission from [47], Nature Publishing Group, 2016).

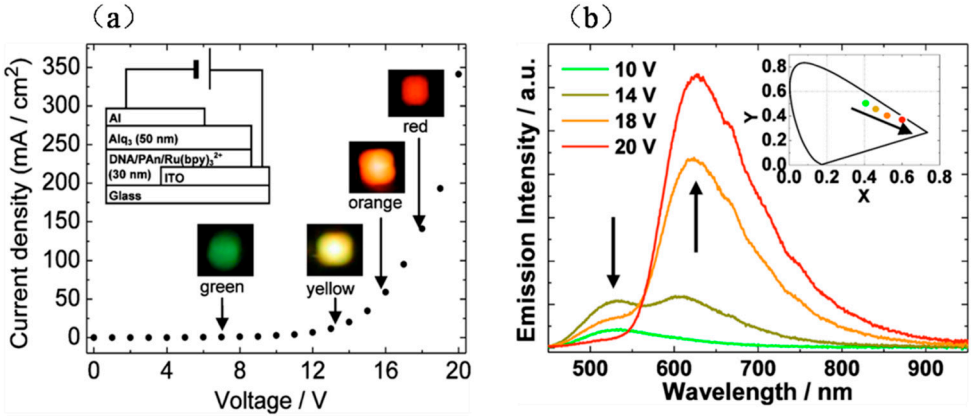

Figure 8. (**a**) Current–voltage characteristics of DNA based OLED (the inset shows a schematic configuration of the device); (**b**) EL spectral of OLED device with different bias voltages (the inset shows the CIE emission color with different voltages) (Reproduced with permission from [48], AIP Publishing, 2010).

Interestingly, DNA also exhibits the possibility of application in photovoltaic devices because of its wide bandgap, adjustable length, and so on. Jayme et al. [49] reported DNA and DNA-PEDOT:PSS coatings could be applied as hole-transport materials in the preparation of dye sensitized solar cell and promoted an increase in the device conversion efficiency. Dagar et al. [50] integrated DNA nanolayers in polymer solar cells, which resulted in power conversion efficiencies increased by ~110% and rectifying ratios by two orders of magnitude. Yusoff et al. [51] investigated perovskite solar cells with DNA hole-transport layer and observed that the device lifetime significantly improved. Besides using DNA as the nanolayer in solar cells, Kumar et al. [52] and Ensslen et al. [53] also demonstrate using DNA as structural element for chromophore assemblies in future solar cells, which may also encourage new applications of DNA as a long term, sustainable, green energy source.

5. Conclusions

The solution-processed DNA complexes are often deposited simply by drop-casting, spin-coating onto glass through self-assembly technique. With excellent thermal stability, high transparency, and a large band gap, DNA shows a significant promise in various applications. Especially in OTFT based devices, DNA can act as the gate-dielectric or quasi-ferroelectric dielectric to contact with

semiconducting layers, to allow control of charge accumulation and transport during operations. Besides, DNA can also be the contact insertion layers to enhance charge injection efficiency by taking advantage of dipole moments. In addition, DNA and the derivate have also exhibited interesting functions in enhancing device performance in other thin film devices, such as OLEDs and solar cells. Therefore, as inexpensive, earth abundant, and biodegradable polymers, DNA would be expected to show more potential in the future green electronics.

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