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Fabrication of non-doped red organic light emitting diode using naturally occurring Curcumin as a donor-acceptor-donor (D-A-D) emitting layer with very low turn-on voltage

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Abstract

The fabrication of a non-doped red organic light-emitting device (OLED) has been developed using naturally occurring curcumin as an emissive layer of OLED. In this research, the electroluminescent spectrum (EL) of curcumin as a donor-acceptor-donor (D-A-D) chromophore was obtained at wavelength of 612 nm. Also, the maximum external quantum efficiencies (EQE) for this device was measured and found to be 0.029 %. The device was characterized by low driving voltage below 1 V at a current density of 7 mA/cm^2. High color purity with Commission International de l'Eclairage coordinates was measured and found to be (0.544, 0.441). Quantum mechanical calculations were achieved base on DFT/B3LYP/6-311G (d,p) level of theory to rationalize the optical properties, indicating the optimized energy and computing the HOMO-LUMO values. Good conformity was observed between theoretical and experimental data.

Keywords*:* OLED; curcumin; donor-acceptor-donor (D-A-D); electroluminescent (EL); low turn on voltage

1. Introduction

Nowadays, the fabrication of non-doped organic light-emitting diodes (OLEDs) have received the increasing interest since they are an alternative method for fabrication process of red OLEDs, which often achieved by less convenient doping process (Hung& Chen, 2002; Fuhrmann & Salbeck, 2003; Chen, 2004). The ideal doping level in the fabrication of dopant-based red OLED is normally low (< 1-2 wt %) and confined in a narrow range (1 wt %)(Tang et al., 1989; Suzuki & Hoshino, 1996; Kido et al., 1994; Bulovic et al.,1998; Xie et al., 2001; Liu et al., 2003). Practically, the fabrication of dopant-based red OLEDs is not convenient for the rapid and mass production of OLEDs since the low concentration of doped material usually is employed for construction of OLEDs.

Recent studies have shown that there are three most successful approaches for reducing the energy band gap in organic compounds including: (*i*) extending the π-conjugation, (*ii*) tuning bond-length alternation in the π-system by stabilizing a quinoidal character, and (*iii*) introducing appropriate donor-acceptor functionalities in a molecule (push-pull effect) (Roncali, 1997; Wudl et al., 1997; Van Mullekom et al, 2001). Among chromophores with low band gap that have been established so far, the donor-acceptor (D-A) type of chromophores are particularly of interest since their band gap levels and other properties can be easily tuned by considering the proper donors and acceptors functionalities in chromophores. The D–A structure is almost a common feature found in many red or deep-red fluorescent chromophores (Chakraborty et al., 2005; Thomas et al., 2004; Kato et al., 2004).

In general, the D–A chromophores used in OLEDs emitting layer are fall into three main categories encompass: (*i*) single head donor-acceptor chromophores (D-A), (*ii*) double-head donor-acceptor-donor (D-A-D) chromophores, and (*iii*) double-head acceptor-donor- acceptor (A-D-A) chromophores. Some examples representing D-A structure: compounds **1** (Sharbati et al., 2010) and **2** (Guo et al., 2012), D-A-D structure: compounds **3** (Kulhánek& Bureš, 2012) and **4** (Takahashi et al., 2014) and A-D-A structure: compounds **5** (Sharbati et al., 2011) and **6** (Yao et al., 2014) are shown in Fig. 1.

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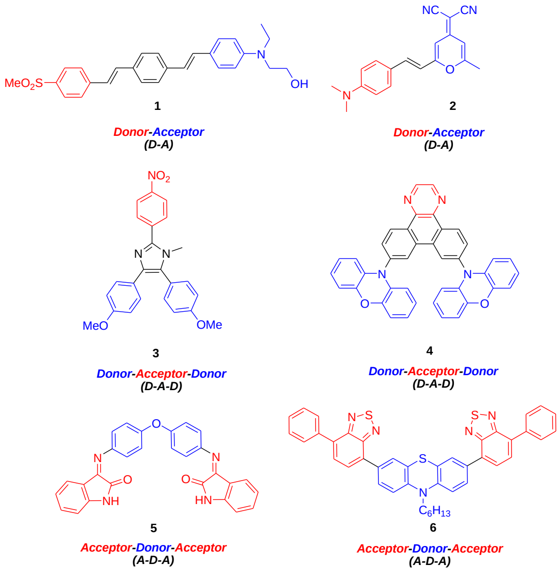

Fig. 1. The chemical structure of some D-A, D-A-D and A-D-A chromophores

Π-conjugated donor-acceptor-donor (D-A-D) oligomers and polymers have been extensively investigated for their controlled optoelectronic properties and also the linear and nonlinear optical effects (Mullen & Wegner, 1998; Skotheim & Reynolds, 2007). In the field of nonlinear optics, D-A-D molecules are special since they provide a large two-photon cross section that can be applied in three-dimensional patterning via photo-polymerization and optical limiting (Albota et al., 1998). Recently, Yang *et al.* reported the near infrared OLEDs based on two D-A-D molecules (Yang et al., 2008). Due to this research, the energy gap in D-A-D molecules can be tuned by changing the strengths of the acceptor component. The considerable electroluminescence properties and high external quantum efficiencies are known advantages for fabricated OLEDs based on D-A-D emitters, and extensive research in this field is now emerging (Yang et al., 2008).

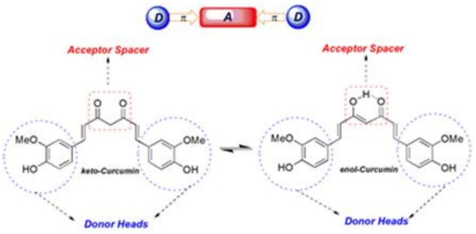

Fig. 2. The chemical structure of two curcumin's tatumers as D-A-D chromophores

Curcumin [1,7-bis(4-hydroxy-3-methoxyphenyl)-1,6-heptadiene-3,5-dione] (Fig. 2) is a naturally occurring yellow pigment obtained from the rhizomes of the plant Curcuma longa (Ahsan & Hadi, 1998). It is a common ingredient used in spices, cosmetics and the traditional medicine in Asia for treatment wounds, infections, and other skin problems (Hatcher et al., 2008). Various animals (Qureshi et al., 1992) and human studies (Lao et al., 2006) have proved that curcumin is extremely safe even at very high doses. The pharmacological safety and efficacy of curcumin makes it a potential compound for treating and prevention of a wide variety of human diseases (Unnikrishnan & Rao,1995; Anto et al., 1998; Strimpakos & Sharma, 2008; Anto et al., 1996; Tønnesen et al., 1987; De Clercq, 2000; Anand et al., 2008; Cui et al., 2007; Baum & Ng, 2004; Yang et al.,2005; Venkatesan et al., 2000; Srivastava et al., 1985; Nirmala & Puvanakrishnan, 1996; Arun& Nalini, 2002; Deodhar et al., 1980).

From a chemistry standpoint, curcumin is a bis-α,β-unsaturated β-diketone (commonly called diferuloyl methane), which exhibits the keto-enol tautomerism (Fig. 2) (Anand et al., 2007).

Although in recent decades numerous applications were developed for curcumin, mostly for its comestible, cosmetic and medicinal applications; however, to the best of our knowledge, there is no report for the use of curcumin in electrical devices especially optoelectronic instruments. In line with our continual research in fabrication of new OLEDs based on donor-π-acceptor oligomers; (Sharbati et al., 2011; Sharbati et al., 2011) hereby, we report the fabrication of a new red-OLEDs using naturally occurring curcumin as a donor-acceptor-donor (D-A-D) chromophore (Fig. 2). Curcumin used in this research was purchased from Aldrich Company but it can be freshly extracted from turmeric powder due to established procedure (Revathy et al., 2011).

2. Experimental Setup, Results and Discussion

2.1. Fabrication of OLED and Experimental Results

The absorption and photoluminescence (PL) spectra of curcumin was measured as a solution in pure methanol which is shown in Fig. 3.

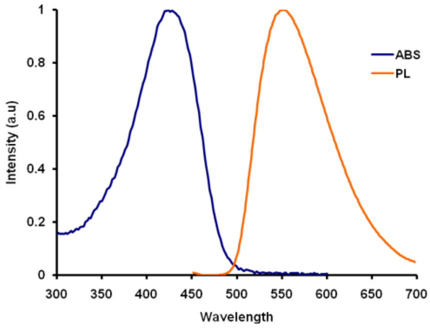

Fig. 3. ABS and PL spectra of curcumin in MeOH

The absorption spectra were recorded using a Perkin-Elmer Lambda 25 UV-Vis spectrometer. The PL spectrum was attained by excitation at the absorption maxima and recorded by an ISA SPEX Triax 180 spectrograph which was coupled to a spectrum-1 liquid nitrogen-cooled charge-coupled device detector. All measurements were performed in air at ambient temperature (room temperature). The absorption and PL spectra peaks were recorded at 424 and 555 nm, respectively.

In this experiment, a thermal evaporator instrument manufactured by Edwards Company for vaporizing organic as well as cathode layers was used. This instrument had the capability to apply the current into element up to 5 A. Also in this instrument, by the use of a rotary and a diffusion pumps in a chamber, the vacuum is generated and is able to generate the required vacuum in the range of 1×10^{-3} to 1×10^{-6} Torr. For vaporizing the materials with this instrument, the electric current is passed from a tungsten element (manufactured by R.D. Mathis Company, Model: B8A-3X.030W) and boat (manufactured by R. D. Mathis Company, Model: C1-AO) inside of this element that becomes warm. By warming the boat, the material inside (organic compound or Al metal used as a cathode) is vaporized.

Each of these materials was deposited separately in such a way that after deposition of each layer, including organic or cathode, the chamber cap was opened and the next material was inserted in a chamber and then the chamber cap was closed and the vacuum was applied to deposit the new layers.

The 3 mm diameter circular devices were made on 2 cm × 2 cm ITO slides. The materials were deposited on ITO by applying a thermal evaporator at 2×10^{-6} Torr, under vacuum condition. Finally, using a shadow mask made of stainless steel, Al cathode was deposited by thermal evaporation in vacuo at 2×10^{-6} Torr.

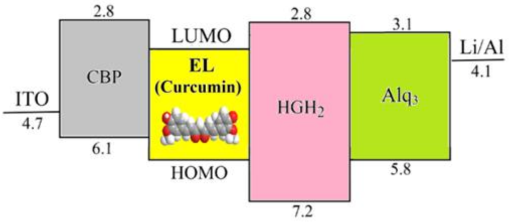

Fig. 4. Schematic energy level (in eV) diagram of multi-layer fabricated OLED using curcumin

The configuration of the multilayer fabricated OLED was ITO/4,4'-Bis(N-carbazolyl)-1,1'-biphenyl (CBP) (45 nm)/ Curcumin (35 nm)/ 1,4-phenylenebis(triphenyl silan) (HGH2) (10 nm)/ Alq_3(35 nm)/LiF(1 nm) /Al (100 nm), as shown in Fig. 4. The electrode work functions and HOMO/LUMO energy data values for CBP, Alq_3, and HGH2 were obtained from the literatures (Xue & Forrest, 2004; Hill et al., 2000).

To facilitate the electron injection, we applied Alq_3 and HGH2 (both purchased from Sigma-Aldrich Company) as electron transporting materials. Using these materials, the potential barriers between Al and Curcumin are decreased and provide more electrons in lowering the turning on voltage for recombination in emissive layer. HGH2 can not only facilitate the electron carriers injection from cathode to emissive layer but also can block the hole layer due to its large HOMO energy level. LiF on Al cathode was also used to assist the electron injection.

The EL spectra were measured using a spectrum analyzer model S2000 made by Ocean Optics Corporation.

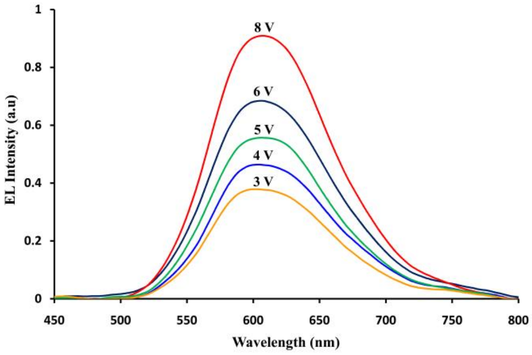

Fig. 5. Normalized EL spectra of curcumin OLED with a non-doped emissive layer

Figure 5 shows the EL spectra of device at J = 1 mA/cm^2. The EL peak emission wavelength is at λ = 612 nm for D-A-D based device which indicates 57 nm red-shift in comparison with its corresponding PL peak in solution.

Figure 5 also shows the EL spectra of curcumin OLED for different applied voltages with peaks at λ=612 nm. The same EL spectral peaks were observed at different bias voltages, showing the stable spectral characteristics against applied voltages.

Figure 6 indicates the CIE 1931 coordinate diagram simulated by MATLAB R2008a software in which x, y and r, g, b coordinates are (0.544, 0.441) and (0.7098, 0.2812, 0.0091), respectively. As it is well demonstrated, in visible light spectrum region that was indicated for curcumin OLED, approximately 71% red, 28% green and 1% blue colour was found.

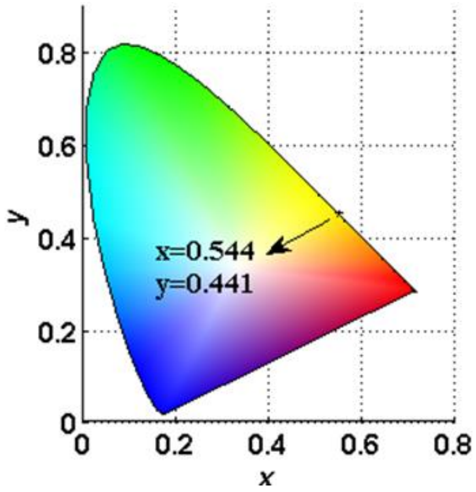

Fig. 6. CIE 1931 coordinates diagram for curcumin OLED light spectrum

Figure 7 shows the current density (J) versus voltage (V) characteristics for devices based on D-A-D oligomer in two different thicknesses using a Keithley 2400 current-voltage source. These characteristics were obtained by the method described in literature (Sharbati et al., 2010).

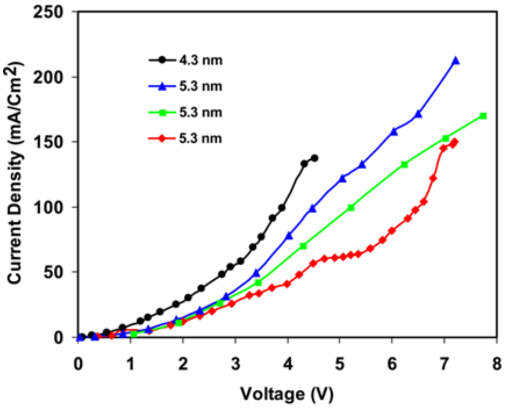

Fig. 7. The Current Density (*J*) versus voltage (*V*) for curcumin OLED

The effects of donor-acceptor on energy band-gap and also operating voltage have been investigated. As discussed earlier, the major chromophores employed in OLEDs are based on donor-acceptor species with a large delocalized p-electrons system. Therefore, this arrangement provides an efficient intramolecular charge-transfer (ICT) and enables further fine-tuning of polarizability of the chromophore (Moona & Kim, 2009). In this regard, the presence of electron donating and withdrawing groups facilitates the injection hole and electron injection properties into π-conjugated system, respectively. As a result, this arrangement can remarkably lower the turn-on voltage.

As can be seen in Fig. 7, the experimental results strongly confirmed the above discussion. In this Figure, the fabricated OLED with 4.3 nm thickness was turned on in very low voltage below 1 volt (≈0.5 V) and the three OLEDs with 5.3 nm thickness turned on in about 1 volt.

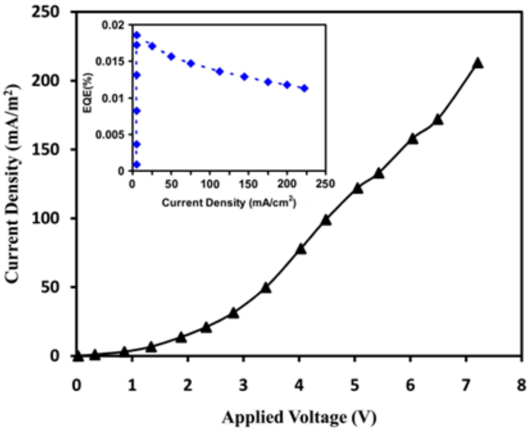

Fig. 8. Current density *J* as a function of the voltage *V* and external quantum efficiency (EQE) of device as function of *J*

The external quantum efficiency (EQE) was derived due to our previous method (Sharbati et al., 2011). The EQE of fabricated device for 5.3 nm thickness and related I-V curve is shown in Fig. 8. The fabricated curcumin OLED has a maximum EQE of ηEQE=0.029% which gradually decreased to ηEQE=0.017%.

The lower efficiencies for D-A-D based devices is attributed to much higher fluorescent quenching of the NIR emitter beside their longer emission wavelength (lower energy band gap and thus lower photon energy).

The EQE of devices at low current densities are remarkably higher. Such a phenomenon is quite usual in red-emitting OLEDs because of emissive quenching by charged or electro-generated triplet species in the device (Young et al., 2002; Wu et al., 2002). The red- or NIR-emissive materials usually have a smaller band gap and they are more susceptible to be altered by charges rather than the impact of other factors.

In addition to the technical drawback such as thermal evaporation, there was a low ability to deposit active materials on each layer without removing the vacuum in chamber. This may cause many defects in structure of OLEDs. One way for improving EQE for these OLEDs consider multilayer structure in emissive layer and also the use of an appropriate host material; however, this was difficult to apply since the host material often has a marginal co-evaporation ability.

In general, curcumin has the remarkable advantage of being able to be used as a red fluorescent chromophores in OLEDs from different aspects: (*i*) curcumin has an EL peak emission wavelength at λ =612 nm and this wavelength is comparable to our previously reported chromaphores and also by others, for instance: 630 and 700 nm (Sharbati et al, 2011), 600 to 905 nm (Sharbati et al, 2010), 700 nm (Yao et al., 2014), 486 to 529 nm (Takahashi et al, 2014), 566 to 678 (Lee et al., 2011) and 692 to 815 (Yang, et al., 2008), and (*ii*) in comparison to other established chromophores which mostly are synthetic, less available, expensive and incompatible with ecofriendly protocols, also curcumin is a natural product, non-toxic and cheap. These unique utilities of curcumin make it useful and a potential chromophore in mass production of red-OLEDs.

2.2. Quantum Mechanical Calculations

Quantum mechanical calculations for molecular structures and relative stabilities of different conformations of curcumin were performed using Gaussian 03W package program (Frisch et al., 2003). The hybrid density functional B3LYP (Becke, 1993; Lee et al., 1988) level of density functional theory (DFT) was employed at 6-311G (d,p) basis set.

The energy levels of the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO), and the energy gap between these molecular orbitals (MOs) were calculated using NBO program (version 3.0) implemented in the Gaussian 03 program. GaussView 5.0 graphical user interface (Dennington et al., 2009) for the Gaussian program was used for the visual presentation of molecular structure, MO surfaces and also theoretical absorption spectrum.

According to the calculations for different conformations of the enol forms of curcumin, the enol conformers in which the C-O single and double bonds and the C=C bands of carbon-chains are in the cis position, are considerably more stable than other enol and keto forms. Figure 9 shows the optimized molecular structure for the most stable conformation of curcumin. As the Fig. 9 indicates, the main scaffolds of molecule as well as the residues (except that of CH_3 bond to O atom) are almost coplanar because of proper resonance conjugations, and also having a relatively strong intramolecular hydrogen bond (O···H−O) in central fragment of this enol tautomer.

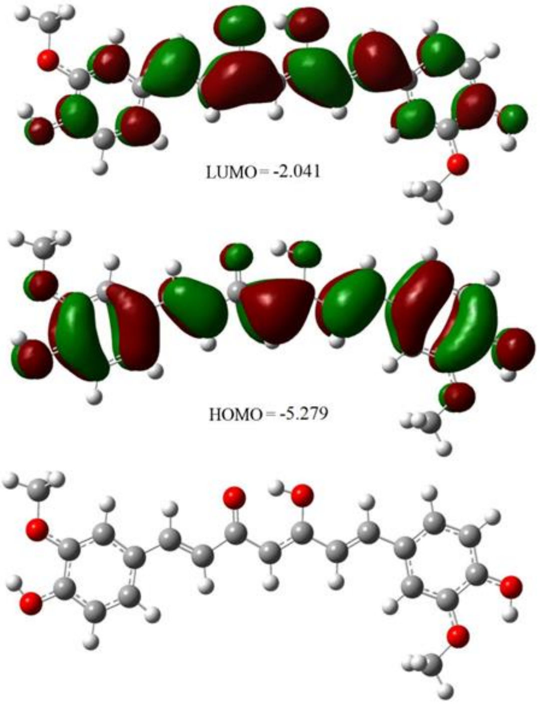

Fig. 9. Optimized molecular structure for the most stable form of curcumin, and its HOMO and LUMO surfaces beside their calculated energies (in eV) at B3LYP/6-311G (d,p) level of theory

As can be observed from structure of curcumin (Fig. 10), two oxygen atoms are responsible for push-pull effect through π-electron delocalization in molecule. The lone-pair of oxygen (LP) in hydroxyl group (donor group) at aryl residue is pushing the electron into π-conjugated system, whereas the electron is pulled out to one of the carbonyl (C=O) group (acceptor group). The difference in hybridization (oxygen hybridization: sp^3 in C-O, versus sp^2 in C=O) of two mentioned oxygens and hence the differences in electronegativity are the main factor for strong push-pull effect (Fig. 10).

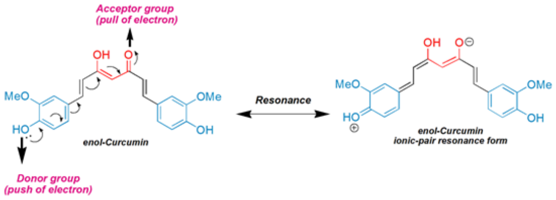

Fig. 10. Push–pull effect through conjugated bonds in curcumin

The HOMO and LUMO level surfaces and their calculated energies at B3LYP/6-311G (d,p) level of theory are also shown in Fig. 9. According to these calculations, the energy gap between HOMO and LUMO levels of curcumin for its absorption (ABS) spectrum is 3.24 eV (383 nm). As mentioned in the previous section, the experimental absorption electronic spectrum peaks at 424 nm (Fig. 3), equivalent to 2.92 eV, signifying about 11% error for the theoretically calculated HOMO-LUMO gap.

To improve the results of the theoretical calculation, time-dependent DFT (TD-DFT) at B3LYP/6-311G (d,p) level of theory was applied to evaluate the HOMO-LUMO gaps from the lowest electronic excitation energy. The optical characteristic of curcumin OLED is summarized in Table 1.

The obtained theoretical electronic absorption spectrum for curcumin is shown in Fig. 11. As TD-DFT calculation indicates, the HOMO-LUMO gaps from the computed first excitation energy is 2.95 eV (420 nm), which shows the best agreement with the experimental result (about 1% error).

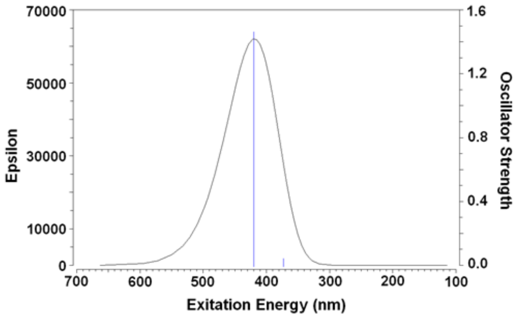

Fig. 11. TD-DFT calculated UV-Vis spectrum for curcumin obtained at B3LYP/6-311G (d,p) level of theory

Table 1. Optical properties of curcumin and fabricated OLEDs based on this oligomer

ABS		PL	EL		EQE$_{max}$	EHOMO (B3LYP)	ELUMO (B3LYP)	ABS Gap (DFT)[b]	ABS Gap (TD-DFT)[c]
λ_{max} (nm)	Gap$_{exp}$[a](eV)	λ_{max} (nm)	λ_{max} (nm)	Gap$_{exp}$[a](eV)	(%)	(eV)	(eV)	(eV)	(eV)
424	2.93	555	612	2.03	0.029	-5.279	-2.041	3.24	2.95

[a] Experimental HOMO-LUMO gaps obtained from ABS and EL spectra (E_{HOMO} and E_{LUMO} are the energy levels of the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO))

[b] The HOMO-LUMO absorption gaps, directly obtained from the energy differences between the HOMO and LUMO

[c] The HOMO-LUMO absorption gaps computed using time-dependent density functional theory method

3. Conclusions

In conclusion, the fabrication of new red-OLED based on curcumin as a naturally occurring D-A-D oligomer has been described. The electrical and optical characteristics of curcumin-OLED have been indicated. HOMO-LUMO values have been measured theoretically using density functional theory (DFT) and they showed the best agreement with the experimental results. A maximum external quantum efficiency (EQE) of ηEQE=0.029% was obtained for device based on A-D-A and device has an emission at 612 nm. The fabricated curcumin-OLED in this study showed very low turn-on voltages at 0.5-1 V. The calculated coordinate value of CIE 1931 from the electroluminescence (EL) spectrum is (0.544, 0.441). The current study on curcumin OLED has proved that for fabricating an efficient red OLED, both properties of chromophores and device configuration are critically important factors.

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