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APPLICATION OF CARBAZOLE DERIVATIVES AS A MULTIFUNCTIONAL MATERIAL FOR ORGANIC LIGHT-EMITTING DEVICES

The object of research is newly synthesized carbazole-derived compounds and organic light-emitting structures based on them. The problem lies in the complex solution of scientific and technical problems of improving the characteristics and stability of organic light-emitting diodes (OLEDs), namely improving the brightness and energy-efficient parameters.

Organic light-emitting structures of blue, blue, and green radiation with color coordinates were formed by the thermovacuum sputtering method and the solution deposition method. The turn-on voltage of the white OLED is 6 V, the maximum brightness of the light-emitting structures was 10,000 cd/m^2. The devices demonstrated a sufficiently high external quantum efficiency of 5 % to 7 %.

This paper reports the multifunctional application of a simple donor-acceptor organic compound, as active and host material in the emission layer of organic light emitting devices. Em1 has been used as active components in OLEDs, where Em1 is the guest emitter (Device A), the acceptor part of the excited emitter (Device B) and the host matrix of the CdSeS/ZnS alloy quantum dot. At least four different OLEDs have been designed and characterized where Em1 plays the role of the guest emitter (Device C). The external quantum efficiencies of devices A–C are characterized by values common to pure fluorescent OLEDs (up to 5 % of the theoretical limit), but these devices sustain low-efficiency roll-off of electroluminescence over a wide range of current densities.

Organic light-emitting diodes based on carbazole-derived compounds, due to their color characteristics, are promising candidates for use in the latest lighting systems. A separate advantage of the data light-emitting structures is a multifunctional application of one compound for different types of light-emitting structures. In addition, organic LEDs on based on carbazole-derived compounds have low energy consumption and are environmentally friendly due to the absence of toxic substances in their architecture, which creates prerequisites for saving energy resources and reducing the industrial burden on the environment.

Keywords: *OLED, exciplex, carbazole-derived compounds, thermally activated delayed fluorescence, CdSeS/ZnS core-shell quantum dots.*

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1. Introduction

Organic light emitting diodes (OLEDs) have great potential in modern displays and organic lighting due to their low power consumption, flexibility, high color quality and potential low cost [1, 2]. OLED technology has been successfully commercialized thanks to significant advances in the synthesis and research of organic small molecule and polymer semiconductors, as well as the design and engineering of efficient device architectures. Many types of light-emitting materials, including fluorescence [3], thermally activated delayed fluorescence (TADF) [4], phosphorescence [5] have been developed to provide high electroluminescence quantum yields due to high intrinsic efficiency, long-term stability, high electrical conductivity and among many other advantages [6], have led to their use as active materials in OLEDs. However, all these elements require chemical diversity according to the structure of the emitter. On the other hand, a unified structural motif is highly desirable to realize multifunctional OLED materials.

For the production of white OLEDs and full-color displays, there is an urgent need to develop materials that emit blue, one of the three primary colors of RGB, in a highly efficient and stable manner [7]. The ideal blue-emitting material has International Commission on Illumination (CIE) coordinates (0.14, 0.08) and is specified in the NTSC (National Television System Committee) standard. However, this is currently far from satisfactory for most blue emitters [8]. Deep blue is arbitrarily defined as having blue electroluminescent emission with (CIE) x, y coordinates x~0.15, y<0.15. So far only a few dark blue emitters with satisfactory chromaticity coordinates and corresponding highly efficient and stable OLEDs have been reported [9].

The problem lies both in the wide bandgap of high-energy blue OLEDs and in the often-unstable mobility of electrons and holes due to their mismatched energy level positions in organic emitter heterostructures. A comprehensive solution to these problems can be achieved by developing high-performance blue emitters with a narrow emission spectral distribution, a small full width at half maximum (fwhm), and a donor-acceptor (D-A) structure capable of carrying both electrons and holes. Such strategies can improve fluorescence efficiency, but may result in a bathochromic effect in the emission spectrum.

In this paper, the AIE-active D-A compound (Em1) [10] and the emission mechanisms occurring in quantum dot-based OLEDs (QLEDs) are investigated.

The aim of this research is to study the multifunctionality of the Em1 compound in organic light-emitting devices and to identification of the mechanism of intramolecular (Em1 compounds) and intermolecular energy transfer between the donor and acceptor (exiplex) compounds. In addition, the transfer of energy from an organic compound to inorganic quantum dots is study. This would indicate that efficient Forster resonance energy transfer (FRET) can take place when Em1 is doped with CdSeS/ZnS alloy quantum dots. Therefore, the possibility of using Em1 as the host component of guest CdSeS/ZnS core-shell quantum dots in the fabrication of the corresponding QLEDs will be found.

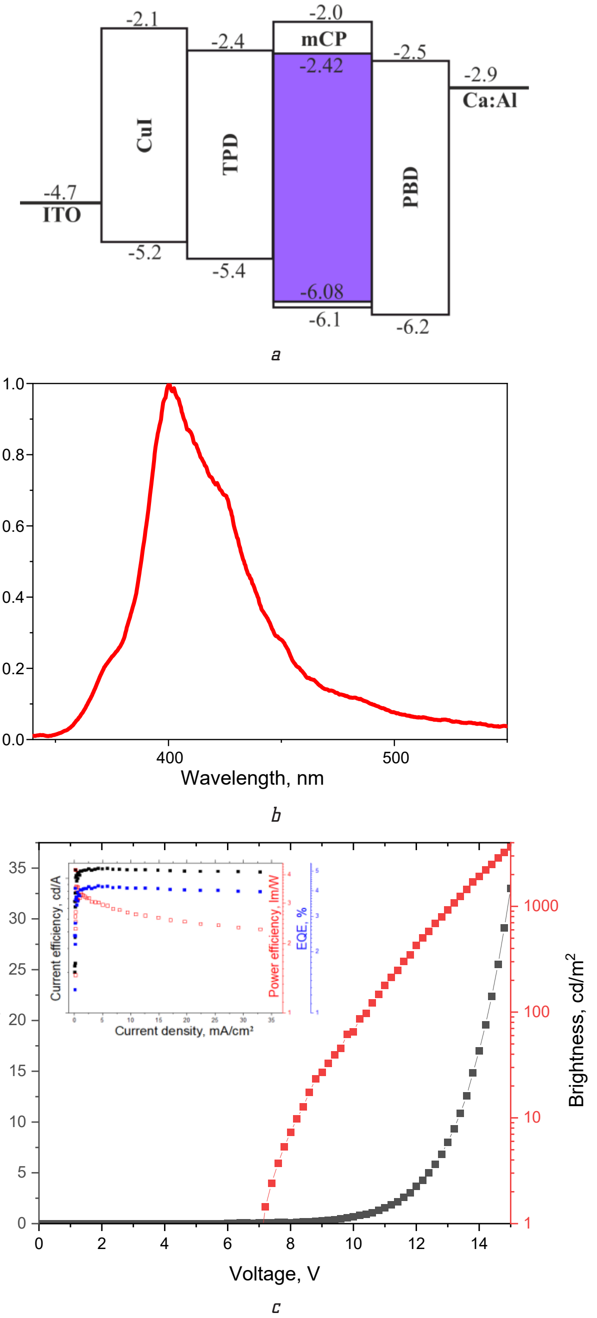
2. Materials and Methods

The electroluminescence properties of Em1 were investigated using conventional OLED structures with emitters doped into the mCP matrix. The device structure and energy diagram are shown in Fig. 1, *a*. The general scheme and layer thicknesses of the fabricated OLEDs are as follows: ITO/CuI (8 nm)/TPD (40 nm)/mCP:Em1 (20 nm, 7 %)/PBD (40 nm)/Ca (50 nm)/Al (200 nm) (device A). The devices were fabricated by gradual deposition of hole and electron transport layers on a pre-cleaned ITO-coated glass substrate under a vacuum of 10^{-5} Torr, co-deposition of the host and dopant layers, and deposition of a metal electrode. ITO was used as the anode material. CuI and N,N′-bis(3-methylphenyl)-N,N′-diphenylbenzidine (TPD) were used to prepare the hole transport layer (HTL). 2-(4-biphenyl)-5-phenyl-1,3,4-oxadiazole (PBD) was used to deposit the electron transport layer (ETL) [11]. HTL and ETL were chosen to confine holes and electrons in the emitting layer, which helps to optimize the performance of OLED. The active area of the resulting devices was 6 mm^2.

The electroluminescence properties of Device B based on stimulated (exciplex) emission from a mixture of Em1 and mMTDATA were studied using the following OLED architecture: ITO/CuI (8 nm)/m-MTDATA (20 nm): Em1 (20 nm)/TSPO1 (10 nm)/TPBI (30 nm)/Ca (50 nm)/Al (200 nm). The energy diagram of device B is shown in Fig. 2. m-MTDATA served as an efficient HTL material and a donor component of the exciplex emission mixture. Thus, Em1 acted as an acceptor for the molecular mixture forming the exciplex. Diphenylphosphine oxide-4-(triphenylsilyl) phenyl (TSPO1) was used as the electron blocking (EBL) material, while 2,2″,2′-(1,3,5-benzinitrile)-tris(1-phenyl-1H-benzimidazole) (TPBi) was used to prepare the ETL layer providing electrons to the active site of device B. Note that the deposition of the donor and acceptor components of the exciplex formation mixture was performed simultaneously from two crucibles with a component mass ratio of 1:1.

Quantum dot (QD)-based LEDs (referred to as QLED or Device C) had the following structure (Fig. 3, *a*): ITO/CuI (8 nm)/Em1:CdSeS/ZnS (wt %, QD = 15 %) (40 nm)/TSPO1 (10 nm)/TPBI (40 nm)/Ca/Al. CuI [12] and TPBI [13] were used as HTL and ETL materials, respectively; the hole blocking layer of TSPO1 [14].

The base materials, Em1 and CdSeS/ZnS QDs, were dispersed in toluene at a ratio of 1 mg per mL. A solution with a spreader concentration of 15 % and a matrix concentration of 85 % was prepared; a layer of a mixture of C Em1 and CdSeS/ZnSCdSeS/ZnS was cast from the dispersion in toluene onto the surface of an ITO substrate with a pre-formed CuI film by centrifugation at 1000 rpm and then annealed at 50 °C.

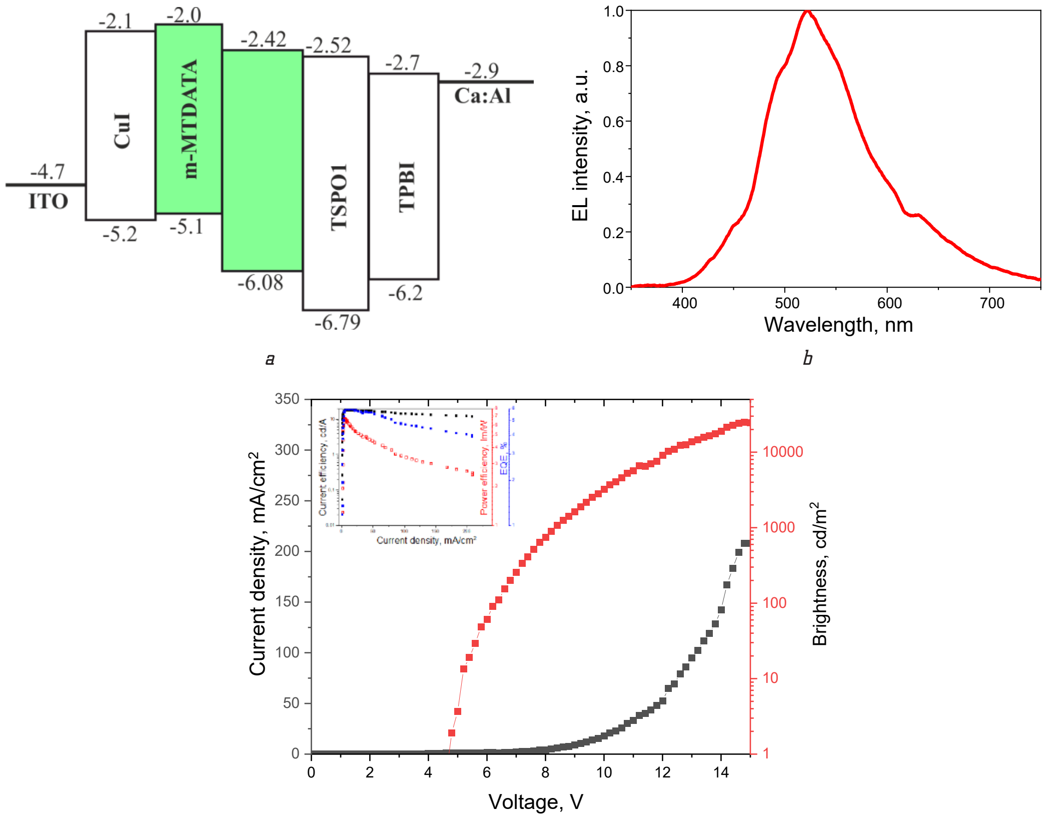


a

b

c

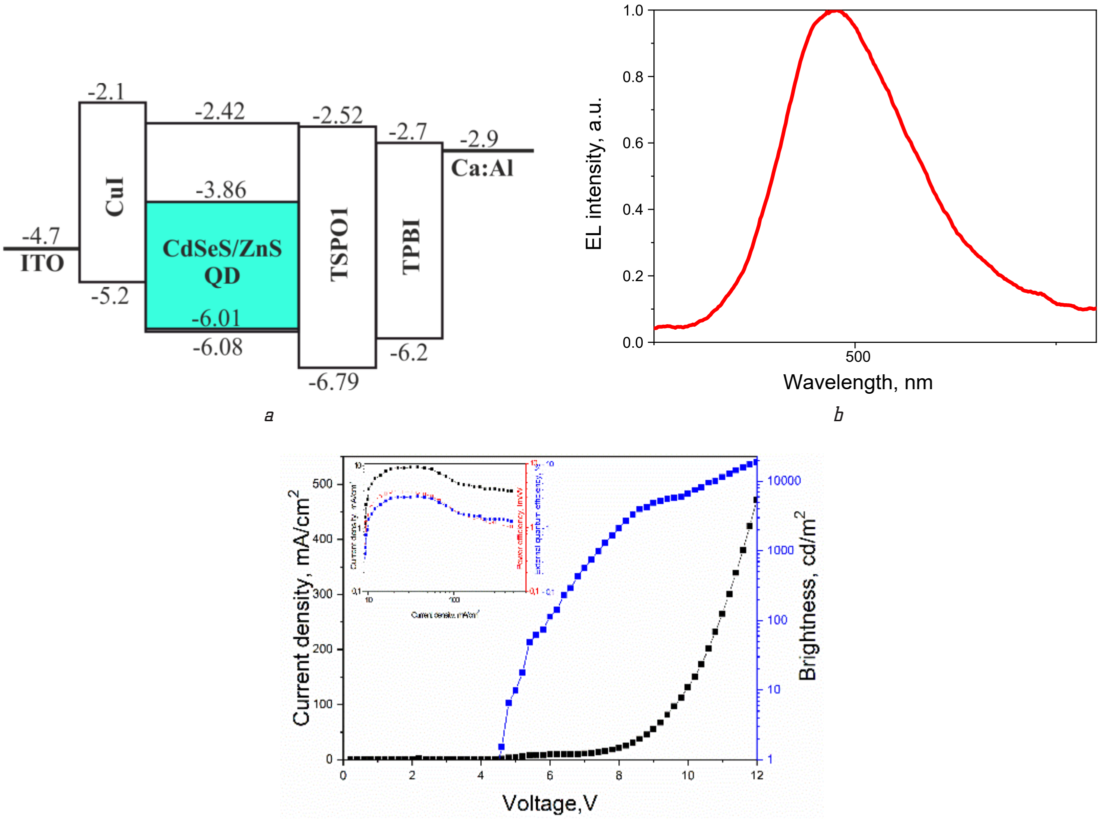
Fig. 1. Diagrams of device A: *a* – energy level; *b* – the corresponding normalized electroluminescence spectrum as well as brightness/current density-voltage; *c* – current efficiency/power efficiency/EQE-current density characteristics



a *b*

c

Fig. 2. Diagrams of device B: *a* – energy level; *b* – the corresponding normalized electroluminescence spectrum as well as brightness/current density-voltage; *c* – current efficiency/power efficiency/EQE-current density characteristics



a *b*

c

Fig. 3. Diagrams of device C: *a* – energy level; *b* – the corresponding normalized electroluminescence spectrum as well as brightness/current density-voltage; *c* – current efficiency/power efficiency/EQE-current density characteristics

3. Results and Discussions

3.1. Fabrication and characterization of host-containing OLEDs. The guest host layer was formed by thermal vacuum co-deposition of mCP with a fluorescence compound Em1 (guest) concentration of 7 wt %. mCP was chosen as the host material because its high singlet excited state (3.6 eV) provides a wide bandgap blue light for efficient FRET. This is due to its good match with OLED emitters (Fig. 1, *a*) [15].

When a bias voltage of 7.4 V was applied to the organic heterostructure, the brightness of device A was 10 cd/m^2 (Fig. 1, *b*). Although this bias voltage is significant, it can be reduced by adding additional electron and hole blocking layers. Device A showed the highest EL efficiency (4.2 % (11.5 cd·A^{-1})) at a current density of 12.5 mA·cm^{-2}. It should be noted that this value is close to the theoretical maximum EQE limit of 5 % expected for fluorescence-based OLEDs [13, 15]. The current, power efficiency and external quantum efficiency characteristics of Device A were stable over a wide range of current density values (Fig. 1, *c*). The maximum luminance was 3600 cd/m^2 at 15 V voltage (Table 1).

Table 1

Characteristics of OLED devices A-C

Device	Von, V	Brightness, cd/m^2	Current efficiency, cd/A	Power efficiency, Lm/W
A	6.8	3600	11.5	3.8
B	4.7	24700	12.0	6.8
C	4.5	18700	9.6	4.6

The electroluminescence spectrum of device A (Fig. 1, *b*) was stable over a wide range of applied voltages; the spectral maximum of the OLED was 413 nm. This observation is in good agreement with the wavelength of the PL intensity maximum of the mCP and Em1 blend film. The OLED exhibited a narrow (fwhm 45 nm) EL spectrum in the range 387–432 nm (Fig. 1, *b*), with no green emission at CIE coordinates (x=0.15, y=0.13) close to dark blue.

3.2. Fabrication and characterization of an exciplex OLED. The exciplex-forming properties of this compound were studied. Exciplexes are usually characterized by a small energy gap between the singlet and triplet ($S1$, $T1$) states of charge transfer (CT) [14]. These states therefore exist in thermal equilibrium at room temperature due to the rapid intersystem transition (ISC) and inverse ICS process. Relaxation to the ground state occurs via a radiative $S1 \rightarrow S0$ transition and the corresponding long-lived luminescent components have TADF properties. This results in the external quantum efficiency (EQE) of m-MTDATA-doped OLED-based Em1 being higher than the 5 % limit for electrofluorescent devices (Fig. 2, *c*, Table 1).

This device B has an electroluminescence spectrum that differs from the photoluminescence spectrum of the Em1 compound. Electrons injected from the cathode overcome an energy barrier of 0.42 eV as they drift from the LUMO level of Em1 (acceptor) to the LUMO level of m-MTDATA (Fig. 2, *a*). Holes injected from the HOMO level of m-MTDATA (donor) to the HOMO level of Em1 have a barrier of 1 eV. Due to this high energy barrier, electrons and holes accumulate in the mixture of Em1 and m-MTDATA. As a result, cross-coupling occurs between electrons at the LUMO of Em1 and holes at the HOMO of m-MTDATA, and the corresponding charge transfer state is responsible for the electroluminescence in device B. The weak shoulder around 450 nm in the electroluminescence spectrum of device B corresponds to the pure exciton emission of m-MTDATA (Fig. 2, *b*); the remaining EL spectrum of device B clearly overlaps with the photoluminescence spectrum of the mixtures of Em1 and m-MTDATA with a maximum around 520 nm (Fig. 2, *b* and Fig. 4).

The exciplex emission of the molecular mixture of Em1 and m-MTDATA films shows a shift of the emission spectrum towards the long wavelength side (Fig. 4); in the case of TADF exciplexes, a small singlet-singlet triplet splitting (ΔEST) is typically observed [15]. To confirm this requirement in mixtures of Em1 and m-MTDATA, PL and phosphorescence spectra of Em1:m-MTDATA were recorded a delay of 5 μs (Fig. 4, inset). Finally, the corresponding PL decay curves reached the millisecond range (Fig. 4, inset), confirming the TADF nature of the photoluminescence due to the long-lived emission decay component of the blended Em1 and m-MTDATA films. According to double exponential fitting (χ_2 is 1.249), the PL decay curves of Em1 and m-MTDATA molecular blend films showed a luminescence lifetime of τ_1 21.3 ns (44 %) and τ_2 98.4 ns (56 %) at room temperature.

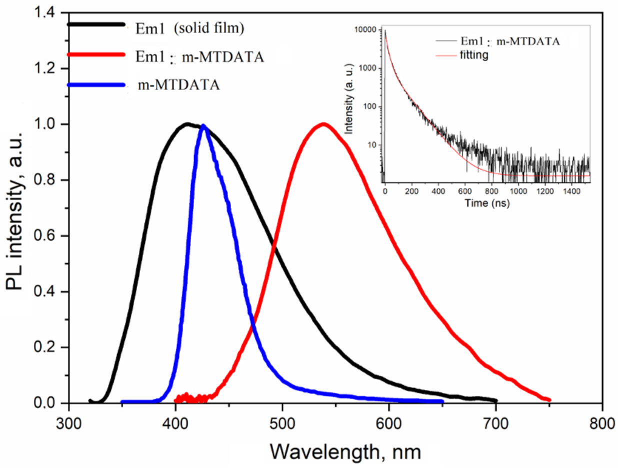


Fig. 4. PL spectra of vacuum-deposited films of m-MTDATA, Em1, and the blend of Em1 and m-MTDATA and PL decay curves (inside)

Long emission lifetimes such as immediate fluorescence and short emission lifetimes such as delayed fluorescence are typical for conventional TADF exciplexes [16, 17].

The brightness of device B was 10 cd/m^2 at a bias voltage of 4.2 V. The external quantum efficiency of this OLED was 5.3 %, slightly above the theoretical limit for fluorescent OLEDs (Table 1). Similar to Device A, the external quantum efficiency and power efficiency of Device B showed relatively stable characteristics over a wide range of current density values (Fig. 2, *c* (inset)). A maximum current efficiency value of 12 cd/A and a maximum luminance of 24700 cd/m^2 were achieved at an applied voltage of 15 V (Fig. 2, *c*). The CIE coordinates of the device (x=0.22, y=0.46) were also very stable under different voltages.

3.3. Fabrication and characterization of the QLED. The study of the functional application of the Em1 compound as a host for inorganic nanoparticles was conducted. The developed QLED architecture is suitable for the deposition of electron-hole pairs in the active layer of Em1 and CdSeS/ZnS mixtures (Fig. 3, *a*). This is due to the small energy barrier (0.1 eV) between the LUMO energy level of the host material and the electron transport material (TSPO1) and the deep HOMO level of TSPO1 (about 6.79 eV), which facilitates the injection of electrons from the host material and the transport of holes to the cathode. The main reasons for choosing CuI as the hole-injecting layer are its –5.2 eV valence band, high conductivity of 0.01^{-1} S/cm and high hole mobility of 20 $\text{cm}^2\ \text{V}^{-1}\text{s}^{-1}$ [18, 19].

The current density-voltage and voltage-intensity curves of QLEDs are shown in Fig. 3, *c*. Device C exhibited a maximum luminance of 18700 cd/m^2 at 12 V (Fig. 3, *c*). The chromaticity coordinates of device C ($x=0.24$, $y=0.37$) are consistent with the PL coordinates of CdSeS/ZnS quantum dots; the QLED showed maximum EL intensity at 491 nm (Fig. 3, *b*). In the current density range of 20–50 mA/cm^2, the maximum external quantum efficiency of the QLEDs is about 2.3 %, decreasing slightly to 1.08 % at 100 mA/cm^2 (Fig. 3, *c* (inset), Table 1).

3.4. Discussion. In recent years, OLED manufacturing technology has reached the stage of practical application, and methods to optimize emission color quality continue to evolve, creating new device architecture approaches to improve energy efficiency and color quality. In this context, the use of OLED technology with it is simplified structure is extremely promising.

The devices exhibit efficient emission characteristics and high color stability, making them very useful for use in modern lighting systems and display technology.

The main limitation of the applicability of the results obtained is the high cost of introducing lighting systems based on the studied compounds into mass production.

Also, the gradual degradation of light-emitting layers during operation leads to a decrease in efficiency and changes in spectral characteristics.

It should be noted that for the further introduction of the obtained OLEDs as key elements of modern lighting systems, complex work needs to be carried out.

In particular, it is necessary to fulfill such complex tasks as the development of the technology of thermal vacuum application of functional light-emitting structures at industrial facilities and obtaining large-area OLED matrices.

Power outages caused by the war in Ukraine may have affected the production technology of light-emitting devices and led to the deterioration of the output parameters of OLEDs.

4. Conclusions

This paper reports the multifunctional application of a simple donor-acceptor organic compound, as active and host material in the emission layer of organic light emitting devices. Em1 has been used as active components in OLEDs, where Em1 is the guest emitter (Device A), the acceptor part of the excited emitter (Device B) and the host matrix of the CdSeS/ZnS alloy quantum dot (at least four different OLEDs have been designed and characterized where Em1 plays the role of the guest emitter (Device C). The external quantum efficiencies of devices A-C are characterized by values common to pure fluorescent OLEDs (up to 5 % of the theoretical limit), but these devices sustain low-efficiency roll-off of electroluminescence over a wide range of current densities. The Em1 compound has intrinsic limitations in photoluminescence efficiency due to the twisted charge transfer nature of the geometry of its fluorescent state. Therefore, in order to increase the luminescence quantum yield of Em1 and improve the illumination properties of devices using Em1 as a luminescence device, it is a priority to prohibit mutual rotation of the donor and acceptor parts of Em1 due to the chemical tuning and structural constraints that occur in the solid state.

Conflict of interest

The authors declare that they have no conflict of interest concerning this research, whether financial, personal, authorship or otherwise, that could affect the study and its results presented in this paper.

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Data availability

The paper has no associated data.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating this work.

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