

Thermally activated delayed fluorescence - as a solution to the low yield problem of the fluorescent OLEDs

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The development of display technologies has begun with the invention of the cinematograph; whereas a next important step was the cathode ray tube. CRT screens reached a high degree of development at the turn of the century, but they have been virtually superseded today by the liquid crystal displays (LCD). Another important step was made when organic LED screens (OLEDs) were introduced, in which only selected pixels are illuminated. The commercial OLED screens are already in production, and interest in the development of this technology is observed by leading electronic companies [1]. It should also be noted that OLED devices can be also successfully used for lighting purposes. [1 ÷ 6]

There are two approaches in design of OLEDs: devices based on small molecules, so-called SMOLED (*Small Molecule Organic Light-Emitting Diode*) or based on polymers, so-called PLED (*Polymer Light-Emitting Diode*) [3, 4, 7, 8]. Conjugated polymers are commonly employed as emitters in polymer LEDs [9, 10]. Decades ago the effects of light on conducting polymers was studied [11, 12], also then the first SMOLED was produced [13]. Since that times an incredible development of organic electronics has been achieved [4, 7]. However, in this article only those solutions that use small molecules will be described.

OLEDs work similarly to classical LEDs where the formation of photons (production of light) is due to recombination of charge carriers: electron (negative) and hole (positive), that are injected by adequate electrodes. Excitation of the emitter molecules, due to recombination of charge carriers, leads to an excited state which can be called an exciton (or optionally exciplex or even excimer, electroplex, or electromer depending on the composition of the light-emitting layer and the processes that occur). Exciton is an unimolecular state, whereas others are bimolecular states. Exciplex and electroplex are composed of two different molecules – donor and acceptor, excimer and electromer are formed by two identical molecules [14, 15]. Regardless of the origin of the excited state there are two possible configurations of the electron spin: singlet (S) with paired electrons and triplet (T) with unpaired electrons. It has been shown that when low molecular weight substances are used as OLED emitters, always 25% of singlet states (S_1) and 75% of triplet states (T_1) are formed [7, 16]. Conventional fluorescent OLED emitters use only singlet states (fluorescence) whereas triplet states (phosphorescence) are somehow wasted, since the yield of the phosphorescence of such a compound is typically negligibly small at room temperature. As a result EQE (*External Quantum Efficiency*) of these electroluminescent diodes does not exceed 5%. EQE is basically a number of photons that escape the OLED per any 100 electrons that are injected into the device [7].

One solution for the problem of low quantum efficiency of the diodes is to use phosphorescent emitters, which due to a strong spin-orbit coupling allow OLED to utilize up to 100% of the excited states in the form of efficient phosphorescence. In a phosphorescent emitter the singlet excited states are effectively converted to triplet states and the quantum yield of phosphorescence can reach 100% [17, 18]. These substances have a number of drawbacks, including a presence of expensive precious metals (usually iridium or platinum) and a low stability of both OLEDs and the pure compounds [19, 20]. A relatively new solution, which can compete with phosphorescent emitters are TADF (*Thermally Activated Delayed Fluorescence*) emitters [21, 22], because both enable to obtain OLEDs with EQE \approx 20% [17, 21]. TADF enables harvesting of both singlet and triplet excited states of the molecule in form of efficient fluorescence. Here the reverse intersystem crossing $T_1 \rightarrow S_1$ occurs, and due to a low singlet-triplet energy barrier, the required energy can be drawn from environment as a heat.

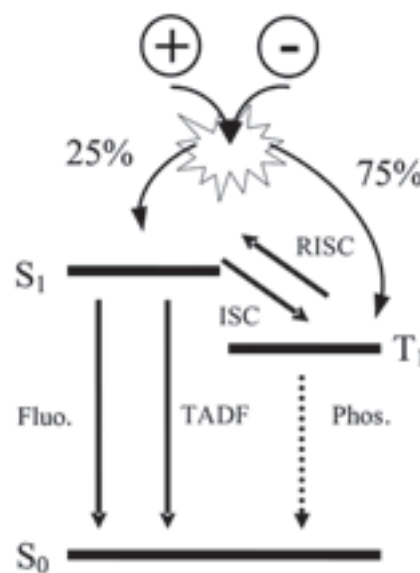


Fig. 1. Schematic diagram of TADF process in an OLED

Anihilation of a hole (+) and an electron (-) in an OLED leads to formation of excited states. Statistically 25% of singlet and 75% of triplet states are formed. TADF allows to recycle the triplet excited states through efficient fluorescence which would be wasted for an inefficient phosphorescence otherwise. S_0 – ground state; S_1 – the lowest singlet excited state; T_1 – the lowest triplet excited state; Fluo. – prompt fluorescence; TADF – thermally activated delayed fluorescence; Phos. – phosphorescence; ISC – intersystem crossing; RISC – reverse intersystem crossing.

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The use of TADF emitters in OLED technology was proposed by Professor Chihaya Adachi and his group from Japan [21, 23]. Currently, other research teams deal with this issue, and among others, Professor Andrew Monkman's group from Durham University in the UK [15, 24, 25]. Also Professor Mieczyslaw Lapkowski's group from Faculty of Chemistry, Silesian University of Technology undertakes research in the area in cooperation with Durham University [26, 27].

TADF emitter is usually a donor-acceptor molecule (exciton emitter) or a complex of two different molecules that is formed when one molecule is excited (exciplex emitter) [15, 21, 22, 24, 25]. Currently exciton and exciplex excited states seem to be the most common states utilized in OLED technology.

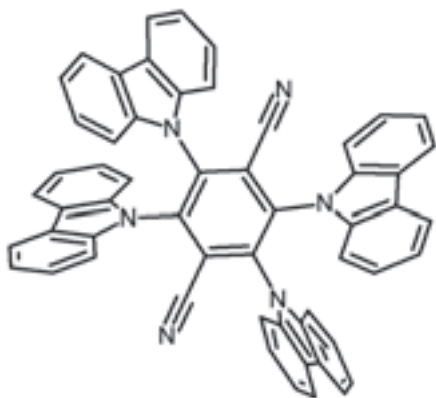


Fig. 2. Simulated geometrical configuration of a representative TADF emitter; donors (carbazole) and acceptor (dicyanobenzene) are not coplanar [21]

Exciton (donor-acceptor) emitter should be used as an example to explain TADF phenomenon. Donors of a TADF are mostly carbazole, phenoxazine, phenothiazine, diphenylamine, and their derivatives. They are connected with the rest of the molecule usually through a nitrogen atom [21, 22, 24, 28÷31]. Emitters reported in literature possess variety of different acceptors. Typically there is one π -conjugated acceptor unit in the molecule and one or more donor units. The main strategy in designing such an emitter is to provide a small integral overlap between HOMO and LUMO – the frontier molecular orbitals. Most molecules are obtained by a design of donor and acceptor aromatic systems which are twisted one to another due to a steric hindrance, which makes conjugation between them difficult, but does not break it completely. As an effect a partial or complete separation of the HOMO and LUMO is obtained, what enables to lower the singlet-triplet energy gap. Therefore the designed molecule can have the S_1 and T_1 states energetically close to each other, which means that difference in energy between them is $\Delta E_{S,T} < 0.5$ eV. The 0.5 eV threshold is an approximate based on the fact that no TADF emitters with higher $\Delta E_{S,T}$ have been reported. In practice, lower $\Delta E_{S,T}$ is better – usually it is ranging from few tens of eV to few hundredths depending on the structure of the molecule. Lower $\Delta E_{S,T}$ increases the rate constant of singlet-triplet reverse intersystem crossing (RISC), which is usually preferred. Due to donor-acceptor structure of a typical TADF emitter charge transfer (CT) state can be formed which has a very low singlet-triplet energy gap [15, 24]. Due to strong contribution of the CT state in exciplex, the lowest value of $\Delta E_{S,T}$ was found [15] for that state, which is approximately 5 meV [21÷25, 28÷31].

The structure of the excited states of TADF emitter is important, because besides $n\pi^*$ and $\pi\pi^*$ states, which are sometimes called locally excited states (LE) due to localization of the excited state only on the part of the molecule (e.g. donor), there is a significant charge-transfer (CT) state present, which is very important in TADF phenomenon (please see below) [24, 29]. Energy of the CT state depends on a polarity of the molecule surrounding. Therefore, with

the increasing polarity of the solvent a bathochromic fluorescence shift in the compound spectrum is observed [24, 32]. The CT state has a very small and difficult to measure $\Delta E_{S,T}$ value, which is its characteristic feature [15, 24]. Depending on a polarity of the environment, energy of the CT state can be located above or below 1LE state. In the second case this leads to a decrease of the $\Delta E_{S,T}$ value, because the CT state becomes the lowest singlet state (S_1) [29]. To obtain OLEDs of satisfying parameters (eg. high EQE) the emitter particles are dispersed in a matrix of another material, which, among others, ensures a good transport of charge carriers [7, 21, 28÷30]. This matrix has a sufficient polarity to reduce energy of the CT state in most cases and thus to reduce the singlet-triplet gap, which is usually preferred.

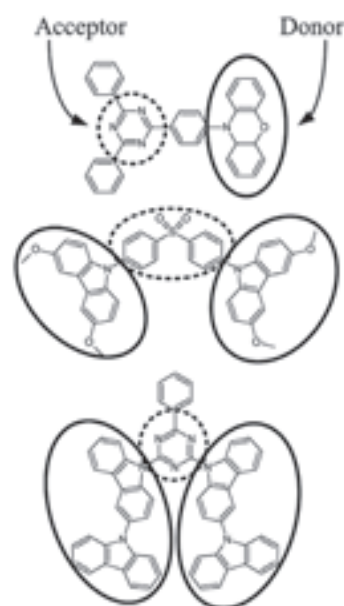


Fig. 3. Chemical structures of the selected TADF emitters. The donor and acceptor units are indicated by solid and dotted lines, respectively [22,28,30]

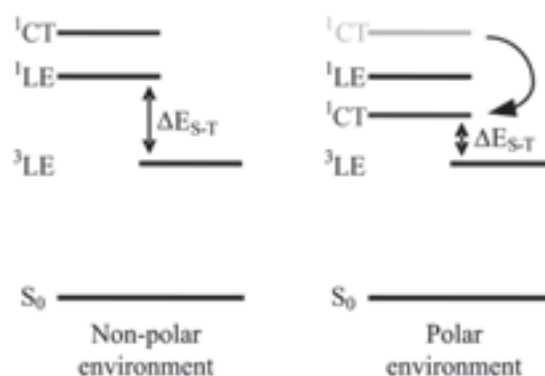


Fig. 4. Example of a typical behavior of electron excited states of a donor-acceptor TADF emitter

The charge transfer (CT) excited state of a TADF emitter is stabilized by polar environment, therefore its energy is lower there than in a non-polar surrounding. As a result the 1CT state may become the lowest singlet excited state of the molecule; 1CT – singlet charge transfer state; 1LE – singlet locally excited state; 3LE – triplet locally excited state; S_0 – ground state $\Delta E_{S,T}$ – singlet-triplet energy gap. CT triplet state was not shown due to negligibly small $^1CT - ^3CT$ energy difference.

The delayed fluorescence differs from a prompt fluorescence in the fluorescence lifetime: a feature of the delayed fluorescence is to have long micro- or millisecond lifetime, whereas prompt fluorescence has

typically nanosecond lifetime [21 ÷ 25]. This is a consequence of TADF mechanism, which includes rate-limiting forbidden $T_1 \rightarrow S_1$ transition. Both intensity and lifetime of the delayed fluorescence depend on temperature – the intensity is higher at higher temperature – and at sufficiently low temperature TADF disappears completely [24]. When TADF was discovered it was found that this process follows Arrhenius equation (1) [33]. Activation energy values calculated from the equation (1) are generally close to $\Delta E_{S,T}$ [24]. Currently many other equations are in use, however all are based on the Arrhenius equation [21, 24].

$$\frac{\varphi_{DF}}{\varphi_P} = c \cdot e^{-\frac{\Delta E}{RT}} \quad (1)$$

where: φ_{DF} – delayed fluorescence (TADF) quantum yield; φ_P – phosphorescence quantum yield; c – pre-exponential factor; R – gas constant, $8.314 \text{ J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$; T – temperature K; ΔE – TADF activation energy $\text{J}\cdot\text{mol}^{-1}$ [33]

Molecules that possess relatively long-lived triplet state which means that it is not strongly affected by non-radiative deactivation, but they have too wide $\Delta E_{S,T}$ gap may show a different kind of delayed fluorescence, TTA (*Triplet-Triplet Annihilation*). The distinction between TADF (E-type fluorescence) and TTA (P-type fluorescence) consists of examination of the kinetics of both processes in the way of how intensity of delayed fluorescence depends on excitation beam intensity. TADF has linear, proportional dependence of delayed fluorescence intensity upon excitation intensity (the process is unimolecular), whereas TTA has quadratic dependence (bimolecular process) [24, 25]. TTA requires a direct interaction of two molecules in a triplet state to form one singlet excited state and one ground state, whereas TADF involves only one molecule.

TADF emitters are now a hot topic due to possibility of obtaining OLEDs with high EQE. TADF emitters which are typical organic molecules composed of elements such as C, H, N, O, S, B are cheaper in the synthesis than phosphorescent emitters, because they do not contain iridium or platinum. There is still a problem in obtaining substances with red fluorescence as most of reported efficient TADF emitters has blue-greenish and green fluorescence. Taking into account that not only charge-transfer compounds can be TADF emitters [31, 34], it can be assumed that as time goes by the TADF phenomenon might be found in many substances that have not yet been examined from this point.

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Aktualności z firm

News from the Companies

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OCHRONA ŚRODOWISKA

ORLEN Południe w programie „Odpowiedzialność i Troska”

ORLEN Południe, w ramach aktywnej polityki dążącej do minimalizacji oddziaływania spółki na otoczenie, podpisało deklarację dobrowolnego przystąpienia do programu „Odpowiedzialność i Troska” (Responsible Care). Poświadczaniem spełnienia wszystkich wymagań jest przyznanie ORLEN Południe przez kapitułę prawa do używania nazwy i logo programu. Poprzez przystąpienie do inicjatywy spółka z Grupy ORLEN zobowiązała się do podjęcia szeregu zadań na rzecz ciągłego zmniejszania uciążliwości dla środowiska naturalnego, dalszej, konsekwentnej poprawy bezpieczeństwa i ochrony zdrowia pracowników. Jednocześnie, przynależność do międzynarodowej inicjatywy Responsible Care, realizowanej obecnie w blisko 60. krajach na świecie, daje ORLEN Południe możliwość wymiany doświadczeń oraz zdobywania wiedzy na wielu płaszczyznach funkcjonowania organizacji biznesowych. ORLEN Południe to już piąta spółka z Grupy ORLEN, która zakwalifikowała się do programu. (kk)

(<http://www.orklenpoludnie.pl/>, 11.05.2016)

NAGRODY, WYRÓŻNIENIA

Order Orła Białego przyznany Prof. Michałowi Kleiberowi

3 maja 2016 r., z okazji Narodowego Świąta 3 Maja, na Zamku Królewskim w Warszawie Prezydent Rzeczypospolitej Polskiej Andrzej Duda wręczył odznaczenia państwowe osobom zasłużonym w służbie państwu i społeczeństwu. Między innymi Orderem Orła Białego został odznaczony prof. Michał Kleiber, członek rzeczywisty PAN, Prezes Polskiej Akademii Nauk w latach 2007–2015. Prof. Michał Kleiber został uhonorowany przez Prezydenta Andrzeja Dudę „w uznaniu znamienitych zasług dla rozwoju polskiej nauki, za wybitne osiągnięcia w pracy publicznej i państwowej”. Prezydent RP dziękował Profesorowi za lata przewodniczenia Polskiej Akademii Nauk i reprezentowania Polski w wielu instytucjach naukowych oraz przynoszenie „swoją postawą chluby Rzeczypospolitej”. Najwyższe państwowe odznaczenie otrzymali także: Irena Kirszenstein-Szewińska, Michał Lorenc, Wanda Półtawska, Zofia Romaszewska i Bronisław Wildstein. (kk)

(<http://www.aktualnosci.pan.pl/>, 3.05.2016)

Dynamiczny rozwój LERG wyróżniony tytułem „Diamenty Forbesa” 2016

LERG znalazł się w gronie laureatów prestiżowego rankingu miesięcznika Forbes „Diamenty Forbesa” 2016. Jest to ranking firm,

które w trzech ostatnich latach najbardziej dynamicznie zwiększały swoją wartość. LERG zajął wysokie 4 miejsce w zestawieniu przedsiębiorstw z województwa podkarpackiego o przychodach przekraczających 250 mln PLN. (kk)

(<http://www.lerg.pl/>, 27.04.2016)

PGE na podium w zestawieniu 200. największych polskich firm

PGE Polska Grupa Energetyczna znalazła się na 3. miejscu opublikowanej przez tygodnik „Wprost” „Listy 200 największych polskich firm”. Tym samym awansowała o jedną pozycję w stosunku do ubiegłorocznej edycji rankingu. (kk)

(<http://www.gkpgge.pl/>, 24.05.2016)

NCBR dofinansuje demonstratory nowych technologii

Narodowe Centrum Badań i Rozwoju ogłosiło wyniki drugiego konkursu w programie „Demonstrator”. Na wsparcie 30. projektów w zakresie prac rozwojowych, które zwieńczone zostaną stworzeniem demonstracyjnych wersji nowych produktów i technologii, NCBR przeznaczy ponad 290 mln PLN. Dzięki wsparciu NCBR będą prowadzone eksperymentalne prace badawczo-rozwojowe, których efektem finalnym będą m.in.: zrobotyzowana uniwersalna linia technologiczna do konfekcjonowania produktów; technologia temperaturowego łączenia tworzyw sztucznych z uwzględnieniem nowych materiałów; e-system wspierający diagnostykę i prowadzenie chorych na padaczkę czy rafinowany, ultraczysty żelazokrzemochrom o kontrolowanej zawartości węgla, azotu, tlenu i wodoru. (kk)

(<http://www.ncbir.pl/>, 13.05.2016)

250 mln PLN na komercjalizację wyników badań

Ministerstwo Nauki i Szkolnictwa Wyższego oraz Narodowe Centrum Badań i Rozwoju ogłaszają wyniki konkursu na dofinansowanie kosztów utrzymania infrastruktury badawczej, służącej komercjalizacji prac B+R. 16 ośrodków badawczych w ciągu pięciu lat otrzyma na ten cel nawet 250 mln PLN. Wnioski w konkursie złożyło 16 ośrodków posiadających infrastrukturę o wartości co najmniej 50 mln PLN, powstałą w ramach Programu Operacyjnego Innowacyjna Gospodarka. Dzięki dobrze przygotowanym aplikacjom, wszystkie projekty otrzymają wsparcie NCBR na prowadzenie badań dla podmiotów zewnętrznych. Na liście beneficjentów konkursu znalazły się m.in. Instytut Chemii Bioorganicznej PAN, Politechnika Warszawska, a także Wrocławskie Centrum Badań EIT+. (kk)

(<http://www.nauka.gov.pl/>, 20.05.2016)

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