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Electronic and thermal properties of compounds bearing diimide, azomethine and triphenylamine units



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ABSTRACT

New triphenylamine containing azomethine diimides and two kinds of poly(azomethine imide)s, i.e., linear and branched were synthesized. These compounds were prepared from two diamines, that is, N,N'-bis(4-amino-2,3,5,6-tetramethylphenyl)phtalene-1,2,4,5-dicarboximide (DAPhDI), N,N'-bis(5-aminonaphtalen)naphthalene-1,4,5,8-dicarboxyimide (DANDI-2) and 4-formyltriphenylamine, 4,4'-diformyltriphenylamine and 4,4'.4"-triformyltriphenylamine. The structures of the compounds were characterized by means of FTIR, ¹H NMR spectroscopy and elemental analysis; the results show an agreement with the proposed structure. Thermal properties of prepared azomethine diimides and polymers were evaluated by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). Obtained compounds exhibited high thermal stability with 5% weight-loss temperatures above 390 °C. Azomethine diimides exhibited glass-forming properties with high glass-transition temperatures 216 and 308 °C. Optical properties of the prepared compounds were investigated by UV-vis and photoluminescence (PL) measurements. All compounds emitted blue light in NMP solution and in solid state as blend with PMMA. The electrochemical properties, that is, orbital energies and resulting energy gap were estimated based on cyclic voltammetry (CV). All synthesized material showed reversible reduction process, furthermore AzPhDI and AzNDI showed partially reversible oxidation process. Electrochemical band gap was found in the range 1.23–1.70 eV. Low molecular weight model compounds were tested as bipolar host materials in blue phosphorescent organic light emitting diodes (OLEDs). The devices exhibited turn-on voltages of about 5.5 V and maximum brightness of 40-220 cd/m².

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1. Introduction

Organic light-emitting diodes (OLEDs), have found commercial application, including displays of mobile phones and TVs. However, there is still a need to improve their efficiency and, above all, to extend working time. Materials that can be used in OLEDs should have the following properties: luminescence minimum of 100 cd/m², a relatively low voltage (less than 10 V), good thermal stability and high glass transition temperature and the resistance to oxygen and water [1]. For full-color display or lighting, the device requires red, green, and blue emissions with relatively equal stability, efficiency, and color purity. However, the performance of saturated deep-blue OLEDs is rather poor as compared to those of green and red ones [2,3]. Thus many investigations are focused on

compounds emitting blue light, which exhibited thermal stability and are suitable for solution process. As blue light emitters mainly complexes of iridium(III) are tested [4]. Host materials for blue emitters play a very important role in determining the device performance [2]. Its task is maintaining a balance between electrons and holes, preventing aggregation of dopant and trapping excitons on the emitter dopant. Triphenylamine (TPA) and its derivatives are often used in OLEDs. Such compounds can be used as hole transporting materials (HTM) [5,6], host materials [2] and blue emitting materials [2]. Ding and Alameh [7] reported OLED devices where Alq₃ was emitting layer and showed that the presence as HTM of N,N'-di(naphthalene-1-yl)-N,N'-diphenylbenzidine) (NPB) reduces the operating voltage. Thangaraju and co-workers [8] reported yellowish-green OLED, where they used 4,4',4"-tris(carbazole-9-yl)triphenylamine (TcTa) as HTM and iridium(III) complex [Ir(msippy)₃] as emissive layer (EML). The drive voltages of 7.5 V and 10.5 V at 1000 cd/m^2 were reached. Shih and co-workers [9] synthesized tris[4-(9-phenylfluoren-9-yl)phenyl]amine (TFTPA)

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which was an excellent host material for two emitters. Blue emitting devices prepared of FIrpic and TFTPA showed 12% of efficiency, while red emitting OLED exhibited very low turn-on voltage (2.6 V). Wang et al. [3] reported 4,4'-(9,9,9",9"-tetraoctyl-terfluorene-9',9'-diyl)bis(N,N-diphenylaniline) (TFDPA) which was a multifunctional material. First fluorene group were responsible for the deep blue emission, and second, attached TPA improved holetransporting ability. TFDPA was used as emitting material in blue OLED devices with a maximum external quantum efficiency of 2.7%. Authors used TFDPA as host material different Ir complexes, to obtain two-color-based OLED emitted white light (WOLED). The highest EQE (9.3%) was observed for $Ir(2-phq)_3$ as dopant. We can find also red-emitting Ir(III) complexes incorporating hole-transporting TPA groups [10,11]. N,N'-di-1-naphthalenyl-N,N'-diphenyl-[1,1':4',1":4",1"'-quaterphenyl]-4,4"'-diamine (4P-NPD) was used as blue emitter in hybrid white OLED with a power efficiency at 1000 cd m⁻² of 22 lm W⁻¹ [2]. Pu et al. synthesized polymers containing TPA and oxadiazole units showing high electroluminescence efficiency [12]. Also a dendrimeric and starshaped compounds containing TPA were applied in OLED devices [13,14]. For example, small molecular star-shaped oligophenylenes, with triphenylamine in central position were electropolymerized to obtain 3D cross-linked films of the polymers, which was applied as hole-injection layers in OLEDs [14]. Iwan and Sek in their review [15] showed a large number of compounds containing triphenylamine units, which were applied in OLED devices. Among many discussed compounds we can find also polyazomethines, such as polymer obtained from 4,4'-diaminotriphenylamine and terephthalic aldehyde, which was applied, together with Alq₃ as emitting material, in green OLED [15,16]. Also polyazomethines synthesized from 4,4'-diformyltriphenylamine and aromatic diamines (benzene-1,4-diamine or 2,3,5,6-tetramethylbenzene-1,4diamine) were used as HTM in devices where emitting layer was Alq₃, and turning voltage was observed about 6 V [15,17].

Imides and polyimides, because of their excellent thermal and mechanical, as well donor properties also found an application in optoelectronics [18,19]. Dautel et al. [20] synthesized and applied in OLED devices, new chiral polyimide and model compound. Driving voltage for devices obtained from polymer was significantly lower (6 V) than for the device with emitting layer comprising a low molecular weight compound (20 V). Zhu et al. [21] reported new naphthalene imides consisting of carbazole units as emitting materials in single-layer devices [21]. They showed that it is possible to tuning electroluminescence, by a slight change in the structure. Hwang et al. in their review [13] showed emitting materials based on imides, for example dendrimers consisted of naphthalimide cores, poly(aryl ether) dendrons, and carbazole or oxadiazole groups, and perylene-3,4,9,10-tetracarboxydiimides with aromatic, dendritic arms attached to the bay positions.

In this paper we described new compounds containing in their structure TPA units, diimide core and imine linkages, that is, (poly)azomethine diimides. Their thermal, optical (absorption and emission in UV–vis range) and electrochemical properties were examined. Moreover, low molecular weight compounds were tested as bipolar host materials in blue phosphorescent organic light emitting diodes (OLEDs). First time such type of compounds was investigated as materials for OLED.

2. Experimental details

2.1. Materials

Benzene-1,2,4,5-tetracarboxylic dianhydride (pyromellitic dianhydride, PMDA), 1,4,5,8-naphthalenetetracarboxylic dianhydride (NDTA), 2,3,5,6-tetramethyl-1,4-phenylenediamine, 1,4-naphtha-

lene diamine, 4-formyltriphenylamine, 4,4'-diformyltriphenylamine, 4,4',4"-triformyltriphenylamine, N,N-dimethylacetamide (DMA), N,N-dimethylformamide (DMF) and toluene-4-sulfonic acid (PTS) trifluoroacetic acid (TFA), N-methyl-2-pyrrolidinone (NMP), chloroform were purchased from Aldrich Chemical Co and used as received. N,N'-bis(4-amino-2,3,5,6-tetramethylphenyl)phtalene-1,2,4,5-dicarboximide (DAPhDI) was synthesized according the procedure describe in our previous work [22].

2.2. Synthesis of diamine

N,*N'*-*bis*(5-*aminonaphtalen*)*naphthalene*-1,4,5,8-*dicarboxyimide* (*DANDI*-2): NTDA (3 mmol, 0.805 g), 1,4-naphthalene diamine (15 mmol, 2.3730 g) and imidazole (23.25 mmol, 1.5 g,) were added to 30 ml dry pyridine and refluxed under argon atmosphere. After 6 h, this mixture was cooled to room temperature. The diamine precipitate was collected by filtration and washed with water and then with hot methanol. The resulting diamine was dried in vacuum. Yield: 65%, violet solid.

¹H NMR (DMSO-d₆, δ (ppm)): 5.92 (s, NH₂, 4H), 6.73 (d, Ar–H, 2H), 6.95 (dd, Ar–H, 2H), 7.16 (t, Ar–H, 2H), 7.55 (t, Ar–H, 2H), 7.61 (t, Ar–H, 2H), 8.27 (d, Ar–H, 2H), 8.76 (s, 4H). FTIR (KBr (cm⁻¹)) v: 3438, 3365 (–NH₂ stretch), 1708, 1673 (C=O imide stretch), 1630 (–NH₂ deformation), 1344 (C–N stretch), 769 (imide ring deformation). Anal. Calcd. for (C₃₄H₂₀N₄O₄) (548,55 g/ mol) C 74.45; H 3.67; N 10.21; found: C 74.08; H 3.66; N 10.28.

2.3. Synthesis of azomethine diimide

Synthesis of azomethine diimides (AZPhDI and AZNDI): Diamine DAPhDI (0.5 mmol, 0.255 g) or diamine DANDI-2 (0.5 mmol, 0.2743 g), monoaldehyde: 4-formyltriphenylamine (1 mmol, 0.273 g) and 0.2 ml of trifluoroacetic acid was added to 4 ml DMA and heated (160 °C) under argon atmosphere. After 16 h, this mixture was cooled to room temperature and after night the resultant product was precipitated. The precipitant was filtrated, washed several times with hot methanol and dried at 100 °C in vacuum. Yield: AZPhDI: 60%, yellow solid, AZNDI: 50%, green solid.

AzPhDI: ¹H NMR (CDCl₃, δ (ppm)) 2.07 (s, CH₃, 12H), 2.10 (s, CH₃, 12H), 7.12 (dd, Ar—H, 8H), 7.18 (dd, Ar—H, 8H), 7.32 (dd, Ar—H, 8H), 7.77 (d, Ar—H, 4H), 8.07 (s, CH=N, 2H), 8.54 (s, phenylene, 2H). FTIR (KBr (cm⁻¹)) v: 1776, 1730 (C=O imide stretch), 1636 (CH=N), 1346 (C—N stretch), 733 (imide ring deformation). Anal. Calcd. for (C₆₈H₅₆N₆O₄) (1021,21): C 79.98; H 5.53; N 8.23, found: C 77.37; H 5.85; N 8.10.

AzNDI: ¹H NMR (DMSO-d₆, δ (ppm)): 8.65 (s, CH=N), 8.79 (s, naphthalene); FTIR (KBr (cm⁻¹)) v: 1715, 1678 (C=O imide stretch), 1623 (CH=N), 1340 (C–N stretch), 769 (imide ring deformation). Anal. Calcd. for ($C_{72}H_{46}N_6O_4$) (1059,17): C 81.65; H 4.39; N 7.93, found: C 81.76; H 4.40; N 7.82.

2.4. Synthesis of poly(azomethine imide)s

General procedure: Diamine (DAPhDI (0.5 mmol, 0.2550 g) or DANDI-2 (0.5 mmol, 0.2743 g)) and dialdehyde (4,4'-diformyltriphenylamine (0.5 mmol, 0.1506 g)) or trialdehyde (4,4',4''-triformyltriphenylamine (0.32 mmol, 0.1050 g)) were dissolved in DMA and 0.1 ml TFA was added. Mixture was heating (160 °C) under argon atmosphere. After 16 h, this mixture was cooled to room temperature. The precipitate was collected by filtration extracted with boiling methanol (Soxhlet apparatus) and dried in vacuum. Polymers prepared from DANDI-2 were insoluble and have not been investigated.

DAPhDI condensed with 4,4'-diformyltriphenylamine resulted in polymer *poly*AzPhDI-I while DAPhDI condensed with tris-(4Download English Version:

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