



Preparation and characterization of new aliphatic-tailed five- and six-membered azomethine-diimides



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H I G H L I G H T S

- New aliphatic-tailed five- and six-membered azomethinediimides were synthesized.
- Five-membered azomethinediimides exhibited liquid crystalline properties in wide temperature range.
- They emitted blue light in NMP solution and in blends with PMMA.
- They underwent reversible reduction and irreversible oxidation processes as evidenced by CV.
- They exhibited electrochemical narrow energy band gaps in the range 1.37–2.09 eV.

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New azomethine-phthalic (AzPhDIs) and azomethine-naphthalene (AzNDIs) diimides were synthesized in condensation reaction of diamines containing phthalic or naphthalene diimide core and two aldehydes: 4-(4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,11-heptadecafluoroundecyloxy)benzaldehyde and 4-octadecyloxybenzaldehyde. The phase behavior of azomethine-diimides was examined by differential scanning calorimetry (DSC) and polarized optical microscopy (POM). AzPhDIs exhibited liquid crystalline properties in wide temperature range, whereas AzNDIs were not liquid crystalline. Thermal stability was investigated by thermogravimetric analysis (TGA). Azomethine-diimides exhibited high decomposition temperatures in the range between 360 and 401 °C. Optical properties were studied by UV–vis and photoluminescence (PL) measurements. Azomethine-diimides emitted blue light in NMP solution and blends with PMMA. The investigated compounds were electrochemically active and underwent reversible reduction and irreversible oxidation processes as evidenced by cyclic voltammetry (CV). Azomethine-diimides exhibited LUMO and HOMO levels in the range of –3.87 to –4.2 eV and –5.56 to –6.02 eV, respectively.

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1. Introduction

Organic materials, both low molecular weight compounds and polymers, have been the subject of recent research in the area of photonics and electronics. The important advantages of organic compounds are light-weight, good processability and tunability of

their properties by structure modifications [1,2]. The considerable attention has been focused on conjugated systems called organic semiconductors both from the basic research and application standpoint. Compounds with imide groups are particularly interesting as potential organic semiconductors. They possess excellent thermal stability, good chemical and oxidative resistance and exhibit n-type semiconducting properties because of electron-accepting character of imide ring [3,4]. Taking into consideration the imide unit structure, imides can be divided into two groups, that is compounds with five- and six-membered imide rings. Six-

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membered diimides obtained from naphthalenetetracarboxylic dianhydride or perylenetetracarboxylic dianhydride have been investigated as materials for field-effect transistors [3,5–7], photovoltaic devices [8,9] and electroluminescent diodes [10,11], whereas five-membered (phthalic) diimides are only occasionally investigated as semiconductors [12–14]. Azomethines, known as imines and Schiff bases, are also attractive group of conjugated compounds. Due to the presence of imine bond ($-N=CH-$) they are isoelectronic to their carbon analogues and therefore can be an alternative to conventional conjugated materials [15]. In contrast to diimides, these compounds exhibit hole-transporting properties and have been studied as p-type materials [16–19]. Azomethines exhibit high chemical, oxidative and reductive resistance and interesting photophysical properties [16,20] and can be exploited for various applications in electronic and optoelectronic devices, such as light emitting diodes [17,18], solar cells [17,18], liquid crystal displays [19], sensors [21] and as electrochromic materials [22,23]. Recently, considerable attention has been focused on the compounds consisting of both electron-donating and electron accepting (D/A) moieties [24–26]. The selection of donor and acceptor unit allows to modify the energy levels of the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO), energy gap and charge transport ability depending on the desired application [5,26]. Donor-acceptor system can be created by introduction of imine bonds into the structure of imide compound. Various azomethine-diimides, which are the compounds containing both imine bonds and imide rings, were presented in our previous works [27–32].

In this paper, we report the synthesis and study of thermal (DSC, POM, TGA), optical (UV–vis, PL) and electrochemical (CV) properties of new azomethine-phthalic diimides and azomethine-naphthalene diimides. The properties of the obtained compounds and their analogues synthesized earlier in our laboratory [27,30] are compared. The synthesized azomethine-phthalic diimides exhibit liquid crystalline properties. Thermotropic liquid crystals offer a variety of unique properties, such as improved charge transfer ability and have received great attention during the years due to their practical applications [33].

2. Experimental

2.1. Materials

Benzene-1,2,4,5-tetracarboxylic dianhydride (pyromellitic dianhydride, PMDA), 1,4,5,8-naphthalenetetracarboxylic dianhydride (NTDA), 1,5-diaminonaphthalene, N,N-dimethylacetamide (DMA), N,N-dimethylformamide (DMF), trifluoroacetic acid (TFA) and pyridine were purchased from Aldrich Chemical Co. Imidazole, 4-(4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,11-heptadecafluoroundecyloxy) benzaldehyde and 4-octadecyloxybenzaldehyde were supplied from Fluka. N,N'-bis(5-aminonaphthalen)naphthalene-1,4,5,8-dicarboxyimide (DANDI-2) was prepared according to the procedure described in our previous work [31].

2.2. Synthesis of N,N'-bis(5-aminonaphthalen)phthalene-1,2,4,5-dicarboxyimide (DAPhDI-2)

PMDA (0.6544 g, 3 mmol) and 1,5-diaminonaphthalene (2.3730 g, 15 mmol) were added to 30 mL of DMF and refluxed under argon atmosphere. After 3 h, the mixture was cooled to room temperature and poured into 150 mL of water. The diamine precipitate was collected by filtration, washed with hot methanol and dried. Brown powder. Yield: 35%.

$^1\text{H NMR}$ (DMSO- d_6 , δ , ppm): 5.98 (s, NH_2 , 4H), 6.77 (d, Ar H, 2H), 6.93 (dd, Ar H, 2H), 7.24 (t, Ar H, 2H), 7.54 (dd, Ar H, 2H), 7.63 (t, Ar

H, 2H), 8.31 (d, Ar H, 2H), 8.48 (s, $\text{H}_{[\text{phenylene}]}$, 2H). FTIR (KBr, cm^{-1}): 3478, 3392 ($-\text{NH}_2$ stretch), 1773, 1721 ($\text{C}=\text{O}$ imide stretch), 1633 ($-\text{NH}_2$ deformation), 1375 ($\text{C}-\text{N}$ stretch), 726 (imide ring deformation).

2.3. Synthesis of azomethine-phthalic diimides (AzPhDIs)

Diamine DAPhDI-2 (0.1246 g, 0.25 mmol), aldehyde (4-(4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,11-heptadecafluoroundecyloxy) benzaldehyde (0.2910 g, 0.5 mmol); or 4-octadecyloxy benzaldehyde (0.1875 g, 0.5 mmol)) and a few drops of trifluoroacetic acid were added to 4 mL of DMA and heated (160 °C) under argon atmosphere. After 19 h the mixture was cooled to room temperature and poured into 20 mL of water. The precipitate was collected by filtration, washed with hot methanol and dried.

DAPhDI-2 condensed with (4-(4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,11-heptadecafluoroundecyloxy)benzaldehyde gave compound AzPhDI-1 and DAPhDI-2 condensed with 4-octadecyloxy benzaldehyde resulted in compound AzPhDI-2.

AzPhDI-1: Gray powder. Yield: 27%. $^1\text{H NMR}$ (CDCl_3 , δ , ppm): 2.17 (m, CH_2 , 4H), 2.37 (m, CH_2 , 4H), 4.15 (m, CH_2-O , 4H), 7.03 (m, Ar H, 4H), 7.11 (m, Ar H, 2H), 7.43 (m, Ar H, 2H), 7.51 (m, Ar H, 4H), 7.63 (m, Ar H, 2H), 7.98 (m, Ar H, 4H), 8.48 (s, $\text{CH}=\text{N}$, 2H), 8.56 (m, Ar H, 2H), 8.62 (s, $\text{H}_{[\text{phenylene}]}$, 2H). FTIR (KBr, cm^{-1}): 3070 ($\text{C}-\text{H}$ aromatic), 2962, 2924, 2868 ($\text{C}-\text{H}$ aliphatic), 1774, 1729 ($\text{C}=\text{O}$ imide stretch), 1624 ($\text{CH}=\text{N}$), 1374 ($\text{C}-\text{N}$ stretch), 729 (imide ring deformation).

AzPhDI-2: Gray powder. Yield: 17%. $^1\text{H NMR}$ (CDCl_3 , δ , ppm): 0.89 (t, CH_3 , 6H), 1.31 (m, CH_2 , 52H), 1.38 (m, CH_2 , 4H), 1.50 (quint, CH_2 , 4H), 1.83 (quint, CH_2 , 4H), 4.06 (t, CH_2-O , 4H), 7.02 (d, Ar H, 4H), 7.11 (d, Ar H, 2H), 7.43 (dd, Ar H, 2H), 7.51 (t, Ar H, 2H), 7.55 (dd, Ar H, 2H), 7.63 (t, Ar H, 2H), 7.97 (d, Ar H, 4H), 8.48 (s, $\text{CH}=\text{N}$, 2H), 8.59 (d, Ar H, 2H), 8.62 (s, $\text{H}_{[\text{phenylene}]}$, 2H). FTIR (KBr, cm^{-1}): 3069 ($\text{C}-\text{H}$ aromatic), 2921, 2851 ($\text{C}-\text{H}$ aliphatic), 1778, 1727 ($\text{C}=\text{O}$ imide stretch), 1623 ($\text{CH}=\text{N}$), 1375 ($\text{C}-\text{N}$ stretch), 726 (imide ring deformation).

2.4. Synthesis of azomethine-naphthalene diimides (AzNDIs)

Diamine DANDI-2 (0.1371 g, 0.25 mmol), aldehyde (4-(4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,11-heptadecafluoroundecyloxy) benzaldehyde (0.2910 g, 0.5 mmol); or 4-octadecyloxy benzaldehyde (0.1875 g, 0.5 mmol)) and a few drops of trifluoroacetic acid were added to 4 mL of DMA and heated (160 °C) under argon atmosphere. After 19 h the mixture was cooled to room temperature and poured into 20 mL of methanol. The precipitate was collected by filtration, washed with hot methanol and dried.

DANDI-2 condensed with (4-(4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,11-heptadecafluoroundecyloxy)benzaldehyde gave compound AzNDI-1 and DANDI-2 condensed with 4-octadecyloxy benzaldehyde resulted in compound AzNDI-2.

AzNDI-1: Brown powder. Yield: 57%. $^1\text{H NMR}$ (CDCl_3 , δ , ppm): 2.19 (m, CH_2 , 4H), 2.36 (m, CH_2 , 4H), 4.16 (t, CH_2-O , 4H), 7.05 (d, Ar H, 4H), 7.09 (dd, Ar H, 2H), 7.46 (m, Ar H, 4H), 7.58 (d, Ar H, 2H), 7.69 (t, Ar H, 2H), 8.01 (d, Ar H, 4H), 8.50 (s, $\text{CH}=\text{N}$, 2H), 8.59 (d, Ar H, 2H), 8.92 (s, $\text{H}_{[\text{naphthalene}]}$, 4H). FTIR (KBr, cm^{-1}): 3070 ($\text{C}-\text{H}$ aromatic), 2956, 2879 ($\text{C}-\text{H}$ aliphatic), 1718, 1680 ($\text{C}=\text{O}$ imide stretch), 1626 ($\text{CH}=\text{N}$), 1343 ($\text{C}-\text{N}$ stretch), 769 (imide ring deformation).

AzNDI-2: Brown powder. Yield: 46%. $^1\text{H NMR}$ (CDCl_3 , δ , ppm): 0.87 (m, CH_3 , 6H), 1.27 (m, CH_2 , 52H), 1.49 (m, CH_2 , 8H), 1.82 (m, CH_2 , 4H), 4.07 (m, CH_2-O , 4H), 7.01 (m, Ar H, 4H), 7.07 (m, Ar H, 2H), 7.46 (m, Ar H, 4H), 7.57 (m, Ar H, 2H), 7.68 (m, Ar H, 2H), 7.98 (m, Ar H, 4H), 8.49 (s, $\text{CH}=\text{N}$, 2H), 8.59 (m, Ar H, 2H), 8.91 (s, $\text{H}_{[\text{naphthalene}]}$, 2H). FTIR (KBr, cm^{-1}): 3067 ($\text{C}-\text{H}$ aromatic), 2923, 2851 ($\text{C}-\text{H}$ aliphatic), 1717, 1679 ($\text{C}=\text{O}$ imide stretch), 1629 ($\text{CH}=\text{N}$), 1340

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