

A Review on Perylene-3,4,9,10-Tetracarboxylic Acid Diimide Molecules

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Abstract

The aim of this article is to inform about perylene-3,4,9,10-tetracarboxylic acid diimide (PDI) derivatives. Also, it is to give short and simple information about optical properties and using fields. Perylene-3,4,9,10-tetracarboxylic acid diimide derivatives known as perylene diimides are material of highly conjugated molecule. PDIs exhibit strong absorptions within the range of 400-450 nm (B band) and 500-700 nm (Q band). Perylene diimide dyes show generally characteristic absorption peaks at 458, 490, 526 nm. These absorption peaks are matched by the corresponding emission peaks at 540, 576, 624 nm, respectively. It has donor core and acceptor carbonyl groups. Perylene diimide has good electron mobility, high fluorescence quantum yields, strong absorption in the visible region, photostability, thermal stability, good semi-conductivity. These properties benefit them application of electronic material. Applications of PDIs are in the field of photo-electronic material, dye lasers, organic light-emitting diodes, and chemosensing materials.

Keywords–Dye, Organic based-solar cells (DSSC), perylene diimide, perylene monoimide, sensor

1 Introduction

Perylene-3,4,9,10-tetracarboxylic acid diimide derivatives known as perylene diimides are abbreviated as PDIs. PDI-chromophoric dye is one of the excellent materials of highly conjugated molecule [1]. It has donor core and acceptor carbonyl groups. Perylene-3,4,9,10-tetracarboxylic dianhydride (PTCDA) is used to synthesis the perylene-3,4,9,10-tetracarboxylic acid diimide. PDIs first reported in 1913 are known Pigment Red 179. Figure 1 shows molecular structures of PDI, PTCDA and Pigment Red 179.

Perylene diimides exist in a wide range of colors from red to violet, and even shades of black. Pigment Red 149 is a yellow shade red material, while Pigment Red 178 and Pigment Red 179 are of blue-shade red color. Figure 2 shows molecular structures of Pigment Red 149 and Pigment Red 178.

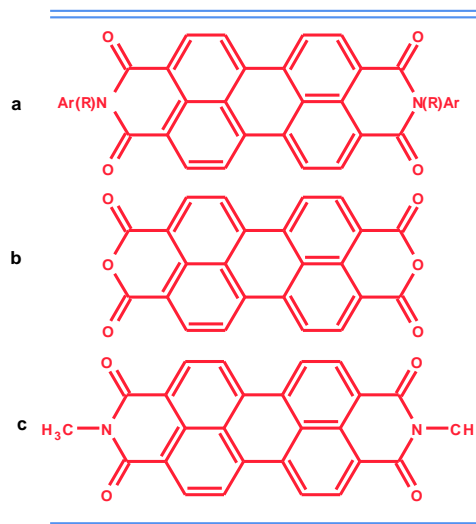


Figure 1. Molecular structures a) perylene diimide (PDI) b) perylene-3,4,9,10-tetracarboxylic dianhydride (PTCDA) c) pigment red 179 (N,N'-dimethyl PDI)

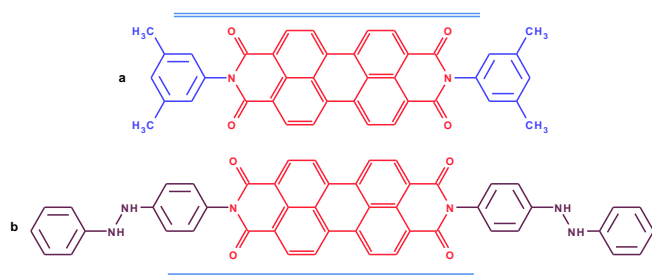
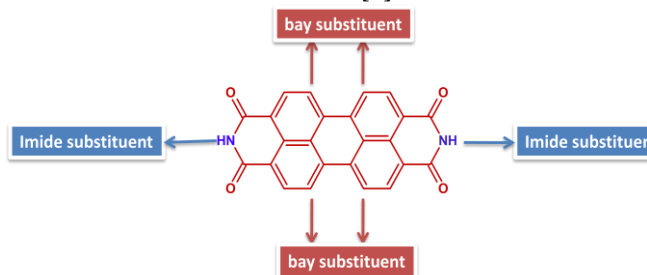


Figure 2. Molecular structures a) pigment red 149 b) pigment red 178

Different PDI derivatives are obtained from both imide and bay position. Some properties of PDI are tuned by modification of the substituent. While substituents on the imide nitrogen atoms have little effect on optical and electronic properties, it affects solubility. Bay substituents at the 1,6,7 or 12 positions affect electronic and optical properties and alter photo-physical properties of PDI [2,3]. The color of perylene diimides is tuned by variation of the substituent of the imide functions [4].

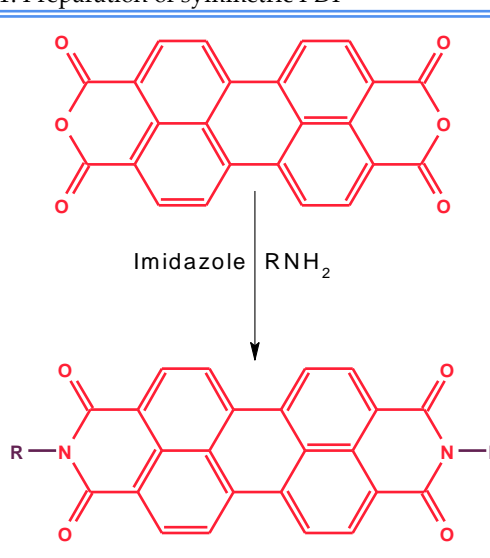


While PDI was initially used exclusively as industrial pigments as red vat dyes, nowadays applications of PDI are in the field of photo-electronic material, dye lasers, organic light-emitting diodes (OLED) [4,5], chemosensing materials [6], biosensing [7]. It has electron transporting character. Also, perylene diimides have been widely studied for biochemical and pharmacological purpose because of considering as potential antitumor drugs acting as telomerase inhibitors [8]. Perylene diimide has good electron mobility [9], high fluorescence quantum yields [10], strong absorption in the visible region [11], photostability [12-14], thermal stability [15-18], large extinction coefficients, high electron affinity [4] and good semi-conductivity [19]. These properties benefit them application of electronic material [15]. PDIs exhibit strong absorptions within the range of 400-450 nm (B band) and 500-700 nm (Q band) [20] owing to their extended conjugated- π systems. The

fluorescence quantum yields of PDIs are near 1.0 [14].

Symmetrical PDI derivatives are prepared by using perylene-3,4,9,10-tetracarboxylic dianhydride (PTCDA) in solvents such as molten imidazole or quinoline by using anhydrous zinc acetate as a catalyst (Scheme 1). Appropriate PDI derivative in high yield forms the reaction between PTCDA and an alkyl/aniline amine. The soluble symmetrically N,N'-substituted PDS are commonly obtained in the same way by using long chain alkyl groups. PDI molecules have insolubility problem in organic solvents [7].

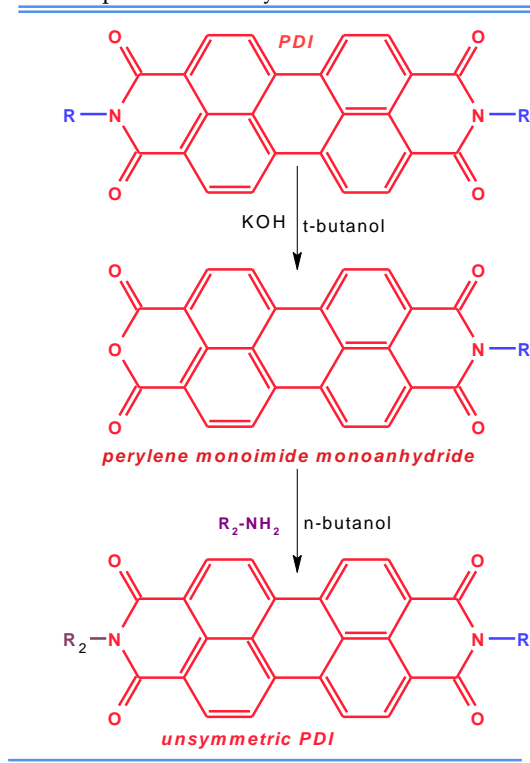
Scheme 1. Preparation of symmetric PDI



Unsymmetrical PDIs are synthesized by using PDI. Firstly, perylene monoimide monoanhydride is obtained to partial hydrolysis. Then, different amine is used for imidization to obtain desired asymmetrical PDI (Scheme 2).

Bay-substituted-PDI derivatives are prepared with introduction of aryl or aryloxy groups into the 1,6,7 or 12 (bay) positions of PDI. Solubility of PDI molecule increases. Also electronic and optical properties of PDI alter. PDI derivatives have good solubility in halogenated solvents such as dichloromethane, chloroform and chlorobenzene. Conjugated groups in the bay positions generally lead to bathochromically shifted spectra [2].

Scheme 2. Preparation of unsymmetric PDI

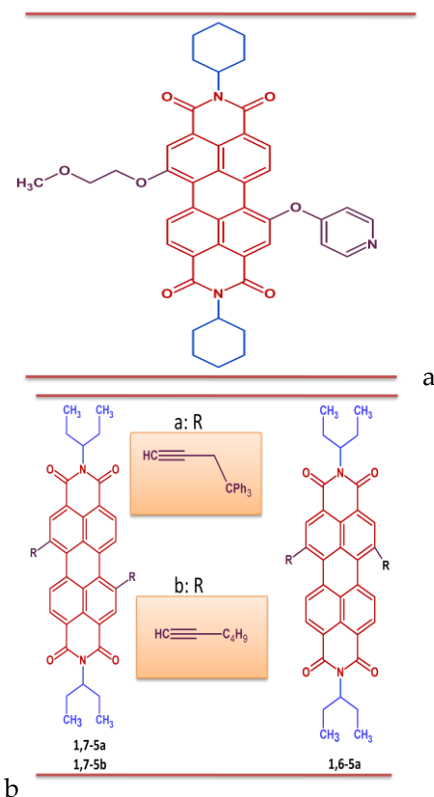


Perylene diimide dyes show generally characteristic absorption peaks for the perylene diimide at 458, 490, 526 nm, which are denoted to their (0,2), (0,1), and (0,0) electronic transitions, respectively. These absorption peaks are matched by the corresponding emission peaks at 540, 576, 624 nm, respectively. The emission spectra of PDI fulfill the mirror image conditions to the absorption spectra in solutions. The perylene compounds show two reversible one electron reduction peaks. First reversible reduction wave was observed in the range of -0.38/ -0.42 V and the second reduction potential was observed in the range of 0.57/-0.62 V. The reduction mechanism for the perylene diimide derivatives is known that electron transfer processes forms a radical anion occurs by the addition of an electron to the lowest LUMO and then the dianion with addition of a second electron to the same orbital. Redox behavior of the perylene diimide molecules is not affected significantly by N-substituent [21].

In this paper, the structure, optical properties and application of some PDI derivatives from literature are reviewed.

Authors reported synthesis some of bay-perylene

diimide derivatives (Figure 3). Zhang et al. have synthesized unsymmetrical PDI derivatives functionalizations at the 1,7 positions. It has absorption spectrum in the range of 556, 524 and 404 nm in Dichloromethane (DCM). It exhibits strongly fluorescent. The fluorescence peak is at 590 nm. It shows the stronger electronic interactions between the electron-deficient core perylene and electron-rich alkoxy groups [22]. Handa et al. have synthesized 1,6 and 1,7 perylene diimides (PDIs) to obtain soluble molecule. It is possible to synthesis soluble perylene diimide via synthetic modification to N-alkyl or aryl PDIs and/or by modification of the "bay" region (1,6,7,12 positions) [7]. PDI derivatives substituted at the bay positions are obtained by using C-O, C-C and C-N coupling. The steric hindrance and perylene core twisting induced by the bay substituents could prevent Π - Π^* stacking, and thus reduce luminescence quenching. Sivamurugan et al. have reported a series of perylenediimide derivatives with electron accepting substituents at the bay positions of the perylene core. Reaction yields are high (70-80%). Author reported that perylene derivatives were found to be thermally stable up to ca.440 °C with 5% weight loss temperature ranging from 436-460 °C [23].



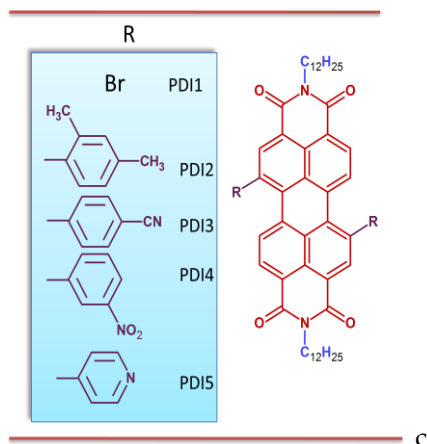


Figure 3. Molecular structures of bay-perylene dimide derivatives synthesis by a)Zhang et.al. b)Handa et al. c) Sivamurgan et al.

The highly soluble bay-perylene dimide containing α and β linked thiophene group have synthesized by Balaji et al. (Figure 3). Oligothiophenes are donor structure with charge transport properties. Both molecules exhibit two absorption maxima with a weak low energy absorption band around 650 nm. Author reported that the absorption spectrum of α -linked bay perylene dimide is blue-shifted compared to the β linked bay-perylene dimide. Author explained that this may be due to the branched nature of the thiophene degment, which prevents the effective conjugation of the pentathiophene. PDI molecules exhibit fluorescence properties but α -linked and β linked bay perylene thiophene dimide derivatives have not fluorescence properties. This is possible to photoinduced intramolecular electron transfer between the oligothiophene moiety and the perylene core [24]. The solubility of PDI molecules is limited. Its solubility can be improved by introducing solubilizing N-substituent with the long-tail or swallow-tail [26]. For a biological application, the water solubility of PDI is essential. Chromophores reported are groups such as sulphonic acid part, quaternized amine groups, polyethylene glycol [27].

Figure 5 shows a typical absorption and emission spectrum of PDI molecules. Perylene diimide dyes show generally characteristic absorption peaks for the perylene diimide at 458, 490, 526 nm. These absorption peaks are matched by the corresponding emission peaks at 540, 576, 624 nm, respectively. The emission spectra of PDI fulfill the mirror image con-

ditions to the absorption spectra in solutions.

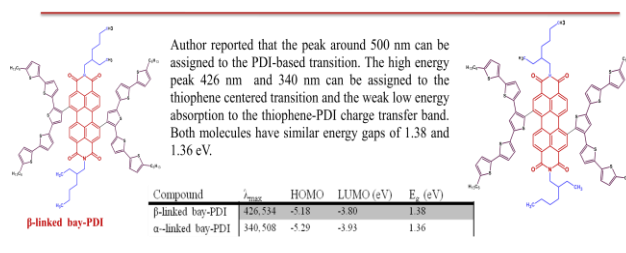


Figure 4. Molecular structures of bay-perylene dimide derivatives synthesized by Balaji et al.

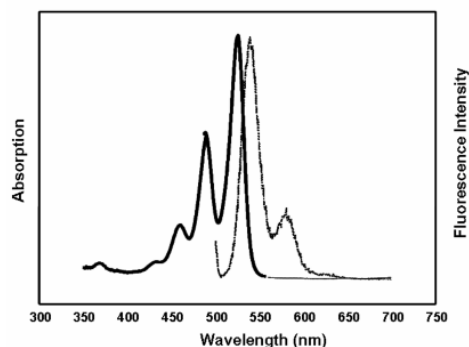


Figure 5. UV-Vis absorption (solid line) and emission (dashed line) spectra of a typical PDI in chloroform

PDI solution concentration in solvent is prepared as 10^{-5} M to prevent reabsorption and concentration quenching. Absorbance values must be lower than 0,2. Generally, perylene dimide molecules have three absorption bands ($\lambda= 458$ nm, 490 nm, 526 nm) and high absorptivite coefficient ($\epsilon=10^5$). They have strong absorption capacity at the visible region. As the conjugation increases, bathochromic shift occurs. Fluorescence quantum yield is about 1 [25]. The electronic nature of aryl substituents in the 2,5,8,11-positions also has a significant impact on the optical properties of PDIs [2].

Wang et al. have synthesized fluorinated on the bay position of perylene diimide. Fluorine has electron affinity. Author reported that highly fluorinated materials display a variety of interesting properties such as thermal and chemical stability, low surface energy and high resistance to oxidation. Author reported that BFPDIs showed an increase in the first reduction potential, indicating that the electron accepting power increased in strength. LUMO levels of BPPDI,

BFPDI-1 and PFPDI-2 were calculated as -3.85 eV, -4.00 eV and -4.02 eV, respectively. The decreased LUMO levels are ascribed to the electron-deficient fluorinated substituents on the bay of perylene diimides. Three PDI molecules are highly soluble in water. So absorption spectra were taken in both water and toluene. Absorption bands of BPPDI measured in toluene are at 551 nm and 518 nm. Absorption bands measured in toluene are at 542 nm for BFPDI-1 and 539 nm for BFPDI-2. It shows blue-shifted. Absorption spectra values taken in water are 557 nm for BPPDI, 550 nm for BFPDI-1 and 548 nm for BFPDI-2. It shows red-shifted [28].

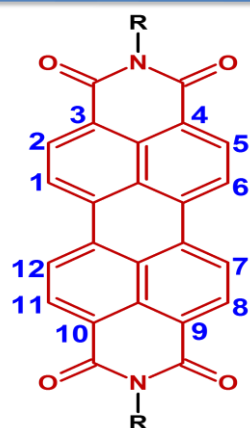


Figure 6. Chemical structure of PDI showing the numbering of the positions in the ring system

It is reported synthesis of unsymmetric triazine-linked porphyrine perylene diimide molecule. Also, porphyrin molecules are sensitizer for chemical and biological sensors. Perylenes are common electron acceptor in porphyrin donor-acceptor systems due to their low LUMO. The intense absorption bands of perylene derivatives are between porphyrin's Soret band and Q bands, thus perylene can enhance the absorption efficiency of the sunlight and is thought to be better acceptor candidate.

The LUMO and HOMO energy levels for HTPP-PDI are -4.05 eV and -5.68 eV, respectively. Both HOMO and LUMO levels of porphyrin derivatives are higher than those of perylene derivatives, indicating a possible electron transfer from porphyrins to perylene. It is reported that HTPP-PDI, the HOMO locates on the porphyrin moiety while the LUMO locates on the perylene moiety [29].

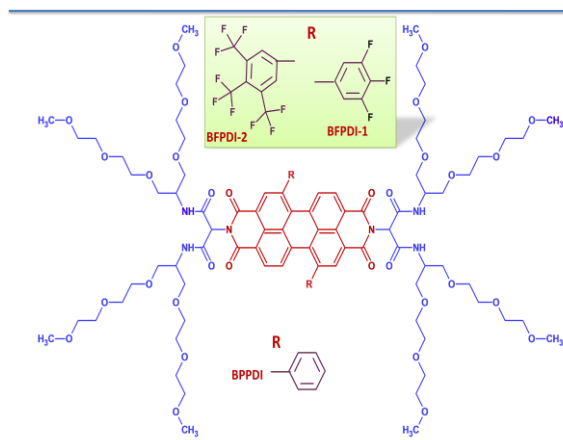


Figure 7. Molecular structures of fluorinated-perylenediimide derivatives synthesized by Wang et al.

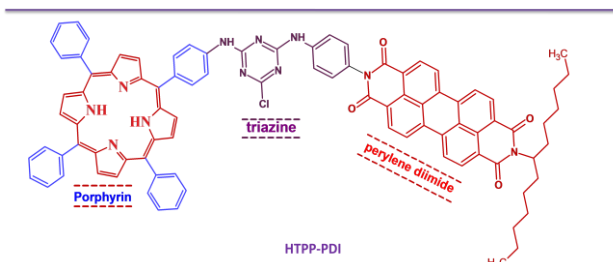


Figure 8. Molecular structure of porphyrin-perylenediimide derivatives synthesized by Zhou et al.

Perylene diimides are used as a sensor. It is reported that rhodamine-perylenediimide fluorescent probe has been synthesized for cell imaging. The perylene diimide chromophore plays as an electron acceptor, the rhodamine units serve as electron donors and the ethylene-diamine plays as spacer [30]. Zhao et al. have used cationic-perylenediimide-G-quadruplex complex based fluorescent biosensor for detection of Pb^{2+} [31]. Hg^{2+} sensor was developed by Cheng et al. [32]. It is reported that perylene diimide derivatives are used for Cu^{2+} probe [33].

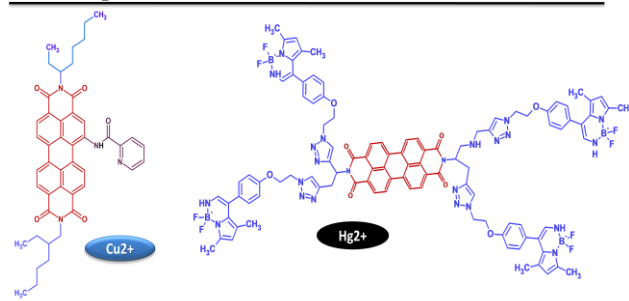


Figure 9. Molecular structures of perylene diimide derivatives for sensors.

Solar cells, also called photovoltaic cells, are an electronic device used to generate electricity directly from sunlight. Dye-sensitized solar cells (DSSCs) have attracted alternative to the conventional photovoltaic cell due to low-cost electricity production from sun light, high power conversion efficiency for solar energy. Perylene diimide derivatives are used for DSSC [34]. Figure 10 shows some perylene derivatives for DSSC and power conversion efficiency (PCE), electrochemical, optical properties [35-37].

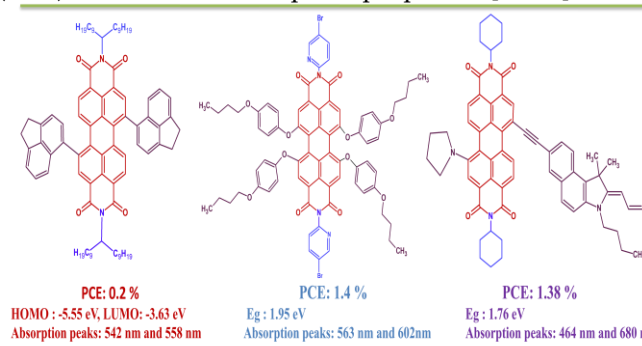


Figure 10. Molecular structures of perylene diimide derivatives for DSSC

Unsymmetrically N-Butyl-N'-(6-hydroxyhexyl) perylene-3,4,9,10-tetracarboxylic acid diimide (N-BuHHPDI) has been synthesized in high yield by Tahir et al. Perylene-3,4,9,10-tetracarboxylic dianhydride (PTCDA) is partial hydrolyzed with KOH. Then, it is obtained potassium salt. Perylene monoimide is obtained by using n-butyl amine. Other side of perylene molecule is anhydride. Then, different amine is used for imidization to obtain desired asymmetrical PDI [38].

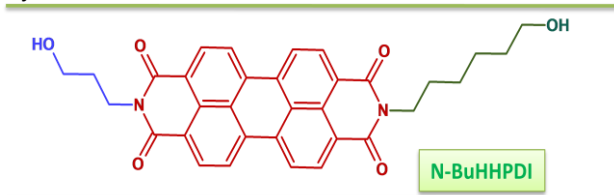


Figure 11. Molecular structure of PDI derivatives synthesized by Kozma et al.

CONCLUSION

Organic dye perylene-3,4,9,10-tetracarboxylic acid diimide derivatives are also known as perylene diimides. Perylene diimide dyes show generally characteristic absorption peaks for the perylene diimide at 458, 490, 526 nm, which are denoted to their (0,2), (0,1), and (0,0) electronic transitions, re-

spectively. These absorption peaks are matched by the corresponding emission peaks at 540, 576, 624 nm, respectively. The perylene compounds show two reversible one electron reduction peaks. Nowadays applications of PDI are in the field of photo-electronic material, dye lasers, organic light-emitting diodes, chemosensing materials, biosensing.

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