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A Novel Fluorescent Disperse Dye based on N-Polyamidoamine Dendrimer-1,8-Naphthalimide: Synthesis, Characterization and Dyeing Properties on Polyester Fibres

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Abstract

Polyamidoamine dendrimer of zero generation was prepared by a divergent synthesis scheme using the reagent excess method starting from ethylenediamine by consecutive Michael addition and ester amidation reaction. In order to obtain the fluorescent dye, the amino end groups of synthesized dendrimer were reacted with 4-amino-1,8-naphthalimide via aromatic nucleophilic substitution reaction. The chemical structures of prepared dye and its corresponding intermediates were studied by FTIR, 1HNMR, 13CNMR and UV-Visible spectroscopic techniques. Synthesized dye was applied as a disperse dye on polyester fabric by high temperature exhaustion dyeing and relevant characteristics of dyed fabrics including dye ability, build up, wash and light fastness were evaluated. Also the position of color in CIELAB coordinates (L*, a*, b*, h°, C*) was assessed.

Keywords: Polyamidoamine dendrimer; 1,8-Naphthalimide; Polyester fabric; Dyeing properties; Fluorescent

Introduction

Dendrimer is a nanometric, three-dimensional and spherical macromolecular with various branches [1,2]. These class of macromolecules were first synthesized and investigated by Tomalia et al [3-5]. Dendrimers consist of three major parts:

- Core which can be an atom or a molecule,
- Branches exceeded from the core and repetition of these branches creates internal layers called generation,
- External functional agents which determine the use of these unique macromolecules [6,7].

Dendrimers have the ability to lock other molecules in their structure and release these molecules in target places. Besides in order to have external chemical agents they can carry and attach other molecules [8]. Polyamidoamine (PAMAM) dendrimers are a new class of these polymeric materials having received much attention due their unique molecular architecture and high concentration of functional end groups. Most of the PAMAMs consist of an ethylenediamine (EDA) core in which a polyamidoamine repetitive segment extends in all directions. The specifics of these dendrimers are their amino end groups which can easily react with various molecules [3]. Because of their excellent structure high functional hydrophilic surface area and good stability of PAMAMs they have used in many fields, in pharmaceutical industry as electrochemical biosensors and drug delivey, environment and textile industry [7,9-13].

The conjugated PAMAMs by aromatic groups emit fluorescence via the aromatic moieties and the luminescent wavelength changes with different aromatic groups. The design and modification of the PAMAM dendrimer with such fluorescent chromophoric systems has created interesting properties and new areas of applications [1,4].

Chemically, 1,8-naphthalimide is a polar molecule with electron deficiency in the aromatic rings and an absorption band $\pi \rightarrow \pi^*$ type in the near-UV range [14]. Strong fluorescence and good photostability of the 1,8-naphthalimide derivatives have caused them more applicable in a wide range of areas, as fluorescent dyes, fluorescent pH sensor. Anti-bacterial agent, anticancer agent, liquid crystal displays and fluorescent sensors for specific metal cations [15-21].

Zero and second generations of PAMAM dendrimer have recently been reacted with 1,8-naphthalimide units in their periphery and applied as photoinduced electron transfer (PET) sensor of protons and of transition metal ions [22]. But, meanwhile, to the best of our knowledge, no research has been performed so far to prepare textile dyes using modification of dendrimer with naphthalimide compounds. The present paper deals with the synthesis, characterization and modification of PAMAM dendrimer with 4-amino-1,8-naphthalimide and its application as a novel fluorescent disperse dye on polyester fabric.

Experimental

Materials and methods

Polyester fabrics (PET) (Twill weave, warp: 136 threads/inch, weft: 72 threads/inch) were used for dyeing throughout the study. Acenaphthene, ethylenediamine (EDA) and methyl acrylate were of analytical grade and purchased from Aldrich Company. The UV–Visible spectra of the synthesized dye was measured on a Cecil 9200 double beam spectrophotometer. NMR spectra were recorded on a Bruker DRX AVANCE spectrometer at 400 MHz and 125 MHz for 1HNMR and 13CNMR respectively, using a dual 5 mm probe head. All the measurements were carried out in DMSO-d6 and tetramethylsilane

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as a solvent and internal standard, respectively. FTIR spectra were measured on a Perkin Elmer Spectrum One, using the KBr pellet technique at a 4 cm-1 resolution. Thin layer chromatography (TLC) analysis followed on silica gel (Fluka F60 254 20 × 20; 0.2 mm) using the solvent system methanol/dichloromethane 1:10 (v/v) as eluent. Melting points (m.p) were determined using a Perkin Elmer Pyris 6 differential scanning calorimeter (DSC). The colorimetric data of the dyed fabrics were obtained using a Gretag Macbeth 7000A spectrophotometer (D65 illumination, 10° observer).

Synthesis of 4-amino-N-PAMAM dendrimer-1,8-naphthalimide (ANPN) and corresponding intermediates

In order to achieve ANPN, acenaphthene and ethylenediamine (EDA) were used as starting materials for preparation of chromophoric system and dendrimer, respectively. For the synthesis of 4-amino-1,8-napthalic anhydride the reactions including nitration, oxidation and reduction were performed on acenaphthene according to our previous works [1,4]. PAMAM dendrimer of zero generation (G0) was prepared by a divergent synthesis scheme using the reagent excess method starting from EDA by consecutive Michael addition and ester amidation reaction [5]. To prepare the full generation of dendrimer, EDA was dissolved in the methanol at the beginning of reaction and followed by cooling down the mixture in an ice bath. Then methyl acrylate was added dropwise at continuous stirring to the dendrimer solution under Argon gas. The mixture was kept at ambient temperature for 168 hours. The excess of methyl acrylate and solvent were removed under vacuum at temperature below 50 °C resulting in a nearly colourless viscous syrup. 1,8-Naphthalimide-conjugated dendrimer was synthesized from obtained zero generation PAMAM dendrimer, which possesses four primary amine groups at periphery. For this purpose, 5-nitro-1,8-naphthalic anhydride was reacted with the primary amine end groups of dendrimer in ethanol solution by the condensation reaction (Figure 1). In this reaction 4-nitro-N-PAMAM-1,8-naphthalimide (NNPN) (Figure 2) was synthesized. Treatment of NNPN with stannous chloride in ethanol using hydrochloric acid leads to produce ANPN (Figure 3) [4]. The completion of reactions was determined by TLC on silica gel (Fluka F60 254 20 × 20; 0.2 mm) with a mixture of methanol and dichloromethane (1:10 (v/v)) as eluent. Yield: 76 %; m.p: 345 °C; FTIR (KBr) cm^{-1}: 3395 (N-H str. Secondary amine), 3076 (C-H str. aromatic), 2953 (C-H str. aliphatic), 1675 and 1636 (C=O str. Carbonyl group), 1614 (v amide I), 1577 (v amide II); 1HNMR (DMSO-d6, 400 MHz, ppm): 7.78-8.64 (20H, aromatic), 7.8 (4H, NHCOCH3), 4.08 (8H, NCH2), 3.34 (8H, CH2NH), 3.03 (4H, CONH), 2.49 (20H, NCH2CH2CO), 1.68 (12H, NHCOCH3); 13CNMR (DMSO-d6, 125 MHz) δ (ppm): 170.04, 164.19, 163.62, 153.32, 140.69, 132.01, 131.19, 126.8, 124.5, 122.08, 118.4, 109.7, 40.62, 40.2, 39.99, 39.36, 24.54 (Figures 1-3).

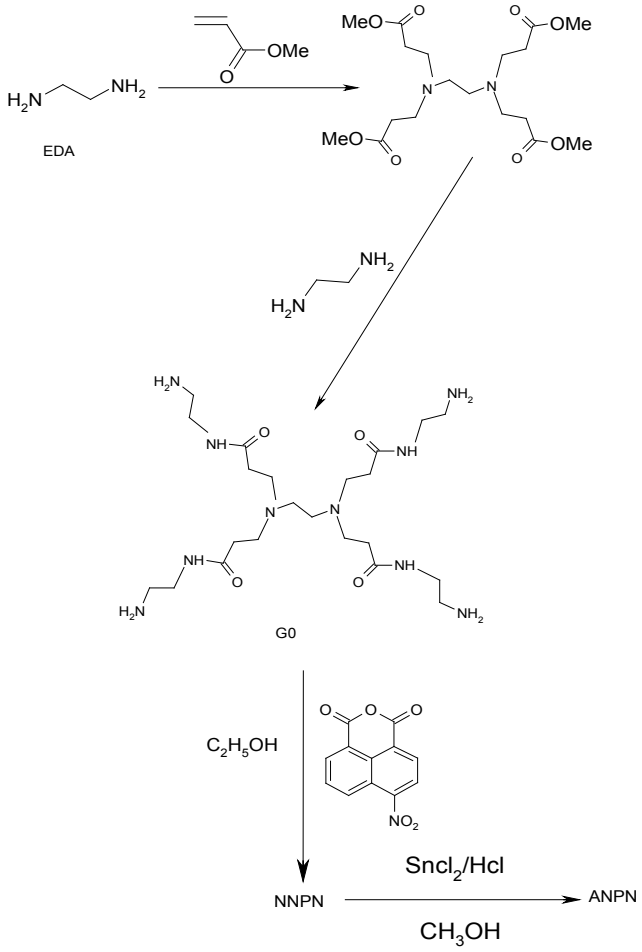

Figure 1: Synthesis pathway of PAMAM, NNPN and ANPN.

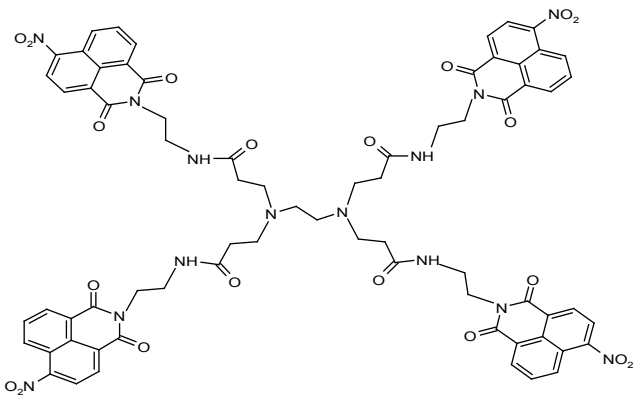

Figure 2: Chemical structure of synthesized NNPN.

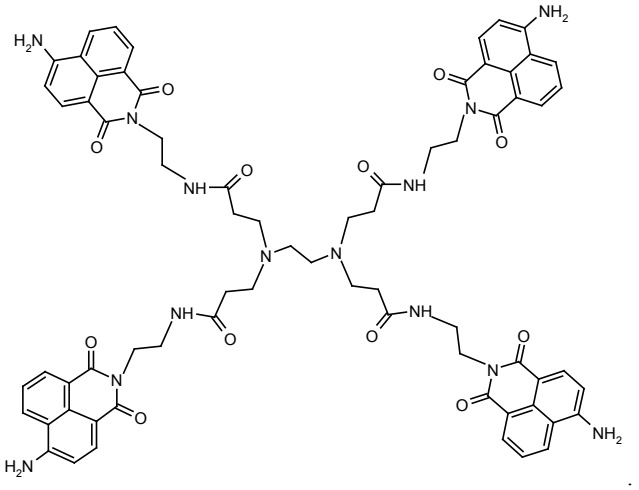

Figure 3: Chemical structure of synthesized ANPN.

Preparation of dye dispersion and dyeing procedure

In order to obtain a dispersion of synthesized dye, dye (0.5 g), dispersing agent (1 g) (Lyoprint EV, Ciba-Geigy) and water (2 ml) were added to a mortar and milled for 60 min; the ensuing mixture was diluted with 20 ml water, transferred to a ball mill and milled for 24 h. The volume of the dispersion was adjusted to 100 ml and filtered through a 5 µm Micro-Prazisions Sieb Fritsch. Dyeing on polyester fabric was carried out by reported dyeing procedure in our previous investigation [22,23]. Before dying, polyester fabrics (1 g) were pretreated with 5 g/l nonionic detergent (Lotensol, Hansa) at 80°C for 20 min, using a Liquor-to-Goods ratio (L:G) of 50:1 and the procedure was followed by rinsing and drying of fabrics. Dyeing was performed in laboratory model glycerin bath of metallic beaker-dyeing machine (Nasaj Sanat Yazd) using L:G of 50:1, pH 4-5 (acetic acid) at 0.1, 0.3, 0.5, 0.7, 1, 1.5, 2 and 4% on the weight of fabrics (o.w.f.) according to the demonstrated dyeing profile in Figure 4.

In order to remove the adsorbed dyes from the dyed fabrics, reduction clearing bath was employed at a L:G of 50:1 using sodium hydrosulphite (2 g/l), sodium hydroxide (1 g/l) and non-ionic detergent (1 g/l) for 20 min at 50°C. The reduction cleared samples were rinsed and allowed to dry.

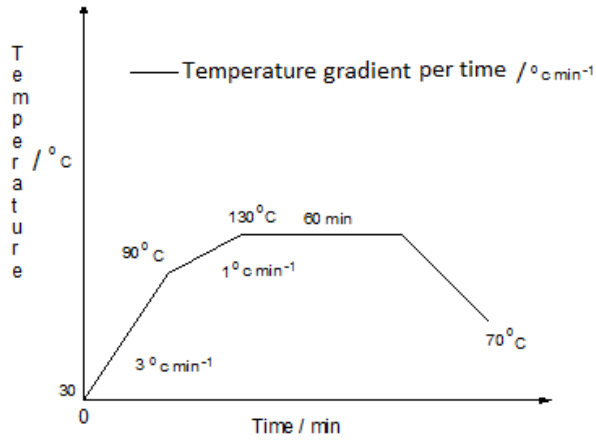

Figure 4: Dyeing profile of polyester fabrics.

Results and Discussion

The chemical structure of synthesized dye was characterized and

confirmed by FTIR, 1HNMR and 13CNMR spectroscopy. Data were presented in the experimental section. The primary amino groups of the initial PAMAM dendrimer gave two absorption bands at 3450 and 3352 cm^{-1}. These bands were disappeared after the modification of the dendrimer with 5-nitro-1,8-naphthalic anhydride in NNPN. The presence of nitro groups in NNPN molecule were confirmed by two bands at 1526 cm^{-1} and 1343 cm^{-1}. Elimination of the peaks corresponding to the nitro groups from the FTIR spectra of NNPN and appearance of stretching peaks due to the formed primary amines in the FTIR spectra of ANPN at 3420 and 3360 cm^{-1} confirmed the accuracy of amination reaction (Scheme 1). FTIR spectras of PAMAM dendrimer of zero generation, before and after modification with 5-nitro-1,8-naphthalimide are compared in Figure 5.

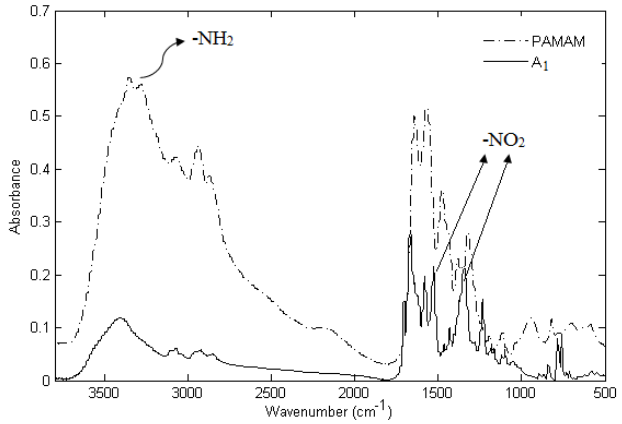

Figure 5: FTIR spectra of PAMAM and NNPN.

Solvatochromism is the ability of chromophores to change their colors in the solution depending on the environmental conditions, such as solvent polarity. The compounds, which incorporate both electron-donating and electron-accepting groups linked by π-conjugated cores, are promising candidates of solvatochromic fluorophores owing to their inherent dipole moments [4]. As a preliminary study of photophysical properties, we measured UV-visible spectroscopy of synthesized ANPN in five organic solvent with different polarity. As seen in Fig. 3 the polarity of used solvents has affected upon the position of the maximum absorption wavelength (ΔλA=21 nm) and increasing of solvent polarity faced it with a bathochromic shift indicating positive solvatochromism. Same solvatochromism effect has been observed for couple of naphthalimide based dyes synthesized in our previous work [1,4]. The molar extinction coefficients of the synthesized dye was 37792 M-1 cm^{-1} at wavelength of maximum absorption which represent charge transfer $\pi \rightarrow \pi^*$ electron transfer at S0→S1 transition. This finding is consistent with the data for compounds of similar nature that have been previously reported [24-26]. The molar extinction coefficient of synthesized dye which indicate the color strength is more than dyes based on 1,8-naphthalimide synthesized to date [27,28] (Figure 6).

For elaborating on the build up of the synthesized dye, the K/S was calculated using the Kubelka-Mank equation (Eq. 1).

$$\frac{K}{S} = (1-R)^2 / 2R$$

Where K, S and R, are absorption coefficient, scatter coefficient and reflectance of dyed sample, respectively.

Build up curve (K/S vs. concentration) of synthesized dye from which it is apparent that it generally reached saturation at concentration approximately 1.5% o.w.f. representing its good build up on polyester fabric (Figure 4). K/S is taken as criteria for dye ability of polyester fibres. The dyed polyester fabric was tested for wash fastness according to ISO-105 test [29,30] using detergent and 2% sodium carbonate. The change in shade and staining of adjacent fabrics were assessed using gray scales. A light fastness was carried out in accordance with the ISO-105 test [31]. Both of the wash fastness (4-5 to 5) and light fastness (7-8) were excellent (Figure 7).

The position of color in CIELAB coordinates including L^*, a^*, b^*, h° and c^* values were assessed by reflectance spectrophotometer and are shown in Table 1 with selected K/S of the dyed polyester. It can be understood from written data in Table 1, synthesized dye has golden yellow hue on polyester fabric (Table 1).

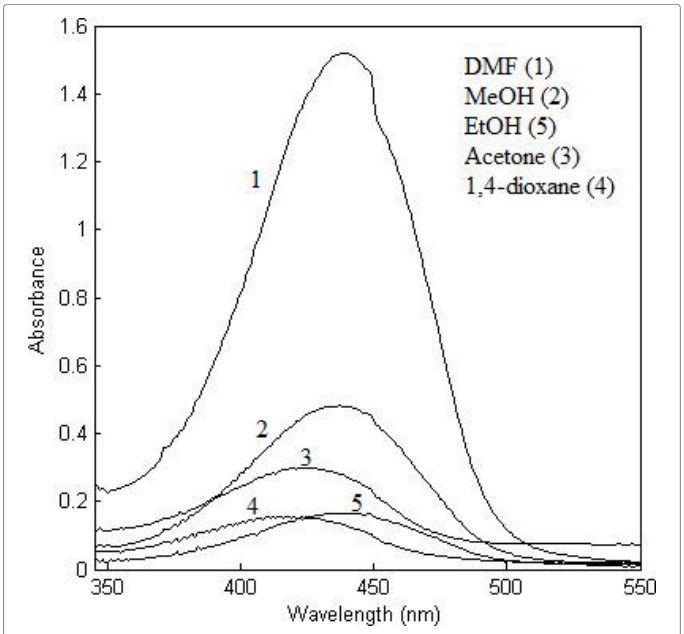

Figure 6: Absorption spectra of ANPN in different organic solvent.

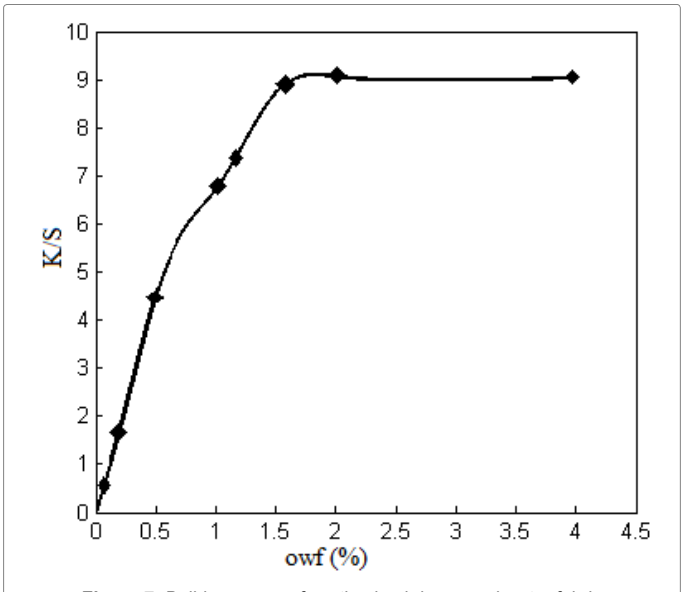

Figure 7: Build up curve of synthesized dye on polyester fabric

Bath No.	Dye (o.w.f)	L*	a*	b*	c*	h°	(k/s)
1	0.1	86.338	-5.711	28.390	28.958	281.37	0.568
2	0.3	84.180	-3.635	42.988	43.141	274.83	2.526
3	0.7	81.196	1.461	50.141	50.162	88.33	4.825
4	1	79.899	4.579	53.866	54.060	85.14	5.936
5	1.2	76.566	8.325	59.678	60.255	82.05	6.412
6	1.6	74.851	12.946	62.137	63.471	78.22	8.934
7	2	71.162	15.195	63.234	65.034	76.48	9.023
8	4	68.536	17.279	63.341	65.655	74.73	9.115

Table 1: Spectrophotometric data of dyed polyester with synthesized dye at 1/1 standard depth.

Conclusions

A novel fluorescent disperse dye based on N-PAMAM dendrimer-1,8-naphthalimide was synthesized and studied using FTIR, 1HNMR, 13CNMR and UV-Visible spectroscopic techniques. Synthesized dye was applied on polyester fabric by high temperature exhaustion dyeing and relevant characteristics of dyed fabrics including dye ability, build up, wash and light fastness were evaluated. Furthermore, the spectral properties of dye were investigated both in solution and on polyester fabrics. Also the position of color in CIELAB coordinates (L^*, a^*, b^*, H^*, C^*) was assessed. The results obtained have clearly demonstrated that the synthesized ANPN can dye the polyester fabric with high efficiency, good build up, excellent wash and light fastness.

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