Research Paper



Mesomorphism behaviour and photoluminescent properties of new asymmetrical 1,2-di(4-alkoxybenzylidene) hydrazines

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Abstract

 $\{1-[4-(n-Alkoxy)]-2-(4'-decyloxy)$ benzylidene $\}$ hydrazines (*n*-alkoxy = O(CH₂)_{*n*}H, *n* = 1–9, 12, 16 or 18), an asymmetrical series of 1,2-disubstituted hydrazines, were prepared in a simple two-step procedure as a part of our continuing work in evaluating hydrophobic azine compounds as photoluminescent liquid crystalline materials. The compounds were characterized spectroscopically and their liquid crystalline behaviour and luminescent properties were evaluated using polarized light optical microscopy, differential scanning calorimetry and X-ray powder diffraction techniques. The studies revealed that all of these compounds are liquid crystalline materials exhibiting photoluminescent properties in the crystalline and liquid crystal states.

Keywords

Mesomorphism behaviour, photoluminescent properties, asymmetrical azines, 1,2-disubstituted hydrazines



Introduction

Azine compounds have attracted considerable interest in several areas: in medical and biological applications,^{1,2} in the preparations of metal complexes^{3,4} and in some other applications.^{5,6}

Over past decades, liquid crystalline and photoluminescent liquid crystalline materials have attracted remarkable attention and interest due to their major contribution to the modern technology of LCDs. Over the past three years, a wide range of liquid crystalline or photoluminescent liquid crystalline compounds with fascinating properties have been prepared.^{7–18} We are interested in the synthesis of azine-type derivatives as photoluminescent mesogenic materials following a simple and practical method. In very recent reports we have described the preparation, liquid crystal and photoluminescent properties of 1,2-bis[4-(*n*-alkoxy)benzylidene]hydrazines (*n*-alkoxy = $O(CH_2)_nH$, n = 1-10, 12, 16 or 18), a symmetrical series of

1,2-disubstituted hydrazines¹⁹ and of {1-(4-propyloxy)-2-[4'-(*n*-alkoxy)]benzylidene}hydrazines (*n*-alkoxy = O(CH₂)_{*n*}H, *n* = 1, 2, 4–10, 12, 16 or 18)²⁰ and {1-[4-(*n*-alkoxy)]-2-(4'-dodecyloxy)benzylidene}hydrazines (*n*-alkoxy = O(CH₂)_{*n*}H, *n* = 1–10, 16 or 18),²¹ two asymmetrical series of 1,2-disubstituted hydrazines. Here we report the preparation, mesogenic behaviour and photoluminescent properties of {1-[4-(*n*-alkoxy)]-2-(4'-decyloxy)benzylidene}hydrazines (*n*-alkoxy = O(CH₂)_{*n*}H,

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Scheme 1. General synthetic diagram for the preparation of asymmetrical 1,2-di(4-alkoxybenzylidene) hydrazines.

n = 1-9, 12, 16 or 18), another closely related asymmetrical series of 1,2-disubstituted hydrazines.

transition band could be clearly observed. This peak might be hidden by the $\pi - \pi^*$ band.

Results and discussion

Synthesis and characterization of the compounds

Since we planned to make a comparison between the liquid crystal and photoluminescent properties of the previously synthesized asymmetrical series of 1,2-disubstituted hydrazines, we designated them Series I and II and the new compounds described here as Series III. The structures of Series I (**3a–I**; $\mathbf{R} = \mathbf{C}_{3}\mathbf{H}_{7}, \mathbf{R}^{1} = \mathbf{C}_{n}\mathbf{H}_{2n} + 1, n = 1, 2, 4-10, 12, 16,$ 18), Series II (4a–l; $R = C_{12}H_{25}$, $R^1 = C_nH_{2n}+1$, n =1-10, 16, 18) and Series III (5a–l; $R = C_{10}H_{21}$, $R^1 =$ $C_nH_{2n}+_1$, n = 1-9, 12, 16, 18) are shown in Scheme 1, which also shows the simple two-step procedure for the preparation of Series III. This involved (1) the preparation of a series of 4-alkoxybenzaldehydes (2a-m; R $C_nH_{2n}+_1$, n = 1-10, 12, 16, 18) via base-catalyzed alkylation (KOH/RBr) of 4-hydroxybenzaldehyde (1) and (2) the reaction of hydrazine hydrate with a 1:1 mixture of 4-decyloxybenzaldehyde (2j; $R = C_{10}H_{21}$) and a 4-(*n*-alkoxy)benzaldehyde (**2a–i**, **k–m**; $R^1 = C_n H_{2n} + 1$, n = 1-9, 12, 16, 18) in an acidic medium and under ambient conditions. It is important to mention that besides the formation of an asymmetrical azine as a major and desired product, two different symmetrical azines are formed as by-products (minors). The crude compounds were purified by column chromatography using *n*-hexane/ethyl acetate (19:1) as eluent.

The prepared compounds were characterized by their spectral data (IR, UV-Vis, NMR and mass spectra), in addition to microelemental analysis. The ¹H NMR and ${}^{13}C{}^{1}H$ NMR spectra of the prepared compounds showed signals corresponding to the various proton and carbon nuclei. The spectral data for each compound are listed in the Experimental section and two examples of ¹H NMR spectra are shown in Figure 1. The FTIR spectra of the prepared compounds displayed several characteristic bands, mainly due to C-H, C=C, C=N and C-O functional groups.²² All the UV-Vis spectra of asymmetrical azine compounds showed two absorption peaks in the ranges 240-244 and 331–334 nm. The first peak (located at a lower wavelength) was assigned to the n- σ^* transition, while the second one (more intense and located at higher wavelength) was attributed to a $\pi - \pi^*$ transition. No peak attributable to the $n - \pi^*$

Liquid crystalline behaviour of the compounds

The liquid crystalline behaviour of asymmetrical azine Series III (5a-l) was investigated using polarized light optical microscopy (POM) and confirmed with differential scanning calorimetry (DSC) and X-ray powder diffraction (XRD). Table 1 shows the details of the phase transition temperatures and thermodynamic data for these compounds. The observation of the mesophase textures under the polarized light optical microscope showed that the prepared compounds exhibited different notable liquid crystalline behaviour which can be summarized as follows: (1) all the compounds are mesomorphic materials displaying enantiotropic liquid crystal properties, six examples of which are shown in Figure 2; (2) all the compounds exhibited nematic mesophase upon heating and cooling. Upon heating, only nematic-thread like texture (N_{Th}) is clearly detected in all of these compounds. Upon cooling from the isotropic liquid, nematic-droplets (N_D) and N_{Th} like textures were observed; (3) compounds 5c to 5l displayed smectic A (SmA) (typical fan-shaped texture) either upon heating or upon both heating and cooling (5d-f showed the mesophase upon heating, while the rest of the compounds exhibited the mesophase upon both heating and cooling); (4) SmA mesophase appeared in 5e, 5k and 5l with wide ranges of temperatures: 64–103°C for 5e upon heating, 113–86°C for 5k upon cooling and 115-96°C for 5I upon cooling; (5) Schlieren-like textures were detected in 5a (upon heating and cooling). This behaviour is significantly different from that of Series I (3a-I) and slightly different from that of Series II (4a-I) which have been reported in our previous work.20,21 Compounds **3a-h** of Series I exhibited only nematic type textures, while 3i-I displayed both smectic and nematic mesophases (for more details see ref. 20). By contrast, in Series II (4a-l), only N_{Th} texture was clearly detected in all of the compounds upon heating. Upon cooling the isotropic liquid, compounds 4a-c showed N_D and N_{Th} like textures, whereas the rest of the compounds (4d-l) exhibited SmA mesophase (typical fan shaped mesophase) in addition to N_D and N_{Th} textures (for more details see ref. 21). The difference basically depends on the length of alkyl chain. For example, in Series I (3a-I), the constant alkyl chain is propyloxy (short tail) whereas constant dodecyloxy and



Figure 1. ¹H NMR spectra of (a) 5e and (b) 5i.

decyloxy alkyl chains (long tails) feature in Series II (**4a–l**) and Series III (**5a–l**) respectively. In general, a nematic droplet-like texture was formed upon cooling the mesogenic compounds from the isotropic phase. Then, the nematic thread-like texture appeared as a result of joining these droplets together.^{18–21,23} The enthalpy of the clearing point of asymmetrical azine compounds Series III (**5a–l**) was in the range 0.93–2.09 kJ mol⁻¹. This further confirmed the nematic to isotropic transition.^{18–21,23–25}

The DSC thermograms of all compounds of Series III (5a–l) (except 5a and 5b) displayed a couple of intense melting peaks (which can be attributed to transitions from different states of crystalline solid into nematic,

passing through a smectic mesophase) followed by a weak clearing peak detected at higher temperatures which was mainly due to the transition from nematic mesophase to the isotropic liquid. Two examples, **5a** and **5i**, are shown in Figure 3. This was also the case with almost all of the Series I (**3a–I**) and Series II (**4a–I**) compounds (for more details see refs 20 and 21). The thermograms of **5a** and **5b** were slightly different, showing an intense melting peak (which can be attributed to the transitions from different states of crystalline solid into nematic mesophase) followed by a weak clearing peak for the transition from nematic mesophase to the isotropic liquid.

No.	Compound	Phase transitions $T_{max}{}^\circ C$ (ΔH/ KJ.mol^1) [ΔS/ J.mole ⁻¹ .K ⁻¹] on first heating	Phase transitions $T_{max}{}^{\circ}C$ (ΔH/ KJ.mol^-I) [ΔS/ J.mole I.K-I] on first cooling
_	Sa	Cr–Sch 85.2 (18.11) [50.56] Sch–N–I 140.9 (2.46) [5.94]	I–N 138.5 (-1.49) [-3.62] N–Sch101.7 (-7.76) [-20.71] Sch–Cr 68.5 (-11.87) [-34.76]
2	5 b	Cr–N 98.5 (17.97) [48.37] N–I 148.5 (0.93) [2.21]	I–N 144.0 (-1.74) [-4.17] N–Cr 85.3 (-18.57) [-51.83]
m	Σ	Cr–SmA 80.0 (6.11) [17.31] SmA–N 93.7 (5.55) [15.13] N–I 136.7 (0.93) [2.27]	I–N 127.5 (-1.85) [-4.62] N–SmA 90.9 (-4.00) [-11.0] SmA–Cr 83.8 (-6.63) [-18.58]
4	5 d	Cr–SmA 67.3 (10.99) [32.30] SmA–N 99.1 (9.99) [26.85] N–I 138.8 (1.40) [3.40]	I–N 134.2 (-2.00) [-4.91] N–SmA–Cr 91.9 (-18.90) [-51.80]
Ŋ	Se	Cr ₁ -Cr ₂ -SmA 63.7 (12.39) [36.80] SmA-N 102.1 (13.37) [35.64] N-I 135.0 (1.01) [2.48]	I–N 131.4 (-1.91) [-4.72] N–SmA–Cr ₂ –Cr ₁ 96.5 (-25.33) [-68.55]
9	Sĩ	Cr ₁ -Cr ₂ -SmA 61.1 (13.02) [38.97] SmA-N 104.9 (9.47) [25.06] N-I 136.6 (1.44) [3.52]	I–N 130.8 (-1.78) [-3.41] N–SmA–Cr ₂ –Cr ₁ 99.5 (-22.41) [-60.16]
٢	50	Cr ₁ – Cr ₂ –SmA 63.8 (16.67) [49.50] SmA–N 108.3 (17.17) [45.03] N–I 133.9 (1.90) [4.67]	I–N 129.2 (-2.01) [-5.00] N–SmA–Cr ₂ –Cr ₁ 102.0 (-21.22) [-56.60]
ω	Sh	Cr ₁ – Cr ₂ –SmA 64.3 (14.71) [43.61] SmA–N 115.4 (17.71) [45.60] N–I 135.1 (1.59) [3.90]	I–N 129.9 (-1.82) [-4.52] N–SmA–Cr ₂ –Cr ₁ 107.4 (-22.45) [-56.60]
6	ū	Cr ₁ -Cr ₂ -Cr ₃ 75.0 (17.72) [50.92] Cr ₃ -SmA- N 119.8 (21.47) [54.66] N-1 132.4 (1.26) [3.11]	I– N 128.8 (-2.09) [-5.20] N–SmA–Cr ₃ 112.3 (-23.34) [-60.58] Cr ₃ –Cr ₂ –Cr ₁ 53.9 (-17.36) [-53.10]
0	S	Сг ₁ –Сг ₂ –Сг ₃ 75.4 (20.60) [59.13] Сг ₃ –SmA–N _{Th} 107.7 (12.88) [33.83] N _{Th} –N _D –I 123.5 (1.34) [3.38]	I–N _D I 19.4(-1.89) [-4.82] N _D –N _{Th} I 11.1 (-1.22) [-3.18] N _{Th} –SmA–Cr ₃ 101.0(-19.12)[-51.12] Cr ₃ –Cr ₂ –Cr ₁ 61.8 (-20.96) [-62.60]
=	Sk	Cr ₁ — Cr ₂ —SmA 83.0 (18.44) [51.80] SmA—N—I 120.1 (15.90) [40.45]	I–N–SmA 114.5 (-16.43) [-42.40] SmA–Cr ₂ 87.5 (-16.78) [-46.55] Cr ₂ –Cr ₁ 58.2 (-18.31) [-55.28]
12	5	Cr ₁ — Cr ₂ —SmA 91.7 (17.32) [47.49] SmA—N—I 119.1 (16.23) [41.39]	I–N–SmA 116.2 (-16.20) [-41.62] SmA–Cr ₂ 95.8 (-15.99) [-43.36] Cr ₂ –Cr ₁ 76.7 (-18.02) [-51.53]
Cr: Crystallin	e solid, SmA: Smectic A mes	ophase, SmC: Smectic C mesophase, Sch: Schlieren, N: Nematic mesophase, N _D : Nematic drople	sts- like texture, N _{Th} : Nematic thread-like texture, I: Isotropic liquid.

Table 1. Phase transition temperatures and thermodynamic data for Series III (**5a–I**) determined by DSC.

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Figure 2. Texture mesophases of some compounds obtained by POM: (a) Schlieren-like texture of **5a** at 133°C upon heating (b) Nematic thread-like texture of **5e** at 120°C upon heating (c) Nematic droplets-like texture of **5f** at 119°C upon cooling (d) Nematic droplets-like texture of **5h** at 117°C upon cooling (e) Fan-shaped texture of **5k** at 105°C upon cooling (f) Fan-shaped texture of **5l** at 100°C upon cooling.

The clearing temperatures of Series III (**5a–I**) compounds were found to decrease with increasing chain length. This revealed that the melting process appears to be governed by the random motion of chains, and is less affected by the van der Waals interactions between chains.^{26–28} This behaviour is quite similar to that of Series I (**3a–I**) and Series II (**4a–I**) compounds. Figure 4 shows a plot of the clearing temperatures as a function of the length of terminal alkoxy chain for asymmetrical azine compounds Series I (**3a–I**), Series II (**4a–I**) and Series III (**5a–I**). It can be clearly seen from the figure that the Series II compounds (**4a–I**) have the lowest clearing temperatures, while Series I compounds (**3a–I**) have the highest. In all three series of compounds, the odd–even effect can be clearly seen from this figure. Moreover, the effect was more pronounced for the shorter propyloxy terminal chain of Series I (blue line).

X-ray powder diffraction

The mesomorphic structure of some Series III compounds (5e-1) as liquid crystalline was studied by powder XRD. The diffractograms were measured at room temperature and at the mesophase temperature for each of these compounds (as an example, the XRD spectra of **5k** at room temperature and at 105°C are shown in Figure 5). The



Figure 3. DSC curves of (a) **5a** and (b) **5i**; red line: heating cycle, blue line: cooling cycle. Note. Coloured version of this figure is available online.

diffractogram measured at room temperature of **5k** (Figure 5(a)) displayed a lamellar structure exhibiting several intense reflections in the range $2\theta = 4-29^{\circ}$, whereas an intense reflection (in a small-angle region) and a broad halo (displayed in the wide-angle region as an indication of

liquid-like chain motion) appeared in the diffractograms measured at the mesophase temperatures (e.g. Figure 5(b)). The position and layer distance of the main peaks are listed in Table 2. It can be seen from this table that the layer distances of the mesophase are slightly longer than those



Figure 4. Clearing temperatures versus alkyl chain length for Series I (3a–I) (blue line), Series II (4a–I) (red line) and Series III (5a–I) (black line).

Note. Coloured version of this figure is available online.



Figure 5. XRD spectra of 5k at (a) room temperature and (b) 105°C.

observed in the crystalline state, suggesting a structural change from interdigitated alkyl chains to non- or partially-interdigitated alkyl chains.^{18,20,21,29}

Photophysical studies

Photoluminescence studies of Series III compounds (**5a–I**) were carried out in the solid state and compared with those of Series I compounds (**3a–I**) and Series II compounds (**4a–I**). These studies revealed that all of these compounds are luminescent at room temperature exhibiting an intense fluorescence band in the near UV region which is attributed to a ligand-centred transition. This band, in general, is located in the wavelength range 495–501.1 nm ($\lambda_{exc} = 248$ nm). For more details regarding the data of the fluorescence spectra of Series III compounds (**5a–I**), see the Experimental section; while for that of Series I compounds (**3a–I**) and Series II compounds (**4a–I**), see refs 20 and 21.

In order to demonstrate the luminescent behaviour of the mesomorphic compounds of Series III (5a–I) in the liquid

Table 2.	X-Ray	diffraction	data foi	· some	mesogenic
compound	ds.				

No.	Compound	Temperature (°C)	Position/2θ (°)	d spacing (Å)
1	5e	25	8.17	7.87
		25	10.41	6.93
		25	21.07	3.80
		25	24.11	3.00
		80	3.39	22.43
		80	20.07ª	3.75
2	5f	25	8.28	8.04
		25	10.36	7.05
		25	21.01	3.88
		25	24.18	3.03
		85	3.38	22.77
		85	20.09ª	3.80
3	5g	25	8.30	8.13
	C	25	10.42	7.04
		25	21.15	3.98
		25	24.16	3.07
		85	3.33	23.02
		85	20.06ª	3.81
4	5h	25	8.29	8.48
		25	10.43	7.08
		25	21.00	4.01
		25	24.11	3.09
		90	3.39	23.18
		90	20.03ª	3.85
5	5i	25	8.29	8.88
		25	10.28	7.14
		25	21.04	4.02
		25	24.17	3.10
		90	3.38	23.47
		90	20.06ª	3.87
6	5i	25	8.13	9.26
	-,	25	10.44	7.22
		25	20.99	4.09
		25	24.09	3.12
		95	3.42	24.57
		95	20.11ª	3.95
7	5k	25	8.35	10.58
		25	10.38	8.51
		25	21.00	4.23
		25	24.12	3.69
		105	3.34	26.40
		105	20.00ª	4.44
8	51	25	8.23	12.01
		25	10.17	10.32
		25	21.04	5.88
		25	24.11	4.77
		100	3.39	29.11
		100	20.08ª	6.09

^aBroad halo.

crystal state, the fluorescence spectra of these compounds were studied at different temperatures, ranging from the crystalline solid state to the isotropic liquid. Furthermore, the emission spectra were also recorded on cooling from the isotropic liquid until the re-solidification temperature (the spectra of **5**1 shown in Figure 6). Upon heating, all the

Figure 6. Normalized fluorescence spectra of **51** in the solid state at various temperatures: (black line) at room temperature, (blue line) at 50°C upon heating, (green line) at 70°C upon heating, (wine line) at 70°C upon cooling from the isotropic, (orange line) at 90°C upon heating, (red line) at isotropic liquid. Note. Coloured version of this figure is available online.

mesomorphic compounds showed almost similar behaviour of the emission band. As the temperature increases, the intensity of the fluorescence decreases (temperature-dependent emission) and it almost disappears at the clearing temperature. The emission maximum was found to be maintained in both the liquid crystal state and the crystalline solid state. This may indicate that neither the mesophase viscosity nor the structural changes affect the excited state. Upon cooling from the isotropic, the emission tends to be recovered and increases in intensity. These results are consistent with our previous work^{18,20,21} and others reported in the literature³⁰ which allow us to emphasize that the liquid crystal state does not quench the emission.

Experimental

All reagents and solvents were obtained from Sigma-Aldrich Co. and were used as supplied without further purification. Solvents were distilled from the appropriate drying agent immediately prior to use. 4-(n-Alkoxy)benzaldehydes $(n-\text{alkoxy} = O(CH_2)_n H, n = 1-10, 12, 16 \text{ or } 18)$ (2a-m), {1-(4-propyloxy)-2-[4'-(*n*-alkoxy)]benzylidene} hydrazines (*n*-alkoxy = $O(CH_2)_nH$, n = 1,2,4-10, 12, 16 or 18) (Series I, 3a-I) and $\{1-[4-(n-alkoxy)]-2-(4'-dodecy$ loxy)benzylidene}hydrazines (*n*-alkoxy = $O(CH_2)_nH$, *n* = 1-10, 16 or 18) (Series II, 4a-l) were prepared according to our previous work.¹⁹⁻²¹ The purity of the prepared compounds was determined by thin-layer chromatography using silica gel as stationary phase and n-hexane/ethyl acetate (16:1) as eluent. Column chromatography was carried out using silica gel 60 (200-300 mesh). Elemental analyses (C, H and N) were determined using an EuroEA 3000 Elemental Analyzer. A Varin Cary 100 Conc. spectrophotometer was used to measure the electronic spectra of the prepared compounds in the region 200-800 nm at room temperature for a 5 \times 10⁻⁵ M solution of the sample in

dichloromethane. A RF-6000 Shimadzu spectrofluorimeter equipped with a ThermoNeslab RTE7 bath was used to measure the fluorescence emissions of all the prepared compounds in the solid state. IR spectra were obtained in the region 4000-600 cm⁻¹ using an 8400S-FT-IR Shimadzu spectrophotometer. NMR spectra were recorded on a Bruker AV-II 500 MHz spectrometer (1H NMR at 500 MHz, ¹³C NMR at 125 MHz) in CDCl₃ using TMS as internal standard. Chemical shifts (δ) are given in ppm and coupling constants (J) are given in hertz (Hz). Mass spectra of the compounds were obtained using an Orbitrap LTQ XL-Thermo Fisher scientific mass spectrometer. DSC thermograms were measured in a nitrogen atmosphere at a heating and cooling rate of 5.0°C min⁻¹ using a Linseis STA PT-1000 equipment, which is precalibrated (the equipment was provided with an autocool system). Liquid crystalline mesophases of the compounds were obtained using a PW-BK5000R microscope equipped with a HS-400 (KER3100-08S) heating stage. The X-ray diffractograms at various temperatures were recorded on an XRD-6000 Shimadzu x-ray diffractometer equipped with an Anton Paar HTK1200 heating stage using Cu-K α radiation ($\lambda = 1.54178$ Å).

Synthesis of { I-[4-(n-Alkoxy)]-2-(4'-decyloxy) benzylidene}hydrazines (Series III, 5a–I); general procedure

Hydrazine hydrate (0.125 g, 2.5 mmol) was added to a mixture of 4-decyloxybenzaldehyde (**2j**; $\mathbf{R} = C_{10}H_{21}$) (0.66 g, 2.5 mmol) and a 4-(*n*-alkoxy)benzaldehyde compound (2.5 mmol) in ethanol (30 mL). To this mixture, acetic acid (100%, 0.1 mL) was added. The reaction mixture was stirred at room temperature for 1 h, during which a precipitate formed. This was filtered off, washed with cold water and purified by column chromatography using *n*-hexane/ ethyl acetate (19:1) as eluent. CAUTION: Hydrazine is a suspected carcinogen and therefore should be handled with appropriate precautions.

[1-(4-Methoxy)-2-(4'-decyloxy)benzylidene]hydrazine (5a; $R = C_{10}H_{21}$, $R^1 = CH_3$): Prepared from 4-methoxybenzaldehyde (2a; $R = CH_3$) (0.34 g) to give (5a; R = $C_{10}H_{21}$, $R^1 = CH_3$; light yellow solid; yield 60%; m.p. (clearing temperature) 141°C; $R_f = 0.43$; UV-Vis (λ /nm) $(\varepsilon_{\text{max}}/\text{dm}^3 \text{ mol}^{-1} \text{ cm}^{-1}): (241) (3700) (n-\sigma^*), (332) (13800)$ $(\pi-\pi^*)$; fluorescence (λ_{exc}^{max}/nm) (λ_{exc}^{max}/nm) : (248.0) (497.9); IR (v/cm⁻¹): 3057 (w, v (C-H) aromatic), 2955 (w, v_{as} (C–H) of CH₃ group), 2918 (s, v_{as} (C–H) of CH₂ group), 2873 (w, v_s (C–H) of CH₃ group), 2850 (m, v_s (C– H) of CH₂ group), 1624 (s, υ (C=N)), 1604, 1575 and 1508 (s, m, s, υ (C=C) aromatic), 1249 (vs, υ (C-O)); ¹H NMR (δ/ppm) : 0.96 (3H, t, J = 7.0 Hz, CH₃), 1.19–1.36 (14H, m, CH₂), 1.79 (2H, m, CH₂), 3.83 (3H, s, OCH₃), 4.05 $(2H, t, J = 6.9 \text{ Hz}, \text{OCH}_2), 6.88 (4H, d, J = 8.8 \text{ Hz}, \text{aro-}$ matic protons), 7.73 (4H, d, J = 8.8 Hz, aromatic protons), 8.59 (2H, s, CH=N); ¹³C NMR (δ/ppm): 10.5 (CH₃), 22.2 (CH₂), 26.6 (CH₂), 29.4 (CH₂), 29.8 (CH₂), 31.8 (CH₂), 56.2 (OCH₃), 69.4 (OCH₂), 114.2 (aromatic C–H), 126.7 (aromatic C), 130.4 (aromatic C–H), 161.2 (CH=N), 161.2 (aromatic C-O); MS m/z: 394. Anal. calcd for



C₂₅H₃₄N₂O₂: C, 76.10; H, 8.69; N, 7.10; found: C, 76.03; H, 8.54; N, 7.01%.

[1-(4-Ethoxy)-2-(4'-decyloxy)benzylidene]hydrazine (5b; R = $C_{10}H_{21}$, $R^1 = C_2H_5$): Prepared from 4-ethoxybenzaldehyde (2b; R $= C_2H_5$ (0.38 g) to give (**5b**; $R = C_{10}H_{21}$, $R^1 = C_2H_5$); light yellow solid; yield 61%; m.p. (clearing temperature) 149°C; $R_f=0.46$; UV-Vis (λ /nm) (ϵ_{max} /dm³ mol⁻¹ cm⁻¹): (242) (3220) $(n-\sigma^*)$, (333) (13940) ($\pi-\pi^*$); fluorescence (λ^{max}_{exc}/nm) (λ^{max}_{ems}/nm) nm): (248.0) (498.0); IR (ν /cm⁻¹): 3066 (w, ν (C–H) aromatic), 2956 (w, v_{as} (C–H) of CH₃ group), 2920 (s, v_{as} (C–H) of CH₂ group), 2874 (w-m, v_s (C–H) of CH₃ group), 2850 (m, v_s (C–H) of CH₂ group), 1622 (m, v (C=N)), 1605, 1575 and 1508 (s, m, m, υ (C=C) aromatic), 1247 (vs, υ (C–O)); ¹H NMR (δ/ppm): 0.91 $(3H, t, J = 7.0 \text{ Hz}, \text{CH}_3), 1.08 (3H, t, J = 7.0 \text{ Hz}, \text{CH}_3), 1.20-1.44$ $(14H, m, CH_2), 1.84 (2H, m, CH_2), 3.95 (2H, t, J = 6.9 Hz,$ OCH_2), 4.08 (2H, t, J = 6.9 Hz, OCH_2), 6.93 (4H, d, J = 8.9 Hz, aromatic protons), 7.74 (4H, d, J = 8.9 Hz, aromatic protons), 8.58 (2H, s, CH=N); ¹³C NMR (δ/ppm): 10.7 (CH₃), 14.81 (CH₃), 22.6 (CH₂), 26.3 (CH₂), 29.3 (CH₂), 29.7 (CH₂), 31.8 (CH₂), 63.3 (OCH₂), 69.2 (OCH₂), 114.4 (aromatic C–H), 126.4 (aromatic C), 130.3 (aromatic C-H), 161.2 (CH=N), 161.5 (aromatic C-O); MS *m/z*: 408. Anal. calcd for C₂₆H₃₆N₂O₂: C, 76.43; H, 8.88; N, 6.86; found: C 76.24, H 8.72, N 6.73%.

[1-(4-Propyloxy)-2-(4'-decyloxy)benzylidene]hydrazine (5c; $R = C_{10}H_{21}$, $R^1 = C_3H_7$): Prepared from 4-propyloxybenzaldehyde (2c; $R = C_3H_7$) (0.41 g) to give (5c; $R = C_{10}H_{21}$, $R^1 = C_3H_7$); light yellow solid; yield 67%; m.p. (clearing temperature) 138°C; $R_f = 0.49$; UV-Vis (λ /nm) $(\varepsilon_{\text{max}}/\text{dm}^3 \text{ mol}^{-1} \text{ cm}^{-1})$: (242) (5180) (n– σ^*), (333) (26650) $(\pi-\pi^*)$; fluorescence (λ^{max}_{exc}/nm) (λ^{max}_{ems}/nm) : (248.0) (495.0); IR (v/cm⁻¹): 3067 (w, v (C–H) aromatic), 2955 (m, v_{as} (C–H) of CH₃ group), 2918 (s, v_{as} (C–H) of CH₂ group), 2872 (w-m, v_s (C–H) of CH₃ group), 2851 (m, v_s (C–H) of CH₂ group), 1622 (m, v (C=N)), 1605, 1574 and 1508 (s, m, s, υ (C=C) aromatic), 1248 (vs, υ (C-O)); ¹H NMR (δ/ppm): 0.82 (3H, t, *J* = 7.3 Hz, CH₃), 0.96 (3H, t, *J* = 7.3 Hz, CH₃), 1.19-1.33 (14H, m, CH₂), 1.75 (4H, m, CH₂), 3.90 (2H, t, J = 6.6 Hz, OCH₂), 3.93 (2H, t, J = 6.6 Hz, OCH₂), 6.89 (4H, d, J = 8.8 Hz, aromatic protons), 7.70 (4H, d, J = 8.8 Hz, aromatic protons), 8.54 (2H, s, CH=N); ¹³C NMR (δ /ppm): 10.5 (CH₃), 14.1)CH₃), 22.4 (CH₂), 26.04 (CH₂), 29.3 (CH₂), 29.6 (CH₂), 32.0 (CH₂), 68.2 (OCH₂), 69.6 (OCH₂), 114.8 (aromatic C-H), 126.8 (aromatic C), 130.1 (aromatic C-H), 161.1 (CH=N), 161.7 (aromatic C-O); MS m/z: 422. Anal. calcd for C₂₇H₃₈N₂O₂: C, 76.74; H, 9.06; N, 6.63; found: C, 76.54; H, 9.01; N, 6.50%.

[1-(4-Butyloxy)-2-(4'-decyloxy)benzylidene]hydrazine (**5d**; $R = C_{10}H_{2l}$, $R^1 = C_4H_9$): Prepared from 4-butyloxybenzaldehyde (**2d**; $R = C_4H_9$) (0.45 g) to give (**5d**; $R = C_{10}H_{21}$, $R^1 = C_4H_9$); light yellow solid; yield 64%; m.p. (clearing temperature) 140°C; $R_f = 0.52$; UV-Vis (λ /nm) (ε_{max}/dm^3 mol⁻¹ cm⁻¹): (241) (3221) (n- σ^*), (331) (13780) (π - π^*); fluorescence (λ^{max}_{exc}/nm) (λ^{max}_{ems}/nm): (248.0) (497.1); IR ($\nu/$ cm⁻¹): 3066 (w, ν (C–H) aromatic), 2956 (w-m, ν_{as} (C–H) of CH₃ group), 2920 (s, ν_{as} (C–H) of CH₂ group), 2872 (w-m, ν_{s} (C–H) of CH₃ group), 2850 (m, ν_{s} (C–H) of CH₂ group), 1622 (m, ν (C=N)), 1604, 1573 and 1508 (s, m, m, ν (C=C) aromatic), 1247 (vs, ν (C–O)); ¹H NMR (δ /ppm): 0.88 (3H, t, J = 7.0 Hz, CH₃), 0.94 (3H, t, J = 7.0 Hz, CH₃), 1.25–1.52 (16H, m, CH₂), 1.80 (4H, m, CH₂), 4.00 (2H, t, J = 6.8Hz, OCH₂), 4.04 (2H, t, J = 6.8Hz, OCH₂), 6.97 (4H, d, J = 8.8 Hz, aromatic protons), 7.79 (4H, d, J = 8.8 Hz, aromatic protons), 8.59 (2H, s, CH=N); ¹³C NMR (δ /ppm): 13.8 (CH₃), 19.2 (CH₃), 22.8 (CH₂), 26.1 (CH₂), 29.2 (CH₂), 29.4 (CH₂), 29.7 (CH₂), 31.9 (CH₂), 67.9 (OCH₂), 68.2 (OCH₂), 114.7 (aromatic C–H), 126.7 (aromatic C), 130.2 (aromatic C–H), 161.1 (CH=N), 161.7 (aromatic C–O); MS *m/z*: 436. Anal. calcd for C₂₈H₄₀N₂O₂: C, 77.02; H, 9.23; N, 6.42; found: C, 77.13; H, 9.15; N, 6.34%.

[1-(4-Pentyloxy)-2-(4'-decyloxy)benzylidene]hydrazine (5e; $R = C_{10}H_{2l}$, $R^1 = C_5H_{1l}$): Prepared from 4-pentyloxybenzaldehyde (2e; $R = C_5H_{11}$) (0.48 g) to give (5e; R = $C_{10}H_{21}$, $R^1 = C_5H_{11}$; light yellow solid; yield 60%; m.p. (clearing temperature) 135°C; $R_f = 0.54$; UV-Vis (λ /nm) $(\varepsilon_{max}/dm^3 \text{ mol}^{-1} \text{ cm}^{-1})$: (242) (3140) (n- σ^*), (332) (16500) $(\pi-\pi^*)$; fluorescence (λ^{max}_{exc}/nm) (λ^{max}_{ems}/nm) : (248.0) (498.0); IR (v/cm⁻¹): 3074 (w, v (C-H) aromatic), 2956 (w, v_{as} (C–H) of CH₃ group), 2922 (s, v_{as} (C–H) of CH₂ group), 2872 (w, v_s (C–H) of CH₂ group), 2854 (m, v_s (C–H) of CH₂ group), 1622 (m, v (C=N)), 1604, 1573 and 1508 (s, m, s, v (C=C) aromatic), 1249 (vs, υ (C–O)); ¹H NMR (δ/ppm): 0.86 $(3H, t, J = 7.1 Hz, CH_3), 0.92 (3H, t, J = 7.1 Hz, CH_3), 1.22-$ 1.51 (18H, m, CH₂), 1.79 (4H, m, CH₂), 3.99 (2H, t, J = 6.8 Hz, OCH₂); 4.03 (2H, t, J = 6.8 Hz, OCH₂), 6.925 (4H, d, J = 8.8 Hz, aromatic protons); 7.74 (4H, d, J = 8.8 Hz, aromatic protons), 8.58 (2H, s, CH=N); ¹³C NMR (δ/ppm): 12.1 (CH₃), 18.1 (CH₃), 22.7 (CH₂), 26.4 (CH₂), 29.3 (CH₂), 29.5 (CH₂), 29.6 (CH₂), 32.0 (CH₂), 67.9 (OCH₂), 68.2 (OCH₂), 114.7 (aromatic C-H), 126.7 (aromatic C), 130.3 (aromatic C-H), 161.1 (CH=N), 161.7 (aromatic C-O); MS m/z: 450. Anal. calcd for C₂₉H₄₂N₂O₂: C, 77.29; H, 9.39; N, 6.22; found: C, 77.20; H, 9.22; N, 6.04%.

[1-(4-Hexyloxy)-2-(4'-decyloxy)benzylidene]hydrazine $(\mathbf{5f}; R = C_{10}H_{21}, R^1 = C_6H_{13})$: Prepared from 4-hexyloxybenzaldehyde (**2f**; $R = C_6 H_{13}$) (0.52 g) to give (**5f**; $R = C_{10} H_{21}$, $R^1 = C_6 H_{13}$; light yellow solid; yield 62%; m.p. (clearing temperature) 137°C; $R_f = 0.57$; UV-Vis (λ /nm) (ε_{max} /dm³ mol⁻¹ cm⁻¹): (242) (3250) (n– σ^*), (331) (17950) (π – π^*); fluorescence (λ_{exc}^{max}/nm) (λ_{ems}^{max}/nm): (248.0) (498.1); IR (v/cm^{-1}) : 3068 (w, v (C–H) aromatic), 2955 (w-m, v_{as} (C–H) of CH₃ group), 2920 (s, v_{as} (C–H) of CH₂ group), 2876 (w, v_{s} (C–H) of CH₂ group), 2850 (m, v_{e} (C–H) of CH₂ group), 1622 (m, υ (C=N)), 1604, 1575 and 1508 (s, m, m, υ (C=C) aromatic), 1246 (vs, υ(C–O)); ¹H NMR (δ/ppm): 0.90 (6H, t, J = 7.0 Hz, CH₃), 1.30–1.51 (20H, m, CH₂), 1.81 (4H, m, CH_2), 3.99 (2H, t, J = 6.9 Hz, OCH_2), 6.94 (4H, d, J = 8.9Hz, aromatic protons), 7.76 (4H, d, J = 8.9 Hz, aromatic protons), 8.57 (2H, s, CH=N); ¹³C NMR (δ/ppm): 10.7 (CH₃), 15.0 (CH₃), 22.5 (CH₂), 26.5 (CH₂), 29.4 (CH₂), 29.5 (CH₂), 29.9 (CH₂), 31.9 (CH₂), 67.9 (OCH₂), 68.2 (OCH₂), 114. 8 (aromatic C-H), 126.7 (aromatic C), 130.3 (aromatic C-H), 161.1 (CH=N), 161.6 (aromatic C-O); MS m/z: 464. Anal. calcd for C₃₀H₄₄N₂O₂: C, 77.54; H, 9.54; N, 6.03; found: C, 77.33; H, 9.42; N, 5.84%.

[1-(4-Heptyloxy)-2-(4'-decyloxy)benzylidene]hydrazine (**5g**; $R = C_{10}H_{21}$, $R^1 = C_7H_{15}$): Prepared from 4-heptyloxybenzaldehyde (2g; $R = C_7 H_{15}$) (0.55 g) to give (5g; $R = C_{10}H_{21}$, $R^1 = C_7H_{15}$); light yellow solid; yield 63%; m.p. (clearing temperature) 134 °C; $R_f = 0.60$; UV-Vis (λ /nm) (ϵ_{max} /dm³ mol⁻¹ cm⁻¹): (241) (3340) (n $σ^*$), (332) (13670) (π-π*); fluorescence ($λ^{max}_{exc}/nm$) (λ_{ems}^{max}/nm) : (248.0) (496.9); IR (ν/cm^{-1}): 3074 (w, ν (C-H) aromatic), 2955 (w-m, v_{as} (C-H) of CH₃ group), 2920 (s, v_{as} (C–H) of CH₂ group), 2873 (w, v_s (C–H) of CH₃ group), 2850 (m, v_s (C–H) of CH₂ group), 1622 (m, v (C=N)), 1604, 1575 and 1508 (s, m, m, υ (C=C) aromatic), 1249 (vs, υ (C–O)); ¹H NMR (δ /ppm): 0.90 (6H, t, J = 7.1 Hz, CH₃), 1.30-1.52 (22H, m, CH₂), 1.81 (4H, m, CH₂), 4.00 (4H, t, J = 6.8 Hz, OCH₂), 6.94 (4H, d, J = 8.8 Hz, aromatic protons), 7.74 (4H, d, J = 8.8 Hz, aromatic protons), 8.58 (2H, s, CH=N); ¹³C NMR (δ/ppm): 10.3 (CH₃), 14.6 (CH₃), 22.3 (CH₂), 26.7 (CH₂), 29.3 (CH₂), 29.5 (CH₂), 29.8 (CH₂), 31.9 (CH₂), 67.9 (OCH₂), 68.3 (OCH₂), 114.8 (aromatic C-H), 126.7 (aromatic C), 130.3 (aromatic C-H), 161.1 (CH=N), 161.7 (aromatic C–O); MS m/z: 478. Anal. calcd for C₃₁H₄₆N₂O₂: C, 77.78; H, 9.69; N, 5.85; found: C, 77.64; H, 9.61; N 5.73%.

[1-(4-Octyloxy)-2-(4'-decyloxy)benzylidene]hydrazine (**5h**; $R = C_{10}H_{21}$, $R^1 = C_8H_{17}$): Prepared from 4-octyloxybenzaldehyde (**2h**; $R = C_8 H_{17}$) (0.59 g) to give (**5h**; R = $C_{10}H_{21}$, $R^1 = C_8H_{17}$); light yellow solid; yield 62%; m.p. (clearing temperature) 135°C; $R_f = 0.62$; UV-Vis (λ /nm) $(\varepsilon_{\text{max}}/\text{dm}^3 \text{ mol}^{-1} \text{ cm}^{-1})$: (241) (3350) (n– σ^*), (331) (13790) $(\pi-\pi^*)$; fluorescence (λ^{max}_{exc}/nm) (λ^{max}_{ems}/nm) : (248.0) (498.0); IR (v/cm⁻¹): 3057 (w, v (C-H) aromatic), 2955 (w-m, v_{as} (C–H) of CH₃ group), 2918 (s, v_{as} (C–H) of CH₂ group), 2878 (w, v_s (C–H) of CH₃ group), 2850 (m, v_s (C– H) of CH₂ group), 1622 (m, υ (C=N)), 1604, 1573 and 1506 (s, m, m, υ (C=C) aromatic), 1247 (vs, υ (C–O)); ¹H NMR (δ/ppm) : 0.88 (6H, t, J = 7.0Hz, CH₃), 1.25–1.48 (24H, m, CH₂), 1.79 (4H, m, CH₂); 4.01 (4H, t, *J* = 6.8 Hz, OCH₂), 6.93 (4H, d, J = 8.8 Hz, aromatic protons), 7.74 (4H, d, J = 8.8 Hz, aromatic protons), 8.57 (2H, s, CH=N); ^{13}C NMR (δ/ppm): 10.3)CH₃), 14.9)CH₃), 22.4 (CH₂), 26.6 (CH₂), 29.35 (CH₂), 29.52 (CH₂), 29.90 (CH₂), 31.92 (CH₂), 67.91 (OCH₂), 68.38 (OCH₂), 114.72 (aromatic C-H), 126.7 (aromatic C), 130.2 (aromatic C-H), 161.1 (CH=N), 161.6 (aromatic C-O); MS m/z: 492. Anal. calcd for C₃₂H₄₈N₂O₂: C, 78.00; H, 9.82; N, 5.69; found: C, 77.88; H, 9.73; N, 5.62%.

[1-(4-Nonyloxy)-2-(4'-decyloxy)benzylidene]hydrazine (**5i**; $R = C_{10}H_{21}$, $R^1 = C_9H_{19}$): Prepared from 4-nonyloxybenzaldehyde (**2i**; $R = C_9H_{19}$) (0.62 g) to give (**5i**; $R = C_{10}H_{21}$, $R^1 = C_9H_{19}$); light yellow solid; yield 64%, m.p. (clearing temperature) 132 °C; $R_f = 0.64$; UV-Vis (λ /nm) (ε_{max} /dm³ mol⁻¹ cm⁻¹): (242) (3870) (n– σ^*), (332) (14250) (π – π^*); fluorescence (λ^{max}_{exc} /nm) (λ^{max}_{ems} /nm): (248.0) (497.0); IR (ν /cm⁻¹): 3052 (w, ν (C–H) aromatic), 2956 (w-m, ν_{as} (C–H) of CH₃ group), 2918 (s, ν_{as} (C–H) of CH₂ group), 2872 (w, ν_s (C–H) of CH₃ group), 2848 (m, ν_s (C– H) of CH₂ group), 1622 (m, ν (C=N)), 1604, 1575 and 1508 (s, m, m, ν (C=C) aromatic), 1247 (vs, ν (C–O)); ¹H NMR (δ /ppm): 0.87 (6H, t, J = 7.0Hz, CH₃), 1.25–1.48 (26H, m, CH₂), 1.78 (4H, m, CH₂), 3.99 (4H, t, J = 6.9 Hz, OCH₂), 6.925 (4H, d, J = 8.5 Hz, aromatic protons), 7.74 (4H, d, J = 8.5 Hz, aromatic protons), 8.58 (2H, s, CH=N); ¹³C NMR (δ /ppm): 10.3 (CH₃), 15.0 (CH₃), 22.4 (CH₂), 26.7 (CH₂), 29.4 (CH₂), 29.5 (CH₂), 29.9 (CH₂), 31.9 (CH₂), 67.9 (OCH₂), 68.4 (OCH₂), 114.7 (aromatic C–H), 126.7 (aromatic C), 130.3 (aromatic C–H), 161.1 (CH=N), 161.6 (aromatic C–O); MS *m*/*z*: 506. Anal. calcd for C₃₃H₅₀N₂O₂: C, 78.21; H, 9.94; N, 5.53; found: C, 78.03; H, 9.72; N, 5.44%.

[1-(4-Dodecyloxy)-2-(4'-decyloxy)benzylidene]hydrazine (5j; $R = C_{10}H_{21}$, $R^1 = C_{12}H_{25}$): Prepared from 4-decyloxybenzaldehyde (**2j**; $R = C_{12}H_{25}$) (0.73 g) to give (**5j**; $R = C_{10}H_{21}$, $R^1 = C_{12}H_{25}$; light yellow solid; yield 64%; m.p. (clearing temperature) 124°C; $R_f = 0.68$; UV-Vis (λ /nm) (ε_{max} /dm³ mol⁻¹ cm⁻¹): (240) (7100) (n– σ^*), (333) (26070) (π – π^*); fluorescence ($\lambda^{\text{max}}_{\text{exc}}/\text{nm}$) ($\lambda^{\text{max}}_{\text{ems}}/\text{nm}$): (248.0) (499.1); IR (v/cm⁻¹): 3057 (w, v (C–H) aromatic), 2950 (w-m, v_{as} (C–H) of CH₃ group), 2918 (s, v_{as} (C–H) of CH₂ group), 2878 (w, v_{s} (C–H) of CH₃ group), 2855 (s, v_s (C–H) of CH₂ group), 1622 (m, v(C=N)), 1600, 1573 and 1508 (s, m, m, v (C=C) aromatic), 1247 (vs, υ (C-O)); ¹H NMR (δ/ppm): 0.91 (6H, t, J = 7.0 Hz, CH₃), 1.03 (3H, t, J = 7.0 Hz, CH₃), 1.31–1.52 $(32H, m, CH_2)$, 1.80 (4H, m, CH₂); 4.00 (4H, t, J = 6.8 Hz, OCH_2), 6.96 (4H, d, J = 8.8 Hz, aromatic protons), 7.76 (4H, d, J = 8.8 Hz, aromatic protons), 8.59 (2H, s, CH=N); ¹³C NMR (δ/ppm): 10.3 (CH₃), 14.9 (CH₃), 22.3 (CH₂), 26.7 (CH₂), 29.3 (CH₂), 29.5 (CH₂), 29.8 (CH₂), 31.9 (CH₂), 67.9 (OCH₂), 68.3 (OCH₂), 114.7 (aromatic C-H), 126.7 (aromatic C), 130.3 (aromatic C-H), 161.1 (CH=N), 161.6 (aromatic C-O); MS m/z: 548. Anal. calcd for C₃₆H₅₆N₂O₂: C, 78.78; H, 10.28; N, 5.10; found: C, 78.61; H, 10.20; N, 5.03%.

[1-(4-Hexadecyloxy)-2-(4'-decyloxy)benzylidene] *hydrazine* (**5**k; $R = C_{10}H_{21}$, $R^1 = C_{16}H_{33}$): Prepared from 4-hexadecyloxybenzaldehyde ($2\mathbf{k}$; R = C₁₆H₃₃) (0.87 g) to give (**5k**; $R = C_{10}H_{21}$, $R^1 = C_{16}H_{33}$); light yellow solid; yield 62%; m.p. (clearing temperature) 120°C; $R_f = 0.71$; UV-Vis (λ /nm) (ϵ_{max} /dm³ mol⁻¹ cm⁻¹): (241) (4080) (n $σ^*$), (333) (17740) (π–π*); fluorescence (λ^{max}_{exc}/nm) (λ_{max}^{max}/nm) : (248.0) (498.1); IR (ν/cm^{-1}): 3066 (w, ν (C-H) aromatic), 2955 (w-m, v_{as} (C–H) of CH₃ group), 2918 (s, v_{as} (C–H) of CH₂ group), 2871 (w, v_s (C–H) of CH₃ group), 2850 (m, v_s (C–H) of CH₂ group), 1624 (m, v (C=N)), 1606, 1575 and 1510 (s, m, m, v (C=C) aromatic), 1251 (vs, v (C–O)); ¹H NMR (δ /ppm): 0.87 (6H, t, J = 7.0 Hz, CH₃), 1.21–1.45 (40 H, m, CH₂), 1.79 (4H, m, CH₂), 3.99 (4H, t, J = 6.8 Hz, OCH₂), 6.93 (4H, d, J = 8.8 Hz, aromatic protons), 7.75 (4H, d, J = 8.8 Hz, aromatic protons), 8.59 (2H, s, CH=N); ¹³C NMR (δ/ppm): 10.4 (CH₃), 14.8 (CH₃), 22.4 (CH₂), 26.7 (CH₂), 29.3 (CH₂), 29.6 (CH₂), 30.0 (CH₂), 31.9 (CH₂), 67.9 (OCH₂), 68.3 (OCH₂), 114.7 (aromatic C-H), 126.7 (aromatic C), 130.3 (aromatic C-H), 161.1 (CH=N), 161.6 (aromatic C-O); MS m/z: 604. Anal. calcd for C₄₀H₆₄N₂O₂: C, 79.42; H, 10.66; N, 4.63; found: C, 79.36; H, 10.57; N, 4.55%.

[1-(4-Octadecyloxy)-2-(4'-decyloxy)benzylidene] hydrazine (**5**I; $R = C_{10}H_{21}$, $R^1 = C_{18}H_{35}$): Prepared from 4-octadecyloxybenzaldehyde (**2**I; $R = C_{18}H_{35}$) (0.94 g) to give (**5**I; $R = C_{10}H_{21}$, $R^1 = C_{18}H_{35}$); light yellow solid; yield 64%, m.p. (clearing temperature) 119°C; $R_f = 0.73$; UV-Vis (λ /nm) (ε_{max} /dm³ mol⁻¹ cm⁻¹): (244) (3980) $(n-\sigma^*)$, (334) (17200) $(\pi-\pi^*)$; fluorescence $(\lambda^{\max}_{exc}/nm)$ (λ_{max}^{max}/nm) : (248.0) (498.3); IR (ν/cm^{-1}): 3070 (w, ν (C-H) aromatic), 2955 (w-m, v_{as} (C–H) of CH₃ group), 2918 (s, v_{as} (C–H) of CH₂ group), 2878 (w, v_s (C–H) of CH₃ group), 2848 (s, v_s (C–H) of CH₂ group), 1624 (m, v (C=N)), 1606, 1575 and 1508 (s, m, m, v (C=C) aromatic), 1251 (vs, v (C–O)); ¹H NMR (δ /ppm): 0.87 (6H, t, J = 7.0 Hz, CH₃), 1.32-1.53 (44H, m, CH₂), 1.80 (4H, m, CH₂), 3.98 (4H, t, J = 6.9 Hz, OCH₂), 6.93 (4H, d, J = 8.8 Hz, aromatic protons), 7.76 (4H, d, J = 8.8 Hz aromatic protons), 8.57 (2H, s, CH=N); ¹³C NMR (δ/ppm): 10.3 (CH₃), 14.9 (CH₃), 22.4 (CH₂), 26.7 (CH₂), 29.3 (CH₂), 29.6 (CH₂), 29.9 (CH₂), 31.9 (CH₂), 67.9 (OCH₂), 68.3 (OCH₂), 114.7 (aromatic C-H), 126.8 (aromatic C), 130.3 (aromatic C-H), 161.1 (CH=N), 161.5 (aromatic C-O); MS m/z: 633. Anal. calcd for C₄₂H₆₈N₂O₂: C, 79.69; H, 10.83; N, 4.43; found: C, 79.60; H, 10.68; N, 4.29%.

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