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Binary systems of non-mesogens with naphthalene derivatives

Yong fang Yao^{a,b,c}, H. N. Patel^d, Ashish Kumar Prajapati^e, Chetan B. Sangani^d, and Yong-Tao Duan^a

^aHenan Provincial Key Laboratory of Pediatric Hematology, Children's Hospital Affiliated to Zhengzhou University, Henan Children's Hospital, Zhengzhou Children's Hospital, Zhengzhou, China; ^bSchool of Pharmaceutical Science, Zhengzhou University, Zhengzhou, Henan, China; ^cMinistry of Education of China, Key Laboratory of Advanced Drug Preparation Technologies (Zhengzhou University), Zhengzhou, China; ^dShri Maneklal M. Patel Institute of Sciences & Research, kadi sarva vishwavidyalaya, Gandhinagar, Gujarat, India; ^eChemistry Department, Faculty of Science, The M.S. University of Baroda, Vadodara, India

ABSTRACT

Binary mixtures with both the components are non-mesogenic becomes an interesting and enlighten feature when they exhibit mesomorphism at ambient temperature. In the present study, we report five binary systems where two non-mesogenes exhibit smectic and/or nematic mesophases at ambient temperature over a certain range of concentration. All the binary systems contain one non-mesogenic naphthalene derivative. The study provides many binary systems of non-mesogens which exhibit mesophases at ambient temperatures. The terminal nitro chain is found to be responsible for the induction of both the nematogenic and smectogenic tendency in the present investigation of binary systems with two 'compatible' component.

KEYWORDS

Binary mixtures; component; naphthalene; nematogenic; nonmesogenic; smectogenic

Introduction

"Liquid crystals with low crystal-mesophase transition temperatures and with wide phase length have been the primary goal of organic chemists in recent years. Since the occurrence of a single component, room temperature liquid crystal with a wide mesophase range is rare. It is necessary to mix two or more low melting materials which may give room temperature mesomorphic mixture with wide mesophase length.

In general binary systems exhibiting mixed mesomorphism can be of three types: (i) where both the components are mesomorphic, (ii) Where only one component is mesomorphic and (iii) Where both the components are non-mesomorphic. Several workers [1–8] studied these types of mixed mesomorphism and discussed the structure-properties relationship in binary mixtures. Mesogenic binary systems of swallow-tailed and a double swallow-tailed compound [9], swallow-tailed and laterally branched liquid crystals [10], bi-swallow-tailed and a terminally-polar compound [11], as well as double-swallow tailed and calamitic electron acceptor compounds [12], are also appeared in the

literature. Kramarenko *et al.* [13–15] presented a phase diagram with a wide TGB phase region for certain binary liquid crystalline systems and then established the influence of the chiral dopant molecular structure on the type of phase diagram and the TGB regions. Dabrowski and Brodzik [16] discussed the influence of aliphatic chain length on the induction of smectic - A phase in polar bicomponent mixtures.

Good numbers of research papers are reported on binary mixtures in which induced mesophases are exhibited because of intermolecular hydrogen bonding [17–29]. It is well known that hydrogen bonds may be formed between carboxyl (-C = O-) and amide (-NH-) groups in poly (amides). Hydrogen bond formation between a carboxylate acceptor and a urea donor, and in some similar systems, was also reported recently [30–33].

Early studies on binary mixtures have suggested the formation of mixed mesomorphism from the pairs of compounds which are non-mesogenic by them [34-36]. Bennet and Jones [37] reported that 4-methoxy benzoic acid and 4-ethoxy benzoic acid which are non-mesogenic, do exhibit nematic mesophase in their binary mixtures. Lohar [38] has confirmed this result and showed that mixtures of the two acids exhibit mixed mesomorphism over a small range of temperature and concentration. Gupta and Vora [39] have reported a few binary mixtures where both the components are non-mesogenic exhibit mesomorphism with the non-linear tendency of the transition temperature curve. Vora et al. [40] observed the induced mesomorphism in the binary mixture were both the components belong to different homologous series. One of the components possesses a nitro group while another component is an ester with $-CH_3$ or $-COOC_4H_9$ terminal group. Physical properties of individual mesogens may or may not undergo modifications in their mixtures. Sometimes, the modifications become characteristic, thereby making the study of mixtures important. Earlier studies [40-46] have suggested the formation of mixed mesomorphism by mixing compounds where none, ore, or both of them are mesogens. The emergence of the mesophase [47], increase or decrease in the mixed mesomorphic ranges and thermal stabilities and study of the factors that influence the modifications have received a greater attention. The binary mixtures of mesogens have provided better formulations for applications in the different field [48]. Dave & Patel re-reported binary systems of structurally similar and dissimilar mesogens and nonmesogens. They also reported four binary systems consisting of structurally dissimilar mesogens and the study of their effect on mixed mesomorphism, arising due to the differences in structural characteristics of these components. Further, Dave et al. reported four binary systems consisting of azo-ester mesogens with nonmesogenic and mesogenic carboxy esters to examine the properties of mixed mesophases [49].

Prajapati *et al.* reported [50] the induction of smectic mesophase in the binary mixtures of esters having terminal broken alkyl chain in one of the components. Mahajan *et al.* [29] have reported induction of mesophases by intermolecular hydrogen bonding in a few binary mixtures were both the components are non-mesogenic by themselves. Recently, Prajapati and Pandya have reported [51] six binary systems where two nonmesogens exhibit smectic and/or nematic mesophase at ambient temperature over a wide range of concentration. With the continuation of previous work in naphthalene derivative; the series of 2'-Napthayl-4-*n*-alkoxy benzoates, there was methoxy to pentyloxy derivatives exhibit non-mesogenic same in 4-*n* octyloxy and hexadecyloxy phenyl



Scheme 1. Synthetic route to compound (i) to compound (vi).

azo-2'methalene. So above combination of non-mesogenus were prepared. They exhibited mesophase in present reported work.

Binary mixtures with both the components are non-mesogenic becomes an interesting and enlighten feature when they exhibit mesomorphism at ambient temperature. In the present study, we report few binary systems where two non-mesogenes exhibit smectic and/or nematic mesophases at ambient temperature over a certain range of concentration. All the binary systems contain one non-mesogenic naphthyl derivative.

Experimental

Preparation of compounds

The Scheme 1 shows the detail of the synthetic route for the preparation of following present compounds.

- i. 2-Naphthyl-4'-*n*-propyloxybenzoate (3 BN).
- ii. 2-Naphthyl-4'-*n*-butyloxybenzoate (4 BN).
- iii. 2-Naphthyl-4'-n-pentyloxybenzoate (5BN).
- iv. 4-(4'-n-Butyloxybenzoyloxy) nitrobenzoate (4 BNB).

Were synthesized by the condensation of respective 4-*n*-alkoxy benzoic acid with 2-naphthol [compound (i) - (iii)], 4-nitro phenol [compound (iv)] respectively by following the procedure described by Dave and Vora [52].

- i. 4-*n*-Octyloxy phenyl azo-2'-naphthalene (8PN).
- ii. 4-*n*-Hexadecyloxyphenylazo-2'naphthalene (16PN).

Were synthesized [53] by alkylation of 4-hydroxyphenylazo-2'naphthalene which in turn is obtained by diazotization and coupling of 2'-amino naphthalene with phenol.

2'-Naphthyl-4-*n*-pentyloxybenzoate:[5BN]

IR spectrum (KBr) ν_{max}/cm^{-1} : 2919, 1726 (-COO-), 1600, 1470, 1428, 1312, 1240, 1170, 840, 825, 735 cm⁻¹.

4-(4'-n-Butyloxybenzoyloxy) nitrobenzoate [4 BNB].

IR spectrum (KBr) ν_{max}/cm^{-1} : 2918, 1724 (-COO-), 1602, 1510 (-NO₂)1472, 1428, 1311, 1241, 1176, 842, 824, 738 cm⁻¹.

4-*n*-Octyloxy phenylazo-2'-naphthalene (8PN).

. IR spectrum (KBr) ν_{max}/cm^{-1} : 2950, 2835, 1610 (-N = N-) 1590, 1505, 1470, 1415, 1270, 1150, 875, 830, 755 cm $^{-1}$

Preparation of binary mixtures

Both the components were weighed accurately in known proportion and melted together in fusion tubes. The mixtures were thoroughly mixed to obtain a homogeneous mixture. The melt was quenched and the solid obtained was ground finely and was used for determining transition temperatures.

The transition temperatures were determined by using a Leitz Laborlux 12 POL microscope provided with a heating stage. The transition temperatures were determined on heating as well as cooling.

System No.	Component A		Component B
1	3BN	+	4BNB
11	4BN	+	4BNB
III	5BN	+	4BNB
IV	8PN	+	4BNB
V	16PN	+	4BNB

Results and discussion



Plate 1.1. Photomicrograph of system III for 37.92 mole% at 51 °C.



Plate 1.2. Photomicrograph of system IV for 08.66 mole% at 52 °C.

Table 1	1.	Transition	temperatures	(°C)	for	system-I.
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Values in the parenthesis indicate monotropy.

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Table 2. Transition temperatures (°C) for the system-II.

[A]

C₄H₀O

	[B] (-NO ₂			
Mole % of A	Cr		N		I		
00.00	•	60			•		
06.61	•	70	(•	54)	•		
13.74	•	76	(•	56)	•		
21.45	•	83	(•	60)	•		
29.82	•	92	(•	64)	•		
38.93	•	98	(•	68)	•		
48.88	•	106			•		
59.80	•	110			•		
71.83	•	114			•		
85.16	•	116			•		
100.00	•	120			•		

000

Table 3. Transition temperatures (°C) for system-III.

	[A]	C ₅ H ₁₁ O	- Co				
	[B]	C ₄ H ₉ O			NO ₂		
Mole % of A	Cr		SmA		N		I
00.00	•	60					•
06.35	•	64			(•	61)	•
13.24	•	47			•	67	•
20.74	•	57			•	74	•
28.93	•	55			•	75	•
37.92	•	85	(•	57)	(•	71)	•
47.81	•	90			(•	70)	•
58.76	•	92			(•	68)	•
70.96	•	94			(•	65)	•
84.61	•	95			(•	61)	•
100.00	•	96					•

Values in the parenthesis indicate monotropy.

The following ten binary mixtures having both the components non-mesogenic are studied.

The transition temperatures of the system I-V are given in Tables 1–5 respectively, the plots of transition temperatures versus mole percent are given in Figures 1–5.

System I: 3BN + 4BNB

	ton temperatures				
	[A] C ₈ H ₁	70-	-N=N		
	[B] C ₄ H ₉		coo	NO ₂	
Mole % of A	Cr		N		I
00.00	•	60			•
00 66			,		-
00.00	•	63	(•	56)	•
17.94	•	63 69	(• (•	56) 62)	•
17.94 27.27	• •	63 69 72	(• (• (•	56) 62) 70)	•
17.94 27.27 36.84	• • •	63 69 72 87	(• (• (•	56) 62) 70) 84)	• • •
17.94 27.27 36.84 46.66	• • • •	63 69 72 87 89	(• (• (• (•	56) 62) 70) 84) 85)	• • •
17.94 27.27 36.84 46.66 56.75	• • • •	63 69 72 87 89 90	(• (• (• (• (•	56) 62) 70) 84) 85) 85)	• • • • •
17.94 27.27 36.84 46.66 56.75 67.12	• • • •	63 69 72 87 89 90 92	(• (• (• (• (•	56) 62) 70) 84) 85) 85) 85)	• • • • •
00.00 17.94 27.27 36.84 46.66 56.75 67.12 77.77	• • • • •	63 69 72 87 89 90 92 94	(• (• (• (• (• (•	56) 62) 70) 84) 85) 85) 85) 85) 86)	• • • • • • • • •
00.00 17.94 27.27 36.84 46.66 56.75 67.12 77.77 87.73	• • • • •	63 69 72 87 89 90 92 94 95	(• (• (• (• (• (•	56) 62) 70) 84) 85) 85) 85) 86) 86)	• • • • • • • • • • • • • • • • • • • •

Table 4. Transition temperatures (°C) for system-IV.

Values in the parenthesis indicate monotropy.

Table 5. Transition temperatures (°C) for system–V.





Mole % of A 00.00	Cr		I		
	•	60			•
06.90	•	65	(•	63)	•
14.29	•	73	(•	70)	•
22.24	•	78	(•	76)	•
30.79	•	80	(•	78)	•
40.02	•	86	(•	83)	•
50.02	•	87	(•	85)	•
60.89	•	88	(•	85)	•
72.74	•	90	(•	85)	•
85.72	•	92	(•	86)	•
100.00	•	95			•

Values in the parenthesis indicate monotropy.



Figure 1. Phase behavior of system I.



Figure 2. Phase behavior of system II.



Figure 3. Phase behavior of system III.

Reference to binary phase diagram (Figure 1) exhibits an induced monotropic nematic phase between 06.89 to 39.99 mole percentage concentrations of 3BN. The nematic isotropic transition temperature curve is extrapolated on the side where monotropic



Figure 4. Phase behavior of system IV.



Figure 5. Phase behavior of system V.

nature is observed up to certain concentration range [36,37, 54–57]. The latent transition temperature obtained by extrapolation for 4BNB is 51° C.

System II: 4BN + 4BNB

Reference to binary phase diagram (Figure 2) exhibits an induced monotropic nematic phase between 06.61 to 38.93 mole percentage concentrations of 4BN. The nematic isotropic transition temperature curve is extrapolated on the side where monotropic nature is observed up to certain concentration range [36,37, 54–57]. The latent transition temperature obtained by extrapolation for 4BNB is 52 °C.

System III: 5BN + 4BNB

Reference to binary phase diagram (Figure 3) exhibits an induced monotropic nematic phase between 06.35 to 84.81 mole percentage concentration of 5BN and monotropic smectic A appears only at 37.92% concentration of 5BN. The latent transition temperature obtained by extrapolation for 4BNB is 51 $^{\circ}$ C.

System IV: 8PN + 4BNB

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Reference to binary phase diagram (Figure 4) shows that the system is non-mesogenic when the concentration of 8PN is low but the monotropic nematic phase is induced from 08.6 to 87.73 mole percentage concentration of 8PN. The latent transition temperature obtained by extrapolation for 4BNB is 52 °C.

System V:16PN + 4BNB

Reference to binary phase diagram (Figure 5) shows that the system is non-mesogenic when the concentration of 16PN is low but the monotropic nematic phase is induced from 06.90 to 85.72 mole percentage concentration of six PN. The latent transition temperature obtained by extrapolation for 4BNB is 50 °C.

Conclusion

The study provides several binary systems of non-mesogens which exhibit mesophases at ambient temperatures.

The terminal nitro chain is found to be responsible for the induction of both the nematogenic and smectogenic tendency in the present investigation of binary systems with two 'compatible' component i. e. one with an ester central linkage with naphthalene moiety and the other with an ester central linkage having a nitro chain. In the present work, even though the two components are 'compatible' i.e. one having central azo linkage and other with an ester central linkage induced nematic/smectic phase in the binary system **IV** and **V** binary system. The study provided the few binary systems of non-mesogens which exhibited liquid crystalline properties at ambient temperatures.

Values in the parenthesis indicate monotropy.

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