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Phase behaviour of the binary mixtures of substituted analogues of LC compounds of the type 4-(4′-substituted phenylazo)-1-naphthyl-4″-alkoxybenzoates

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Abstract

Binary mixtures made from components of the four series of the molecular structure 4-$C_nH_{2n+1}O$–C_6H_4–COO–$C_{10}H_6$–N=N–C_6H_4–X, in which both components bear the same alkoxy group (n = 8, 10, 12, or 14) but of different substituent (X = CH_3O, CH_3, Cl, or NO_2), were prepared and characterized for their phase behaviour. Transition temperatures of mixtures were measured by differential scanning calorimetry and identified by polarized-light microscopy. The dependence of the phase behaviour of mixed systems upon differences in the electronic nature of the terminal group, X, attached to both components as well as the length of the alkoxy group, was investigated from the phase diagrams constructed for the various binary combinations. The nematic order parameters are in accordance with the conclusions drawn.

Keywords: 4-(4′-Substituted phenylazo)-1-naphthyl-4″-alkoxybenzoates; Phase diagrams; Nematic phase

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1. Introduction

In a rod-shaped LC compounds, lateral substitution decreases the thermal stability of both solids and mesophases (smectic and nematic) [1–6]. Generally, it decreases the breadth of the unsubstituted molecule and, consequently, the long narrow molecules will be forced apart, so reducing the strength of the intermolecular lateral attractions. The stability of both solid- and meso-phases, depends to greater or lesser extent on these cohesive forces in order to maintain the parallel orientations of the molecules. Expectedly, a side substituent which bridges the 2- and 3-positions, as in the 1,4-naphthalene derivatives, the bulk of these molecules is so great that it reduces its melting temperature (T_m) and unfortunately, its mesophase stability (T_C) too. Dave et al. [7–10] synthesized a number of Schiff's base homologous series having naphthalene moiety in the molecular core. The effect of chemical composition on 1,4-, 1,5-, and 2,6-substituted naphthalene derivatives has also been studied [11–14]. Prajapati et al. [15–17] prepared series of compounds based on the terminal 2-amino-naphthalene and found them possessing only the nematic phase.

On the other hand, 3-ring molecules are known to possess relatively high mesophase stability compared to the corresponding 2-ring molecules. Furthermore, comparison of the two 3-ring molecules, bearing identical terminal substituents, namely, the unsymmetrically substituted benzoates of hydroquinone [18], and the substituted phenylazophenyl 4-alkoxy benzoates [19], revealed that the replacement of one of the ester group in the former by an azo group to give the latter compounds, resulted in an increase in the stability of the mesophase. Thus, the azo/ester 3-ring molecules were chosen in which the central benzene ring is replaced by a laterally protruded 1,4-naphthalene moiety aiming to obtaining compounds with low melting temperatures and still retaining adequately high mesophase stability. In a previous work, such compounds of structure **I** have been prepared and their phase behaviour investigated in the pure state [20]. All compounds investigated were found to be only nematogenic and, irrespective of the terminal polar group X, the mesophase stability of compounds was found to decrease systematically upon increasing the chain length of the other terminal, alkoxy, group. Alternatively, there was no systematic variation of their melting temperatures with the chain length. Comparison of the 1,4-

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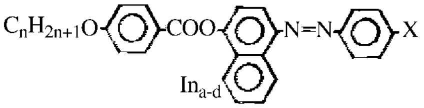
naphthalene (**I**) with the 1,4-phenylene (**II**) analogues revealed that, the lateral inclusion of the extra-fused benzene ring results in a reduction in the mesophase stability of the compounds.

Looking forward to obtaining LC materials with low melting points and reasonable mesophase stability, attention is directed towards binary mixtures of analogues bearing substituents X of different polarities aiming, hopefully, to obtaining eutectic compositions with low melting temperatures. The goal of the present study is to prepare series of binary mixtures made from any two analogues bearing different substituent X, but of the same alkoxy-chain length; and the study of their effect on the mixed meso- and solid-phase behaviour.



$In_{a\text{-}d}$

I8, n = 8; **I**10, n = 10; **I**12, n = 12; **I**14, n = 14
a, X = CH_3O; b, X = CH_3; c, X = Cl; d, X = NO_2

2. Experimental

2.1. Materials

Chemicals have been purchased from the following companies: Aldrich, Wisconsin, USA, E. Merk, Darmstadt, Germany, and Fluka, Buchs, Switzerland.

2.2. Physical characterization

Calorimetric measurements were carried out using a PL-DSC of Polymer Laboratories, England. The instrument was calibrated for temperature, heat and heat flow according to the method recommended by Cammenga et al. [21]. DSC measurements were carried out for small samples (2–3 mg) placed in sealed aluminum pans. All of the thermograms have been achieved at a heating rate of 10 °C/min in inert atmosphere of nitrogen gas (10 ml/min). Transition temperatures were estimated as the maximum of the corresponding peak.

Transition temperatures were checked and types of mesophases identified (by the contact method) for all mixtures prepared, with a standard polarized-light microscope PLM (Wild, Germany) attached to a home made hot-stage.

For phase diagrams, binary mixtures of any two analogues, covering the whole composition range, were prepared by melting the appropriate amounts of each component, stirring to give an intimate blend, and then cooling with stirring to room temperature. The transition temperatures obtained for the prepared blends, as measured by both DSC and PLM, agreed within 2–3 °C. In the phase diagrams, constructed by plotting transition temperatures versus mixture composition, the symbol “≡” denotes nematic–isotropic, “◯” solid–mesophase transitions, “≡” eutectic temperature and “·” order parameter.

3. Results and discussion

Transition temperatures and enthalpies of the investigated compounds, as reported previously [20], are summarized in Table 1. In the table, the following symbols are given, respectively, for the various transition temperatures and enthalpies: crystal–nematic ($T_{C\text{–}N}$ or T_m and $\Delta H_{C\text{–}N}$), nematic–isotropic ($T_{N\text{–}I}$ or T_C and $\Delta H_{N\text{–}I}$), and the mesomorphic ranges ($\Delta T = T_C - T_m$). It can be seen from Table 1 that all compounds investigated are of high melting (T_m) and clearing (T_C) points. In all binary mixtures investigated only nematic phases were detected.

Table 1
Transition temperatures (°C) and transition enthalpies (kJ/mol) of the homologous series ($\mathbf{I}n_{a\text{–}d}$)

Compound number	X	T_m ($T_{C\text{–}N}$)	ΔH_m ($\Delta H_{C\text{–}N}$)	T_C ($T_{N\text{–}I}$)	ΔH_C ($\Delta H_{N\text{–}I}$)
I8_a	CH_3O	112.7	20.2	178.1	0.71
I10_a		108.6	22.6	167.3	0.67
I12_a		104.8	13.8	153.0	0.56
I14_a		109.6	25.6	142.5	0.33
I8_b	CH_3	139.0	21.2	149.8	1.01
I10_b		105.7	17.8	148.2	0.63
I12_b		115.0	15.0	139.1	0.42
I14_b		121.9	11.8	137.3	0.17
I8_c	Cl	106.1	21.7	160.5	0.88
I10_c		98.8	23.3	138.9	0.71
I12_c		105.8	24.9	128.9	0.46
I14_c		97.4	24.5	130.1	0.42
I8_d	NO_2	152.2	19.4	184.8	0.67
I10_d		121.4	23.5	177.6	0.46
I12_d		130.4	17.9	168.1	0.29
I14_d		131.2	22.6	161.0	0.08

3.1. Binary mixtures of components bearing different electron-donating substituents ($\mathbf{I}_a/\mathbf{I}_b$)

Fig. 1a–d collects the phase diagrams constructed for the four binary systems prepared from the CH_3O- and CH_3-substituted components in each of the four series **I**8 to **I**14, respectively. Fig. 1 shows that, in all of the four systems, the mesophase–isotropic transition temperatures (T_C) vary linearly with composition. Such simple binary phase behaviour is expected, since both components of any mixture bear a substituent with comparable electronic nature, i.e., both have an electron-donating nature. With respect to their solid phase, all diagrams, except that of the system **I**12_a/**I**12_b, exhibit eutectic behaviour in their solid–mesophase transition temperatures (T_m). The mixed solid phase of the dodecyloxy ($n = 12$) substituted analogues (**I**12_a/**I**12_b), exhibit solid solution throughout the whole composition range, indicating a compatible crystalline structure between both components. In the solid phase of these systems, there is an irregular trend in the crystalline lattice of the solids, evidenced by the irregular variation of both T_m and ΔH_m (see Table 1), with increasing the alkoxy-chain length. Thus, in the lower homologues, $n = 8$ and 10, the CH_3O- and CH_3-substituted derivatives form heterogeneous mixtures which exhibit eutectic behaviour. Increasing the chain length to $n = 12$, resulted in compounds of compatible crystalline structures. Compatibility is again lost in the homologues with $n = 14$.

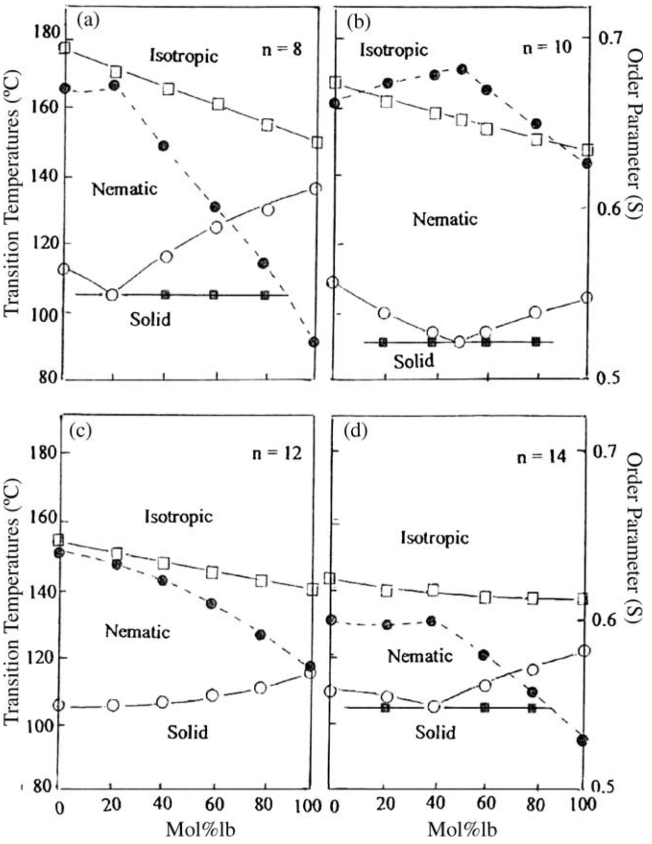


Fig. 1. Phase diagrams for the binary mixtures of the methoxy- and methyl-substituted analogues: (a) **I**8$_a$/**I**8$_b$, (b) **I**10$_a$/**I**10$_b$, (c) **I**12$_a$/**I**12$_b$, and (d) **I**14$_a$/**I**14$_b$.

3.2. Binary mixtures of components bearing different electron-withdrawing substituents (I$_c$/I$_d$)

The phase diagrams of the binary systems **I**$_c$/**I**$_d$ are illustrated in Fig. 2a–d. Referring to Fig. 2, the phase diagrams of the chloro- with the nitro-substituted analogues, each, show near linear T_C-composition dependence in their mixed nematic phase. This results, again, reflects the nearly similar behaviour effected by substituent of closely related electronic nature, i.e., both are of electron-withdrawing nature. The crystal–nematic transition temperatures, as a function of temperature, behave in a different manner, which vary according to the alkoxy-chain length, *n*. Thus, for the analogues, *n* = 8, it seems that both components **I**8$_c$ and **I**8$_d$ possess compatible crystalline structures and, consequently, their phase diagram exhibits regular variation of T_m with composition. For higher homologues, the situation is quite different, components possess different crystalline structures thus gave mixtures that exhibit eutectic behaviour.

3.3. Binary mixtures of components one bearing an electron-donating (I$_a$ or I$_b$) and the other bearing electron-withdrawing substituents (I$_c$ or I$_d$)

The phase diagrams constructed for binary mixtures of components one bearing electron-donating (CH_3O or CH_3) group; i.e. **I**$_a$ or **I**$_b$, and the other bearing electron-withdrawing (Cl or NO_2) group, i.e. **I**$_c$ or **I**$_d$, are presented in Figs. 3–6. Figs. 3 and 4, which illustrates, respectively, the phase diagrams of binary mixtures composed from the CH_3O-, or CH_3-substituted derivatives, as the first component, and the chloro-derivative as the second, show that all phase diagrams in both systems, irrespective of the alkoxy-chain length, show more or less, linear composition dependence of T_C. These results reflect the similar mesophase behaviour effected by the CH_3O (or CH_3) and chloro substituents. Their crystal–nematic transitions behave differently; where different substituent, X, seemed to result in compounds of different crystalline structures. Thus, in the system **I**$_a$/**I**$_c$, for *n* = 8 (Fig. 3a), addition of **I**8$_c$ to its methoxy analogue (**I**8$_a$) disrupts the crystal lattice of **I**8$_a$ accompanied by a sudden decrease in the melting point (T_m). Alternatively, addition of **I**8$_a$ to **I**8$_c$ again leads to a decrease in T_m as a result of crystal lattice modification. There exists a range of composition where a linear T_m-dependence was observed indicating the formation of solid solution, within this range of composition. Increasing the alkoxy-chain to *n* = 10 and 12, resulted in compounds of compatible crystal structure indicated by the gradual variation of T_m with composition (Fig. 3b and c). Further increase in *n* (up to 14) has led to a system that exhibits eutectic solid behaviour together with an intermediate composition range of solid solution. This is evidenced, as will be shown below, by the regular variation of order parameter within this range of composition. Such irregular

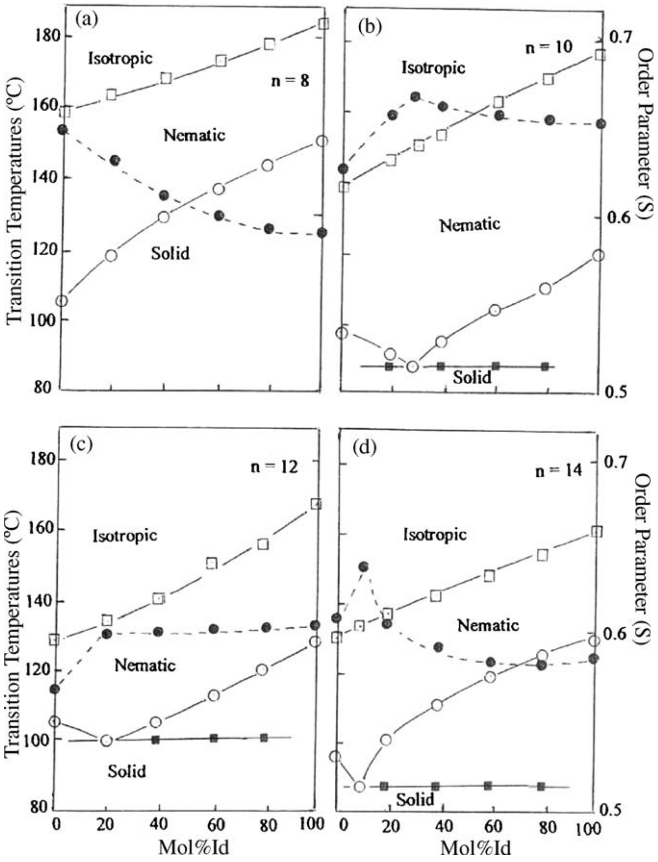


Fig. 2. Phase diagrams for the binary mixtures of the chloro- and nitro-substituted analogues: (a) **I**8$_c$/**I**8$_d$, (b) **I**10$_c$/**I**10$_d$, (c) **I**12$_c$/**I**12$_d$, and (d) **I**14$_c$/**I**14$_d$.

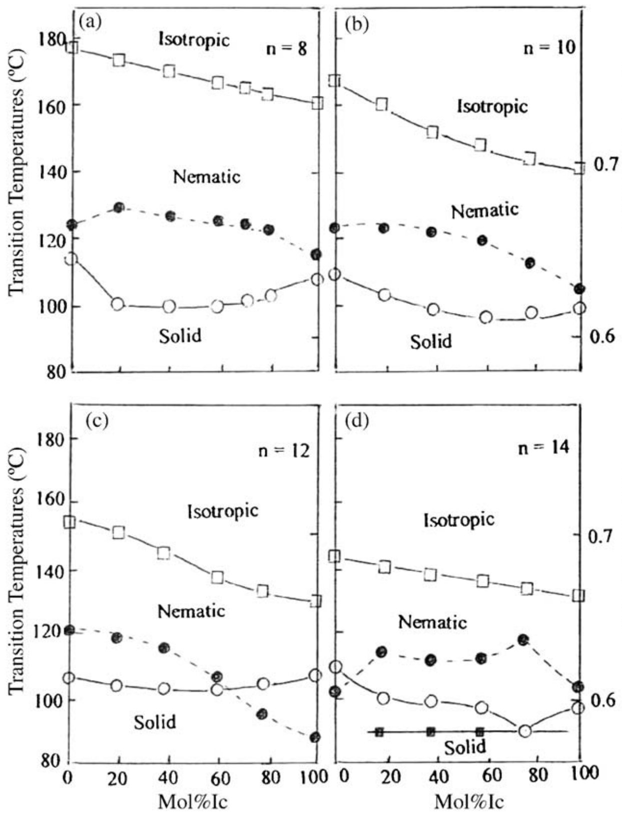


Fig. 3. Phase diagrams for the binary mixtures of the methoxy- and chloro-substituted analogues: (a) **I**8_a/**I**8_c, (b) **I**10_a/**I**10_c, (c) **I**12_a/**I**12_c, and (d) **I**14_a/**I**14_c.

behaviour, upon increasing *n*, can be explained in terms of lattice energy that is dependent on variation in molecular structure.

The corresponding phase diagrams for the systems $\mathbf{I}_b$/$\mathbf{I}_c$ (Fig. 4) indicates that, irrespective of the alkoxy-chain length, the stability of the nematic phase (T_C) is linearly dependent on composition, while their solid phase behaviour is chain-length dependent. A solid solution is formed, throughout the whole composition range, in the lowest homologues ($n = 8$), indicating the resemblance of crystal structures between the two components **I**8_b and **I**8_c. As "*n*" increases, compatibility is lost and eutectic behaviour was observed for higher homologues ($n = 10$–14). The discrepancy in behaviour between the two mixed systems $\mathbf{I}_a$/$\mathbf{I}_c$ and $\mathbf{I}_b$/$\mathbf{I}_c$ may be ascribed to differences in crystal structures of pure components of $\mathbf{I}_a$ and $\mathbf{I}_b$ as indicated previously from their own binary mixtures $\mathbf{I}_a$/$\mathbf{I}_b$.

The phase diagrams for the remaining binary systems, namely, $\mathbf{I}_a$/$\mathbf{I}_d$ and $\mathbf{I}_b$/$\mathbf{I}_d$ are depicted, respectively, in Figs. 5 and 6. Fig. 5a–d, which illustrates the phase diagrams of the binary mixtures composed from the CH_3O- and NO_2-substituted analogues, show that they are of typical eutectic type with T_C-composition exhibiting but slightly negative deviation from the linear dependence. This behaviour indicates that differences in the electronic nature between the CH_3O and NO_2 groups resulted in differences in the molecular arrangement in molecules possessing them and, consequently, their mixed systems show non-linear behaviour either in the solid or mesophases.

Surprisingly observed in the phase diagrams (Fig. 6) of the corresponding system $\mathbf{I}_b$/$\mathbf{I}_d$, constituting the CH_3- and NO_2-substituted analogues, compatibility of their solid phase, irrespective of the alkoxy-chain length, and negative deviation from the linear composition-dependence of T_C, were observed. Such negative deviation increases as n increases, indicating the disturbance in the molecular arrangement in the mesophase of one component by the addition of the second, thus decreasing its mesophase stability, T_C. The irregular trend in the mesomorphic range, which increases first as *n* is increased from 8 to 10, and then decreases in higher homologues can be attributed to the irregularity in the melting points. Again, the discrepancy observed between the two closely related binary systems, $\mathbf{I}_a$/$\mathbf{I}_d$ and $\mathbf{I}_b$/$\mathbf{I}_d$, is again attributed to the difference in the crystalline structures between components $\mathbf{I}_a$ and $\mathbf{I}_b$.

3.4. Nematic-order stability and miscibility

If a nematogenic compound is melted, the molecules are free to move whereby the positions of their ends are disorganized, whereas the parallel alignments of the molecules persists. At the solid–nematic transition, cohesive forces weaken profoundly [11]. Thus, although substantial variation in the nature of layer crystal lattices upon melting is very unprobable, nevertheless, a nematogenic arrangement may change greatly from the crystal of one homologue to another and not necessarily in a regular

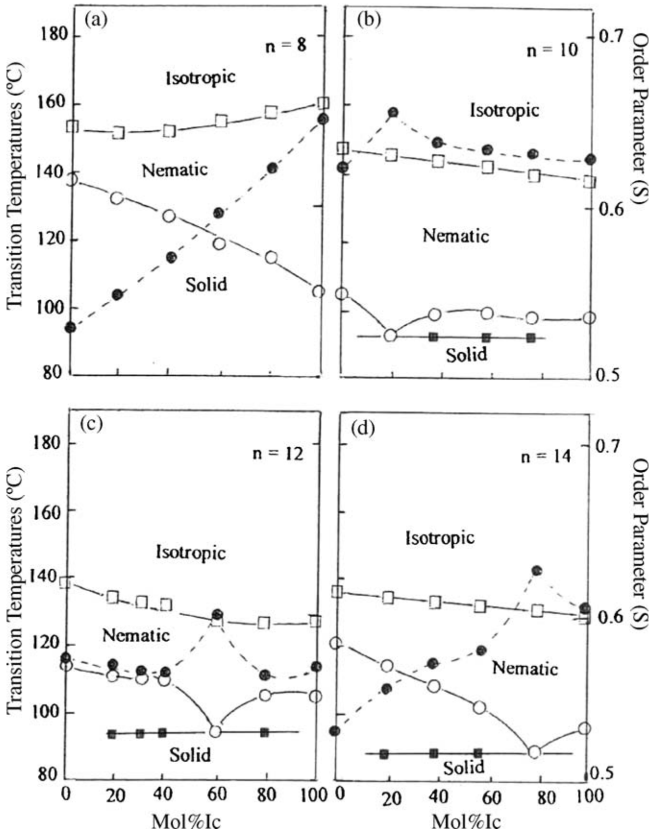


Fig. 4. Phase diagrams for the binary mixtures of the methyl- and chloro-substituted analogues: (a) **I**8_b/**I**8_c, (b) **I**10_b/**I**10_c, (c) **I**12_b/**I**12_c, and (d) **I**14_b/**I**14_c.

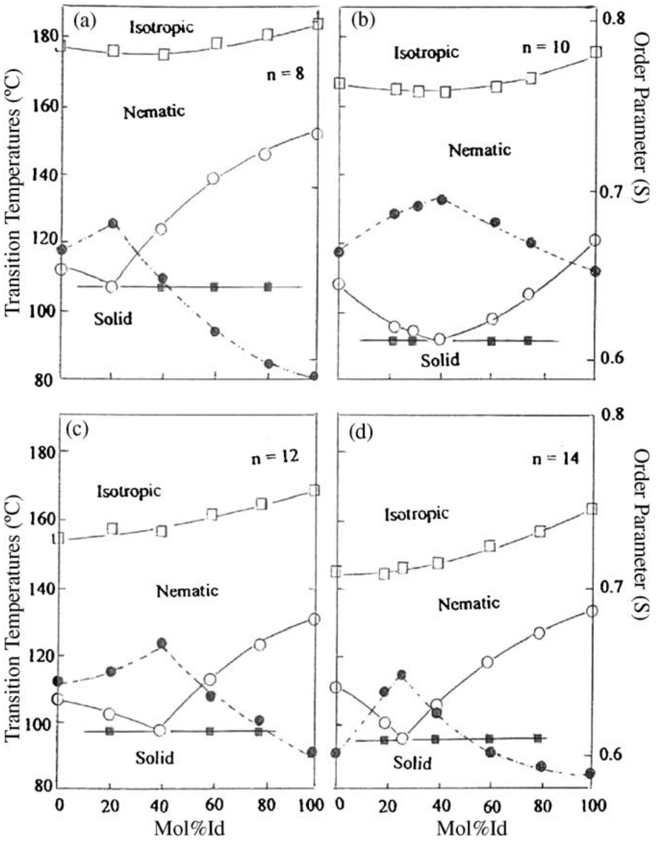


Fig. 5. Phase diagrams for the binary mixtures of the methoxy- and nitro-substituted analogues: (a) **I**8_a/**I**8_d, (b) **I**10_a/**I**10_d, (c) **I**12_a/**I**12_d, and (d) **I**14_a/**I**14_d.

fashion (see Table 1). Certainly, the solid-crystalline structure will affect the strength of the intermolecular forces of attraction and, therefore, the melting point of the nematogenic compound will depend on the exact nature of the crystal packing. In this respect, the nematic order parameter (*S*), just at the melting point, can be used to evaluate changes in the crystal structure in mixtures, if it is calculated as a function of mixture composition. This is made possible using Eq. (1), deduced by Hikmet and Lub [22]:

$$S = [1 - 0.98(Tm/TC)]^{0.22} \tag{1}$$

The *S* values calculated are drawn, as a function of mixture composition, in their corresponding phase diagrams. In this way, one may investigate the composition-dependence of the nematic mesophase arrangement, just at the melting temperature, without giving chance for the molecules to move. It has to be mentioned that in Eq. (1), T_m replaces *T*. Assuming an uncertainty in T_m and *T* equals ±1 K, the expected uncertainty in *S* does not exceed ±0.002.

As expected, the nematic order parameter, *S*, at $T = T_m$, varies with composition in a fashion coincident with that of T_m, except that *S* is higher for mixtures with lower T_m. This is due to the fact that the order parameter (*S*) for a mesophase decreases as the temperature is increased. Thus, for the mixtures of differently substituted analogues (Figs. 1–6), let us first take the mixtures of the methoxy- and methyl-substituted analogues as examples. For alkoxy-chain length $n = 8$, 10, and 14 carbons (Fig. 1a, b and d), the components of each mixture are of incompatible crystal structures and eutectic points are observed associated with peak values of *S*. For the mixture **I**12_a/**I**12_b (Fig. 1c), *S* (at T_m) varies more or less gradually with composition indicating the compatibility of the molecular arrangements of both components either in the solid or in the nematic phase. Compatibility of solids to form solid solution was observed before, either in the binary mixtures of the homologues of the investigated series [23], or in mixtures of their phenyl analogues, **II** [24]. In the binary mixtures of the methoxy- with the chloro-substituted analogues, for $n = 8$ (Fig. 3a) the addition of one component to the other, up to a certain composition, is accompanied by a drop in T_m and a consequent increase in *S*. This indicates that the presence of one component in the other facilitates the formation of the mesophase by weakening the cohesive forces within the crystal lattice. Within the intermediate composition range, *S* varies more or less linearly with composition indicating, again, the compatibility of both components either in the solid or mesophase. These findings add extra evidence for the formation of solid solution within this intermediate range of composition. A similar behaviour was observed for $n = 14$, except that it passes also through a eutectic composition, accompanied by a maximum *S*. Partial solid solubility was previously observed [23] in the binary mixtures of their homologues. The other mixtures (Fig. 3b and

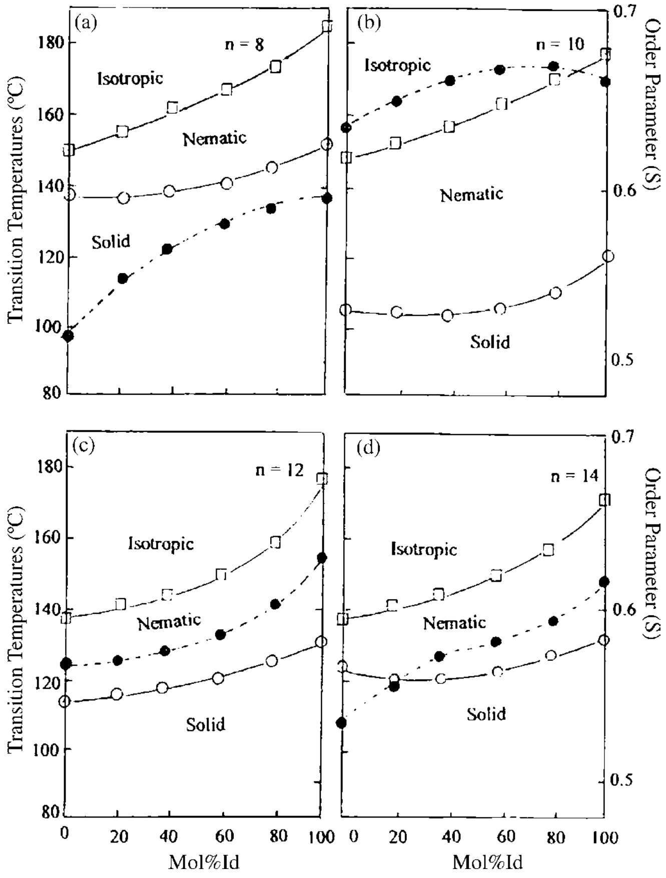


Fig. 6. Phase diagrams for the binary mixtures of the methyl- and nitro-substituted analogues: (a) **I**8_b/**I**8_d, (b) **I**10_b/**I**10_d, (c) **I**12_b/**I**12_d, and (d) **I**14_b/**I**14_d.

c) form solid solution through the whole composition range, while the last mixture (Fig. 3d) possesses eutectic point. Other systems, Figs. 2, 4–6, behave in similar ways but to varying extents.

4. Conclusions

Binary mixtures were prepared from two differently substituted analogues, but bearing the same alkoxy-chain, of the 4-(4′-substituted phenylazo)-1-naphthyl-4″-alkoxybenzoates, and characterized for their binary phase behaviour. All compounds investigated were found to possess relatively high melting and clearing temperatures, indicating strong intermolecular forces of association either in the solid or mesophases. Replacement of the central benzene ring, in the 4-(4′-substituted phenylazo)-1-phenyl-4″-alkoxybenzoates (**II**), by a 1,4-naphthalene moiety to give the investigated naphthalene-based compounds (**I**), has led to: (i) reduction in both melting and clearing points, (ii) the disappearance of the Sm A phase (of **II**), and (iii) disturbance in the solid-crystalline structure. As a result, their mixed phase properties behave in an irregular manner as the alkoxy-chain is lengthened. Two types of T_C-composition dependencies were observed either a linear variation, or slightly negative deviation. On the other hand, dependent on X and n, their mixed solid phases showed three different types of variation with composition:

(a) The two components are of compatible crystalline structures and their phase diagrams showed a gradual variation of T_m (or S) with composition (Figs. 1c, 2a, 3a and b, 4a, and 6a–d). In such type of binary phase behaviour, mixing of components does not lead to improvement in their mesophase properties; that is, it does not reduce T_m.

(b) The two components are of different crystal lattice that is disturbed by the addition of one component to the other. In such case a range of composition was observed in which a solid solution is formed. Such a range is always within the intermediate range of composition (Fig. 3a) and having melting temperatures lower than those of both components. Such a behaviour was also observed [23] in mixtures of the homologues of **I**. This property may have its application since the melting point, within this range of composition, is but slightly dependent on composition.

(c) The two components have incompatible crystal structures and their solid mixture exhibits eutectic behaviour (Figs. 1a, b and d, 2b–d, 3d, 4b–d, and 5a–d). Unfortunately, except for the binary system ($\mathbf{I}_a/\mathbf{I}_d$) that is composed of the methoxy with the nitro analogues ($n = 10$ and 14 carbons), the depression in melting points does not exceed 10 °C for the eutectic composition compared with the lower melting-point component of the mixture. Even in these cases (Fig. 5b and d) the depression in melting points amounts to 17.9 and 17.4 °C, respectively.

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