

Antibacterial Activity and Mesophase Identification of Azomethnine and Azo derivatives

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Abstract

A new mesogenic homologous series of liquid crystalline azomethnine and azo derivatives of 4'-n-alkoxybenzylidene-4-aminoazobenzene has been synthesized and characterized by a combination of elemental analysis and standard spectroscopic methods. In the present series, nematic mesophase commences from n-propyloxy derivative as enantiotropic and persists up to the last member synthesized except in derivative C₁₀. The smectic A (SmA) mesophases commences from n-pentyloxy derivative as enantiotropic and also persists up to n-tetradecyloxy synthesized. Imine linkage (-CH=N-) between central and terminal benzene rings helping to understand the polarizability effect as well as nature of mesophase as stability gets reduced due to the presence of nitrogen in the ring.

The mesomorphic properties of present series are of great importance to evaluate the effects of central linkage on mesomorphism. The mesomorphic properties of present series were compared with other structurally related mesogenic homologous series to evaluate the effects of central linkage on mesomorphism.

Keywords: Antibacterial activity, Mesomorphism, Nematic phase, Smectic phase.

Introduction

Polarity is affected by the presence of terminal groups in the molecule. It has been found that terminally substituted compounds exhibit more stable mesophases than non-substituted mesogenic compounds. Any terminal group which increases the molecular polarizability without significantly increasing the molecular breadth, increases the thermal stability of resulting mesophase. In order to study the effect of terminal substitution on mesomorphism, the present series containing azo Schiff base as central linkage and different terminal substitutions has been synthesized and their mesomorphic properties were studied like molecular breadth, thermal stability of the resulting mesophase etc.

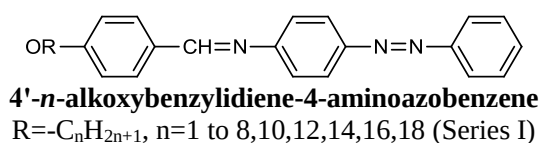
A number of homologous series of mesogenic azo Schiff base liquid crystalline compounds with normal and branched terminal alkyl chains have been reported previously in the literature.^{2-6,11} Molecular geometry is based on polarizability contribution to the dispersive forces which are vital to the formation of liquid crystal. In the present study, a mesogenic

homologous series with azo-schiff base has been synthesized. In this series, number of [4'-n-alkoxybenzylidene-4-aminoazobenzene] compounds have been synthesized to study the effects of azomethnine groups on the different textures smectic and nematic phases.

To investigate the relationship between mesomorphism and chemical constitution, a homologous series of Schiff's base compounds has been synthesized by condensing 4-n-alkoxy-1-naphthaldehydes. New azo ester ferroelectric liquid crystalline compounds derived from chiral S-lactic acid as a terminal moiety have also been reported⁸. Lai and Lin⁹ have synthesized and evaluated azo dye compounds and have shown variation in mesomorphic behaviour with change in alkyl chain length. Liquid crystal polymers that contain an azo-ester linkage in the main chain have also been synthesized¹⁶. Few azo Schiff base liquid crystalline derivatives have been synthesized and studied¹².

Four new azomethnine dyes, (PAZB-6, PAZB-7, PAZB-8, PAZB-9) containing azo and azomethnine groups were prepared and studied¹⁴. By this, new study of anisotropy of thermal and electrical conductivity of coloured stretched PVA films has been undertaken. Azo compounds containing different transition temperature and phase range have great importance in many fields where these can be fed as best applications as photonic devices, electro-optic devices, dye material in dye and textile industries as of having great texture and colouring and texture. Earlier from 19th century, azo Schiff base compounds are of great interest upto this date also. Azo dyes are also being used in liquid crystal display devices for guest-host interactions¹⁰. The systems having azo, azomethnine moieties would be exceptionally promising for wide range of new technological applications.

The general structural formula is in scheme 1:



Scheme 1: General formula of azo Schiff base with different alkoxy chains.

Material and Methods

4-hydroxybenzaldehyde, n-bromo-alkane, potassium carbonate (K₂CO₃), NaNO₂, HCl, dry acetone, dry CCl₄, few drops of acetic acid and ethanol were used as received. Solvents were dried and distilled before use. Microanalysis of the compounds was performed on a Coleman carbon-hydrogen analyzer and the values obtained are in close

agreement with those calculated. IR spectra were determined for KBr pellets using a Shimadzu IR-408 spectrophotometer.

^1H NMR spectra were obtained with a Perkin-Elmer R-32 spectrometer using tetramethylsilane (TMS) as internal reference standard. The chemical shifts are quoted as δ (ppm) downfield from the reference and CDCl_3 as solvent.

The phase assignments and transition temperatures were determined by thermal polarizing optical microscopy (Leitz Laborlux 12 POL) equipped with a heating stage (Linkam Scientific Instruments Ltd., T95-HS). The enthalpies of transitions are reported in Jg^{-1} and they were measured on a differential scanning calorimeter (Mettler TA-4000) at a rate of 5°Cmin^{-1} . The instrument was calibrated using pure indium as a standard.

In the first step, synthesize diazonium salts from primary amines using a typical procedure from the literature¹³ by dissolving 0.002 mole of Na_2CO_3 in 12ml of distilled water and then addition of 0.0057 mole from the primary amine and continue heating until it dissolve completely, then after filtration and cooling using ice bath ($0-5^\circ\text{C}$), then add 0.0054 mole of NaNO_2 while stirring and then add (1.25 ml) of conc. HCl drop by drop. We observe the emission of chlorine gas and get the diazonium salt of such primary amine in excellent yields.

Reagents and conditions: (i) NaNO_2 , HCl , ($0-5^\circ\text{C}$) ; (ii) R-Br, anhydrous, potassium carbonate (K_2CO_3), dry acetone ; (iii) Dry CCl_4 , few drops of acetic acid.

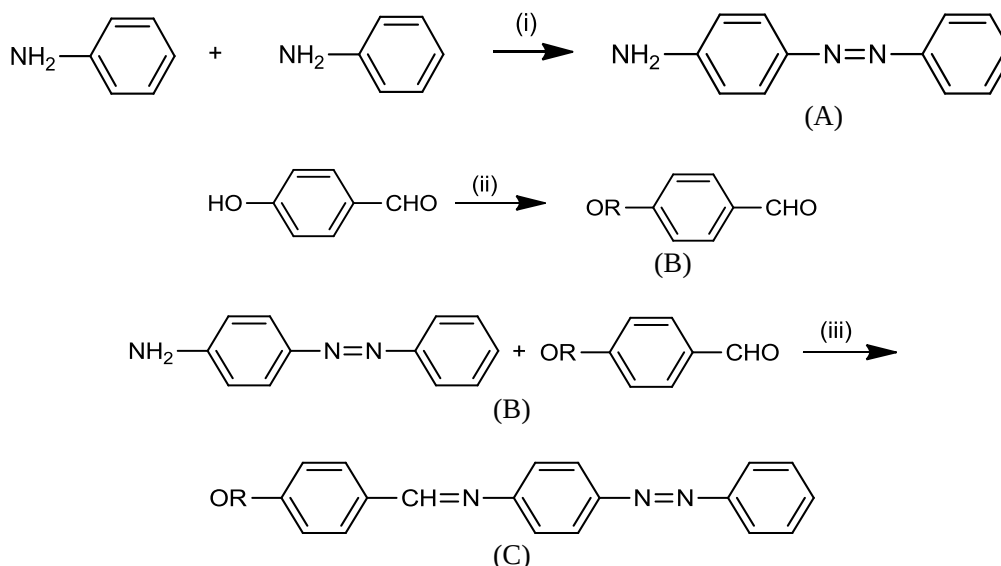
4'-n-alkoxybenzylidene-aminoazobenzene was synthesized by the diazotization method (A). 4-n-alkoxybenzaldehydes (B) were prepared using the method mentioned in the literature¹⁹. 4-n-phenylazoaniline formed in (A) is then allowed to react with 4-n-alkoxybenzaldehyde (B) through a condensation reaction in presence of dry CCl_4 as a solvent.

About 15-20 ml of dry CCl_4 are with 2-3 drops of acetic acid in 250 ml Round bottom flask. Reflux the mixture for around 2 hours at room temperature and then hot filter it, collect the yellow ppts after 15 min, insoluble solid was removed through filtration. After filtration, obtained product is allowed for complete drying. Then, the obtained product was allowed to recrystallize two times from ethanol to get final azo Schiff base compounds as L.C. The purities of all these compounds are checked by thin layer chromatography (Merck kieselgel 60F₂₅₄ precoated plates).

Antibacterial Activity: Antibacterial activity was carried out against sensitive test cultures like *S. aureus* and *E. coli* by agar well diffusion method. Molten nutrient agar plates were seeded with 10^7 to 10^8 suspension of sensitive bacterial strain. Amoxicillin and clavunilic acid ($240\mu\text{g/ml}$) were used as a positive control. The plates were incubated at 37°C for 24-48 hrs. An antibacterial activity was expressed as the diameter of zone of inhibition produced by the compounds under study.

Characterization: The elemental analysis of all the compounds of the present series were found to be satisfactory. Typical spectral data of the representative members were found to be consistent with the proposed structure. The IR and ^1H NMR spectral data were found to be consistent. All the azo Schiff base compounds were characterized by using Shimadzu-IR spectrophotometer.

As a preliminary investigation, the mesophase exhibited by the derivatives of the present series were concluded by the optical microscopic studies. The liquid crystalline phases were observed by using a polarizing optical microscope. Thin film samples were obtained by sandwiching them between a glass slide and cover slips. The space filling model, ball and stick model of dye derivative is shown in figure 1.



Scheme 2: shows the general synthetic root for diazonium salts

Series I: 4'-*n*-heptyloxybenzylidene-4-aminoazobenzene
IR spectrum (KBr) ($\nu_{\max}/\text{cm}^{-1}$): 2935.5, 2859 cm^{-1} (C—H aliphatic), 1609 cm^{-1} (—CH=N of azomethine), 1601 (—N=N—), 1508, 1507.8, 1506 cm^{-1} , 1258.4 cm^{-1} , 1000 cm^{-1} , 857.23 cm^{-1} , 687.35 cm^{-1} ; ^1H NMR(CDCl_3)(300 MHz): δ 0.88 (t, 3H, —CH₃), 1.29–1.67 (m, 10H, 5 x —CH₂—), 4.02 (t, 2H, Ar—O—CH₂—), 6.97 (d, 2H, ArH), 7.29 (d, 2H, ArH), 7.45 (tt, 1H, ArH), 7.52 (d, 2H, ArH), 7.85 (d, 2H, ArH), 7.96 (d, 2H, ArH), 7.98 (d, 2H, ArH), 8.42 (s, 1H, —CH=N—).

Series I: 4'-*n*-dodecyloxybenzylidene-4-aminoazobenzene
IR spectrum (KBr) ($\nu_{\max}/\text{cm}^{-1}$): 2955.5, 2869 cm^{-1} (C—H aliphatic), 1607 cm^{-1} (—CH=N of azomethine), 1598 (—N=N—), 1506, 1504.8, 1502 cm^{-1} , 1248.4 cm^{-1} , 1100 cm^{-1} , 847.23 cm^{-1} , 684.35 cm^{-1} ; ^1H NMR(CDCl_3)(300 MHz): δ 0.88 (t, 3H, —CH₃), 1.26–1.48 (m, 20H, 10x —CH₂—), 4.03 (t, 2H, Ar—O—CH₂—), 6.75 (tt, 1H, ArH), 6.99 (d, 2H, ArH), 7.32 (d, 2H, ArH), 7.53 (d, 2H, ArH), 7.85 (d, 2H, ArH), 7.96 (d, 2H, ArH), 7.98 (d, 2H, ArH), 8.43 (s, 1H, —CH=N—). All the compounds of the present series and the comparable series exhibit mesomorphism.

Results and Discussion

In the present series, nematic mesophase commences from *n*-propyloxy derivative as enantiotropic and persists up to the last member synthesized except in C₁₀. The smectic A

(SmA) mesophase commences from *n*-pentyloxy derivative as enantiotropy and also persists up to *n*-tetradecyloxy synthesized. On heating the higher homologues of series I (*n*=5–14) gave only focal-conic texture characteristic of SmA mesophase. Transition temperatures (°C) of the series I compounds are shown in table 1.

For series A, on cooling the *n*-octyloxy (*n*=8) isotropic liquid of 4'-*n*-alkoxybenzylidene-4-aminoazobenzene (*X* = H)¹⁵ exhibits only schlieren texture of the nematic phase; no focal-conic texture characteristic of SmA mesophase exists. The Schlieren texture of nematic mesophase is heated at 137°C (Series I; *n*=7) and focal conic texture of Sm-A mesophase at 122 °C on heating (Series I; *n*=7) (Figure 2).

When the terminal attractions are more pronounced, the nematic mesophase prevails. In this series, the first two members are non-mesogenic. The overall polarizability is enhanced with the increase of alkyl chain length as the series is ascended i.e. as *n* increases. The ratio of terminal attractions to lateral attractions decreases as the series is ascended; the net result is that the lateral attractions gain predominance over the terminal attractions. The molecules under such conditions will adhere strictly maintaining the layered structure, although they might partially acquire a fluid condition.

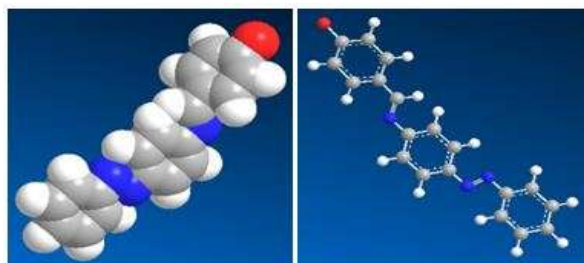


Figure 1: Space filling model, ball and stick model of dye derivative

Table 1
Transition temperatures (°C) of the series I compounds

Compound No.	R = —C _n H _{2n+1} n =	Cr		SmA		N		I
1	1	•	151	—			—	•
2	2	•	150	—			—	•
3	3	•	154	—		•	178	•
4	4	•	158	—		•	177	•
5	5	•	153	•	155	•	160	•
6	6	•	148	•	153	•	154	•
7	7	•	122	•	137	•	165	•
8	8	•	130	•	150	•	162	•
9	10	•	109	—		•	143	•
10	12	•	106	•	126	•	129	•
11	14	•	97	•	125	•	101	•
12	16	•		—	122	•	124	•
13	18	•		—	95	•	99	•

() = monotropic value; Cr = crystalline solid; SmA = smectic A phase; N = nematic phase; I = isotropic liquid phase; •, phase exists; —, phase does not exist

Under such situations, the layered smectic structure is maintained giving rise to a smectic mesophase. As the temperature is further increased, the strict adherence of the molecules in the layered form is weakened due to increased thermal vibrations. The molecules slowly acquire a more fluid nature, although they do not abruptly acquire any randomness.

At a definite temperature the layered structure breaks down but end to end parallel orientation is still maintained giving rise to more fluidity and a nematic threaded texture to the molecules. Thus, polymorphism is exhibited from the fifth member up to the last member of the homologous series. From fifth member onwards, a purely SmA mesophase is exhibited. The overall polarizability of the molecules is enhanced as the chain length of the alkyl chain at one end increases contributing to the exhibition of smectic texture. However, the ratio of terminal to lateral attractions is also low that at a certain stage the thermal vibrations break down the orientation of molecules without giving them a chance for adopting a nematic orientation¹⁸.

Figure 3 shows the plot of transition temperatures against the number of carbon atoms in the alkoxy chain, from which it can be noticed that the crystal–mesophase/isotropic transition temperatures decrease sharply with an initial increase in the length of terminal alkoxy tail which gives

pronounced enantiotropic behaviour from the third member in the series I. The nematic–isotropic (N–I) transition temperatures also decrease with increase in the number of carbon atoms in the alkoxy chain and exhibit the usual odd–even effect for the first four derivatives whereas it exhibits a tendency for rising SmA–N as well as SmA–I transition temperature in the ascending series, which levels off slightly in the middle and higher homologues respectively.

Table 2 provides a comparison of smectic mesophase range, transition temperatures and molecular structure of representative compounds of series I ($n=8$) and series A ($n=8$) respectively, as well as structurally related compounds of series (B–E)². The mesophase range of compound of series I is about 32 °C and SmA–I transition temperature of series I is higher by 71 °C whereas for the compounds of series A, smectic phase does not exist. However, the SmA–I transition temperature for the compound of series I is higher when compared with that of compound of series A. The molecular structure of compound of series I differs from compound of series A only at the lateral. Series I has azo Schiff base, whereas compound of series A has an ester and azo group at lateral position. Gray⁷ explained that a compound that requires more thermal energy to disorganize its parallel molecular arrangement in the nematic phase is more thermally stable.



Figure 2: Microphotographs of texture observed for compound (a) Schlieren texture of Nematic mesophase on heating at 137 °C (Series I; $n=7$). (b) Focal conic texture of Sm-A mesophase at 122 °C on heating (Series I; $n=7$)

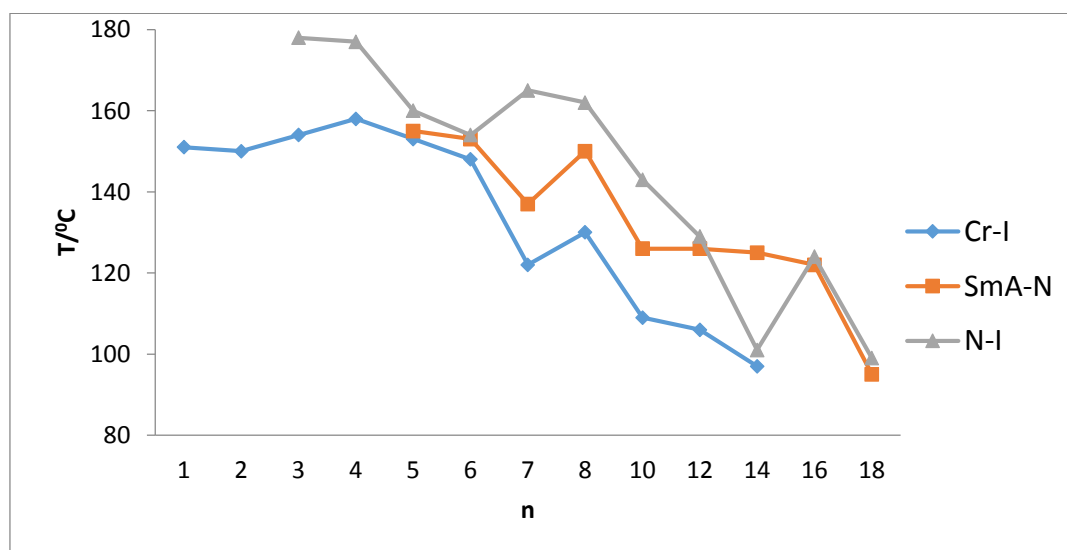


Figure 3: The phase behaviour for series I compounds

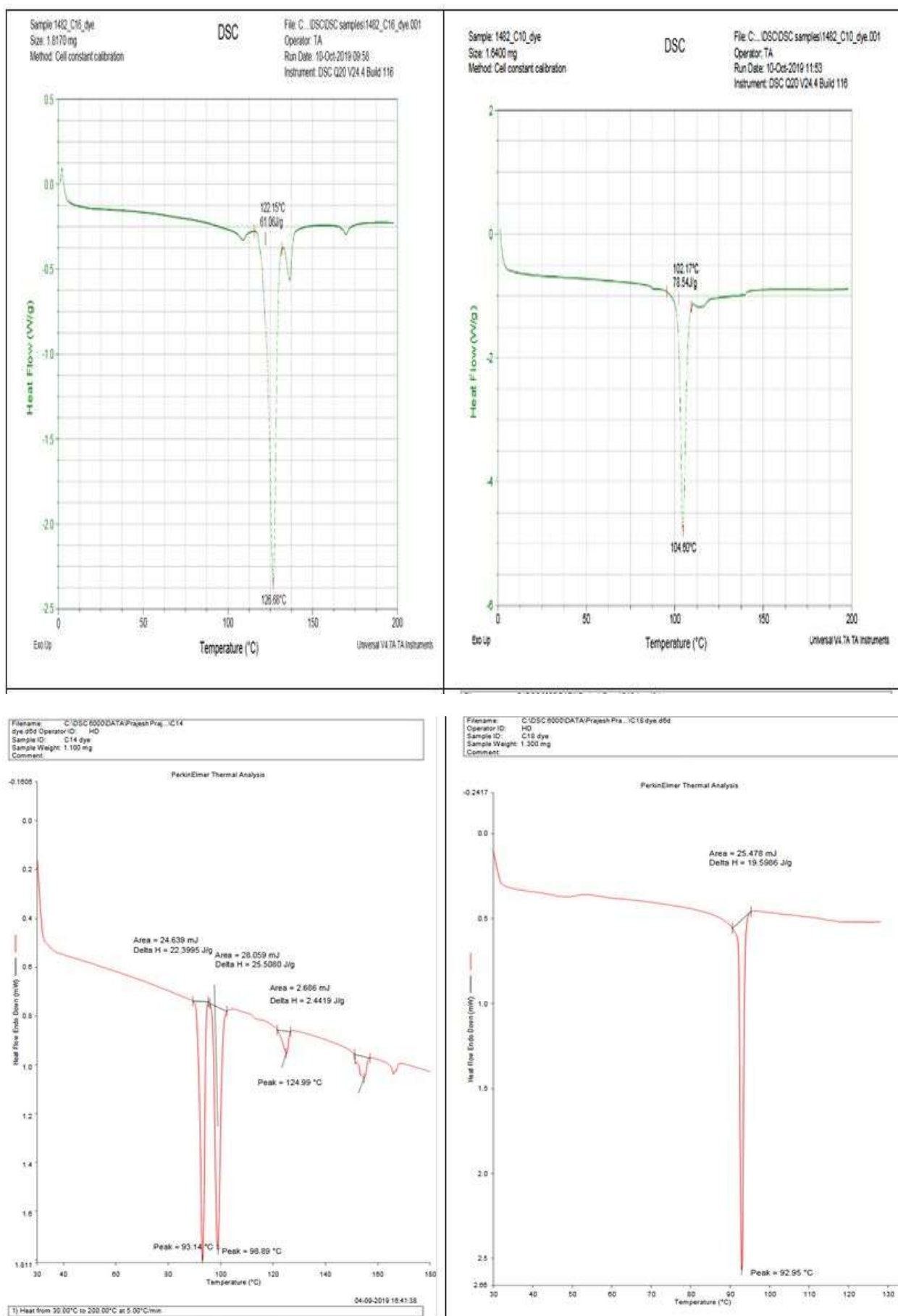


Figure 4(a) and 4(b): Showing DSC isotherms of derivatives

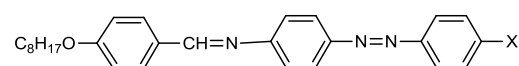
The DSC thermographs are shown in figure 4. The average thermal stability of the compounds of series I, is higher than series A. Both the series have the same terminal substituent at one end and at the other end *n*-alkoxy chain. The two compounds differ only in central linkages as series I has an azomethine(-CH=N-), azo (-N=N-) central linkage.

As mentioned earlier, series I has the central azomethine (-CH=N-) group, whereas series A has the central carboxy (-COO-) group. The oxygen atom of this central carboxy group in the molecule of series A will bump into the non-bonded side of adjacent H of the aromatic ring, thereby causing strain on the molecule. This will cause some twist around the C-O bond and will force the benzene ring out of the plane and will broaden the molecule¹.

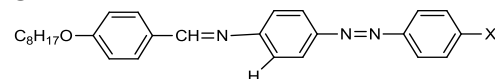
In terms of twist made by the central groups in the molecules in series I, twist occurs due to the H atom of the central azomethine (-CH=N-) group which is very less in comparison with the twist made by the oxygen of the (-COO-) group in series A. Presence of -CH₃ group at the lateral position makes it non coplanar. Thus the thermal stabilities of present series I are maximum than series A and other comparable series. In series A, presence of lateral -CH₃ which is ortho to -COO- linkage increases the breadth and reduces the co-planarity of the molecule. This leads to late commencement of smectic mesophase while in our series, the molecule is coplanar which leads to the early commencement of smectic mesophase.

If we compare with series B, the width of smectic mesophase and the SmA-I transition temperature of series I is higher by 20°C and lower by 84°C just by the reason of presence of Schiff base at both the positions which reduces the coplanarity of the molecule. If we compare with series C, the width of smectic mesophase and the SmA-I transition temperature of series I is lower by 36°C and 70°C, this is because of by changing the positions of the terminal groups. Comparison of series I with series D and E slightly differing at the central groups replaced by the -COO at one of the central position in series D shows that the SmA-I transition temperature is higher by 20°C and smectic phase does not exist and in series E with (-COO) group at both the positions, the SmA-I transition temperature of series I is lower by 41°C and 32°C.

Comparing our series with other ones, we get:

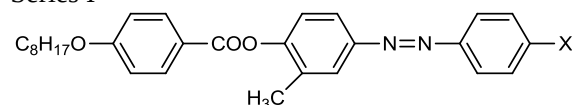


OR



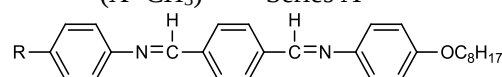
4'-*n*-alkoxybenzylidene-4-aminoazobenzene (X = H)

Series I

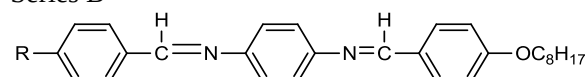


(X=CH₃)

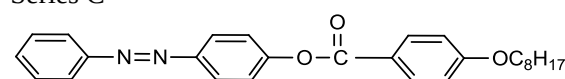
Series A



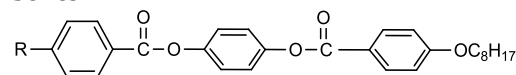
Series B



Series C



Series D



Series E

Series A¹⁹, Series (B-E)⁴

The presence of azo Schiff base at the lateral positions in place of ester group changes the molecular confirmation which leads to decrease the transition temperature. Presence of azo Schiff base makes the molecule more polarized. Aromatic ring attached to azo and Schiff base shows an immense impact on the stability of the molecule. IR and NMR data is helpful in making it very clear how the presence of terminal groups and the alkyl chains changes transition temperature and phase length and corresponding change in thermal stability, phase behavior and polarizability.

Table 2

The smectic mesophase range, phase transition temperatures and comparative molecular structure of compounds of series I (n=8), series A(n=8) and series (B-E) (n=8).

Compound	Mesophase range/ ⁰ C	SmA-I/ ⁰ C	Commencement of smectic phase
I	32	162	C ₃
A	-	91	C ₃
B	12	246	C ₄
C	68	232	C ₆
D	-	142	C ₁₀
E	73	194.9	C ₈

From above mentioned results, it can be observed that not only the geometry of the molecule but also length, breadth and polarizability of the molecule play an important role in studying the mesomorphic behavior of compounds.¹⁹

Antibiotic Sensitivity Test

The inhibitory and antibiotic properties of two different chemical derivatives C8, C12 dyes were studied. Two different chemical compounds stock solutions were prepared in DMSO (Dimethyl sulfoxide). Two compounds were tested for its antibiotic properties or effect against gram positive and gram negative bacteria namely *E. Coli* and *S. aureus*.

aureus. Clavum (Amoxicillin and Clavunilic Acid) was used as control standard in all the plates. Significant zone of inhibition could be observed for control standard in all the plates of *E. coli* as well as *S. aureus*. None of test compounds showed zone of inhibition against *E. Coli* and *S. aureus* at two different concentrations (Figure 5).

Interpretation: From the above experiment, it can be concluded that compound C12 dye and C8 dye re-crystallized showed no zone of inhibition at minimum inhibitory concentration 700 ($\mu\text{g/ml}$) and at 200($\mu\text{g/ml}$) respectively against *S. aureus* and *E. coli* (Table 3).

Table 3
Result table showing zone of inhibition against minimum inhibitory concentration

Compound name	Conc. Of compound ($\mu\text{g/ml}$)	Zone of inhibition(in mm)	
		<i>E.coli</i>	<i>S.aureus</i>
A1	350	--NIL--	--NIL--
A2	175	--NIL--	--NIL--
A3	87.5	--NIL--	--NIL--
S	240	22	30
B1	250	--NIL--	--NIL--
B2	125	--NIL--	--NIL--
B3	62.5	--NIL--	--NIL--
S	240	22	31

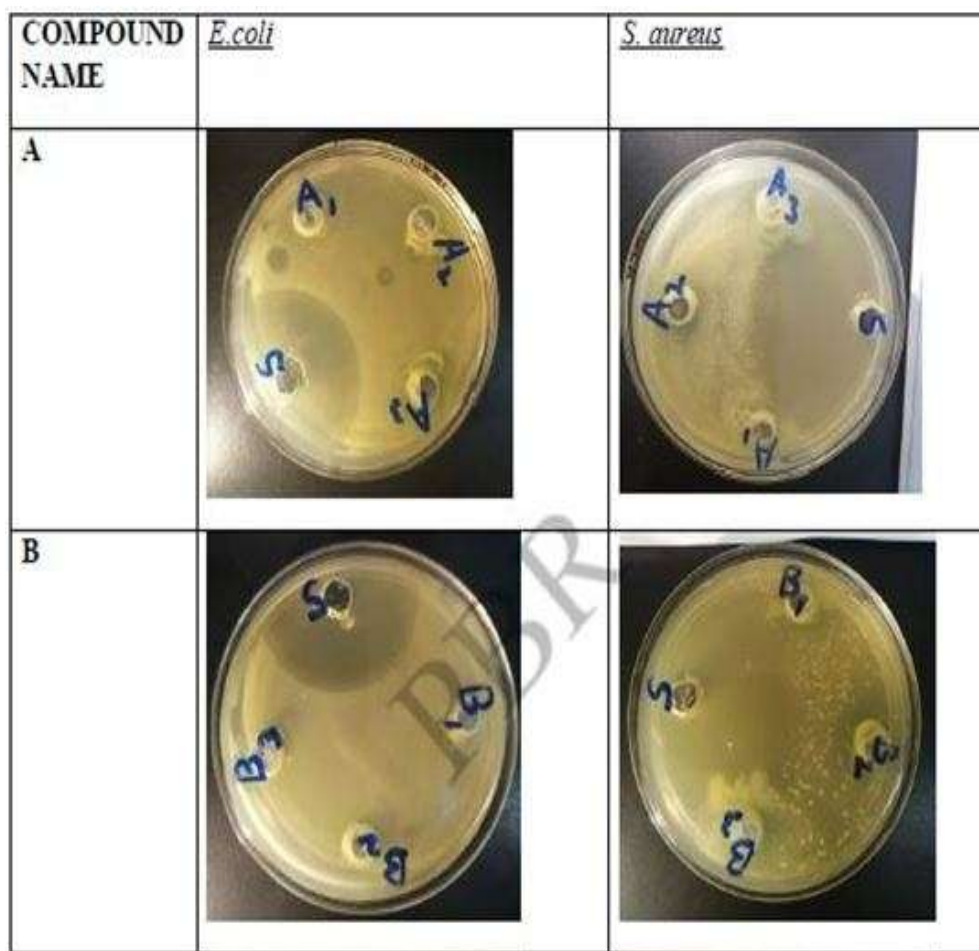
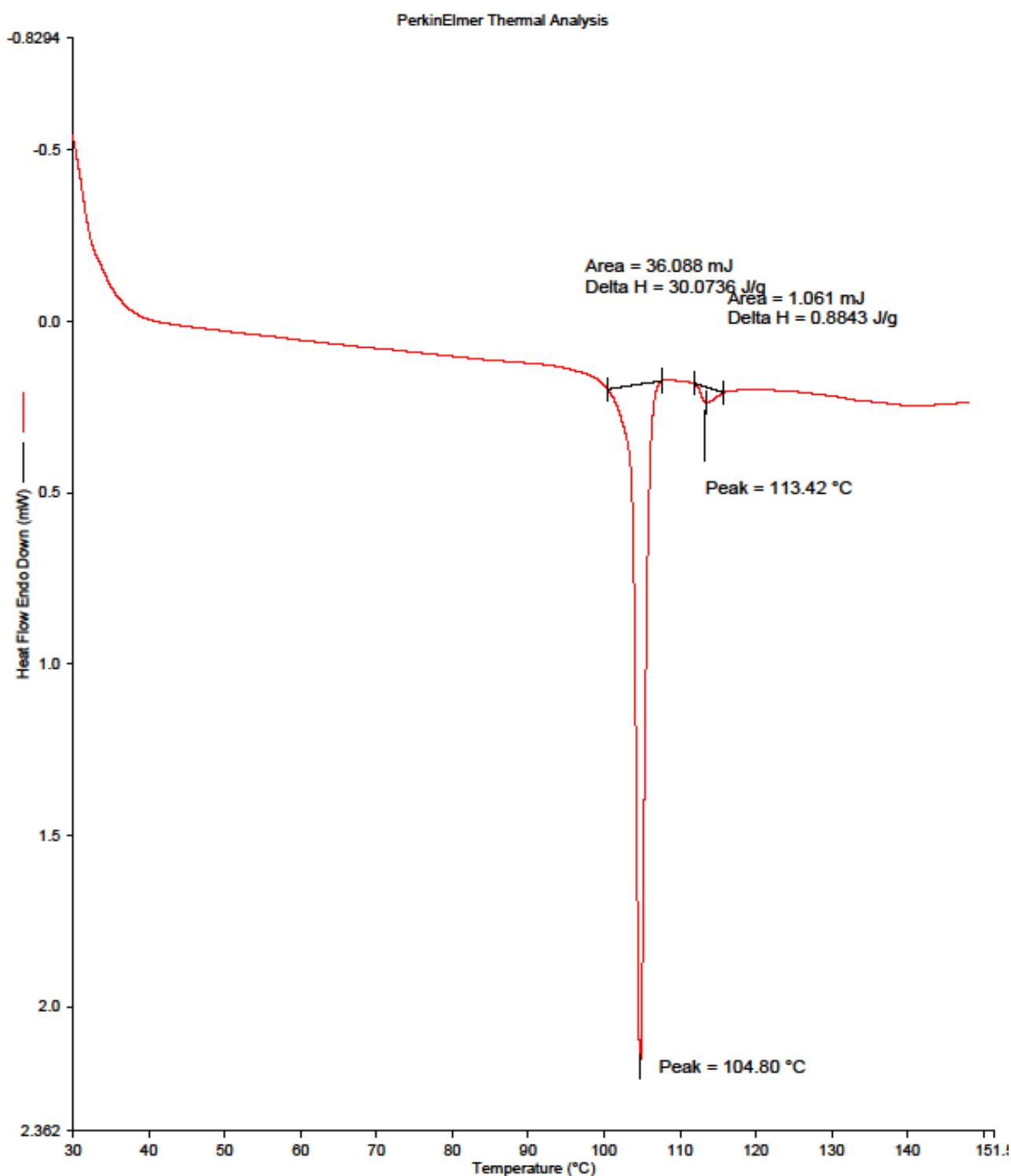


Figure 5: Zone of inhibition

Supplementary Data**DSC of dye derivatives:****(1) C12 dye**

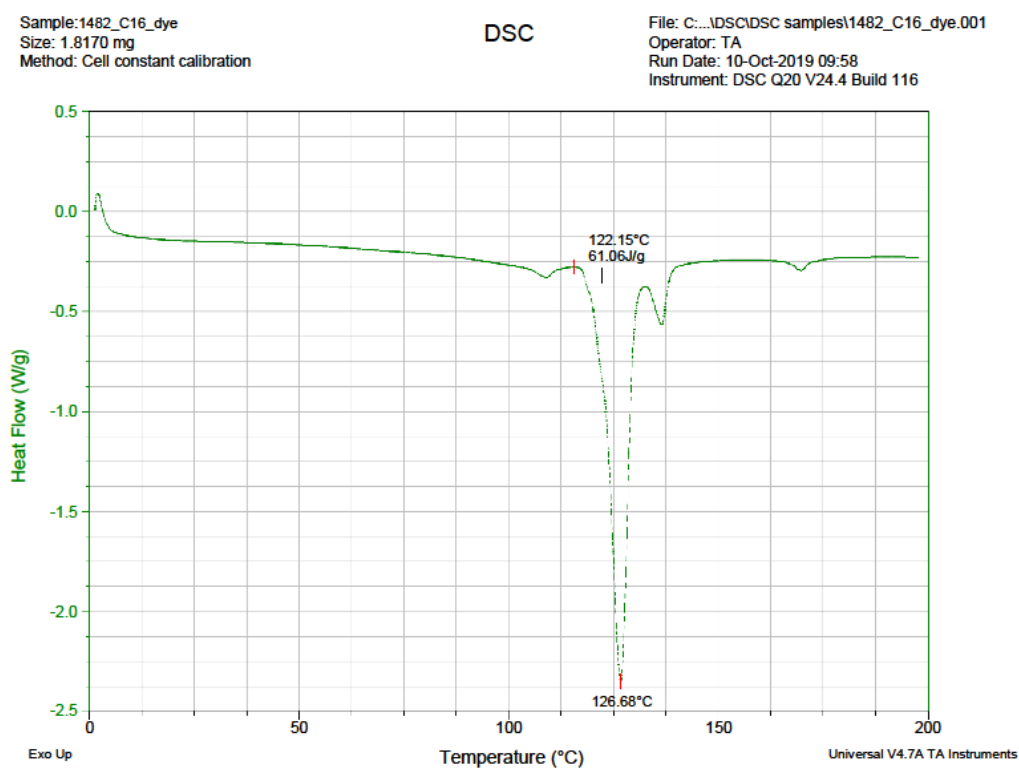
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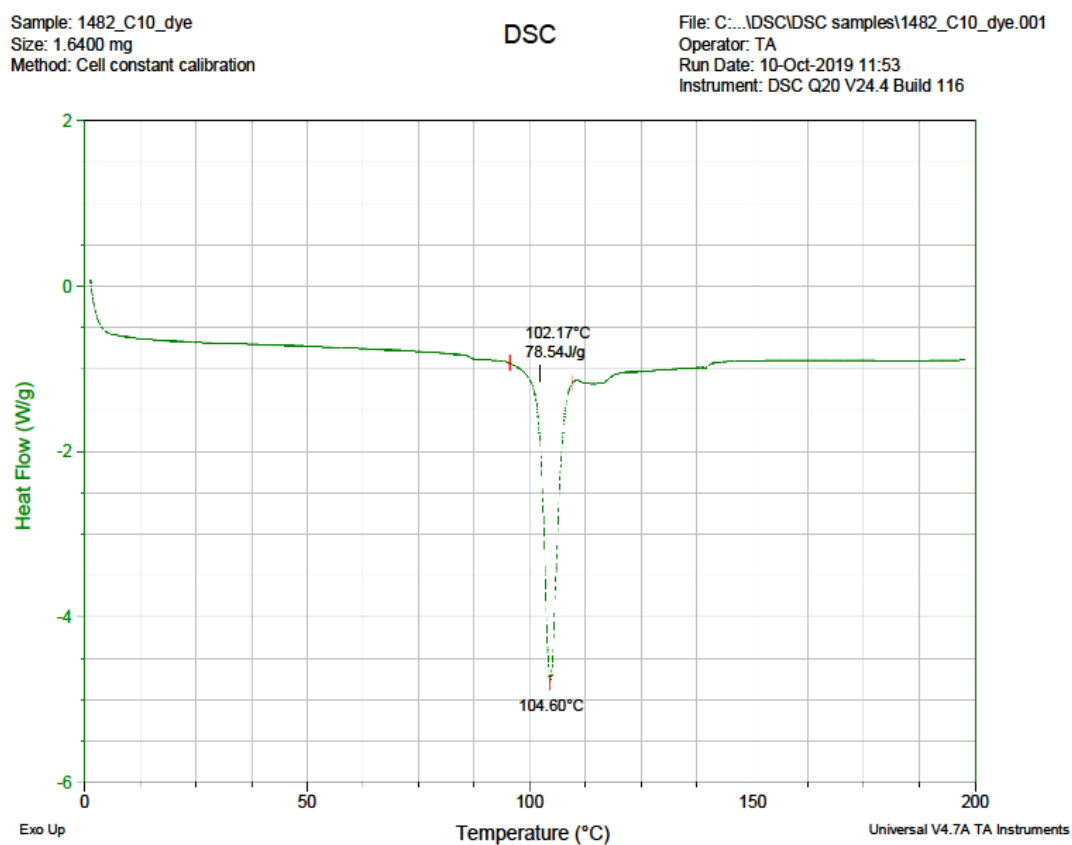
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1) Heat from 30.00°C to 150.00°C at 5.00°C/min

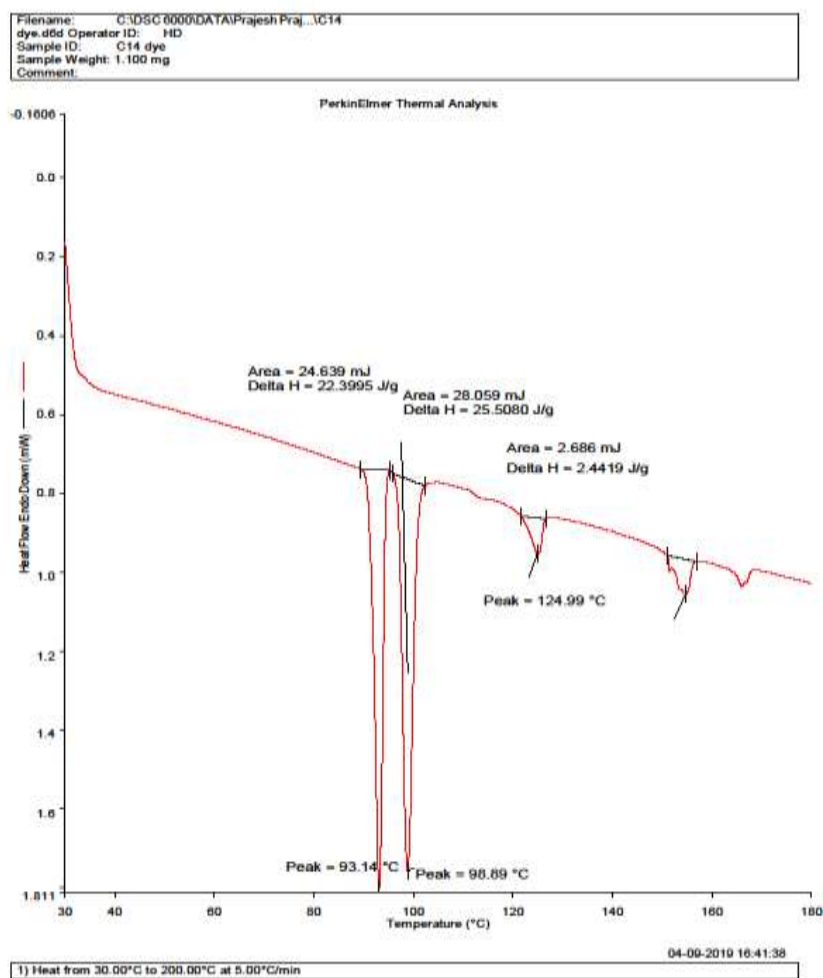
(2) C16 dye



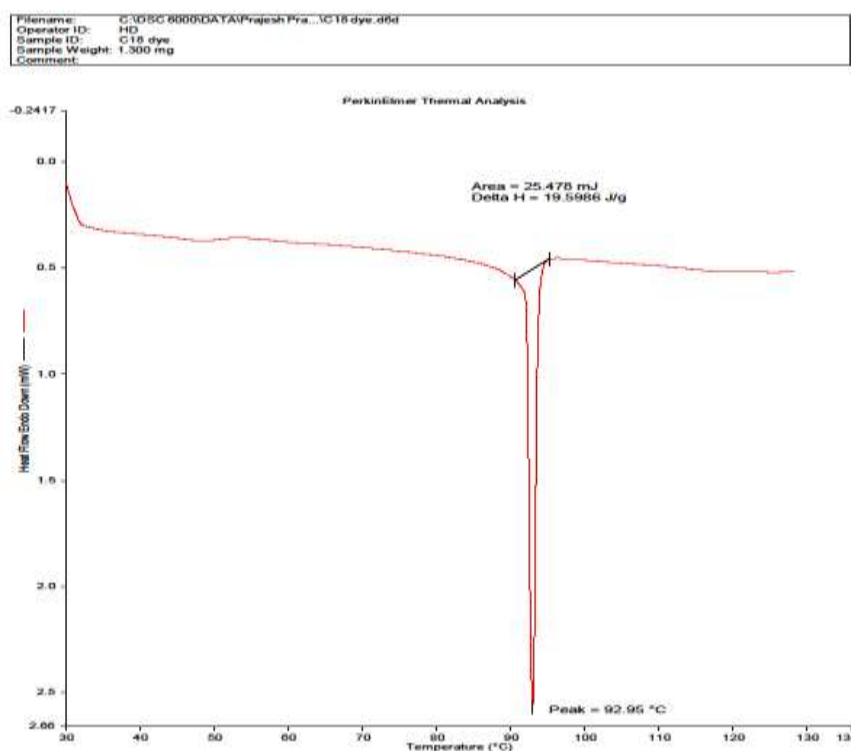
(3) C10 dye



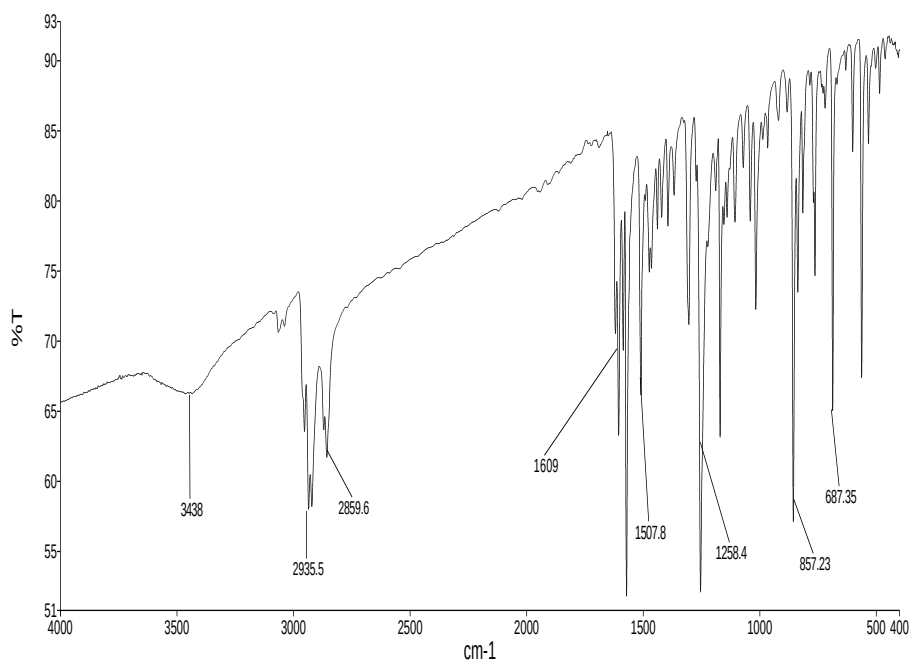
(4) C14 dye



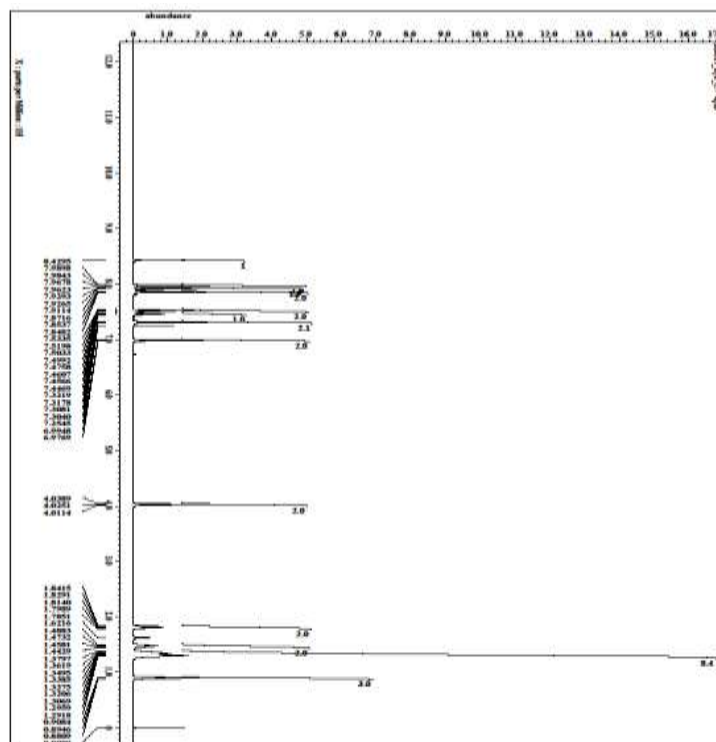
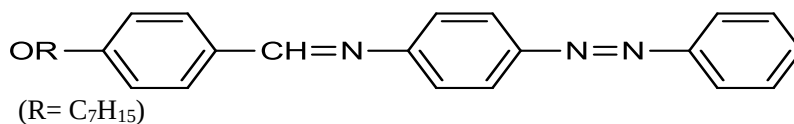
(5) C18 dye



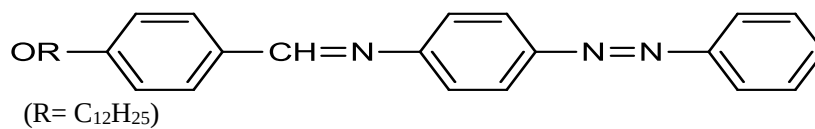
IR: C7 azo dye

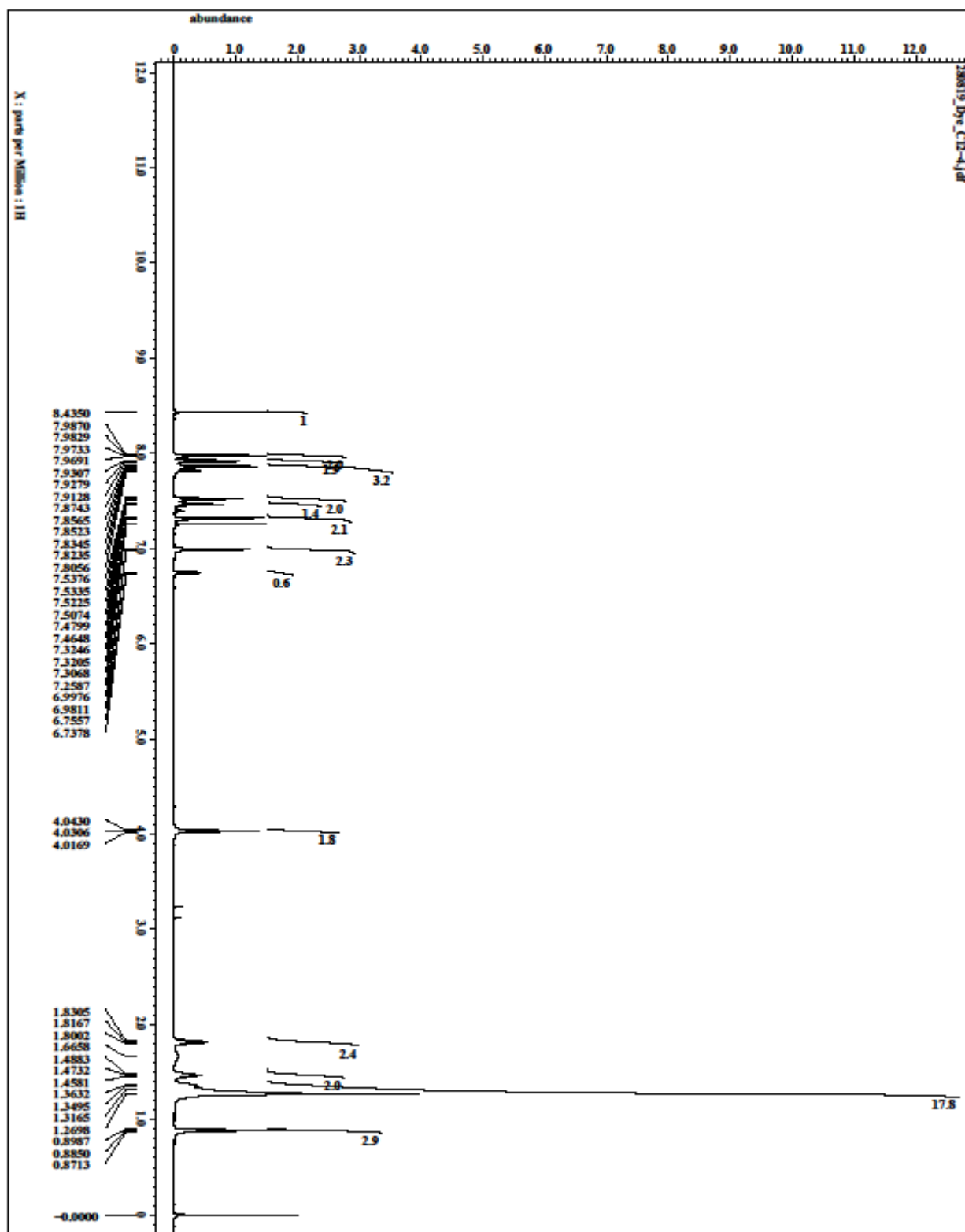


NMR: C7 dye



NMR: C12 dye





Conclusion

From the data presented above, we conclude that in order to check the mesomorphic behavior of the compounds, not only the geometry but also length, breadth and polarizability of the molecule are important factors. Terminal alkyl chain provides a strong mutual attraction. Presence of long

terminal alkyl chain makes contribution towards the thermal stability of smectic A phase as well as nematic phase as azo and Schiff base groups as central core units mainly differ in their length and extension of the electronic conjugation. These derivatives were found to be ineffective to inhibit the growth of several sensitive bacterial strains.

Acknowledgement

The authors thank Prof. Keyur Shah in Shri M. M. Patel Institute of Sciences and Research for providing research facilities. One of the authors, H. N. Patel, thanks GUJCOST (GUJCOST/MRP/2016-17/441) for the grant of minor research project entitled "Synthesis, Characterization and Mesomorphic Properties of Novel Liquid Crystalline Materials".

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(Received 04th January 2021, accepted 04th March 2021)