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Vladimir F. Petrov [a]

[a] LC Works, Camberwell, Australia

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Pyrimidine as a Structural Fragment in Calamitic Liquid Crystals

Vladimir F. Petrov
LC Works, Camberwell, Australia

The effect on the physicochemical and electro-optical properties of introducing the pyrimidine ring into the molecular structure of calamitic liquid crystals is discussed and rationalized in terms of existent theories; a comparison is made with the corresponding nitrogen-containing six-membered ring derivatives and 1,4-phenylene derivatives.

Keywords: electro-optical properties; liquid crystals; physicochemical properties; pyrimidine

1. INTRODUCTION

In continuation of our study of the structure–property relationships in nitrogen-containing liquid crystals (see, for example, the previous publications [1–4]), we present here a review that examines in some detail the effect of the introduction of the pyrimidine ring into the molecular structure of calamitic liquid crystals on the appearance of the mesophases and their physicochemical and electro-optical properties. When possible, the properties of the pyrimidine derivatives are compared with those of the corresponding nitrogen-containing six-membered ring derivatives and 1,4-phenylene derivatives.

It has been demonstrated that structural nonrigidity is a general property of heteroaromatic and aromatic six-membered rings such as pyrimidine, pyrazine, pyridazine, pyridine, 1,2,4-triazine, and benzene [5]. Changes in ring deformability in these molecules are correlated with aromaticity [6] of cyclic conjugated systems and are governed mainly by electronic effects [5]. Particularly, it has been shown that the transition of the pyrimidine ring from a planar equilibrium

Address correspondence to Vladimir F. Petrov, LC Works, 6/68 Brinsley Road, Camberwell, VIC 3124, Australia. E-mail: lcworks@hotmail.com

conformation to a nonplanar structure with a relevant torsion angle value $\pm 20^\circ$ causes an energy increase of less than 2.6 kcal/mol [5]. It should be noted that 18% of the pyrimidine molecules possess considerable nonplanar geometry at every moment of time with values of endocyclic torsion angles up to 15° [5]. It is believed that these structural features of the pyrimidine ring should affect the physicochemical and electro-optical properties of the pyrimidine derivatives.

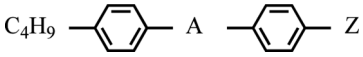
2. MESOMORPHIC PROPERTIES

The phase-transition temperatures of the pyrimidine derivatives and reference compounds are presented in Tables 1–9 where Cr, Sm, Sm*, SmG*, SmF*, SmI*, SmC, SmC*, SmB, SmA, N, Ch, and I denote the crystalline, smectic, chiral smectic, chiral smectic G*, F*, I*, smectic C, chiral smectic C*, smectic B, A, nematic, cholesteric, and isotropic phases, respectively. Values given in parentheses refer to monotropic phase transitions.

The clearing points (nematic–isotropic or smectic–isotropic phase-transition temperatures) of the 2,5-disubstituted pyrimidine derivatives can be lower (compounds **1-1**, **1-2**, **1-4**, **1-7**; **1-8**–**1-10**, **1-12**, **1-13**; **2-1**, **2-2**; **2-10**, **2-11**, **2-15**; **4-12**, **4-13**; **7-1**, **7-3**–**7-6**; **7-2**, **7-4**, **7-6**; **8-2**, **8-3**; **8-5**, **8-6**; **9-1**, **9-2**, **9-4**; **9-5**, **9-6**; **9-9**, **9-10**) and higher (compounds **1-1**, **1-3**, **1-5**, **1-6**; **1-8**, **1-11**, **1-14; 1-9**–**1-14**; **2-9**, **2-13**, **2-15**; **2-12**, **2-14**, **2-15**, **3-1**, **3-2; 3-3**, **3-4**, **3-5**–**3-7**; **3-8**, **3-9**, **3-10**, **3-11; 4-3**–**4-8**; **4-9**, **4-10**; **4-11**, **4-13**; **4-14**, **4-15**; **5-1**–**5-3**; **5-4**–**5-6**; **7-2**, **7-3; 8-4**, **8-6**; **9-1**–**9-3**; **9-7**, **9-8**) in comparison with those of the corresponding reference compounds. The melting temperatures of the 2,5-disubstituted pyrimidine derivatives can be lower (compounds **1-1**–**1-7**; **1-8**–**1-14**; **2-3**, **2-4**; **3-3**, **3-4**; **3-8**, **3-9**; **4-11**–**4-13**; **7-1**, **7-3**–**7-6**; **7-2**, **7-4**–**7-6**; **9-1**–**9-4**; **9-9**, **9-10**) and higher (compounds **2-1**, **2-2**; **2-9**, **2-11**, **2-13**, **2-15**; **2-12**, **2-14**, **2-15**; **3-1**, **3-2**; **3-5**–**3-7**; **3-10**, **3-11**; **4-3**, **4-4**–**4-8**; **4-9**, **4-10**; **4-14**, **4-15**; **5-1**–**5-3**; **7-2**, **7-3**; **8-2**, **8-3**; **8-4**–**8-6**; **9-5**, **9-6**) in respect to those of the corresponding reference compounds. Particularly, the influence of the introduction of the molecular fragment A into the molecular core of compounds presented in Table 1 on their mesomorphic properties can be expressed by the following orders of increasing the clearing temperatures (T_{cl}), melting temperatures (T_m), and nematic ranges ΔT: System (I), shown in Scheme 1.

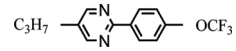
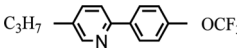
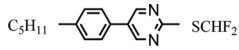
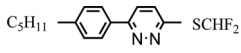
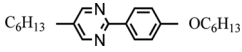
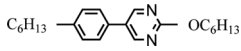
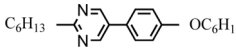
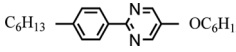
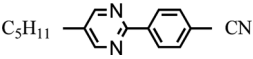
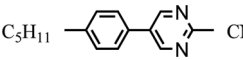
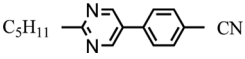
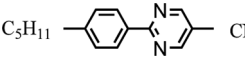
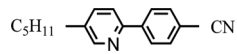
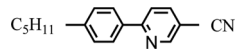
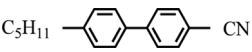
These results reveal that the introduction of the pyrimidin-2,5-diyl fragment into the molecular core of weakly polar system (I) forms low melting liquid crystals with moderate clearing points that show no nematic phase. In the case of strong polar cyano derivatives (I), the introduction of the pyrimidin-2,5-diyl fragment, depending on the

TABLE 1 Mesomorphic Properties of Liquid Crystals:

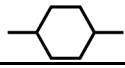


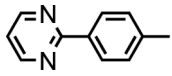
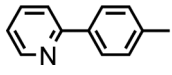
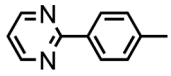
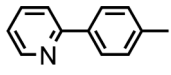
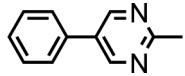
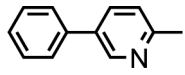
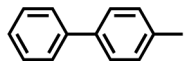
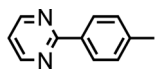
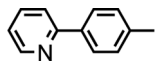
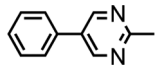
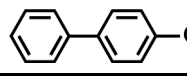
No.	A	Z	Phase transitions (°C)	Ref.
1-1		C_4H_9	Cr 118 SmA 186 I	[7]
1-2		C_4H_9	Cr 205 Sm 213 N 219 I	[8]
1-3		C_4H_9	Cr 161.3 SmC 166.4 N 181.9 I	[9]
1-4		C_4H_9	Cr 161 Sm 198 I	[10]
1-5		C_4H_9	Cr 155 N 182 I	[11]
1-6		C_4H_9	Cr 170 N 172 I	[12]
1-7		C_4H_9	Cr 208 Sm 218 I	[13]
1-8		CN	Cr 93.5 N 244 I	[14]
1-9		CN	Cr_2 69.5 Cr_1 138 Sm 180 N 263 I	[14]
1-10		CN	Cr 129 SmA 139 N 256 I	[15,16]
1-11		CN	Cr 95 N 233 I	[17]
1-12		CN	Cr 143 SmC 164 N 258 I	[11]
1-13		CN	Cr 144 SmC 192 N 261 I	[11]
1-14		CN	Cr 154 N 242 I	[18]

TABLE 2 Physicochemical Properties of Some Liquid Crystals

No.	Compound	Phase transitions (°C)	$\varepsilon_\perp$	$\Delta\varepsilon$	$\Delta\varepsilon/\varepsilon_\perp$	Δn	Ref.
2-1		Cr 43.1 Sm 48.2 I		18.9[a]		0.134[a]	[19]
2-2		Cr 22 Sm 65.1 I		13.2[a]		0.134[a]	[19]
2-3		Cr 50.8 I		14.7[a]			[19]
2-4		Cr 95 I		9.4[a]			[19]
2-5[b]		Cr 31 N 60.5 I	3.3[c]	0.7[c]	0.21[c]	0.158[c]	[20,21]
2-6		Cr 45 SmA 71 I					[22]
2-7		Cr 44 SmB 59 SmA 89 I					[22]
2-8		Cr 49 SmA 77 I					[15]
2-9		Cr 71 N (52) I	10.0[d]	21.3[d]	21.3[d]		[23]
2-10		Cr 10 8.5 I					[23]
2-11		Cr 87.5 I					[23]
2-12		Cr 96 N 109 I	3.4[d]	3.4[d]	1.0[d]		[23]
2-13		Cr 33.6 N 43.5 I	10.9[c]	17.8[c]	1.63[c]		[17,24]
2-14		Cr 47.4 N 68 I	8.0[c]	6.5[c]	0.81[c]		[1,17,25]
2-15		Cr 22.5 N 35 I	8.2[d]	8.3[d]	1.01[d]		[23,26,27]

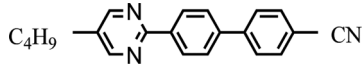
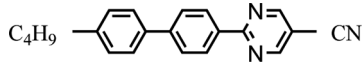
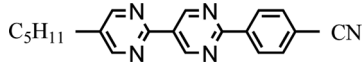
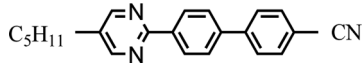
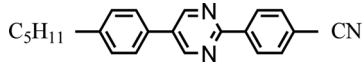
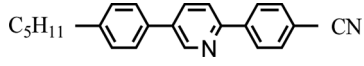
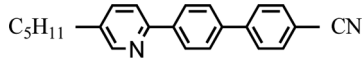
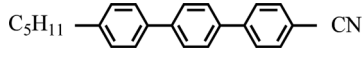
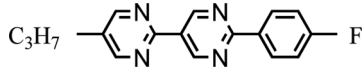
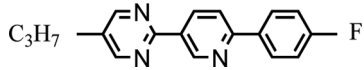
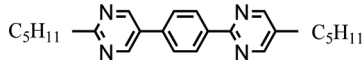
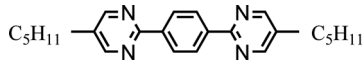
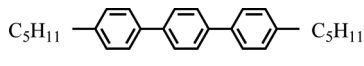
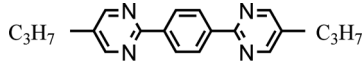
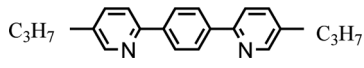
[a]Extrapolated from 10 wt% solution in ZLI-1132 at 20°C.
[b]$K_{33}/K_{11} = 0.68$, $\tau = 0.96$ [27].
[c]$\tau = T_{means}/T_{N-I} = 0.95$; T_{means}, T_{N-1}, K.
[d]$\tau = 0.98$.

TABLE 3 Physicochemical Properties of Liquid Crystals: A  B

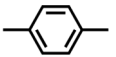
No.	A	B	Phase transitions (°C)	$\varepsilon_\perp$	$\Delta\varepsilon$	$\Delta\varepsilon/\varepsilon_\perp$	Δn	Ref.
3-1	C_4H_9	 $SCHF_2$	Cr 69.1 Sm 111.1 N 131.4 I	5.0[a]	5.5[a]	1.10[a]	0.184[b]	[19]
3-2	C_4H_9	 $SCHF_2$	Cr 41.5 Sm 95 N 119 I	5.7[a]	5.4[a]	0.95[a]	0.144[b]	[19,28]
3-3	C_4H_9	 OCF_2Cl	Cr 30 Sm 130.6 N 135.2 I		16.1[b]		0.142[b]	[29]
3-4	C_4H_9	 OCF_2Cl	Cr 79.5 Sm 121.7 N 130.3 I		12.2[b]		0.134[b]	[29]
3-5	C_5H_{11}	 F	Cr 116 N 158 I		16.9[b]		0.131[b]	[30]
3-6	C_5H_{11}	 F	Cr 98 N 150 I		7.9[b]			[30]
3-7	C_5H_{11}	 F	Cr 94.4 N 152.9 I		4.6[c]		0.098[a]	[31,32]
3-8	C_5H_{11}	 CN	Cr 100.5 N 231 I	3.2[d]	18.1[d]	5.66[d]		[33]
3-9	C_5H_{11}	 CN	Cr 113 N 228 I	3.2[d]	14.0[d]	4.38[d]		[17]
3-10	C_5H_{11}	 CN	Cr 157 SmA (153) N 235.3 I					[34]
3-11	C_5H_{11}	 CN	Cr 96 N 219 I		12.0[b]			[35]

[a] $\tau = 0.95$.
[b] Extrapolated from 10 wt% solution in ZLI-1132 at 20°C.
[c] Extrapolated from 20 wt% solution in liquid-crystalline material at 25°C.
[d] $\tau = 0.75$.

TABLE 4 Physicochemical Properties of Some Liquid Crystals

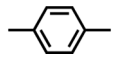
No.	Compound	Phase transitions (°C)	$\Delta\varepsilon$	Ref.
4-1		Cr 112 Sm 212.5 N 262 I		[14]
4-2		Cr_2 99.5 Cr_1 126.5 Sm 216.5 N 274.5 I		[14]
4-3		Cr 186 Sm 264 N	41.2[a]	[36]
4-4		Cr 124 Sm 204.5 N 259.5 I		[14]
4-5		Cr 125 N 241 I		[14]
4-6		Cr 76 N 232.1 I	19.6[b]	[17]
4-7		Cr 90 Sm 162 254 I	21.4[b]	[17]
4-8		Cr 130 N 239 I	13.0[c]	[33,37]
4-9		Cr 199 N 203 I		[36]
4-10		Cr 141 N 184 I		[38]
4-11		Cr 171 Sm 213.4 I		[39]
4-12		Cr 179 N 189.5 I		[40]
4-13		Cr 192 Sm 213 I		[13]
4-14		Cr 185 N 213 I		[40]
4-15		Cr 183 N 208 I		[41]

[a]Extrapolated from 2 wt% solution in liquid-crystalline material at 20°C.
[b]Extrapolated from 10 wt% solution in ZLI-1132 at 20°C.
[c]$\tau = 0.7$.

TABLE 5 Physicochemical Properties of Some Liquid Crystals: A  CN and binary mixtures

No.	A	Phase transitions (°C)	d_1 (Å)	d_2 (Å)	d_2/L	μ (D 25°C)	β (deg)	$\varepsilon_\perp$	$\Delta\varepsilon$	$\Delta\varepsilon/\varepsilon_\perp$	g, $T_{N\text{-}I}$	Δn	k_p (40°C)	Ref.
5-1	C_7H_{15} 	Cr 45 N 51 I		27	1.29	6.7	0	8.6[a]	16.0[a]	1.9[a]	0.450		0.6212[b]	[23,42–44]
5-2	C_7H_{15} 	Cr 30.9 N 47 I	17.9[c]	27.2[c]	1.30[c]	6.1	6	9.2[d]	15.8[d]	1.7[d]	0.495		0.6241	[1,17,24,43,45–47]
5-3	C_7H_{15} 	Cr 28.5 N 42 I		29	1.38	4.7		5.2[d]	11.1[d]	2.1[d]	0.500		0.6507	[26,42,44,45]
5-4	R 	N 51 I		23[c]				8.7[d]	19.6[d]	2.9[d]		0.174[d]		[1]
5-5	R 	N 44.1 I		25.6[c]				10.7[d]	16.0[d]	1.5[d]		0.175[d]		[1]
5-6	R 	N 39 I		27.4[c]				6.0[d]	11.7[d]	2.0[d]		0.184[d]		[1]

Note: **5-4**–**5-6** are binary mixtures (C_5 and C_7 = 40 mol%:60 mol%).
[a] $\tau = 0.98$.
[b] $T_{means} = 44.5$°C.
[c] $T_{means} = T_{N\text{-}I} - 10$°C.
[d] $\tau = 0.95$.

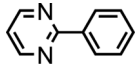
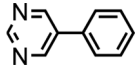
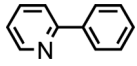
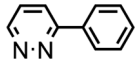
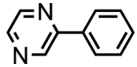
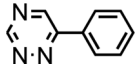
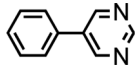
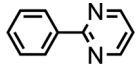
TABLE 6 Physicochemical and Electro-optical Properties of Some Liquid Crystals: A  CN and Binary Mixtures

No.	A	Phase transitions (°C)	η (cP)	γ_1(P 25°C)	E (eV 25°C)	K_{33}/K_{11}	U_{90} (V)	U_{10} (V)	τ_{on}^{1} (ms)	τ_{on}^{2} (ms)	τ_{off} (ms)	Ref.
5-1	C_7H_{15} 	Cr 45 N 51 I	25[a]									[23]
5-2	C_7H_{15} 	Cr 30.9 N 47 I										[24]
5-3	C_7H_{15} 	Cr 28.5 N 42 I	18.1[a]									[23,26]
5-4	R 	N 51 I		1.90	0.565	1.06[b]	1.07	1.48	70	20	90	[1]
5-5	R 	N 44.1 I		1.84	0.540	1.25[b]	1.08	1.54	75	20	105	[1]
5-6	R 	N 39 I		1.10	0.546	1.37[b]	1.30	1.78	85	21	69	[1]

Note: **5-4**–**5-6** are binary mixtures (C_5 and C_7 = 40 mol%:60 mol%). Electro-optical measurements: d = 10 μ, T_{meas} = 25°C, U_{meas} = 3 V.
[a] T_{means} = 38°C.
[b] τ = 0.95.

TABLE 7 Mesomorphic Properties of Liquid Crystals:

C_8H_{17}— A — $OC_5H_{10}C^*H(CH_3)C_2H_5$

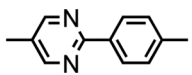
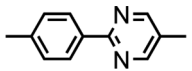
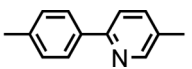
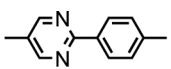
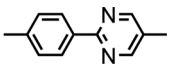
No.	A	Phase transitions (°C)	Ref.
7-1		Cr 13 SmC* 49 SmA 59 I	[48]
7-2		Cr 40.2 SmG* 43.4 SmC* 57.8 SmA 72.3 I	[49]
7-3		Cr 34.4 SmG* (28.5) SmF* 56.2 SmC* 70.5 I	[50]
7-4		Cr 98 Sm (80) SmC* 117 I	[51]
7-5		Cr 49 SmC* 62 I	[52]
7-6		Cr 44.5 SmC* 81.2 SmA 84.6 I	[15]
7-7		I 58 SmA 35 SmC* 29 Cr	[53]
7-8		Cr 16 SmC* 57.5 SmA 59 I	[52]

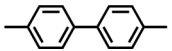
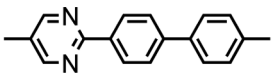
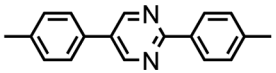
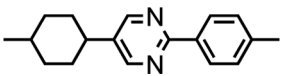
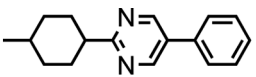
positions of its nitrogens, can create liquid crystals with low melting and moderate clearing points; widest nematic ranges or liquid crystals with moderate melting and highest clearing temperatures; and moderate nematic ranges among compounds of system (I).

Some of these results do not support the suggestion of Maier and Saupe that increasing the anisotropy of polarizability should enhance the nematic thermostability [64]. In this case, the clearing points of compounds, depending on the molecular fragment A, should always follow this dependence: $T_{cl} \rightarrow$ A: tetrazine < pyrimidine < pyridazine < pyrazine < pyridine < benzene [65,66].

As is evident from Tables 1 and 2, the positions of the nitrogen atoms in the pyrimidine ring introduced into the molecular core of liquid crystals significantly affect their mesomorphic properties. So far, pointing the nitrogens of the pyrimidine ring toward the cyanophenyl fragment in three-ring derivatives (compound **4-5**) results in the formation of only a nematic phase, exhibiting low melting and clearing temperatures in comparison with those of the corresponding

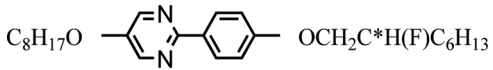
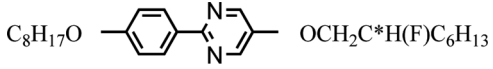
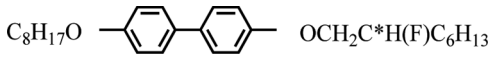
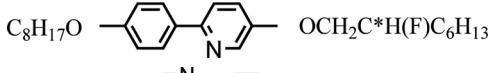
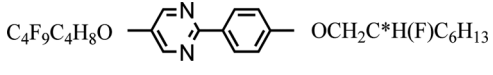
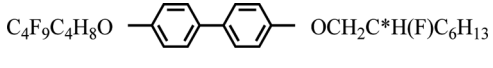
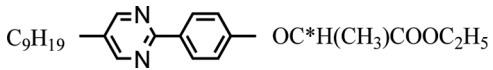
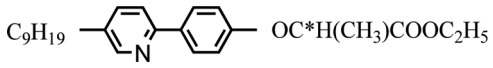
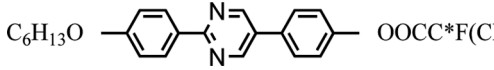
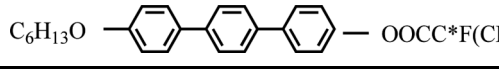
TABLE 8 Physicochemical and Electro-optical Properties of Some Liquid Crystals

No.	Compound	Phase transitions (°C)	Ps – (nCcm^2)	Θ (deg).	τ (μs)	γ_Φ (Pa s)	Ref.
8-1	$C_8H_{17}O$  $C_2H_4C^*H(F)C_6H_{13}$	Cr 35 SmA 82 I	3.6[a]	18[a]	157[a]		[54]
8-2	$C_8H_{17}O$  $C_2H_4C^*H(F)C_6H_{13}$	Cr 74 SmA 82 I	5.0[a]	25[a]	164[a]		[54]
8-3	$C_8H_{17}O$  $C_2H_4C^*H(F)C_6H_{13}$	Cr 66 Sml* 73 SmC* 86 SmA 90 I	11.3[b]		150[b]		[55]
8-4	$C_{10}H_{21}O$  $C_5H_{10}C^*H(CH_3)OC_2H_5$	Cr 10 SmC* 55 I	42.5[c]	32.3[c]		0.19[c]	[56]
8-5	$C_{10}H_{21}O$  $C_5H_{10}C^*H(CH_3)OC_2H_5$	Cr 43 SmC* (13) Ch (27) I	29[d]	39[d]		0.88[d]	[56]

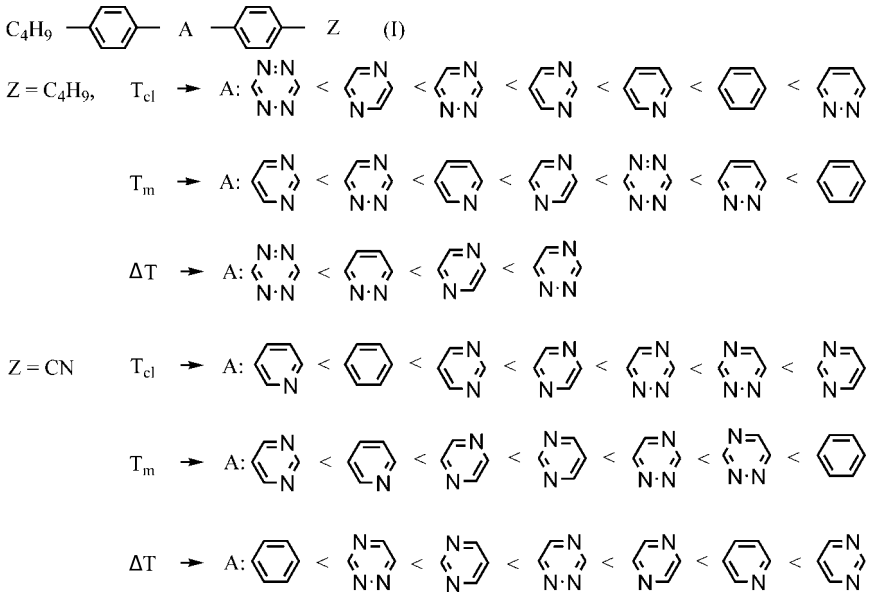
8-6	$C_{10}H_{21}O$  $C_5H_{10}C^*H(CH_3)OC_2H_5$	Cr 31 Sm_2 32 Sm_1 38 SmC^* 48 I	23.4[e]	23.8[e]	0.08[e]	[56]
8-7	$C_8H_{17}O$  $OOCCH_2C^*H(CF_3)C_6H_{13}$	Cr 94 SmC^* 133 SmA 154 I	2.7[f]	302[f]		[57]
8-8	$C_8H_{17}O$  $OOCCH_2C^*H(CF_3)C_6H_{13}$	Cr 75 SmC^* 125 SmA 151 I	2.8[f]	254[f]		[57]
8-9	$C_9H_{19}O$  $OC_5H_{10}C^*H(CH_3)C_2H_5$	Cr 69 SmB 110.8 SmC^* 120.4 SmA 158.7 I				[58]
8-10	$C_9H_{19}O$  $OC_5H_{10}C^*H(CH_3)C_2H_5$	Cr 37 SmB 103 SmA 112 I				[58]

[a,f] Measured in 10 and 5 wt% solution in liquid-crystalline material (LCM) at 25°C.
[b] Measured in 10 wt% solution in LCM at 20°C.
[c,d,e] T_{means} = 20, 10, 42°C, respectively.

TABLE 9 Physicochemical and Electro-optical Properties of Some Liquid Crystals

No.	Compound	Phase transitions (°C)	Ps – (nCcm^2)	Θ(deg).	τ (μs)	Ref.
9-1		Cr 69.9 SmC* 79.6 SmA 96.2 I	80.1[a]		15.1[a]	[59]
9-2		Cr 62.6 SmC* 92 SmA 97 I	94.4[b]		22[b]	[60]
9-3		Cr 113 I				[60]
9-4		Cr 73 Sm* 81.4 SmC* 107.9 I				[60]
9-5		Cr 84.2 SmC* 117.7 SmA 120.6 I	130.2[c]	37.8[c]	18.8[c]	[59]
9-6		Sm_4 47 Sm_3 87 Sm_2 91 Sm_1 97 SmC* 112 SmA 130 I	46.8[d]	31.3[d]	40[d]	[61]
9-7[e]		SmC* 63 SmA 64 Ch 89 I	10.4[f]	29[f]	160[f]	[62]
9-8[e]		SmC* 59 Ch 85 I	9.6[f]	26[f]	140[f]	[62]
9-9		Cr 88 SmC* 130 I	250[g]			[63]
9-10		Cr 172 SmC* 174 SmA 191 I I	87[g]			[63]

[a] $T_{means} = T_{SmC^* - SmA} - 9.6°C$.
[b] $T_{means} = T_{SmC^*} - 10°C$
[c,d,f] T_{means} = 100, 97, 20°C respectively.
[e] Measured as 10 wt% solution in ZLI-3234B.
[g] $T_{means} = T_{SmC^* - SmA} - 2°C$.

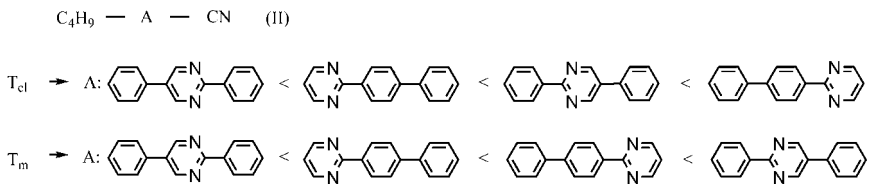


SCHEME 1 Mesomorphic properties of some liquid crystals.

compound **1-9** that has its pyrimidine nitrogens pointed in the opposite direction. In the case of two-ring cyano derivatives, the opposite situation was found (compounds **2-9** and **2-11**). It reveals the importance of the molecular structure of liquid crystals. As is evident from Table 2, pointing the nitrogens of the pyrimidine ring toward the hexyloxy group or hexyloxyphenyl fragment in two-ring hexyl-hexyloxy weakly polar derivatives creates the mesophases, which exhibit lower melting and clearing temperatures (compounds **2-5**–**2-8**). The influence of the position of the pyrimidine ring in the molecular core and the positions of its nitrogens on the mesomorphic properties of three-ring cyano derivatives can be illustrated by system (II), shown in Scheme 2.

It shows that the introduction of the pyrimidine ring as the central ring, with its nitrogens pointed toward the cyanophenyl fragment into the molecular core, forms the nematic phase, which exhibits the lowest melting and clearing temperatures. Similar trends can be observed in system (III) for weakly polar two-ring derivatives **2-5**–**2-8**, as shown in Scheme 3.

It can be seen from Table 4 that increasing the number of the pyrimidine rings in the molecualr core of liquid crystals enhances the melting and clearing temperatures (compounds **4-3**–**4-5**, **4-9**,



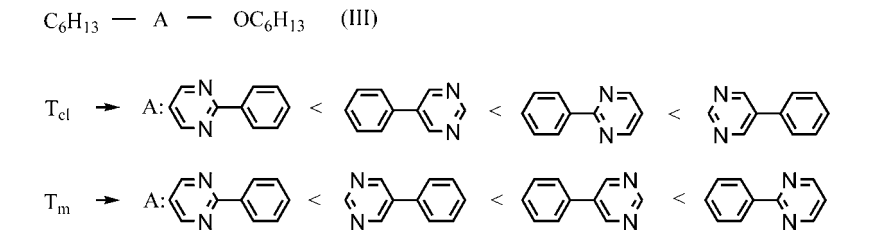
SCHEME 2 Mesomorphic properties of some liquid crystals.

4-10). Interestingly, pointing the nitrogens of two pyrimidine rings toward each other results in the disappearance of the smectic phase and the formation of the nematic phase, exhibiting higher melting-point and lower clearing temperatures in comparison with those of the corresponding derivative that has the nitrogens pointed in the same direction (compounds **4-11**, **4-12**).

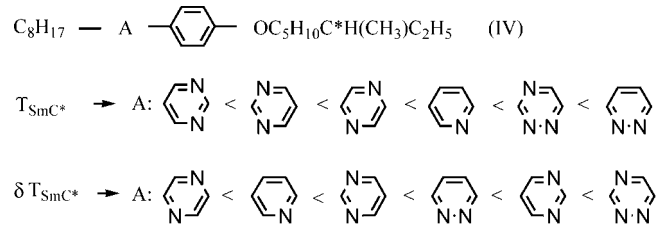
Similar trends can be found for the chiral liquid crystals presented in Tables 7–9 and systems (IV) and (V), shown in Schemes 4 and 5.

Particularly, the introduction of the pyrimidine ring into the molecular core of system (IV) creates liquid crystals showing the lowest smectic C* thermostability and moderate smectic C* range among compounds of this system. From system (V), it follows that pointing the nitrogens of the pyrimidine ring toward the chiral end group and the connection of the pyrimidine ring to the chiral end group forms the smectic C* phase, which exhibits the lowest smectic C* thermostability and range. Similar trends have been reported for other pyrimidine derivatives [67–253].

X-ray diffraction (XRD) of liquid crystals is one useful method for the study of the structure of their mesophases [**147 k, 148 k, see A and E spisok**] [46, 254]. The investigation of polar liquid crystals revealed in some cases the simultaneous existence of two fluctuation-layer structures with incommensurate periods d_1 and d_2, where $d_1 < L$, $L < d_2 < 2L$, and l is a molecular length [46,254]. XRD studies of 5-n-heptyl-2-(4-cyanophenyl)pyrimidine failed to reveal the simultaneous



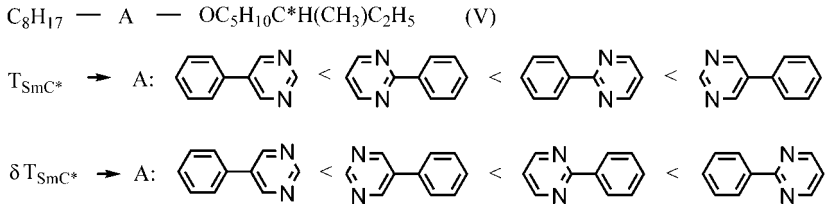
SCHEME 3 Mesomorphic properties of some liquid crystals.



SCHEME 4 Mesomorphic properties of some liquid crystals.

existence of two fluctuation-layer structures with incommensurate periods (compound **5-1**, Table 5) in comparison with that of the corresponding pyridine derivative **5-2**. Similarly, only a dimeric density wave was found for 4-n-heptyl-4′-cyanobiphenyl **5-3**. As is evident from Table 5, 5-n-heptyl-2-(4-cyanophenyl)pyrimidine and its binary mixture with the fifth homologue show more pronounced molecular overlap (d_2/L is smaller) in the formation of the dimers with respect to that of the corresponding pyridin-2,5-diyl and 1,4-phenylene derivatives (compounds **5-1**–**5-3**, mixtures **5-4**–**5-6**). In the case of some weakly polar pyrimidine derivatives, only monolayer formation ($d/L \leq 1$) has been observed [96,101,105,230]. Similar trends have been reported for other pyrimidine derivatives [141–143,164,170,203,212,228]. It should be noted that Refs. [192,224–227] present the results of XRD of some pyrimidine derivatives in their crystalline phases.

It can be proposed that the electronic and geometric structures of the pyrimidine ring and reference fragments [5,6,65,66,190] play a very important role in the intra- and intermolecular interactions [255–257], which affect the packing of the molecules and which predominantly influences mesophase stability [44,255–258]. Anisotropic dispersion interactions, and consequently the anisotropy of polarizability, depending on the electron density distribution in the molecular fragments under consideration, also influence the packing and hence the stability of the mesophases but play a secondary role compared to the steric factors [258]. Other molecular aspects such as the association



SCHEME 5 Mesomorphic properties of some liquid crystals.

[44] or dipole–dipole attraction in polar liquid-crystalline derivatives, which can influence the packing of the molecules, also affect the stability of the mesophases [258].

3. STATIC DIELECTRIC PROPERTIES

The relationship between the dielectric anisotropy $\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$, where $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ are dielectric constants that are parallel and perpendicular, respectively, to the nematic director **n**, and the molecular structure of liquid crystals are described by the theory of Maier and Meier [259]:

$$\Delta\varepsilon = \frac{\mathrm{NhF}}{\varepsilon_0}\left[\frac{\Delta\alpha - \mathrm{F}\mu^2}{\mathrm{kT}(1 - 3\cos^2\beta)\mathrm{S}}\right], \tag{1}$$

where $\mathrm{h} = 3\varepsilon^*/(2\varepsilon^* + 1)$, $\varepsilon^* = (\varepsilon_{\parallel} + 2\varepsilon_{\perp})/3$; $\Delta\alpha = (\alpha_{\parallel} - \alpha_{\perp})$ is the polarizability anisotropy; F is the cavity reaction field; μ is the dipole moment; β is the angle between the molecular long axis and the dipole moment, N is the number of molecules per unit volume, and S is the order parameter.

As is evident from Tables 2–5, the introduction of the pyrimidine ring into the molecular core of liquid crystals significantly increases their dielectric anisotropy in comparison with that of the corresponding pyridin-2,5-diyl, 1,4-phenylene, and other reference derivatives (compounds **2-1**–**2-4**; **2-9**, **2-13**, **2-15**; **3-1**–**3-9**, **3-11**; **4-3**, **4-6**–**4-8**; **5-1**–**5-3**, mixtures **5-4**–**5-6**). These results are due to the enhanced dipole moment of the pyrimidine ring and its coincidence with longitudinal molecular axis (for example, the dipole moments of pyrimidine, pyridine, and benzene diminish in the following order: 2.33 D, 2.21 D, 0 [260,261]). Because the direction of the dipole moment of the pyridazine ring is perpendicular to the longitudinal molecular axis, it lowers the total dipole moment and consequently the dielectric anisotropy of the pyridazine derivative (compounds **2-3** and **2-4**, Table 2). The importance of the positions of the pyrimidine ring in the molecular core and its nitrogens in the ring on the dielectric properties is illustrated by comparing the data of compounds **2-9** and **2-12**. The coincidence of the dipole moments of the pyrimidine ring and cyanophenyl fragment significantly increases the total dipole moment and the dielectric anisotropy in comparison with the opposite case. As can be seen from Table 2, the introduction of the pyrimidine ring into the molecualr core of two-ring alkyl-alkoxy derivatives produces weakly polar liquid crystals exhibiting positive dielectric anisotropy (compound **2-5**).

It has been shown that lowering the ratio $\Delta\varepsilon/\varepsilon_{\perp}$ is favorable for supertwisted nematic display applications [262]. In this case, weakly polar pyrimidine derivatives and strong polar reference compounds

look better than the corresponding strong polar pyrimidine derivatives (compounds **2-5**, **2-9**; **2-9**, **2-13**, **2-15**; **3-1**, **3-2**; **3-8**, **3-9**; **5-1**, **5-2**; **5-4**–**5-6**, Tables 2, 3, 5).

It has been demonstrated that mesogenic molecules possessing strongly polar terminal groups form associated pairs. Both head-to-head and head-to-tail pairing occurs [256], but antiparallel association predominates and reduces the effective dipole moment [44]:

$$\mu_{\text{eff}}^2 = g\mu^2, \tag{2}$$

where g is the correlation factor characterizing the association tendency. For nonassociating systems, g is equal to 1. The data collated in Table 5 reveal that compounds **5-1**–**5-3** exhibit the g values smaller than 1, indicating an antiparallel association, with the smallest g value recorded for the pyrimidin-2,5-diyl derivative **5-2**. Similar trends have been reported for other pyrimidine derivatives [100,109,120,122,128,129,137,139,155, 158–160,178,180,200,221,231]. The dielectric relaxation processes have been studied in Refs. [85,86,198,202,235].

4. OPTICAL PROPERTIES

The phenomenological relation between the refractive index and the electric polarization is defined as [263,264]

$$\frac{(n^{*2} - 1)}{(n^{*2} + 2)} = \frac{N\alpha^*}{3\varepsilon_o}, \tag{3}$$

where the mean polarizability $\alpha^* = (\alpha_{\parallel} + 2\alpha_{\perp})/3$; the mean refractive index $n^{*2} = (n_e^2 + 2n_o^2)/3$; and n_o is the ordinary and n_e is the extraordinary refractive indices. As expected, Equation (3) and polarizability data should explain the optical properties of the pyrimidin-2,5-diyl derivatives presented in Tables 2, 3, and 5. However, it is not so easy because the order of increasing the polarizability of the molecular fragments under consideration (see **Section 2**) is not supported by the order of increasing the optical anisotropy in some cases (see for example, compounds **2-1**, **2-2**; **3-1**–**3-4**). This situation needs more careful consideration including the direct measurements of the optical anisotropy and the study of the molecular packing of the discussed compounds [265,266]. Similar trends have been observed for other pyrimidine derivatives [98,100,109,120,128,129,137,139,178,221,223].

5. VISCO-ELASTIC PROPERTIES

It has been shown that the nematic liquid-crystalline materials for display applications should have a low viscosity for giving the acceptable

response times to liquid-crystalline displays [262,267]. The rotational viscosity γ_1 of a nematic liquid crystal (NLC) is a dissipative coefficient describing the rate of reorientation of the NLC director [268]. The magnitude of the rotational and flow viscosities depend on molecular structure, intermolecular association, and temperature [269,270]: as temperature increases, viscosity decreases [23,269–272]. According to the results on flow and rotational viscosity presented in Table 6, the pyrimidin-2,5-diyl derivatives exhibit higher values of flow and rotational viscosity compared with the corresponding pyridin-2,5-diyl and 1,4-phenylene derivatives (compounds **5-1**, **5-3**; mixtures **5-4**–**5-6**). A more pronounced and stable association tendency of the pyrimidin-2,5-diyl derivatives compared with that of reference compounds (see **Section 3**) may be responsible for increasing their viscosity [273].

In the bulk of NLCs, elastic properties are determined by three elastic constants (Oseen–Frank constants), corresponding to the restoring forces opposing splay (K_{11}), twist (K_{22}), and bend (K_{33}) [274,275]. It has been shown that the elastic constants depend on molecular structure, intermolecular association, and temperature of NLCs [276–278]. The elastic constant ratio K_{33}/K_{11} of liquid crystals is an important parameter for supertwisted nematic liquid crystal displays (STN-LCDs), defining their electro-optical performance [279]. As is evident from Table 6, the pyrimidin-2, 5-diyl cyano derivatives show the lowest value of the ratio K_{33}/K_{11}, which corresponds to the lowest value of the squared dimeric density wave period d_2^2 for binary mixtures **5-4**–**5-6** presented in Table 5 [46]. Similar trends have been demonstrated for other pyrimidine derivatives [98,100,109,122,220–222].

6. MOLECULAR PACKING

It has been shown that liquid-crystalline molecular packing plays a very important role in the creation of mesophases [255,257] and defines their optical properties [263]. The molecular packing coefficient is expressed in [45] as

$$k_p = \frac{N_A V_\rho}{M} \tag{4}$$

where N_A is the Avogadro number, ρ is the density, M is the molecular weight, and V is the intrinsic (van der Waals) volume of the molecule, calculated from the van der Waals volume increments of the individual atoms or by using the average atomic radii and chemical lengths. As is evident from Table 6, the introduction of the pyrimidine ring into the molecular core of two-ring cyano derivatives results in the formation of liquid crystals exhbiting the lowest value of the molecular

packing coefficient in comparison with that of reference compounds (compounds **5-1**–**5-3**). Similar trends have been found for other pyrimidine derivatives [96,179,223,230].

7. COMPARATIVE CHARACTERISTICS OF LIQUID CRYSTALS

Selection of the best components for liquid-crystalline materials and prediction of new chemical structures require comprehensive comparative investigation of the physicochemical and electro-optical characteristics of liquid-crystalline compounds. It has been shown that the dielectric, diamagnetic, viscous, and elastic constants, as well as the nematic–isotropic transitions of solutions of homologues, approximately obey the additive rule [280]. This fact is employed for the comparison of physicochemical and electro-optical properties of the pyrimidin-2,5-diyl and reference derivatives (mixtures **5-4**–**5-6**, Tables 5, 6). This was done by using binary mixtures (containing a pentyl and a heptyl homologue), giving broad nematic ranges with clearing points depending on the molecular structure as observed for the pure compounds in **Section 2**. From Table 6, it follows that the pyrimidin-2,5-diyl cyano derivatives show the lowest threshold U_{90} and saturation U_{10} voltages in comparison with those of reference compounds. These results are consistent with the dependence of the threshold voltage of the twist effect on the dielectric anisotropy and elastic constants [262]:

$$U_{90} \sim \pi \left[\frac{\kappa}{\varepsilon_o \Delta\varepsilon} \right]^{1/2} \tag{5}$$

where κ is the elastic expression, $\kappa = K_{11} + (K_{33} - 2K_{22})/4$.

The lowest twist-effect response times of the 1,4-phenylene cyano derivatives compared with those of the corresponding heteroaromatic derivatives (mixtures **5-4**–**5-6**) can be correlated with the corresponding lowest viscous behavior of these compounds.

8. PHYSICAL PROPERTIES OF THE SMECTIC C* PHASE

The spontaneous polarization P_s of the smectic C^* liquid crystals is an important parameter because of its linear coupling with an applied electrical field, which is the basis of the application of these materials [281]. The polarization is caused by the cramped rotation of the dipoles of the molecules and varies with the position of these dipoles with respect to the chiral group [281]. The spontaneous polarization P_s is a quantity directly related to the response time τ as a switching device [282]:

$$\tau = \frac{\gamma_\varphi \sin^2 \Theta}{P_s E}, \quad (6)$$

where γ_φ is the rotational viscosity, which refers to the rotation about an axis perpendicular to the director **n** and P_s; Θ is the tilt angle; and E is an applied electric field.

According to Scherowsky and Sefkow [283], liquid crystals should have the following structural features to obtain high spontaneous polarization:

1) a high dipole moment perpendicular to the longitudinal molecular axis;
2) short distances between the chiral center, the lateral dipole, and the mesogenic part, respectively; and
3) a restriction of free rotation in the chiral region.

As is evident from Tables 8 and 9, the replacement of the pyridine ring by the pyrimidine ring with its strong dipole moment pointed along the longitudinal molecular axis (see **Section 3**) increases (compounds **9-7**, **9-8**) and decreases the spontaneous polarization (compounds **8-2**, **8-3**, and Ref. [62]). In the case of the replacement of the benzene ring by the pyrimidine one, only an increase of the spontaneous polarization was observed (compounds **8-4**–**8-6**; **9-5**, **9-6**; **9-9**, **9-10**). Similarly, one can find higher tilt angles, rotational viscosities, and response times of the pyrimidin-2,5-diyl derivatives compared with those of reference derivatives [compounds **8-4**–**8-6**; **9-5**, **9-6** (except response times); **9-7**, **9-8**]. As is evident from Table 8, moving on the pyrimidine ring closer to the chiral end group increases the spontaneous polarization and lowers the response times (compounds **8-7**, **8-8**). Lowering the distance between the pyrimidine ring and the chiral center and changing the direction of the dipole moment of the pyrimidine ring in 180° enhances the spontaneous polarization, response times, and tilt angles (compounds **8-1**, **8-2**; **9-1**, **9-2**). It is difficult to compare compounds **8-4** and **8-5** because the measurements of the spontaneous polarization and tilt angle were made at the different absolute temperatures. A lower measuring temperature obviously corresponds to a higher rotational viscosity (compounds **8-4** and **8-5**). Similar trends have been reported for other pyrimidine derivatives [80,83, 84,94,95,102,115,116,147,153,162–164,169,176,177,185,191,202,205,210, 212,214,217,228,236,243,244,247,251,252].

9. CONCLUSION

Systematic studies of the effects of introducing the pyrimidine ring into the molecular structure of calamitic liquid crystals on the creation

of the mesophases and their physicochemical and electro-optical properties have been performed, with attempts to correlate the molecular-level parameters with the observed properties. The information here presented may lead to a better understanding of the nature of liquid crystals.

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