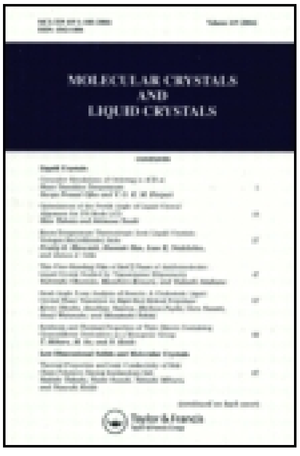

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

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Version of record first published: 20 Apr 2011.

To cite this article: George W. Smith (1978): Some Extended Range Nematic Liquid Crystal Mixtures, Molecular Crystals and Liquid Crystals, 49:1, 27-31

To link to this article: http://dx.doi.org/10.1080/00268947808070322

Mol. Cryst. Liq. Cryst. Vol. 49 (Letters), pp. 27-31

Printed in the United States of America

SOME EXTENDED RANGE NEMATIC LIQUID CRYSTAL MIXTURES

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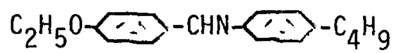
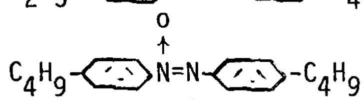
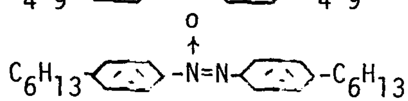
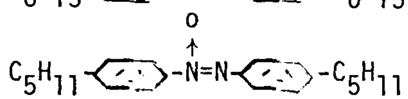
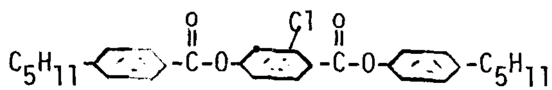
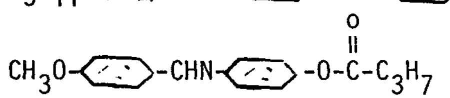
(Submitted for Publication June 1, 1978)

Extending the temperature range of nematic liquid crystals is technologically important for the operation of electrooptic devices in environments where ambient temperatures vary widely. The expected lowest temperature limit (eutectic temperature) of eutectic-forming nematic mixtures is calculable from the equations of le Chatelier, Schröder, and van Laar (CSL). The assumptions basic to the derivation of these equations are discussed elsewhere.[1,2] However, straightforward application of the method makes it apparent that the greatest depression of the eutectic temperature is obtained for components having both low crystal-nematic temperature T_{KN} and small crystal-nematic latent heat L_{KN}.

Thus an expeditious search for extended temperature range liquid crystals starts with measurements of T_{KN} and L_{KN}. Ideally it is also desired to have nematic-isotropic temperature as high as possible. Furthermore, the three CSL assumptions -- no solid solution formation, ideal nematic solution, and temperature-independent L_{KN} values -- must hold in order for the calculation method to work. And finally, one hopes that unexpected and unwanted non-nematic phases will not intervene in the predicted nematic temperature range.

This note reports phase ranges for some liquid crystal mixtures based on components for which thermal parameters have been previously published.[3,4,5] The expected eutectic nematic ranges for the mixtures were calculated using a simple computer program based on the iterative method of Hulme, et al.[5] The nematogens chosen for the mixtures and their thermal parameters are listed in Table I.

TABLE I. Liquid crystals used in mixtures.

Symbol	Chemical Formula	T_{KN}(°C)	L_{KN}(kJ/mol)	T_{NI}(°C)
I		22.6	13.9	46.0
II		36.0	17.2	79.5
III		19.6	13.4	32.5
IV		25.5	17.2	54.3
V		26.6	15.7	67.9
VI		39.6	23.8	123.0
VII		50.5	23.3	111.8
VIII		130.5	17.2	239.0

Expected eutectic compositions and nematic temperature ranges were calculated for nine mixtures (six binaries, two ternaries, and one quaternary). Corresponding mixtures were prepared (with compositions approximating the calculated values as closely as possible) and the phase behavior determined by differential scanning calorimetry and (in some cases) thermal microscopy.[4] The predicted and measured temperature ranges are shown in Table II.

A glance at Table II reveals three kinds of behavior. The measured temperature ranges for mixtures 1, 7, and 9 are in reasonable agreement with the values predicted from the CSL equations, indicating that these systems obey the three fundamental assumptions fairly well. The measured lower limits for mixtures 2, 3, 4, 5, and 8 are all considerably higher than the predicted values. The similarity in molecular structures for the components (particularly III, IV, and V) lead one to suspect the formation of solid solutions.[1] Mixture 6 does not display the expected eutectic behavior due to the formation of a smectic phase (temperature range 93°C to 157°C) at the predicted eutectic composition.[6,7] Indeed component VIII has a marked tendency to form smectics in mixtures, as was revealed by experiments with a number of binaries containing component VIII. It appears that either intervention of an unwanted smectic phase or formation of solid solutions can militate against the existence of wide-range nematic eutectics.

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TABLE II. Liquid crystal mixtures and their predicted and measured nematic temperature ranges.

Mixture	Components	Calculated Eutectic Point and Nematic Range: Mole Fractions	T_{eut}(°C)	T_{NI}(°C)	Measured Nematic Temperature Range(°C)
1	II,VII	0.64,0.36	17.0	92.0	19 to 88
2	I,V	0.55,0.45	-7.0	56.0	-1 to 54
3	IV,V	0.49,0.51	-2.0	61.0	14 to 61
4	III,IV	0.57,0.43	-7.0	42.0	-1 to 42
5	III,V	0.56,0.44	-8.0	48.0	6 to 49
6	VI,VIII	0.62,0.38	24.0	168.0	smectic phase intervenes
7	II,VII,VI	0.46,0.23,0.31	4.0	101.0	6 to 100
8	III,IV,V	0.41,0.28,0.31	-21.0	50.0	1 to 51
9	II,VII,VI,V	0.30,0.13,0.17,0.40	-11.5	86.0	-9 to 82

5. D. S. Hulme, E. P. Raynes and K. J. Harrison, J.C.S. Chem. Comm. 1974, p. 98; also G. W. Gray, K. J. Harrison and J. A. Nash, Pramana, Suppl. No. 1, 381-396 (1975).

6. M. Domon and J. Billard, Pramana Suppl. No. 1, 131-154, (1975), have given a detailed treatment of phase diagrams for binary liquid crystal mixtures. They discuss the existence in mixtures of mesophases which do not occur in either pure component.

7. The smectic phase is not observed for compositions sufficiently far from that calculated for the eutectic. Thus for mole fractions of component VI equal to 0.10 and 0.87 respectively, the following transition temperatures were observed: eutectic (crystal to crystal + nematic), 25°C and ~29°C; excess (crystal + nematic to nematic comesophase), ~115°C and ~35°C; and nematic-isotropic, 226°C and 133°C.