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Temperature effect on refractive indices and order parameter for mixture liquid crystal (UCF)

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ABSTRACT

Liquid crystals are state of substance possesses characteristics between traditional liquid and crystallization. For instance, perhaps liquid crystal away similar to the crystals. There are different types of crystal in the liquid phase .In this research has been the use of high refractive birefringence as well as (UCF), the study of the influence of temperature on the refractive indices at wavelength (623.8 nm), and room temperature. In addition, we used the four – parameters model to study this case, and compared between the experimental results with theoretical. This study is showed, when the temperature increases the ordinary refractive indices (n_o) increases. And both the extraordinary refractive indices (n_e), optical anisotropy (birefringence Δ n) are decreases, also we find the order parameter (S) decreased as the temperature is increased.

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INTRODUCTION

Temperature is play an important role in affecting the LC refractive indices .As the temperature increases, n_e behaves differently from n_o.The derivative of n_e (i.e, $\partial n_e / \partial T$) is always negative. However, $\partial n_o / \partial T$ changes from negative to positive as the temperature exceede the crossover temperature J.Li,S.Gauza,and S.T.Wu,(2004).

For elevated temperature operation of a LC device, temperature – dependent refractive indices need to be modeled accurately. Some semi – empirical models W.H.de Jeu, (1980), I.C.Khoo and R.Normandin,(1985) have been developed for describing the temperature effect on the LC refractive indices. However, a comprehensive model would be much more desirable.

B – Theory:

The classical Clausius – Mossotti equation[4] correlates the dielectric constant (ε)of an isotropic medium with its molecular polarizability (α) at low frequencies . Replacing $\varepsilon = n^2$, the Lorentz – Lorenz equation M.Warenghem and G.Joly,(1991) correlates the refractive index of an isotropic medium with molecular polarizability in the optical frequencies. In principle , these two equations are derived for isotropic media and are not suitable for anisotropic liquid crystals in which dielectric constants ($\varepsilon_{\parallel}$, $\varepsilon_{\perp}$) and molecular polarizabilities $(\alpha_{\parallel}, \alpha_{\perp})$ are all anisotropic in nematic phase.

Vuks made a bold assumption that the internal field in a LC is the same in all direction and gave a semi – empirical equation correlating the refractive indices with the molecular polarizabilities for anisotropic materials M.F.Vuks, (1966).

$$\frac{n_{e,o}^2 - 1}{<n^2> + 2} = \frac{4\pi}{3} N\alpha_{e,o} \qquad (1)$$

In equation (1) ,n_e and n_o are refractive indices for the extraordinary ray and ordinary ray , respectively, Nis the number of molecules per unit volume , $\alpha_{e,o}$ is the molecular polarizability, and $< n^2 >$ is defined as

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$$< n^2 > = \frac{n_e^2 + 2n_o^2}{3} \tag{2}$$

In Eq. (1), n_e and n_o are coupled together so that the relationship between the refractive indices and the corresponding molecular polarizabilities is not clear. To reveal this relationship, we should decouple n_e from n_o by solving Eq.(1). Substituting Eq.(2) to Eq.(1) and separating n_e and n_o, we obtain J.D.Jackson, (1962).

$$n_e = \sqrt{1 + \frac{4\pi N\alpha_e}{1 - \frac{4}{3}\pi N<\alpha>}} \tag{3}$$

$$n_o = \sqrt{1 + \frac{4\pi N\alpha_o}{1 - \frac{4}{3}\pi N<\alpha>}} \tag{4}$$

were $< \alpha >$ is the average polarizability of the LC molecule and is defined as L.M.Bilnov, (1983).

$$< \alpha > = \frac{\alpha_e + 2\alpha_o}{3} \tag{5}$$

when N $<\alpha>$ is small, Eqs.(3) and (4) can be expanded into power series .Retaining the first two terms, we derive J.D.Jackson, (1962).

$$n_e \approx \frac{3\sqrt{2}}{4} + \frac{\sqrt{2}\pi N < \alpha >}{1 - \frac{4}{3}\pi N < \alpha >} + \frac{\frac{2\sqrt{2}}{3}\pi NS(\gamma_e - \gamma_o)}{1 - \frac{4}{3}\pi N < \alpha >} \tag{6}$$

$$n_o \approx \frac{3\sqrt{2}}{4} + \frac{\sqrt{2}\pi N < \alpha >}{1 - \frac{4}{3}\pi N < \alpha >} - \frac{\frac{\sqrt{2}}{3}\pi NS(\gamma_e - \gamma_o)}{1 - \frac{4}{3}\pi N < \alpha >} \tag{7}$$

Birefringence of a LC material is defined as $\Delta n = n_e - n_o$. Subtracting Eq.(7) from Eq.(6), we obtain

$$\Delta n \approx \frac{\sqrt{2}\pi NS(\gamma_e - \gamma_o)}{1 - \frac{4}{3}\pi N<\alpha>} \tag{8}$$

The average refractive index of a LC is defined as

$$< n > = \frac{n_e + 2n_o}{3} \tag{9}$$

Substituting Eqs.(6) and (7) into Eq. (9), we derive

$$< n > \approx \frac{3\sqrt{2}}{4} + \frac{\sqrt{2}\pi N < \alpha >}{1 - \frac{4}{3}\pi N < \alpha >} \tag{10}$$

Substituting Eqs. (8) and (10) back to Eqs. (6) and (7), the refractive indices have the following simple expressions:

$$n_e = < n > + \frac{2}{3}\Delta n \tag{11}$$

$$n_o = < n > - \frac{1}{3}\Delta n \tag{12}$$

In theory ,both n_e and n_o are functions of wave length and temperature. The wavelength effect has been addressed extensively. Here, we focus on the temperature effect. According to our experimental data and fitting results, the average refractive index $<n>$ decreases linearly as the temperature increases J.Li,S.Gauza,and S.T.Wu, (2004).

$$< n(T) > = A - BT \tag{13}$$

Equation (13) has a negative slope. The value of B is around 10^{-4} K^{-1}

On the other hand , birefringence is dependent on the order parameter **S** .Based on Haller 's approximation, the order parameter can be approximated as $\mathbf{S = (1 - T/T_C)^{\beta}}$ .Thus, the temperature – dependent birefringence has the following form Haller,(1985).

$$\Delta n(T) = \Delta(n)_o (1 - \frac{T}{T_C})^{\beta} \tag{14}$$

In Eq.(14),(Δn)$_o$ is the LC birefringence in the crystalline state(or **T** = 0 K), the exponent β is a material constant, and $\mathbf{T_c}$ is the clearing temperature of the LC material under investigation.

Substituting Eqs. (13) and (14) back to Eqs.(11) and (12), we derive the four – parameter model for describing the temperature effect on the LC refractive indices:

$$n_e \approx A - BT + \frac{2(\Delta n)_o}{3}\left(1 - \frac{T}{T_C}\right)^{\beta} \tag{15}$$

$$n_O \approx A - BT - \frac{(\Delta n)_o}{3}\left(1 - \frac{T}{T_C}\right)^{\beta} \tag{16}$$

At the first glance, Eqs.(15) and (16) seem to have four fitting parameters. As a matter of fact ,parameters **A** and **B** can be obtained from fitting the temperature – dependent $< n(T) >$ data at a given wavelength, and parameters $(\Delta n)_o$ and β can be obtained by fitting the temperature – dependent Δn data. Thus, once we have measured the temperature – dependent n_e and n_o, we rearrange the data into $< n(T) >$ and $\Delta n(T)$ and parameters [A,B],and [$(\Delta n)_o$, β] can be obtained fairly straightforwardly.

For direct – view and projection LC displays, the LC mixture employed usually exhibits a low birefringence and high clearing temperature ($T_C > 90\ C^0$).Under the condition that $T << T_C$, the $(1 - T/T_C)^{\beta}$ term in Eqs. (15) and (16) can be expanded into a power series. Keeping the first three terms, we obtain

$$n_e = A_e - B_e T - C_e T^2 \tag{17}$$

$$n_O = A_O - B_O T + C_O T^2 \tag{18}$$

where

$$A_e = A + \frac{2(\Delta n)_o}{3}$$
$$B_e = B + \frac{2(\Delta n)_o \beta}{3T_C} \tag{19}$$
$$C_e = \frac{(\Delta n)_o \beta(1-\beta)}{3T_C^2}$$

And

$$A_O = A - \frac{(\Delta n)_o}{3}$$
$$B_O = B - \frac{(\Delta n)_o \beta}{3T_C} \tag{20}$$
$$C_O = \frac{(\Delta n)_o \beta(1-\beta)}{6T_C^2}$$

Equations (17) and (18) indicate that the LC refractive indices have a parabolic relationship with temperature for the low birefringence and high clearing – point LC material when the operating temperature is far from the clearing point. For n_e , the placket of the parabola is downward, whereas for n_o, the placket is upward Stefano,B. (2006).

2 – Order parameter (S):

1- Direct Haller 's Extrapolation Method:

In homogeneous alignment, a nematic liquid crystal behaves like a uniaxial crystal. The relation between macroscopic order parameter and temperature we will take the relation between macroscopic order parameter and refractive indices in parallel and perpendicular direction of the long molecular axis were obtained by modifying the equation for uniaxial crystal as Singh,*et al*., (1990).

$$n_{\parallel} = \bar{n} + \frac{2}{3} Q . \Delta n \tag{21}$$

$$n_{\perp} = \bar{n} + \frac{1}{3} Q . \Delta n \tag{22}$$

Where $\bar{n}$ is the average refractive index and Δn is the birefringence corresponding to complete alignment and for uniaxial alignment and for uniaxial crystal $n_{\parallel} = n_e$, $n_{\perp} = n_o$ and $\delta n = n_e - n_o$ Musevic,*et al*., (1989), W.H.de Jeu,(1980). From both the equations (21) and (22) we get

$$Q = \frac{n_{\parallel} - n_{\perp}}{\Delta n} = \frac{n_e - n_o}{\Delta n} = \frac{\delta n}{\Delta n} \quad (23)$$

The value of macroscopic order parameter (S) becomes 1 at absolute temperature that is at (0 K^0) $\delta n = \Delta n$. This can be determined by extrapolating δn for T = 0 K^0. This extrapolation is done on the linear portion of the graph drawn between birefringence (δn) versus $lin\ [1 - \frac{T}{T_C}]$ as done by other workers Singh, *et al*., (1990), Arora, *et al*.,(1992).

2 – Modified Vuk′ s Method:

In order to find out the microscopic order parameter (S) the refractive indices n_e and n_o have been analyzed following the method of Haller Haller,I.,(1975) and Horn Horn,R.G. (1978). This method uses Vuk ′ s relation Vuks,M.F.(1966).

$$S\left[\frac{\delta\alpha}{\alpha}\right] = \frac{3\,(n_e^2 - n_o^2)}{(n_e^2 + 2\,n_o^2 - 3)} \quad (24)$$

Where $\delta\alpha$ is the polarizability anisotropy and α is the molecular polarizability. The principal polarizabilities are determined as described by Sarana *et al* Sarna, R.K., *et al*,(1979), Chandel,V.S., *et al*., (2011).

C – Experimental setup:

When the laser beam polarized at 45 degrees with respect to the director of the crystal passes through the sample, it splits itself in an " ordinary ray " and an " extraordinary ray" .Because of the LC birefringence the two beams exit the LC cell traveling in two different directions, by measuring the deviation angle of these rays with respect to the situation in which the (UCF) LC is absent it is easy to retrieve the two refractive indices of the liquid crystal by means of the refractions laws.

To study the temperature effect on refractive indices we put the LC cell in hot stage (see Fig.1), the hot stage is controlled by thermometer and timer.

In Fig. (1), we show the ordinary and extraordinary spots as detected by the detector. Two different situation are presented, so that it is easy to find the position of the two beam maxima. The point where ray R_{ref} (see Fig.2), emerging from the empty wedge, encounters the observation plane π , which perpendicular to R_{ref}, is determined experimentally by translating the detector along the X – axis by means of the millimetric translator until the peak of the spot is exactly in the center of the detector. After that the cell is filled with the (UCF)LC an the two refracted beams R_o and R_e appear (see Fig. 2).

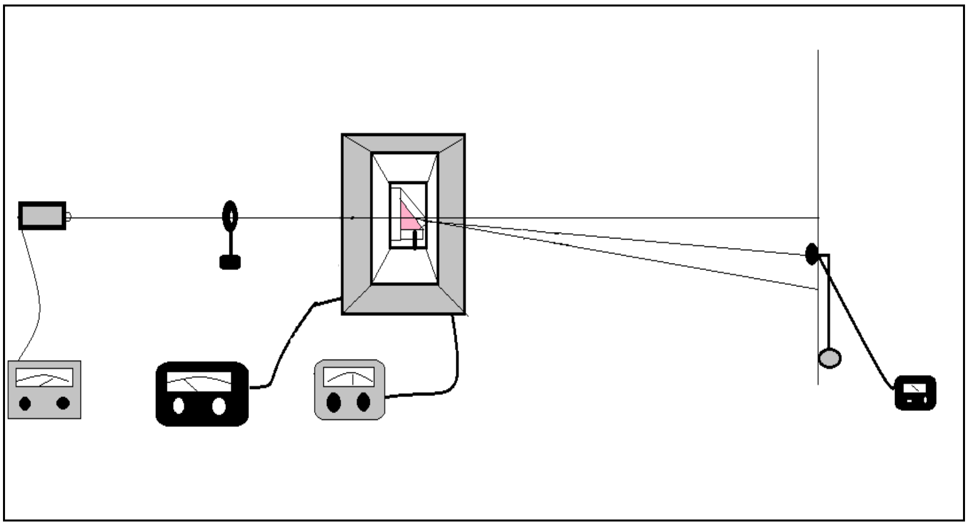

Fig. 1: Schematic diagram of the thermo – optic system

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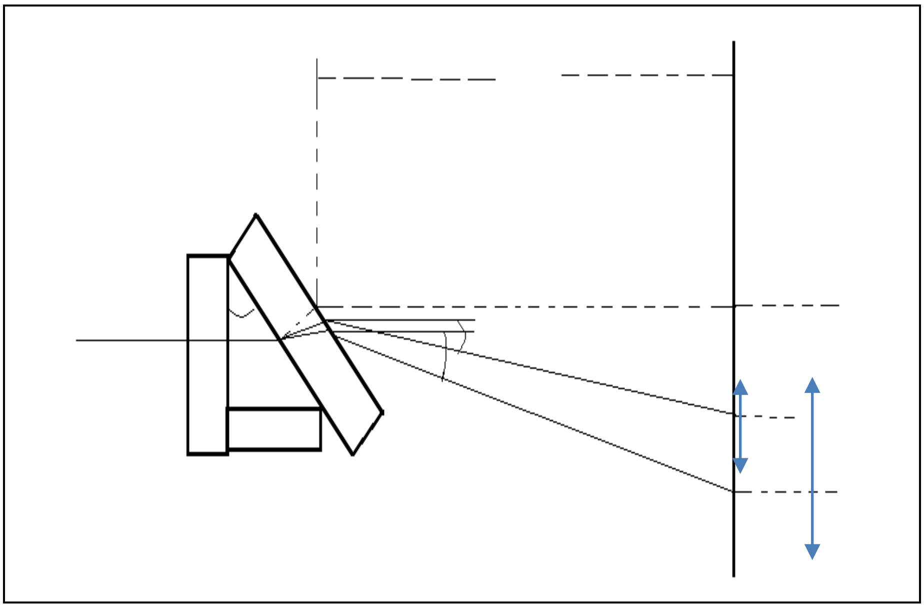

Fig. 2: Liquid crystal wedged cell, the He – Ne laser beam encounters first the substrate A. When the cell is empty only ray R_{ref} emerges. When the wedge is filled the ordinary and extraordinary rays R_o and R_e appear. The angles formed by R_o and R_e with R_{ref} are called θ_o, θ_e) Stefano,B. (2006).

Where θ is the angle of the wedge formed by the two SiO_2 plates θ_o and θ_e are the angles formed by the two beams R_o and R_e with respect to the beam R_{ref} emerging from the wedge when the (UCF) liquid crystal is absent .From elementary geometry it follows

$$\theta_o = \tan^{-1}\frac{X_o}{L} \quad (25)$$

$$\theta_e = \tan^{-1}\frac{X_e}{L} \quad (26)$$

So that

$$n_o = \frac{\sin(\theta + \tan^{-1}(\frac{X_o}{L}))}{\sin(\theta)} \quad (27)$$

$$n_e = \frac{\sin(\theta + \tan^{-1}(\frac{X_e}{L}))}{\sin(\theta)} \quad (28)$$

Each measurement has been repeated many times changing the temperature of the sample in order to reconstruct the temperature dependence of the refractive indices , every time the temperature was changed, we waited about 20 minutes,in order to reach a good thermalization, and the average refractive index is

$$< n(T) > = \frac{(2n_o + n_e)}{3} \quad (29)$$

And the optical anisotropy

$$\Delta n = n_e - n_o \quad (30)$$

To find the relationship between the order parameter (S) and temperature (T) for mixture liquid crystal (UCF), we will take the refractive indices values for extraordinary and ordinary refractive index from temperature effect on refractive indices ,and take the birefringence value for (UCF) LC at ($0\ K^0$) is (Δn = .6827).

RESULT AND DISCUSSION

1- The temperature effect on ordinary refractive index (n_o)

According equation (27), we find the ordinary refractive indices, these data′ s is representation in Fig (3).

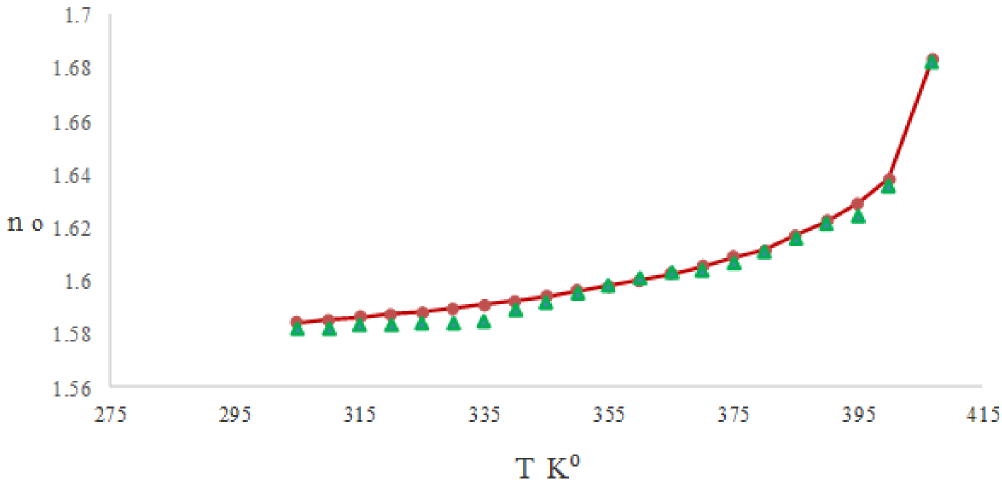

Fig. 3 : Temperature – dependent ordinary refractive index (n_o) for mixture liquid crystal (UCF), at wave length (623.8 nm). Triangles are experimental data for ordinary refractive index[Eq (27)] and solid lines are fittings the four – parameter model [Eq. (16)].

From above the figure, we find the ordinary refractive index is increase when the temperature is increase,these behaver is agreement with many researches same as Jun Li . *et al.*, (2006). To explain this case we can say because of the random motion for rod like molecules is increase when the temperature is increase, therefore, the ordinary beam velocity is decrease.

2 - The temperature effect on extraordinary refractive index (n_e)

Appling the equation (28), we find the extraordinary refractive index (n_e) is decrease when the temperature is increase (see Fig.(4)).

To explain above figure, we find the velocity of extraordinary beam is have larger values, therefore, the extraordinary refractive index is decrease when the temperature value is increase. This result is agreement with other researches same as Thomas,T.,*et al.*, (2006).

3 – Temperature effect on average refractive index < n >:

By using equation (29), and using the ordinary and extraordinary refractive index values, we find the average refractive index values, these data' s is representation in Fig (5). This figure is shown, the average refractive index is decrease linearly as the temperature increase.

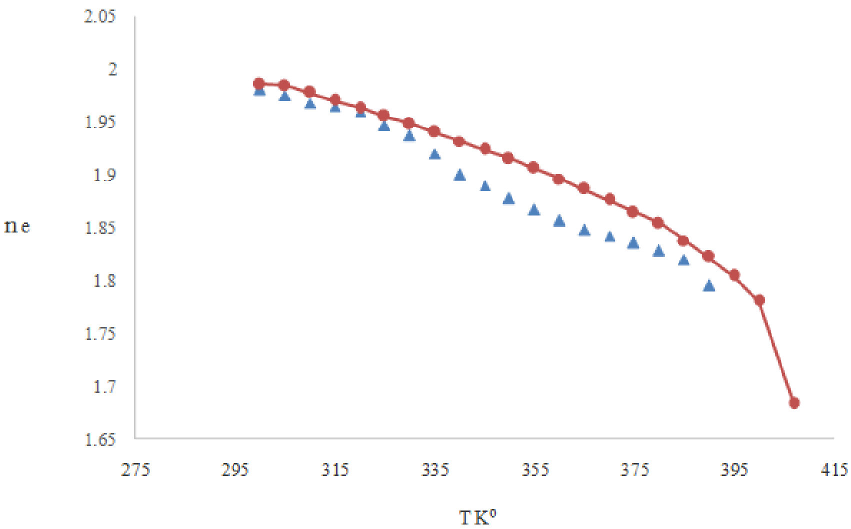

Fig. 4: Temperature – dependent extraordinary refractive index (n_e) for mixture liquid crystal (UCF) at wave length (623.8 nm), triangles are experimental data [Eq.(28)]. Solid lines are fitting using the four – parameter model [Eq. (15)].

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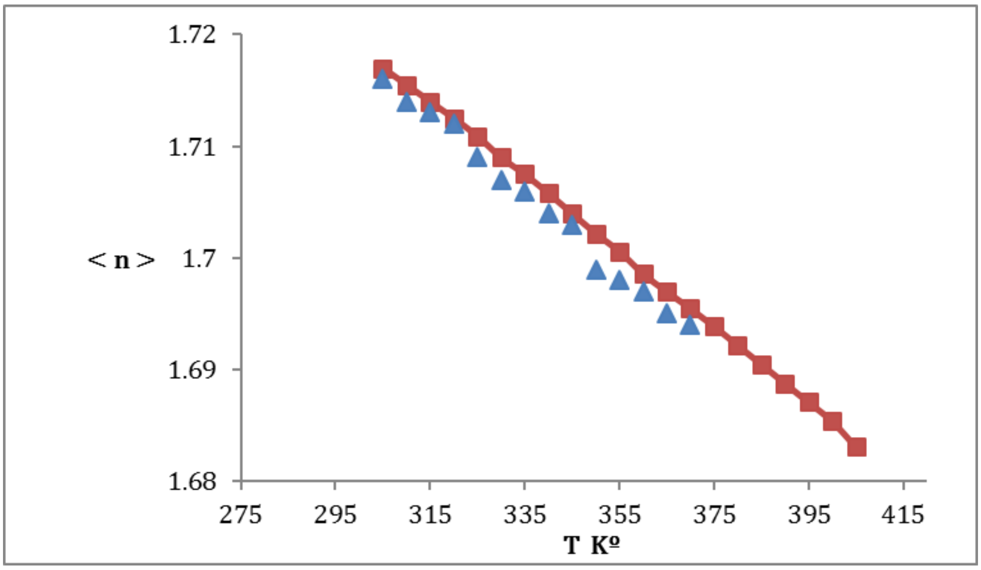

Fig. 5: Temperature – dependent on average refractive index < n > for mixture liquid crystal (UCF) at wave length (623.8 nm).Triangles are experimental data [Eq. (29)], and solid line is fitting using the four – parameter model [Eq. (9)].

4 – Temperature effect on birefringence (Δ n)

According the equation (30), and take results the extraordinary and ordinary refractive index values, we can to find the optical anisotropy (birefringence) values on the temperature effect, and these data′ s can be representation in Fig. (6).

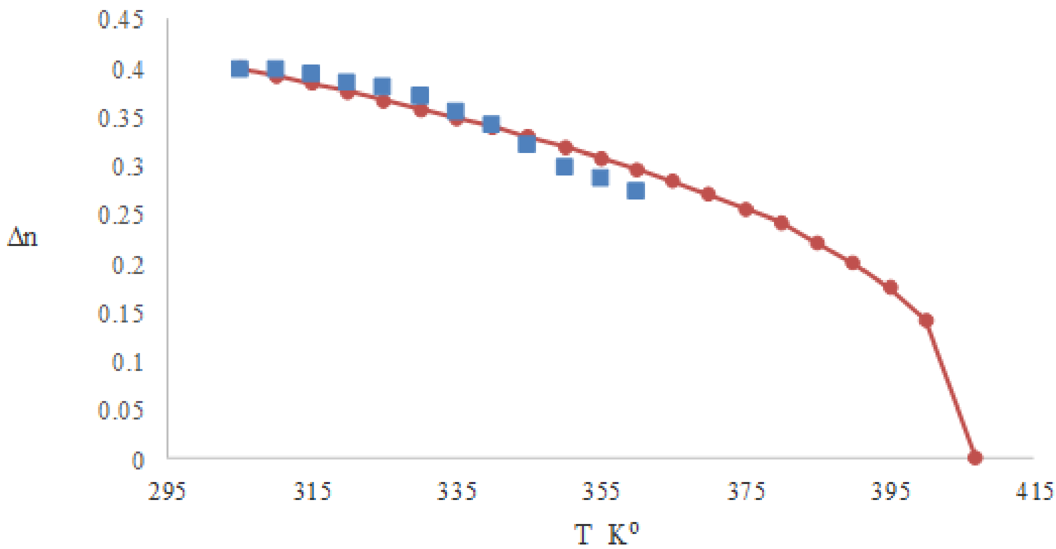

Fig. 6: Temperature – dependent on birefringence (Δ n) for mixture liquid crystal (UCF) at wave length (623.8 nm). Solid squares are experimental data [Eq. (30)],and solid lines are fittings using the four – parameter model [Eqs. (14)].

5 – Temperature effect on order parameter:

By using the equation (23), we can to find the order parameter (S). These data ′ s is representation in Fig.(7).

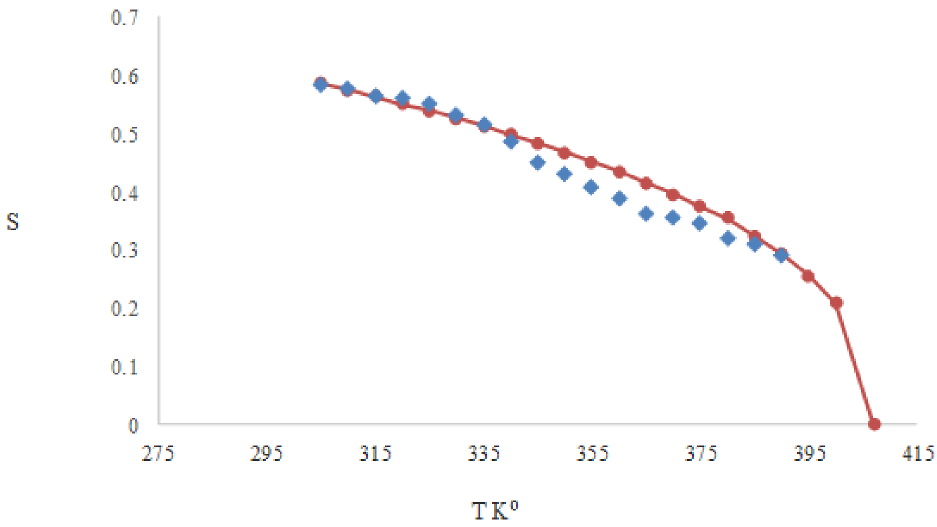

Fig. 7: Relationship between order parameter (S) and temperature (T) for mixture liquid crystal (UCF),at wave length (623.8 nm) .Squares solid is experimental data [Eq. (23)]. Solid line is fittings using the four – parameter model [$S = (1 - \frac{T}{T_C})^{\beta}$].

From above figure we find the order parameter (S) is decrease as the temperature is increase , because of random moving for rod like molecular the liquid crystal, therefore the order parameter is vanish at clearing temperature (T_C).

E – Conclusions:

We using the four – parameter model for describing the temperature effect on refractive indices of mixture liquid crystal (UCF) based on the Vuks equation, at wave length (623.8 nm). Excellent agreement between the experimental data and theory is obtained nearly. We find the ordinary refractive index (n_o) increased when the temperature increased, but the extraordinary refractive index (n_e) and birefringence (Δn) are decreased as the temperature is increased. Also we find the average refractive index < n > decreased linearly when the temperature increased, and we find the order parameter (S) decreased as the temperature is increased.

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