The Effect of Magnetic Field on the Phase Transition of Swollen Liquid Crystal Elastomers

Warsono[1,2], **Yusril Yusuf** [1], **Pekik Nurwantoro**[1] , **Kamsul Abraha**[1]
[1] *Department of Physics, Faculty of Mathematics and Natural Science, Gadjah Mada University, 55281 Yogyakarta, Indonesia*

[2] *Department of Physics Education, Faculty of Mathematics and Natural Science, Yogyakarta State University, 55281 Yogyakarta, Indonesia*

Abstract

The effect of magnetic fields on the swelling of liquid crystal elastomers (LCE) dissolved in liquid crystal (LC) solvent have been studied. The Flory-Huggins model used to calculate the free energy of an isotropic mixing and the Maier-Saupe model used to calculate the free energy of a nematic mixing. Numerical integration method used to calculate the order parameter and the total free energy of system (consists of : nematic free energy, elastic free energy, isotropic mixing free energy and magnetic free energy) and the calculation results graphed as a function of temperatures for various magnetic fields and as function of magnetic fields for various of temperatures. We find that the magnetic field shifts the transition points towards higher temperatures, increases the energy transition, and induces an isotropic phase to paranematic phase.

Key words : M*agnetic field*, swollen liquid crystal elastomers, Flory-Huggins and Maier-Saupe models, free energy, order parameter, numerical integration

1. Introduction

Liquid crystal elastomers (LCE) are hybrid materials composed of liquid crystals (LC) and elastomers. Elastomers are crosslinked polymer chains that have elastic properties and liquid crystals are mesogenic materials that have orientational order properties, so LCE have combination properties of both substances [1, 2, 3, 4, 5]. The coupling between strain and alignment of mesogenic unit, and the responsiveness to the external stimuli such as temperature, electric field, magnetic field, UV light, cause the LCEs materials promise to be suitable for construction of actuators and detectors, and for various application, ranging from micro-pump to artificial muscles [6,7,8]. However, there are some drawbacks of dry LCE in responding to the external fields, for example, it requires a high electric field to induce electromechanical effects. Therefore, one way to increase the responsiveness to the external field is by dissolving LCE in LC solvent. Amazing, the result shows that swollen LCE requires an electric field 200 times lower than the dry LCE to produce electromechanical effects [9]. Accordingly, the swollen LCE could be a good candidate to observe measurable shape changes at low voltages. Based on this, we also sure that the swollen LCE responsiveness to the magnetic fields. By applying a magnetic field, the behavior of swollen LCE will be known, so it can be applied to many applications.

In this paper, we theoretically study the influence of magnetic field on the phase transition of swollen LCE. The phase behavior of the swollen LCE can be seen from the graph of the relationship between free energy and order parameter with the applied magnetic field. We used the Flory Huggins model to calculate the free energy of isotropic mixing and used the Maier-Saupe model to calculate the free energy of nematic mixing. Furthermore, the order parameter is calculated using numerical integration methods. Through analyzing the graph, it can be seen the influence of magnetic field on the phase transitions of swollen LCE.

2. Theoretical Model

2.1. *Free Energy*

In this paper, we assume that swollen LCE is binary mixture systems, with the LCE as solute and LC as solvent. The total free energy of swollen LCE can be expressed as a sum of free energy of isotropic mixing $\boldsymbol{F}_{\text{mix}}$, free energy of nematic ordering $\boldsymbol{F}_{\text{nem}}$, and free energy due to elasticity of nematic network $\boldsymbol{F}_{\mathrm{el}}$, as follows:

$$F = F_{\text{mix}} + F_{\text{nem}} + F_{\text{el}} \tag{1}$$

If the swollen LCE under magnetic field, the total free energy expressed as follow:

$$F_{tot} = F_{\text{mix}} + F_{\text{nem}} + F_{\text{el}} + F_{\text{mag}} \tag{2}$$

The first term $\boldsymbol{F}_{\text{mix}}$ shows the free energy of isotropix mixing. According to the Flory Huggins theory, the free energy is given by [10,11,12,13]:

$$\frac{F_{\textbf{mix}}}{N_t k_B T} = \left[\frac{\varphi_1}{N_1} \ln \varphi_1 + \varphi_2 \ln \varphi_2 + \chi \varphi_1 \varphi_2 \right] \tag{3}$$

$$f_{mix} = \left[\frac{\varphi_1}{N_1} \ln \varphi_1 + \varphi_2 \ln \varphi_2 + \chi \varphi_1 \varphi_2 \right] \tag{4}$$

where φ_1 and $\varphi_2 = 1 - \varphi_1$ are volume fraction of solute molecules (LCE) and solvent molecules (LC), N_1 is number of monomer in polymer chains, χ is Flory-Huggins interaction parameter, N_t is total number of unit cells, k_B is the Boltzman constant, T is absolute temperature and f_{mix} is free energy density of isotropic mixing. The Flory-Huggins interaction parameter χ depends on the temperature through the following relationship:

$$\chi = A_1 + \frac{B_1}{T} \tag{5}$$

where A_1 and B_1 are constants that depend on polymer blend [11].

The second term F_{nem} in Eq.(2) shows the free energy of binary nematic mixtures. Using the Maier-Saupe model, the free energy of nematic mixtures expressed as [14]:

$$\frac{F_{nem}}{N_t kT} = -\Big[\varphi_1 \Sigma_1 + \varphi_2 \Sigma_2 + \tfrac{1}{2} \nu_{11} \varphi_1^2 S_1^2 + \tfrac{1}{2} \nu_{22} \varphi_2^2 S_2^2 + \nu_{12} \varphi_1 \varphi_2 S_1^2 \Big] \tag{6}$$

$$f_{nem} = -\Big[\varphi_1 \Sigma_1 + \varphi_2 \Sigma_2 + \frac{1}{2} \nu_{11} \varphi_1^2 S_1^2 + \frac{1}{2} \nu_{22} \varphi_2^2 S_2^2 + \nu_{12} \varphi_1 \varphi_2 S_1^2 \Big] \tag{7}$$

where Σ_1 and Σ_2 represent the entropy of mesogenic group in solute (LCE) and in solvent (LC), respectively, ν_{11} and ν_{22} are the nematic (Maier-Saupe) interaction parameter of the pure component, ν_{12} represents the cross-interaction parameter between dissimilar mesogens, and f_{nem} is free energy density of nematic mixture.

The nematic interaction parameters, ν_{11}, ν_{22} and ν_{12}, have a relationship with temperature with the following equation [15,16] :

$$\nu_{11} = 4.541 \frac{T_{NI1}}{T} \tag{8}$$

$$\nu_{22} = 4.541 \frac{T_{NI2}}{T} \tag{9}$$

$$\nu_{12} = c \sqrt{\nu_{11} \nu_{22}} \tag{10}$$

where T_{NI1} and T_{NI2} are the temperature of *nematic-isotropic* phase transition of the component1 and the component 2, c is proportionality constant. The entropy of component 1 and 2 can be deduced as [15, 17]:

$$\Sigma_1 = \ln Z_1 - m_1 S_1$$

$$\Sigma_2 = \ln Z_2 - m_2 S_2$$

where Z_1 and Z_2 are the partition function of the component 1 and the component 2, m_1 and m_2 are dimensionless mean field parameter characterizing the strength of potential fields, and S_1 and S_2 are the orientational order parameter of the component 1 and the component 2, respectively.

The elastic free energy F_{el} of swollen LCE can be derived from classical rubber elasticity theory as follows [10, 11, 18, 19]:

$$F_{el} = \tfrac{1}{2} nkT \left[\lambda_{xx}^2 + \lambda_{yy}^2 + \lambda_{zz}^2 - 3 \right]$$

where *n* is number of strand between crosslink, λ_{xx}, λ_{yy}, and λ_{zz} are deformation factor in the *x*, *y*, and *z* directions. If the deformation factors associated with the order parameter of the components of swollen LCE, S_1 and S_2, and the volume fraction of LCE, φ, then the equation of the elastic free energy can be expressed in Eq.(11).

$$\frac{F_{el}}{N_t kT} = \frac{3}{2} \left(c \left(\frac{\varphi_1}{N_1{}^4 A} \right)^{\frac{1}{3}} - \frac{\varphi_1}{N_1} \right) \tag{11}$$

$$f_{el} = \frac{3}{2} \left(c_t \left(\frac{\varphi_1}{N_1{}^4 A} \right)^{\frac{1}{3}} - \frac{\varphi_1}{N_1} \right) \tag{12}$$

where $A = (1 + 2S_1)(1 - S_1)^2$ and $c_t = \left(3 \big/ 2^{\frac{2}{3}} \right)$.

The final term F_{mag} in Eq.(2) shows the free energy due to the magnetic field. When the magnetic field **H** is applied to the mesogen having positive diamagnetic $\Delta\chi = \chi_{||} - \chi_{\perp}$ with the director **n,** the magnetic free energy is given by [20, 21, 22]:

$$F_m = -\tfrac{1}{2} \Delta\chi \int \left(\hat{\mathrm{n}} \bullet \vec{\mathrm{H}} \right)^2 f(\theta) d\Omega$$

$$F_m = -\tfrac{1}{2} \Delta\chi H^2 \left(\int (cos\,\theta)^2 f(\theta) d\Omega \right)$$

$$F_m = -\tfrac{1}{2} k_B T \left(\frac{\Delta\chi H^2}{k_B T} \right) \left\langle (cos\,\theta)^2 \right\rangle$$

$$F_m = -\tfrac{1}{2} k_B T h \left[\tfrac{1}{3} (1 + 2S) \right] \tag{12}$$

where $\left(\hat{n}\bullet\vec{H}\right)=H\cos\theta$, $h=\left(\Delta\chi H^{2}/k_{B}T\right)$ is the dimensionless parameter of the magnetic field, and $\left\langle\left(\cos\theta\right)^{2}\right\rangle=\frac{1}{3}\left(1+2S\right)$. If this is applied to the swollen LCE, which has two kinds of mesogen with different volume fraction, then the total energy is the sum of each component as follows :

$$F_{mag}=F_{m1}+F_{m2}$$

$$\frac{F_{mag}}{N_{t}k_{B}T}=-\frac{1}{2}h\left[\frac{1}{3}\varphi_{1}\left(1+2S_{1}\right)+\frac{1}{3}\varphi_{2}\left(1+2S_{2}\right)\right] \quad (13)$$

$$f_{mag}=-\frac{1}{2}h\left[\frac{1}{3}\varphi_{1}\left(1+2S_{1}\right)+\frac{1}{3}\varphi_{2}\left(1+2S_{2}\right)\right] \quad (14)$$

2.2. *Order Parameter*

Orientational order is the most important feature of liquid crystals. The average directions of the long axes of the rod-like molecules are parallel to each other. Because of the orientational order, liquid crystals possess anisotropic physical properties; that is, in different directions, they have different responses to external fields such as an electric field, a magnetic field, and shear [23, 24]. In mixtures of liquid crystals, the molecules of different components may possess different degrees of nematic ordering. In the mixture, the order parameter expressed by [25, 26]:

$$S_{i}=\left\langle P_{2}\left(\cos\theta_{i}\right)\right\rangle \quad (15)$$

$$S_{i}=\int_{0}^{\pi}P_{2}\left(\cos\theta_{i}\right)f\left(\theta_{i}\right)d\Omega_{i} \quad (16)$$

where $P_{2}(x)=\frac{1}{2}(3x-1)$ is the second Legendre polynomial, θ_{i} is the angle between a reference axis and the director of a mesogen belonging to component *i* (*i*=1 for solute and i=2 for solution), and $d\Omega_{i}=2\pi\sin\theta d\theta$. The function $f(\theta_{i})$ is the normalized orientation distribution function which may be expressed by:

$$f(\theta_{i})=\frac{1}{Z_{i}}\exp\left(-\frac{V(\theta_{i})}{k_{B}T}\right) \quad (17)$$

where Z_{i} is partition function defined as:

$$Z_{i}=\int_{0}^{\pi}\exp\left(-\frac{V(\theta_{i})}{k_{B}T}\right)d\Omega_{i} \quad (18)$$

and $V(\theta_{i})$ is the potential field describes intermolecular interaction. In the Maier-Saupe model, the potential $V(\theta_{i})$ expressed by:

$$V(\theta_{i})=-\alpha S_{i}\left(\frac{3}{2}\cos^{2}\theta_{i}-1\right) \quad (19)$$

$$V(\theta_{i})=-\left(m_{i}k_{B}T\right)\left(\frac{3}{2}\cos^{2}\theta_{i}-1\right) \quad (20)$$

$$m_{i}=\left(\frac{\alpha S_{i}}{k_{B}T}\right) \quad (21)$$

where α is orientational interaction constant and m_{i} is dimensionless mean field parameter. Substitution Eqs.(17), (18), (19), and (21) into Eq.(16), we obtain :

$$S_{i}=\frac{\int_{0}^{\pi}P_{2}\left(\cos\theta_{i}\right)\exp\left(\frac{\alpha S_{i}P_{2}\left(\cos\theta_{i}\right)}{kT}\right)\sin\theta\,\mathrm{d}\theta}{\int_{0}^{\pi}\exp\left(\frac{\alpha S_{i}P_{2}\left(\cos\theta_{i}\right)}{kT}\right)\sin\theta\,\mathrm{d}\theta} \quad (22)$$

$$S_{i}=\frac{\int_{0}^{\pi}P_{2}\left(\cos\theta_{i}\right)\exp\left(m_{i}P_{2}\left(\cos\theta_{i}\right)\right)\sin\theta\,\mathrm{d}\theta}{\int_{0}^{\pi}\exp\left(m_{i}P_{2}\left(\cos\theta_{i}\right)\right)\sin\theta\,\mathrm{d}\theta} \quad (23)$$

The order parameter S_{i} can be obtained by numerically solving of Eq.(23). The average value of order parameter S in binary mixture is given by [27]:

$$S=\varphi_{1}S_{1}+\varphi_{2}S_{2} \quad (24)$$

where S_{1} is order parameter of mesogen in solute and S_{2} is order parameter of mesogen in solvent.

2.3. *Phase Transition under Magnetic Field*

One way to determine the phase transition in the swollen LCE is to calculate the value of the order parameter. The order parameter S_{1}, S_{2}, and S as function of temperature calculated numerically by Eqs.(23) and (24). Under magnetic field, the dimensionless mean field parameter m_{1} and m_{2} obtained by minimizing the total free energy with respect to the order parameter S_{1} and S_{2} as follow:

$$\left(\frac{\partial F_{tot}}{\partial S_1}\right) = 0 \qquad (25)$$

$$\frac{\partial\left(F_{mix} + F_{nem} + F_{el} + F_{mag}\right)}{\partial S_1} = 0$$

$$-N_t kT\varphi_1\left([\nu_{11}\varphi_1 S_1 + \nu_{12}\varphi_2 S_2 - m_1] - 3c\frac{S_1(1 - S_1)}{\varphi_1 A}\left(\frac{\varphi_1}{N_1{}^4 A}\right)^{\frac{1}{3}} + h\right) = 0$$

$$m_1 = \nu_{11}\varphi_1 S_1 + \nu_{12}\varphi_2 S_2 - 3c_t\frac{S_1(1 - S_1)}{\varphi_1 A}\left(\frac{\varphi_1}{N_1{}^4 A}\right)^{\frac{1}{3}} + h \qquad (26)$$

$$\left(\frac{\partial F_{tot}}{\partial S_2}\right) = 0 \qquad (27)$$

$$\frac{\partial\left(F_{mix} + F_{nem} + F_{el} + F_{mag}\right)}{\partial S_2} = 0$$

$$N_t kT\varphi_2\left[\nu_{22}\varphi_2 S_2 + \nu_{12}\varphi_1 S_1 - m_2 + h\right] = 0$$

$$m_2 = \nu_{22}\varphi_2 S_2 + \nu_{12}\varphi_1 S_1 + h \qquad (28)$$

Substitution of Eqs.(8) and (9) into Eq.(26) and Eqs.(9) and (10) into Eq.(27), we get m_1 and m_2 as function of temperature *T* and magnetic field *h* as show in Eqs.(29) and (30).

$$m_1 = 4.541\left(\frac{T_{NI1}}{T}\varphi_1 S_1 + \frac{c\sqrt{T_{NI1}T_{NI2}}}{T}\varphi_2 S_2\right) - 3c_t\frac{S_1(1 - S_1)}{\varphi_1 A}\left(\frac{\varphi_1}{N^4 A}\right)^{\frac{1}{3}} + h \qquad (29)$$

$$m_2 = 4.541\left[\frac{T_{NI2}}{T}\varphi_2 S_2 + \frac{c\sqrt{T_{NI1}T_{NI2}}}{T}\varphi_1 S\right] + h \qquad (30)$$

Substitution of Eqs.(29) and (30) into Eq.(23), and then the results substitute to the Eq. (24) and to the free energy equations, we get the relationship between S, F_{mix}, F_{nem}, F_{el}, F_{mag}, and F_{tot} with temperature *T* under magnetic field *h*.

3. Result and Discussion

In this section, we calculate the average order parameter S, the free energy density of isotropic mixing f_{mix}, the nematic free energy density f_{nem}, the elastic free energy density f_{el}, the magnetic free energy density f_{mag}, and the total free energy density f_{tot} of the swollen LCE as a function of temperature *T* and magnetic field *h*. In these calculations, all the quantities are set as follows:

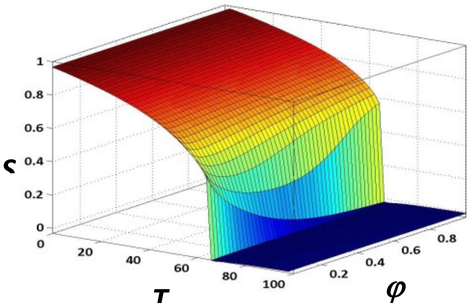

Fig. 1. Order parameter *S* as function of

$N_1 = 200$, $\varphi_1 = \varphi = 0.5$, $T_{NI1} = 70^o C$, $T_{NI2} = 60^o C$, $c = 1$, $A_1 = 0.103$ and $B_1 = -43$.

Figure 1 shows the order parameter S as a function of temperature T and volume fraction φ without applying of magnetic field h. The phase diagram of Nematic-Isotropic transition is *first order transition*.

The effect of magnetic field on order parameter is shown in Figure 2. By applying a magnetic field, the order inside the system is considerably changed. At the same temperature, the increase in the magnetic field causes the increase of the order parameter, which means that the regularity of the system increases [28]. If the magnetic field is increased, the phase transition occurs at a higher temperature and the transition points shift towards higher temperatures. Applying magnetic field in the isotropic region (*S*=0) cause the order parameter increase up to the zero values (*S*>0), see Fig.2(b). It means that the magnetic field induce isotropic phase to paranematic phase [21,29].

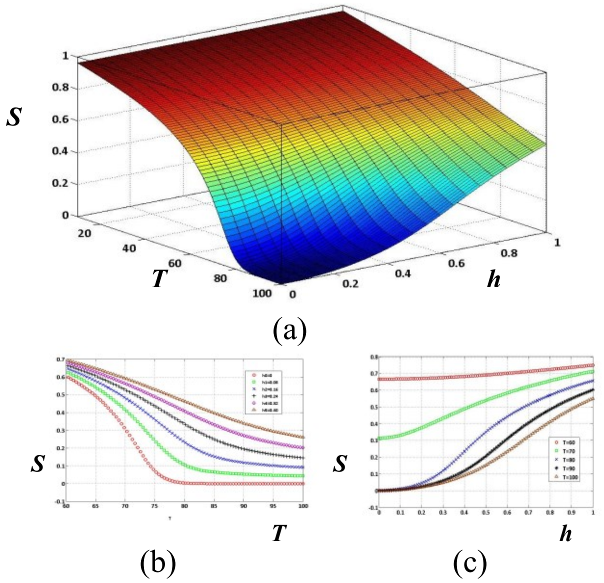

Fig. 2. The relationship between *S*, *T* and *h* (a).*S* as function of *T* and *h*, (b).*S* as function of *T* for various value of *h*, and (c).S as function of *h* for various value of *T*

At low temperatures, the increase in the magnetic field does not significantly affect to the increase of the order parameter because the mesogenic molecules already in the high regularity. In contrast to the high temperature (greater than the transition temperature), changes in the magnetic field caused the order parameter increase drastically as shown in Fig.2(c).

Figure 3, Figure 4, Figure 5, Figure 6, and Figure 7 show the effect of magnetic field on the nematic free energy density f_{nem}, the free energy density of isotropic mixing f_{mix}, the elastic free energy density f_{el}, the magnetic free energy density f_{mag}, and the total free energy density f_{tot} of the swollen LCE.

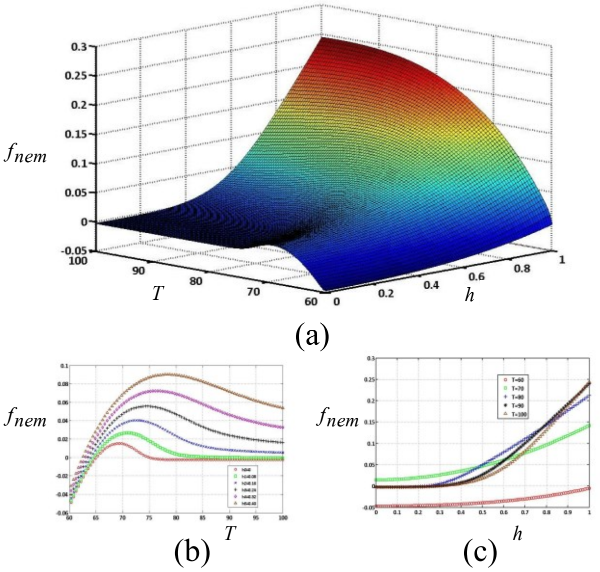

Fig. 3. The relationship between f_{nem}, *T* and *h* (a). f_{nem} as function of *T* and *h*, (b). f_{nem} as function of *T* for various value of *h*, and (c). f_{nem} as function of *h* for various value of *T*

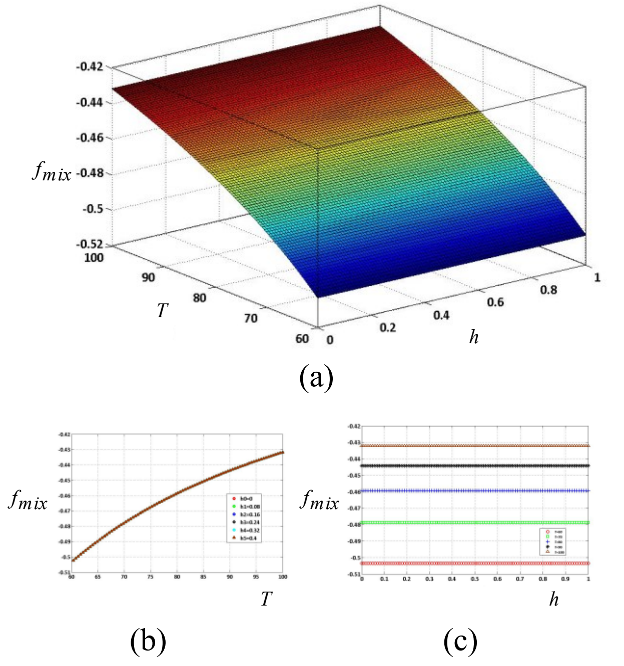

Fig.4. The relationship between f_{mix}, *T* and *h* (a). f_{mix} as function of *T* and *h*, (b). f_{mix} as function of *T* for various value of *h*, and (c). f_{mix} as function of *h* for various value of *T*

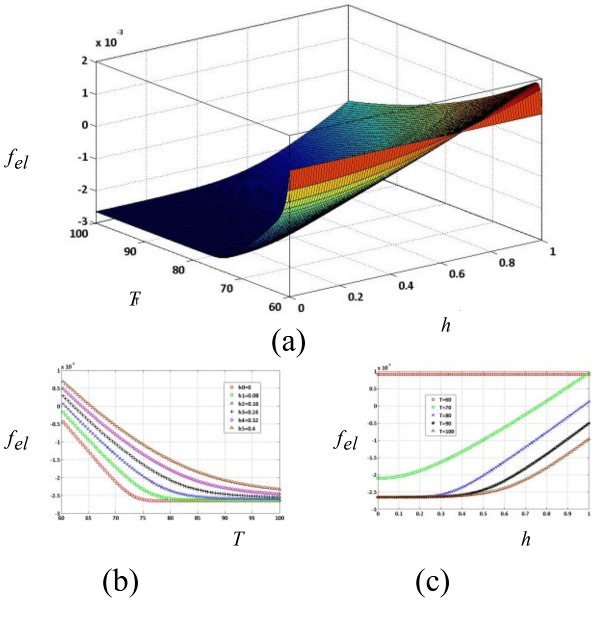

Fig.5. The relationship between f_{el}, *T* and *h* (a). f_{el} as function of *T* and *h*, (b). f_{el} as function of *T* for various value of *h*, and (c). f_{el} as function of *h* for various value of *T*

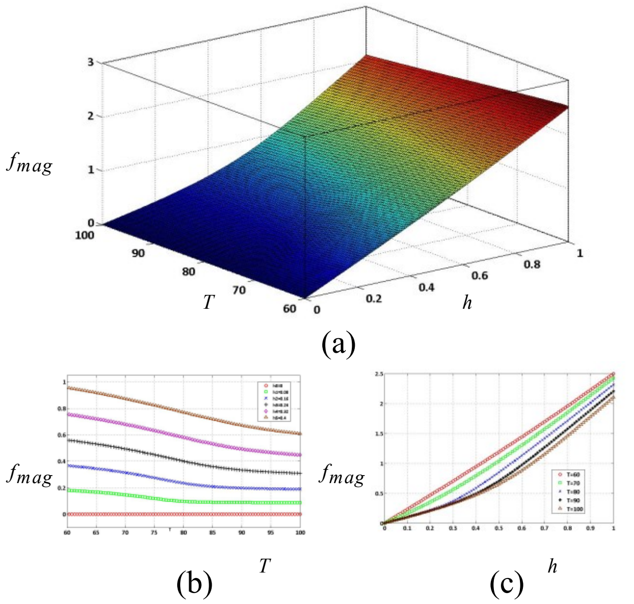

Fig.6. The relationship between f_{mag}, T and h (a). f_{mag} as function of T and h, (b). f_{mag} as function of T for various value of h, and (c). f_{mag} as function of h for various value of T

Based on these graphs, the magnetic field effect on the nematic free energy density f_{nem}, the elastic free energy density f_{el}, and the magnetic free energy density f_{mag}, but has no effect to the free energy density of isotropic mixing f_{mix}. It can be explained that in addition to the free energy density of the isotropic mixture, each of the free energy contains the order parameters as presented in the Eq.(7), Eq.(12), and Eq.(14).

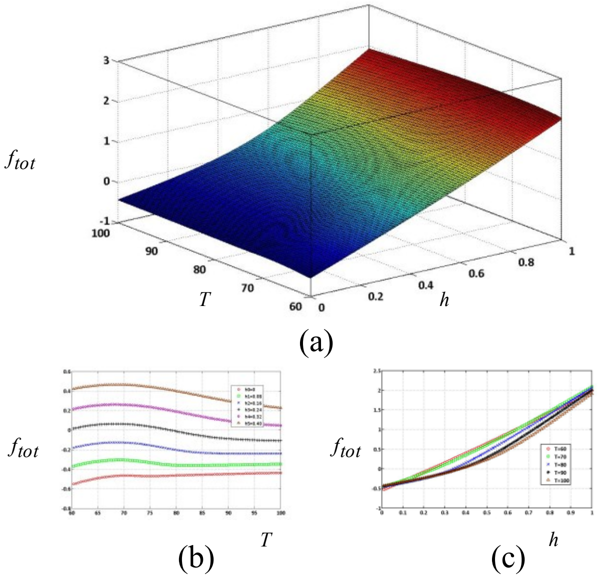

Fig.7. The relationship between f_{tot}, T and h (a). f_{tot} as function of T and h, (b). f_{tot} as function of T for various value of h, and (c). f_{tot} as function of h for various value of T

In general, if the magnetic field is increased, the free energy of the system will increase include the free energy of the phase transition.

4. CONCLUSION

We have studied the effect of magnetic field on the phase transition of the swollen liquid crystal elastomers. Combining the Flory-Huggins model for isotropic mixing and the Maier-Saupe model for nematic mixing of two kinds of mesogens, we used numerical integration method to calculate the order parameter and the free energy of the systems as function of temperature and magnetic field. We find that the magnetic field shifts the transition points towards higher temperatures, induces an isotropic phase to paranematic phase, and increases the free energy transition.

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