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Effect of Graphene Oxide on The Photoluminescence of Antiferroelectric Liquid Crystal Mixtures

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Abstract

UV absorbance study has been performed in spectrum mode, time scan mode and overlay mode. The decrement in UV absorbance has been validated from all these examined modes. The red shift in absorbance bands for pristine antiferroelectric liquidcrystal (AFLC) and graphene oxide (GO) AFLC dispersed system have been observed. The UV absorbance maxima for AFLC dispersed with GO has been shifted from 317.27 to 324.57 nm. This shift is associated with the existence of different GO concentrations in AFLC medium.TheimprovementinluminescenceintensityofAFLC dispersed GO nanoparticles mixtures has been revealed. Theluminescenceintensityof AFLC mixture enhanceswithincreasing concentration of GO for fixedweightof AFLC sample. The variation in the luminescence intensity of AFLC and its dispersed system with GO at different temperatures or phases is examined. Theused Graphene oxide has beensynthesized by using Hummers method.

Keywords: Antiferroelectric liquid crystals, Graphene oxide nanoparticles, UV absorbance, Photoluminescence.

1. Introduction

Recently ample researches have been done with other forms of liquid crystals (LCs) such as ferroelectric liquid crystals (FLC). In FLCs the molecules are arranged in layers and they are tilted with respect to the layer normal. The direction of tilt is opposite in adjacent layers in case of antiferroelectric liquid crystals (AFLCs). The ferroelectric or antiferroelectric phases exhibit polar ordering if the rod-like molecules are chiral. A fluid with overall polar order is considered

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to be highly appreciable for display applications owing to the fast response of the molecules. Ferroelectric liquid crystals (smectic C*) composed of rod-shaped molecules are the first example of such fluid-like phase with polar ordering [1]. Recently, in many articles nano-dispersion studies have been discussed in which FLC has been used as the host medium [2-8].Srivastava et al. presented a switchable grating based on chiral single walled carbon nanotube (SWCNT) doped FLC where the diffraction efficiency of the highly doped system was significantly increased [3]. Photoluminescence (PL) intensity and dielectric properties of FLC are modified by functionalized multiwalled carbon nanotube (MWCNTs) [9]. The alignment of SWCNTs in a FLC medium is investigated through scanning electron microscope (SEM) and their interactions are studied through Raman spectroscopy, FTIR etc. the studies revealed that SWCNTs can be well aligned by smecticLC [10]. S. Ghosh et al. had investigatedCNT-AFLC composite system that yields high luminescence characteristic properties which aim at display purpose [11]. It was found that minute addition of dopant can reduce the switching response between bright/dark conditions by decreasing the rotational viscosity (γ_{rot}) of an AFLC system. Tilt angle was essentially unaltered which was necessary to achieve perfect dark state of an AFLC device. The spontaneous polarization was increased owing to the dominant interaction of the aromatic cores of LC molecules with the honey-comb like pattern of the CNT walls. The SmC_A ordering was improved around the CNTs. S. Roy etal. had found enhancement in photoluminescence of AFLC- CdS nanorods doped system [12]. They had examined the enhancement in PL intensity withthedifferent weight percent concentrationofCdS nanorods into fixedweightofAFLC.

Antiferroelectric liquid crystal (AFLC) (SmC_A*) which is a sub phase of SmC* has even faster response time, easy DC compensation,wide viewing angle and full grey scale compatibility. However, till now their commercial applications have not been successful because of poor alignment which is responsible for poor contrast ratio (CR) and light leakage. The solution to the dark state problem has been resolved by introducing orthoconicAFLC materials (OAFLC) with tilt angle ~45º[13- 16]. Till then several research groups have investigated such OAFLC materials in details [15-16]. However, these OAFLC materials often possess extremely small helical pitch and high rotational viscosity (γ_{rot})etc, which in turn causes various technical issues like elevated driver expenses and delay in switching/response times etc. Most convenient non-synthetic way to address this problem is dispersion of nanomaterials in AFLC host.

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The motivation of this present study is to modify photoluminescence property of an orthoconic AFLC W1000 mixture (fixed amount) by dispersing different concentration of graphene oxide GO in it, thus facilitating its application in displays. Previously many studies have been done by dispersing CNTs in nematic and ferroelectric LC. Here for the first time GO are dispersed in an antiferroelectric LC to improve its photoluminescence properties significantly. Also, LC induced orientation of GO has been achieved as observed from optical micrographs. The study helps to understand the interaction of GO with surrounding anisotropic medium possessing antiferroelectric ordering.

2. Experimental details

2.1 The chemical structure and phase transition of AFLC material:

Antiferroelectric liquid crystal, W1000 mixture has been used in the present work. It is a mixture of two compounds CM1 (52.5% by weight) and CM2 (47.5% by weight) with chemical structure shown below;

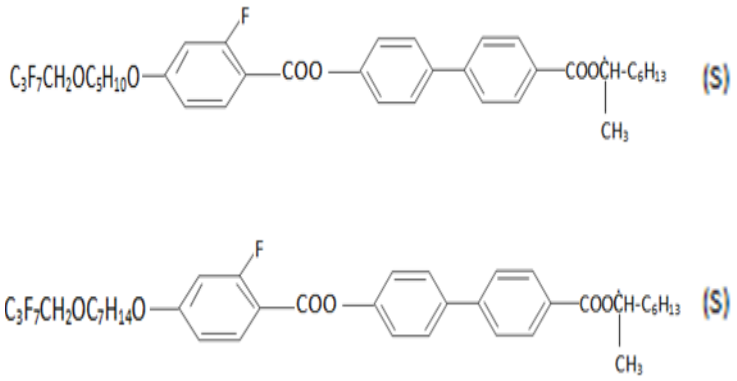

Fig.1. The Chemical Structure of AFLC W1000 mixture

AFLC W1000 mixture has been composed of equimolar binary mixture with two fluorinated isomers (S) forms constitute this mixture.AFLC W1000 mixture has been synthesized by our collaborating group (Department of chemistry, Military University of Technology, Warsaw, Poland) [12, 16]. It exhibits the following phase sequence:SmC_A* ↔ 101.7ºC ↔SmC* ↔ 103.4ºC ↔SmA ↔ 105.7ºC ↔ Iso.AFLC W1000 mixture is highly tilted, long pitch, near Orthoconic mixture [16]. The specifications and descriptionfor this AFLC mixture had already been published by our collaborating research group [12].Graphene oxide was synthesized by

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Hummer's method [17]. The detailed structural characterization of graphene oxide had already been reported in our previous work [18, 19].

2.2 Preparation of AFLC-Graphene oxide composite and sample cell filling

A suspension of graphene oxide was prepared in dimethylformamide (1 mg/ml DMF, Merck chemicals) and sonicated for 3 hours to eliminate agglomerates of graphene oxide [19]. The concentration of graphene oxide was optimized and then dispersed into 1 mg of AFLC to prepare AFLC- graphene oxide composite by taking 0.1% wt/wt (MIX 1) and 0.3 % wt/wt of graphene oxide (MIX 2) in AFLC. The prepared composites were uniformly dispersed by magnetic rotator for 1 hour at isotropic (iso) temperature (120ºC) along with sonication for another 1hour [19]. The solvent was evaporated from the mixtures with slow evaporation rate at an elevated temperature.

At first Graphene oxide has been dispersed into fixed weight of W1000 mixture. Highly conducting, optically transparent ITO substrates were used as electrodes. Photolithography technique was used to mask square pattern (5.0×5.0 mm^2) [19]. The surface treated ITO glass plates were assembled and then treated with silane layer (phenyl trichlorosilane) and washed with propanol. In order to achieve planar aligned cells, the substrates were further treated with nylon 6/6 and rubbed gently in antiparallel manner. The glass substrates are now packed with electrodes facing each other to form capacitor type cell. These cells were calibrated, and their thickness has been evaluated to be around 6µm which was fixed by the mylar spacer.The LC sample cells were filled with pure AFLC and AFLC- graphene oxide composite by capillary action.

3. Measurement techniques

UV visible absorption study of pure AFLC and GO dispersed system have been carried out using UV- VISElico double beam spectrophotometer (SL 210) for a wavelength range of 250-600 nm in spectrum mode, overlay mode and time scan mode [19]. The Photoluminescence spectra of pure and GO dispersed system have been carried out using Cary eclipse fluorescence spectrophotometer (Agilent technologies) in scan mode using 5 nm slit width, 350-550 nm wavelength at different temperatures.

4. Result and Discussion

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Figure 2 shows the variation of UV absorbance with wavelength in spectrum mode for AFLC-GO dispersed system. The reduction in the absorbance has been observed on dispersing GO in AFLC mixture. The absorbance peak for pure AFLC is obtained at 317.2 nm, for MIX 1 at 324.57 nm and for MIX 2 at 324.57 nm respectively which corresponds to π-π^* transition. This shows the red shift in absorbance wavelength, whereas pure GO absorbance had already been reported in our previous work [**19**] which depicts the absorbance peak at 240.44 nm. The variation in UV absorbance of neat W1000 AFLC and AFLC- GO dispersed system is associated with the change in chemical composition and the electronic transitions that causes change in oxygen containing functional groups and C-C conjugated bonds. Since the amount of AFLC is fixed and there is change in the concentration of dopant, therefore by fixing the amount of AFLC we have fixed the scattering area (since at room temperature (RT) SmC^* phase is highly light scattering phase). Therefore, it can be concluded that whatever change in absorbance is taking place, is due to the variation in the dopant concentration. The red shifted absorbance band has been ascribed as the variation in effective refractive index (n_{eff}) of GO because of presence of AFLC mixture. The decrease in UV absorbance is effectively due to the electronic coupling between the homo of the donor and the LUMO of the acceptor molecules. There is a formation of charge transfer complex in the doped system that changes the net charge on GO, resulting in the permanent dipole moment in them. Related to the GO flakes distribution in AFLC mixture, the corresponding shift in wavelength is observed.

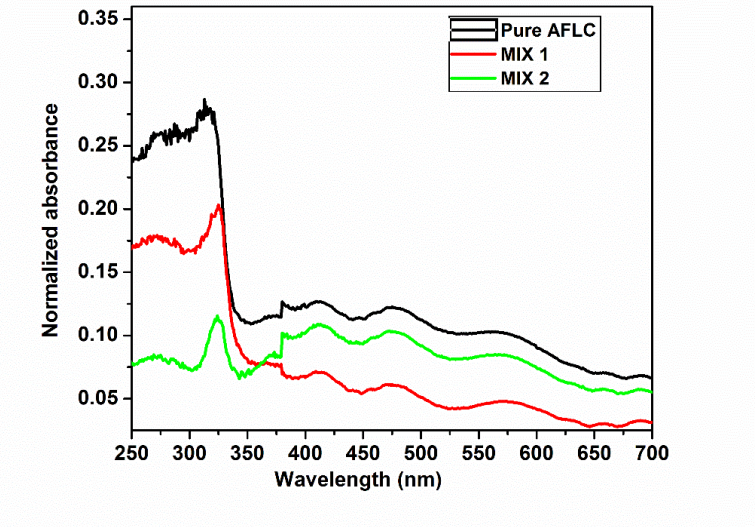

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Fig.2. UV Absorbance spectra for Pure AFLC and its mixture with graphene oxide (GO) in spectrum mode.

UV absorbance study in time scan mode has been shown in figure 3. Inset of the plot shows UV absorbance peak for pure AFLC mixture. UV absorbance in GO dispersed system has been reduced as compared to pure AFLC mixture. It has been observed that for all samples, the absorbance increases and reaches to maximum value for a particular instant of time and then it decreases periodically. The decrease in UV absorbance is also evident from the **figure1** in spectrum mode.

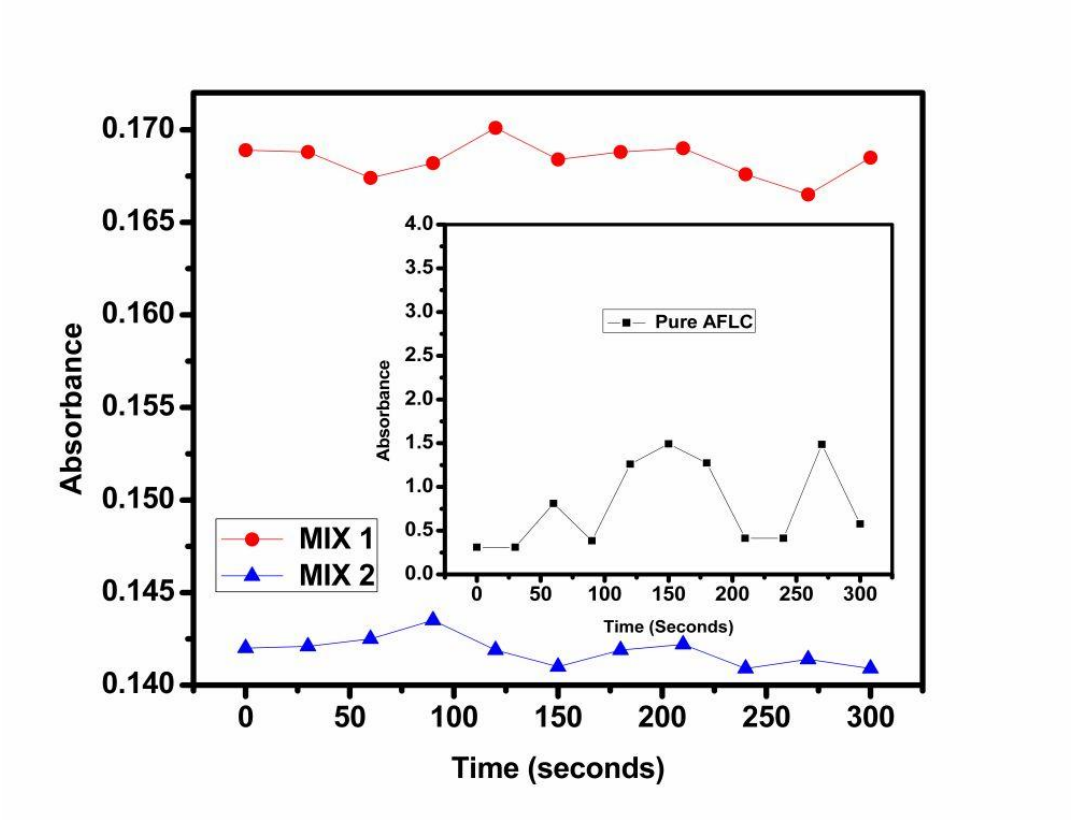

Fig.3. UV Absorbance spectra for Pure AFLC and its mixture with graphene oxide (GO) in Time Scan mode.

To further validate our observation, UV absorbance in overlay mode has been examined as shown in figure 4, which also results the decrement in absorbance with red shift in wavelength by increasing the concentration of GO in AFLC mixture. Here, the absorbance peak has been obtained at 321.43 nm for pure AFLC, 331.61 nm for mixture 1 and 334.24 nm for mixture 2 respectively.

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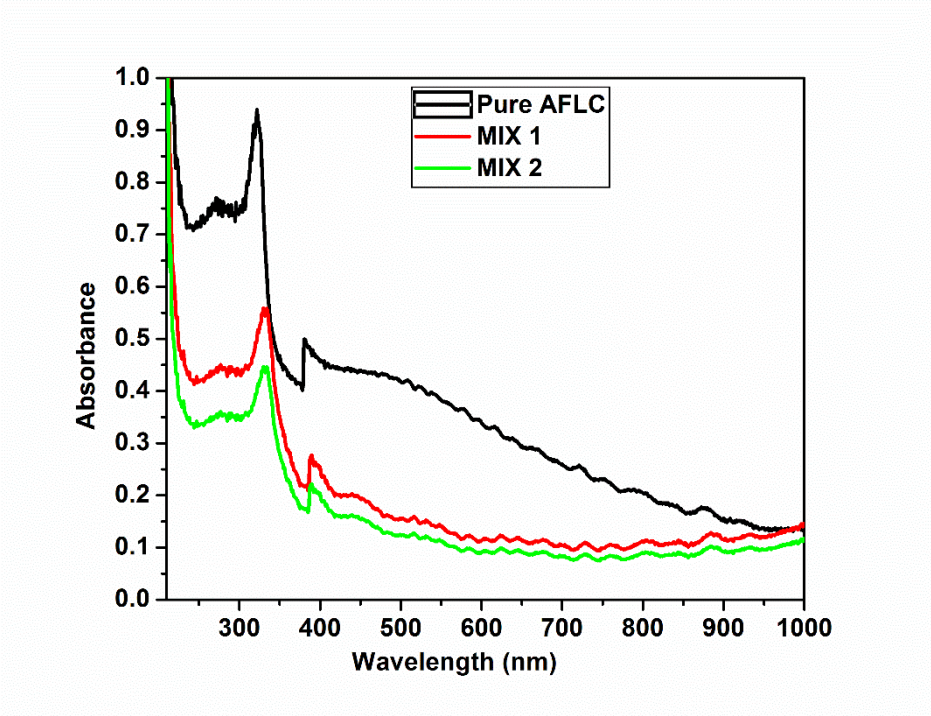

Fig.4. UV Absorbance spectra for Pure AFLC and its mixture with graphene oxide (GO) in Overlay mode.

Figure 5 shows the photoluminescence (PL) intensity variation with wavelength for AFLC W1000 mixture and AFLC- GO dispersed system at room temperature. For pure AFLC mixture, PL emission maxima has been obtained at 421.82 nm, for mixture 1 at 422.20 nm and for mixture 2 at 421.25 nm respectively with their corresponding excitation wavelength as 317.27 nm, 324.57 nm and 324.57 nm. The PL intensity has been suppressed at these PL emission wavelengths. The small peaks at 490 nm and 510 nm has also been observed with enhanced intensity for dispersed system as compared to pure AFLC. The possible reason for this trend might be the fact that the scattering of light decreases with wavelength. As the concentration of GO changes, the light scattering centers are also affected which in turn affect the luminescence yield. The quenching in PL intensity at maximum emission wavelength with the concentration of GO has been associated with the crystal lattice defects or change in the surface topology of the AFLC- GO dispersed system and the non-radiative energy transfer between the mesogenic molecules and GO dopant. Another possible reason might be the change in PL intensity due to change in pH level of the dispersed system. Basic character causes quenching in PL intensity as compared to acidic ones. Inset of **figure 4** shows the GO luminescence peak at 422 nm for

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excitation wavelength 310.23 nm as previously reported in our work [19], The hydrophilic nature of graphene oxide and the occurrence of various other groups such as carboxylic(R-COOH) groups, ketones, lactols, alcohols (R-OH) and epoxides (R-O-R) has influential role on the Vander wall interactions between graphene layers [18]. GO synthesized by hummer's method has oxygen containing functional groups like (-C=O) carbonyl and (-COOH) carboxyl group at sheet edges whereas (C-O-C) epoxy and (-OH) hydroxyl on the basal plane. Ion capturing process in graphene is described as: Hexagonal pattern of carbon atom layers organized in such a way that it resembles membrane like shape containing small holes with oxygen atoms. These circular holes are termed as "crown ethers" which are neutral molecules that traps metallic ions. Dispersion of graphene in polar solvent (containing +ve and –ve ions) is accompanied by the trapping of +ve ions in the "crown ether" pores. The capturing and releasing phenomenon of charge carriers can be triggered by changing the ionic current through bias voltage. These "crown ether" pores stretch and dilate by the amount of force that enhances the +ve ion flow via membrane. The maximum effect is observed on stretching the membrane from all directions while partial effect can be seen by tugging in single direction. There are several factors that affect the ionic flow such as sheet thinness, interaction between ion- pore, ions- surrounding fluid. The delicate balance between ions- surrounding is disturbed on slightly stretching the pores that nullify ion-pore interaction.

The breaking of C- bonds and cleavage of π- network is caused by the oxidation of graphene. The luminescence of GO arises from the radiative recombination of electron- hole pairs confined within sp^2 hybridized carbon clusters which act as luminescent centers. The suppression in luminescence of AFLC-GO dispersed system has been associated with the fact that oxygen containing functional groups induces structural variations. The luminescence mechanism involves not only radiative recombination of electron - hole pairs localized in aromatic domains of different sizes but also the non-radiative recombination process of electron-hole at the cost of radiative recombination, along with surface state and band edge emissions. The introduction of numerous sp^2 hybridized carbon clusters causes increased non-radiative recombination process. $\pi - \pi^*$ states have been confined at -C=O and -C-O-C- groups. The energy variation between these band states givesluminescence intensity. The tuning in luminescence is associated with the change in -C=O and -C-O-C- ratio.

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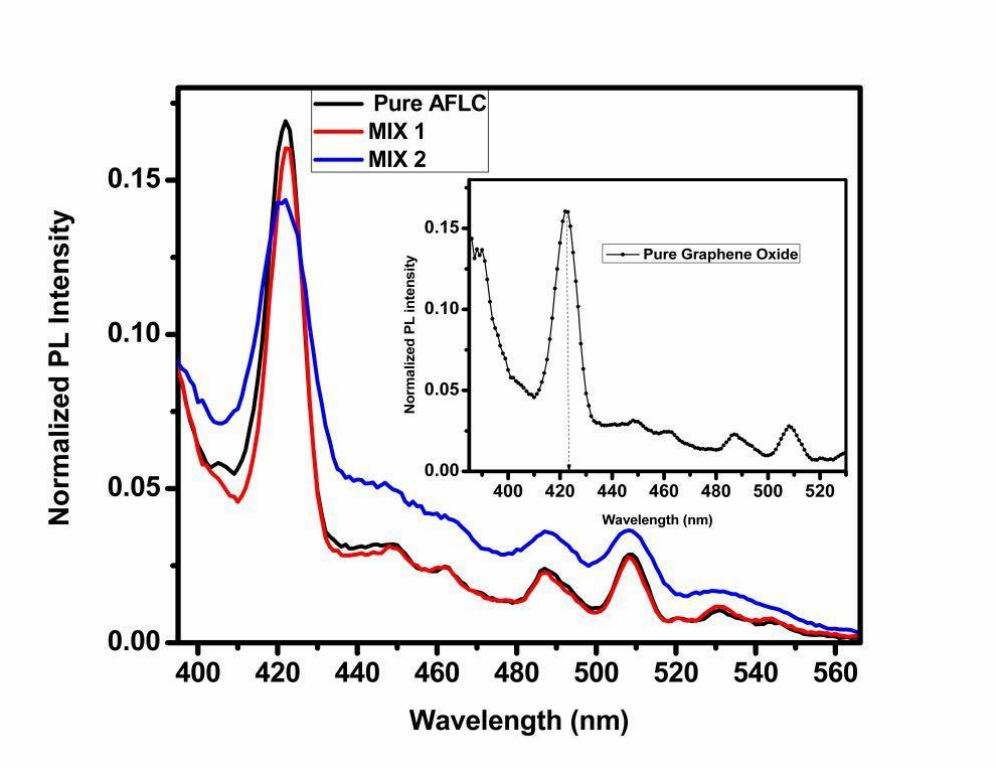

Fig.5. Photoluminescence Spectra for pure AFLC and its mixture with graphene oxide (GO) at room temperature.

Figure 6 shows the graph of PL intensity for pure AFLC on cooling cycle from 110ºC to 40ºC temperature. It shows the variation in PL intensity for isotropic to SmA phase, SmA to SmC^* phase and SmC^* to SmC_A^* phase. The observation reveals the suppression in PL intensity in isotropic phase because of the dilution effect in the liquid crystal medium [**23**]. On cooling to SmA phase, the enhancement in the PL intensity has been observed because of the improvement in the orientation order of liquid crystal molecules on cooling from isotropic to Sm A phase. As in SmA phase, liquid crystal molecules possess layer organized inside the layers and the molecules are parallel (∥) with their long axes perpendicular (⊥) to the phase. The optical uniaxiality in Sm A phase results homeotropic texture which gives the dark state between crossed polarizers. Similarly, on further cooling from smectic A to smectic C^* phase there is no variation in PL intensity has been observed. The layered organization in smectic C phase is formed from the "Lateral stacking interaction" between adjacent molecules. Such "lateral stacking interaction" is produced from the existence of extended aromatic cores which are characterized by a large phenyl system. These layers preserving the thickness are disturbed as

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they slide over one another in order to get adjusted to surface environment. Thus, the layer distortion is responsible for the appearance of optical properties in smectic phases. In SmC^* phase, molecules possess ferroelectricity, the value of spontaneous polarization is dependent relative to the temperature and it increases with the tilt angle on cooling from SmA to SmC^* phase.

On cooling further fromSmC^* to SmC_A^* phase, maximum PL intensity has been observed at 100ºC then it suppresses as 100.7ºC, 90ºC, 101ºC, 95ºC. This observe trend has been associated to the fact that the phase transition temperature of SmC^* to SmC_A^* phase is 101.7ºC therefore at 100ºC, it completely transforms to SmC_A^* phase and SmC_A^* phase exhibits the opposite tilt direction in alternate layers as compared to SmC^* phase.

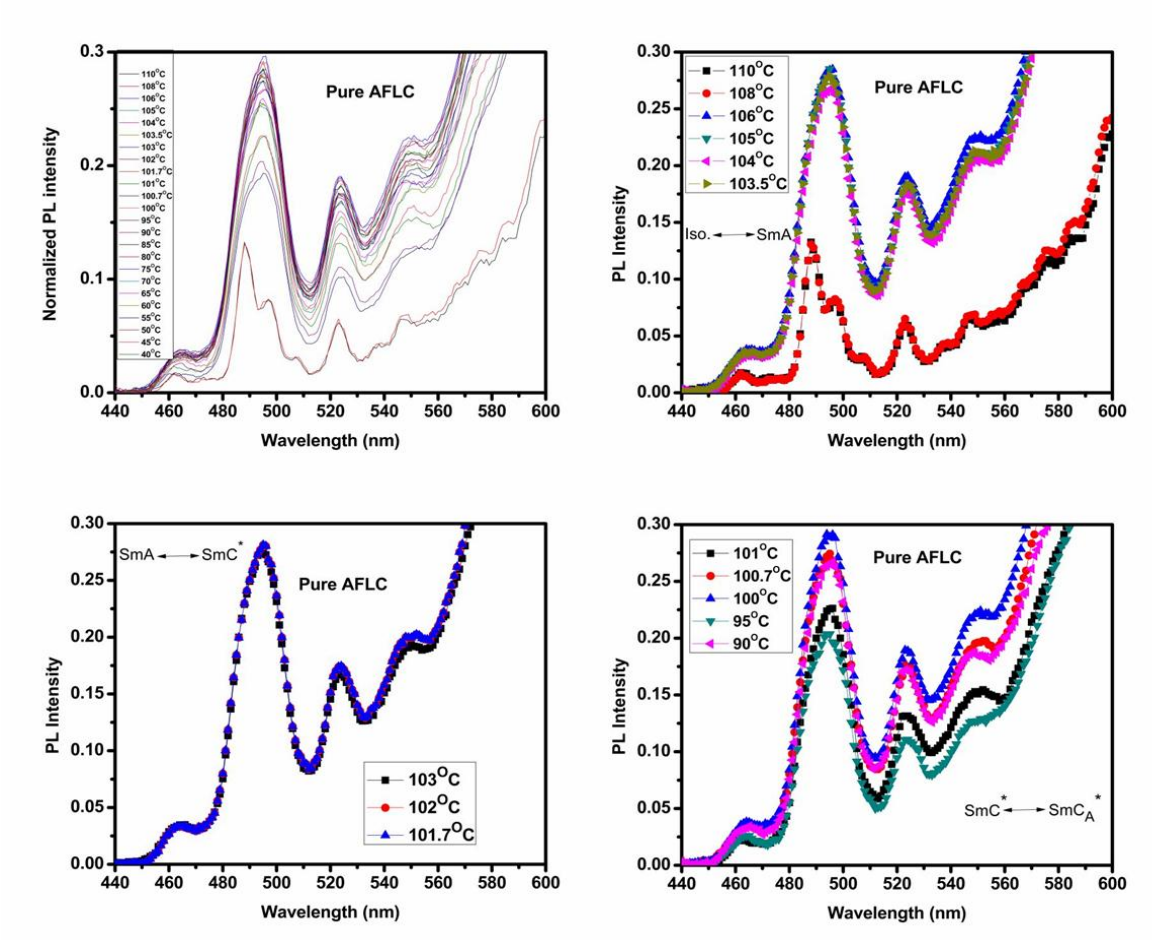

Fig.6 Photoluminescence Spectra for Pure AFLC in cooling cycle from 110ºC to 90ºC.

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Figure7 and **8** shows the luminescence variation with wavelength at different temperatures in cooling cycle from 110ºC to 95ºC for MIX 1 and MIX 2. The enhancement in PL intensity has been observed for GO dispersed system. The high luminescence yield is strongly dependent on GO concentration. We have examined the luminescence of AFLC by changing GO concentrations to 0.2 wt/wt% and 0.3 wt/wt%. AFLC W1000 mixture possesses highly light scattering FLC/SmC* phase therefore, incident photons gets multiplied on exposure to UV light. These incident photons are responsible for exciting GO flakes and we obtain high luminescence intensity. However scattering area remains fixed as the weight of AFLC is kept fixed and high luminescence has been obtained because of increasing concentration of GO in AFLC medium.

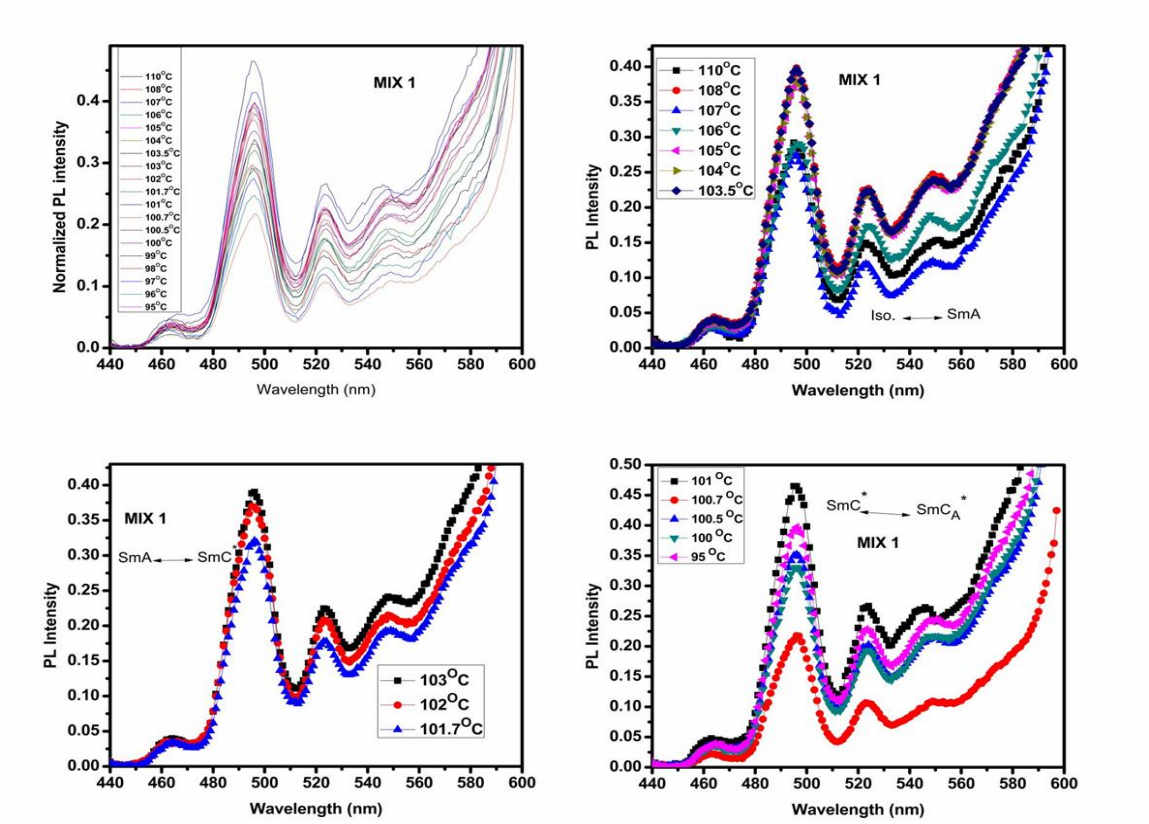

Fig.7Photoluminescence Spectra for MIX 1 in cooling cycle from 110ºC to 90ºC.

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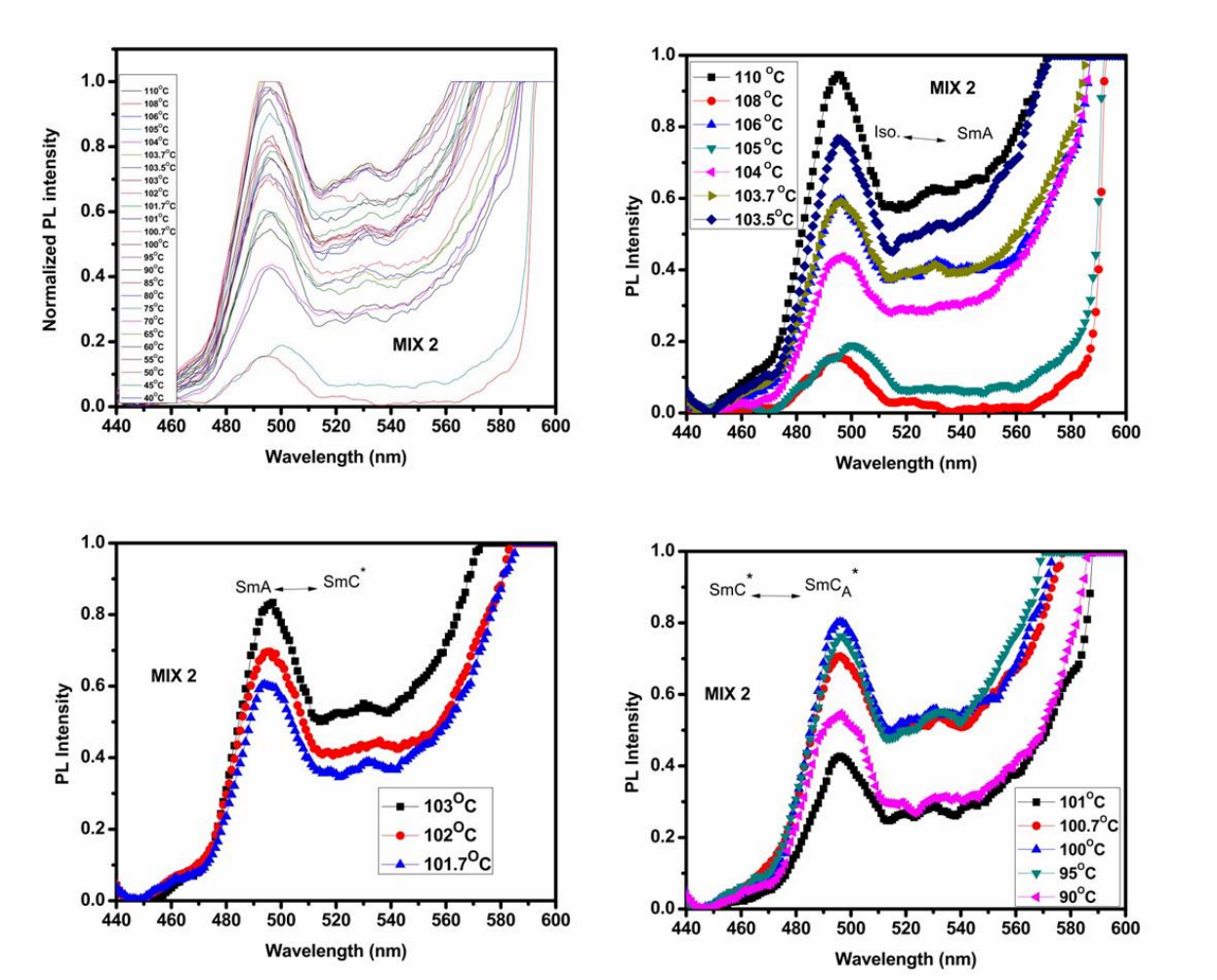

Fig.8 Photoluminescence Spectra for MIX 2 in cooling cycle from 110ºC to 90ºC.

5. Conclusion

Theinfluential effectof GO onphotoluminescence(PL) of AFLC liquid crystal mixture has been examined.The high yield of luminescence for AFLC – GO dispersed system has been found to be stronglydependenton GO concentration in AFLC medium. Luminescence intensity of pure and dispersed system has been found to change with the change in temperature of liquid crystal medium. This has been associated with various phases exhibited by AFLC mixture especially the influence of SmC^* and SmC_A^* phase. The red shifted UV absorbance wavelength has been revealed for GO dispersed AFLC system with decreased absorbance.Thispaperwillprovidetheviews and ideasfor thefabrication of novel photonic and display devices with enhanced features [**24, 25**].

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