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Ultrasonic Measurements and Optical Retardation Studies on Liquid crystalline Materials

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Short running title: Ultrasonic Measurements and Optical Retardation Studies on Liquid Crystalline Materials
Keywords: optical studies; Molecular self assembly; optical retardation; Ultrasonic measurements; adiabatic compressibility: Ultrasonic Measurements and Optical Retardation Studies on Liquid crystalline Materials
Abstract: We report the results of our studies on optical and thermal properties of binary mixture of compounds, viz: voriconazole and ethylene glycol. The mixture exhibits a very interesting lyotropic induced polymorphism such as sma, smc, smc* and sme phases, sequentially when the specimen is cooled from its isotropic liquid phase. They were observed using microscopic technique and also been verified from the results of optical anisotropic techniques. Ultrasonic experimental studies have been carried out to understand the molecular self assembly of the above phases at higher and lower temperatures. Optical retardation has also been discussed.
Keywords: Optical studies; molecular self assembly; optical retardation; ultrasonic measurements; adiabatic compressibility:

I. INTRODUCTION

During the last two decades: the interest in liquid crystalline materials has increased, which has lead for the increase inopto-electronic applications[1]. The ultrasonic properties of liquid crystals provide additional information about the different phases and phase transitions in different compounds. It is well known that the liquid crystals often exhibit polymorphism [2, 3].

However: optical polymer films play a critical role in improving the contrast ration gray scale, viewing angle and brightness in displays. New optical componensation films were used to improve the angular dependence of contrast and gray scale inversion, in which the optical retardation of the discotic molecular construction in hybrid molecular orientation mimicked that of the liquid crystals[4]. In addition other wide viewing LCD monitor technologies have been developed including in-plane switching (IPS) mode, multi-domain vertically aligned (MVA) mode and optically compensated bent (OCB) mode. However the liquid crystal response time in IPS, which does not have negative optical compensations film, still needs to be improved. In the other two modes: high quality uniaxial negative optical compensations films are desired to improve the optical performance of the black state and suppress the light leakage besides a multi-domain photo alignment in each pixel upon exposure to polarized UV can also be achieved in displays, which will provide the displays with improved wide viewing angles and less color shift [5-8].

In the present investigation, our aim iscarried outto study the optical and thermal properties of binary mixtures of liquid crystalline materials namely; Voriconazole and ethylene glycol (EG), which exhibits an lyotropic induced polymorphism such as SmA, SmC, SmC* and SmE phases, sequentially when the specimen is cooled from its isotropic liquid phase. They were observed using microscopic technique and also been verified from the results of optical anisotropic techniques. Ultrasonic experimental studies have been carried out to understand the molecular self assembly of the above phases at higher and lower temperatures. Optical retardation has also been discussed.

II. EXPERIMENTAL STUDIES

In the present work we have been considered the compounds namely: Voriconazole, and Ethylene Glycol was obtained from the Padmashri Scientific, Mysore. It was further purified twice by a re-crystallization method using benzene as a solvent. Mixtures of different concentrations of Voriconazole and Ethylene Glycol were prepared and were mixed thoroughly. These mixtures of various concentrations of Voriconazole, and Ethylene Glycol were kept in desiccators for a long time. The samples were subjected to several cycles of heating, stirring, and centrifuging to ensure homogeneity. The phase transition temperatures of these concentrations were measured with the help of Leitz-polarizing microscope in conjunction with a hot stage. The samples were sandwiched between the slide and cover slip and were sealed for microscopic observations. Refractive index has been measured using Abbe Refractometer. A polarizer has been introduced in Abbe refractometer to block the extraordinary ray, which clears the

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contrast of the boundary line at view of Refractometer. To calculate birefringence Δn following relation has been used $\Delta n = n_e - n_o$. The temperature of Abbes Refractometer is controlled by circulating heated oil using JULABO F-25, refrigerated circulator. The temperature was measured by placing a thermocouple in close vicinity of the sample with an accuracy of +0.1°C. The ultrasonic velocity was measured at 2 MHz using the ultrasonic interferometer UI 601 NPL India. The cell was essentially the same as that supplied with the interferometer except a few modifications for the heating arrangement. The temperature of the cell was controlled by controlling the current flowing through the heating element surrounding the cell. The permitted temperature control was ±0.2°C. The ultrasonic velocity measurements were accurate to ±0.2%. [9, 10]

III. RESULTS AND DISCUSSIONS

A. *Optical Texture Studies*

For the purpose of optical texture studies, the sample was sandwiched between slide and cover glass and then the optical textures were observed using a Leitz polarizing microscope in conjunction with a specially constructed hot stage. Mixtures with concentrations ranging from 10%-70% of Voriconazole in Ethylene Glycol slowly cooled from isotropic melt. The genesis of nucleation starts in the form of small bubbles initially, but which progressively grow radially and form a focal conic fan texture of smectic-A phase in which the molecules are arranged in layers and the texture is shown in Figure 1 (a). This phase appears to be metastable and undergoes slow transformations to give schlieren texture of smectic-C phase as shown in Figure 1(b) on cooling the specimen. The molecular orientation of SmC phase is not energetically stable and then it changes over to SmC* phase, which exhibits a radial fringes on the fans of focal conic textures, these are the characteristics of chiral SmC∗ phase, which as shown in Figure 1(c). In the same way, orientation of the molecules in SmC∗ phase is also not stable and then changes over to focal conic fan texture with radial striation on the fans [11,12], which is the characteristics of smectic-E phase and it is observed as shown in Figure 1 (d).At this phase transition, i.e, from smectic-C* phase to smectic-E phase, it is observed that there is a drastic change in the values of optical anisotropic of the given sample [13, 14].This anomalous behavior is presumably associated with the degree of order of the molecular arrangement in smectic-E phase and then it becomes crystalline phase at room temperature.

B. *Temperature Variations Of Ultrasonic Velocity, Adiabatic And Molar Compressibility*

The periodic squeezing and stretching of a fluid, through the propagation of sound waves, provides considerable information about the internal properties of the matter. The study of ultrasonic propagation in liquid crystals has provided a significant contribution in understanding many interesting properties of these materials. Ultrasonic studies on liquid crystals are one of the several tools generally employed to establish phase transitions in these compounds. As ultrasonic velocity and absorptions are related to the molecular structure of the studies during phase transitions and mesomorphic range provides good information to have an idea regarding the structural behavior of liquid crystals. In order to study the temperature dependent ultrasonic velocity and related parameters such, as adiabatic compressibility and molar compressibility provides information regardingthe phase transitions and pre-transitional effects in liquid crystals. Molecular orientations of one more additive substances increases as well as increasing the orientation order of liquid crystalline phases and also the measured ultrasonic velocity should show a nature of attractive forces existing between the mixtures of given molecules. Data on some of the properties associated with refractive index, ultrasonic velocities, and surface tension find extensive application in chemical engineering process, simulation and molecular dynamics [15].The temperature variation of ultrasonic velocity and adiabatic and molar compressibility in the present case is shown in Figures 2 (a-c).The velocity exhibits an anomalous behavior at the isotropic mesophase transition, while it varies linearly in the isotropic and mesomorphic phases away from transition. The velocity shows a dip at transition. The ultrasonic velocity linearly increases in isotropic phases up to transition with decreasing temperature [16, 17]. An increase in velocity is explained as a decrease in mean distance between the molecules, thereby increasing the potential energy of interaction between molecules. The velocity reaches minimum at transition temperature and increases sharply below the transition, and then it shows a linear increase in mesophase. The change in velocity and other parameters [18] at the transition results from disordered molecular arrangement in isotropic phase to an ordered arrangement of molecules in the liquid crystalline phase with long-range orientational order [19].Variation in adiabatic compressibility [20] is remarkably linear in isotropic and mesomorphic phases, but it shows a step jump at isotropic-mesophase transition. The result of molar compressibility varies linearly with temperature at the isotropic phase transition. In this study, the higher values of thermal expansion coefficient in mesophase than in isotropic phase confirm the tendency of increasing order ofmolecules with decrease in temperature. It is firmly established that the ultrasonic velocity and the related parameters [21] are structure-dependent. Besides, depending on the structure, these are related to intermolecular interactions and degree of molecular order in liquid crystalline mixture.It is well known that in liquid crystalline phases, the molecules are arranged in order, and the

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orderliness increases from isotropic to lyotropic induced polymorphism. These lyotropic induced polymorphism is known to exhibit at different temperatures and at different concentrations of given molecules compared with other conventional liquid crystalline phases such as smectic-A, schlieren texture of smectic-C, SmC* phase and smectic-E phases [22, 23].

C. Optical Phase Retardation

The precise determination of the optical phase retardation and thereby of the birefringence for certain materials (liquid crystal, for example) plays an important role in various fields of research. Photonic switching operation in free space, for example, requires devices that rely on the separation of polarization or the deflection of light beams by means of liquid crystal cells whose birefringence is controlled electrically. The accurate measurement of their optical path difference is useful to optimize such structures. Many techniques have been developed for measuring the index of refraction and/or the birefringence of anisotropic liquid crystalline samples[24, 25].Birefringence is responsible for the appearance of interference colors in liquid crystal display operating with plane-polarized light. Interference between the extraordinary ray and the ordinary ray, which have traveled through the medium with different velocities, gives rise to the colored appearance of given liquid crystalline materials. For the liquid crystals with positive birefringence ($\Delta n>0$) the e-axis is the “slow” axis and the o-axis is the “fast” axis. If the optical axis is not parallel to the surfaces, the extraordinary refractive index has to be replaced by the angle between the direction of propagation and the optical axis. The retardation is wavelength dependent, so that positive and destructive interference occur at different wavelengths, resulting in the suppression of some part of the visible spectrum, and, therefore, a nonwhite color. Moreover, the birefringence is also wavelength and temperature dependent, because the refractive indices also vary with these parameters [26].

The optical retardation values of the given mixture were estimated at temperature dependent optical birefringence for different concentrations and at different thickness of the liquid crystalline materials. Here we have pointed out in our study that the variation of optical retardation with birefringence is dependent upon both the thickness and shape of the molecular aggregation, in addition to their dependence on orientational order. However, we also notice that the optical retardation varies with mole percent and thickens of voriconazole. The variation of optical retardation as function of temperature dependent birefringence in the present case is shown in Figure 3. Mixtures of different concentrations of given material varies with the birefringence value and hence here it is observed that the optical retardation increases with increasing the birefringence and at different thickness of the given materials. However, optical retardation varies significantly upon cooling, which may be due to the transformation of polymorphic induced chiral smectic phase. Changes in optical retardation are expected to be due to changes in the dimension of discs along with changes in the orientational order of the molecule.

IV. CONCLUSIONS

The salient features of this investigation are as follows: The existence of lyotropic induced chiral polymorphic smectic phases have been observed by using microscopic technique in binary mixture of Voriconazole in Ethylene Glycol. The temperature variation of birefringence across chiral smectic-C smectic-C* phase is more predominant than other transitions. Changes in optical retardation value are are expected to be due to changes in the dimension of discs along with changes in the orientational order of the given molecules. The anomalous behavior of liquid crystalline physical properties, such as ultrasonic velocity, adiabatic compressibility, and molar compressibility, is discussed at the isotropic mesosphere transition.

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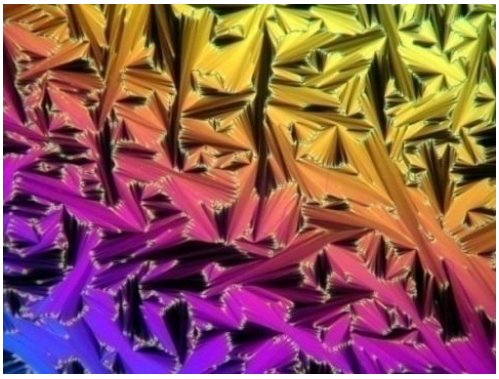
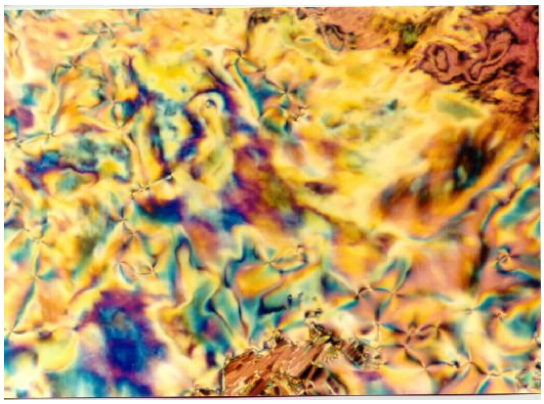
Figure Captions

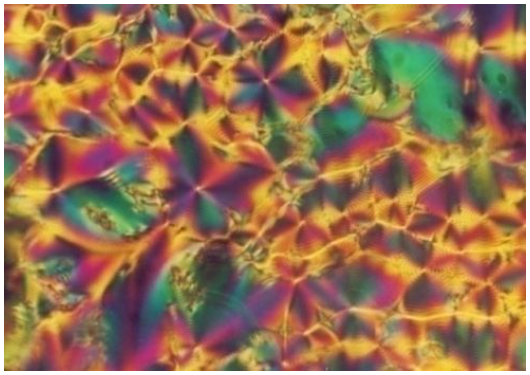
Figure 1. Microphotographs obtained in between the crossed polars.



1) Focal conic fan-shaped texture of SmA phase (250X)



b. Schlieren texture of SmC phase (250X).



c. Radial fringes on the fans of focal conic textures of smectic-C* phase (250X).

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d. Focal conic fans with radial striation of smectic-E phase (250X).

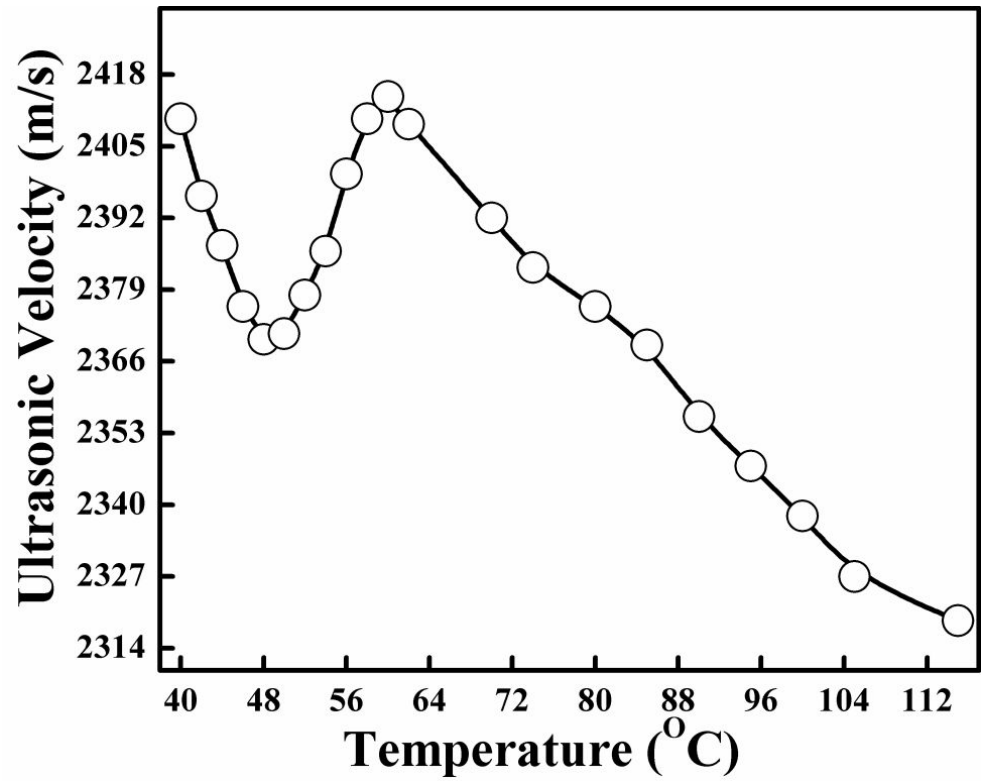

Figure 2(a).Temperature variation of ultrasonic velocity for a sample of Voriconazole in Ethylene Glycol

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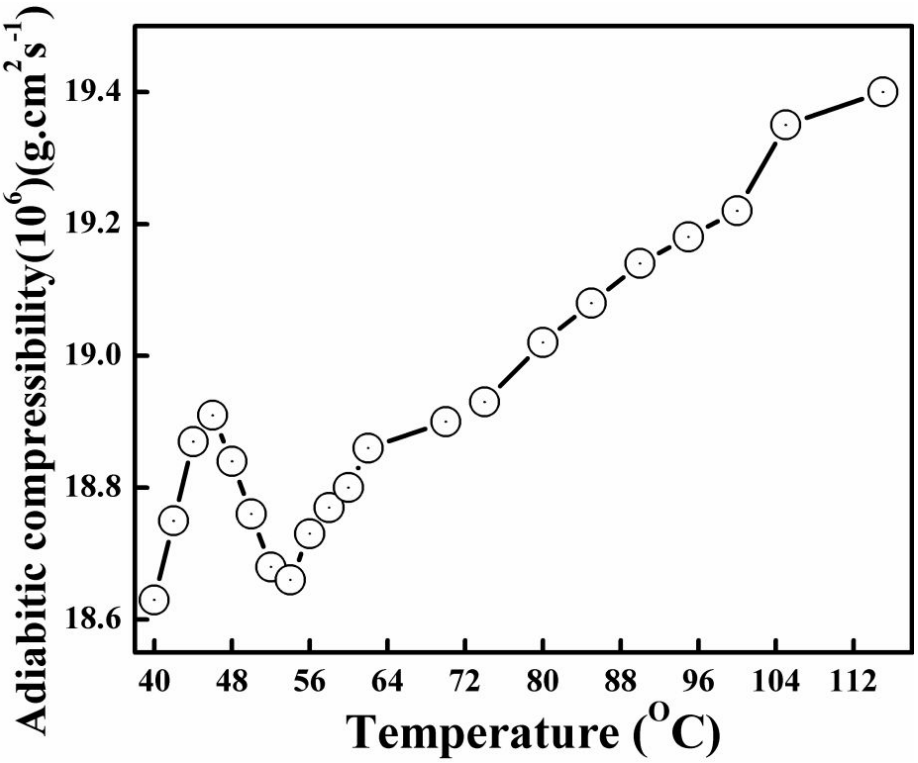

Figure2(b).Temperature variation of adiabatic compressibility for a sample of Voriconazole in Ethylene Glycol

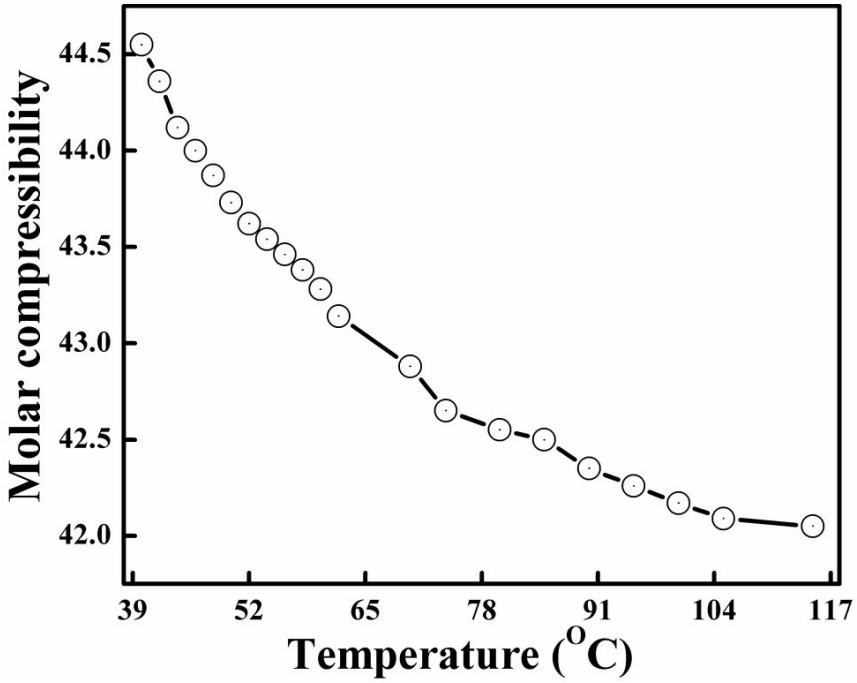

Figure 2(c).Temperature variation of molar compressibility for a sample of Voriconazole in Ethylene Glycol.

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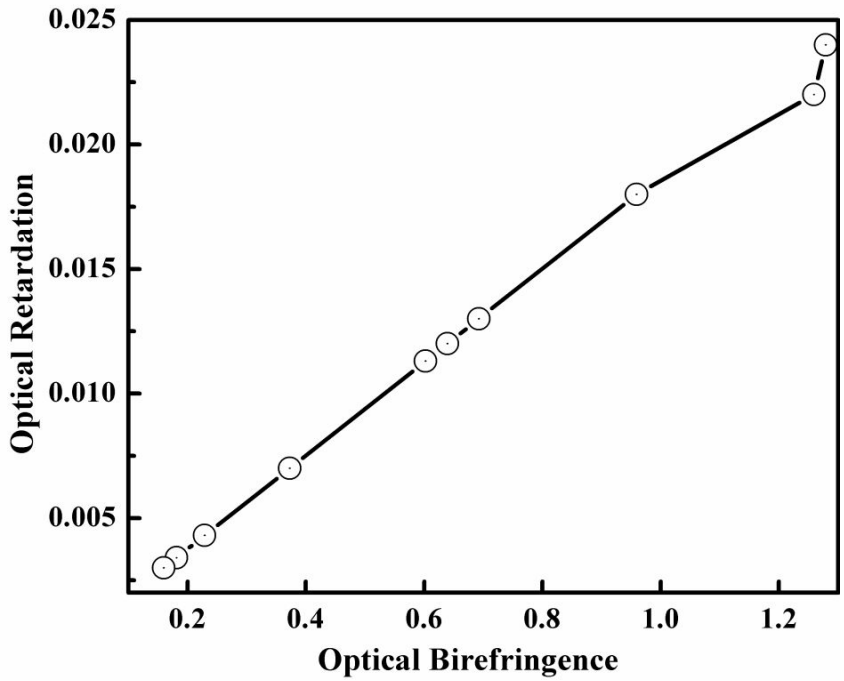

Figure 3.The variation of optical retardation as function of optical birefringence for a sample of Voriconazole in Ethylene Glycol.

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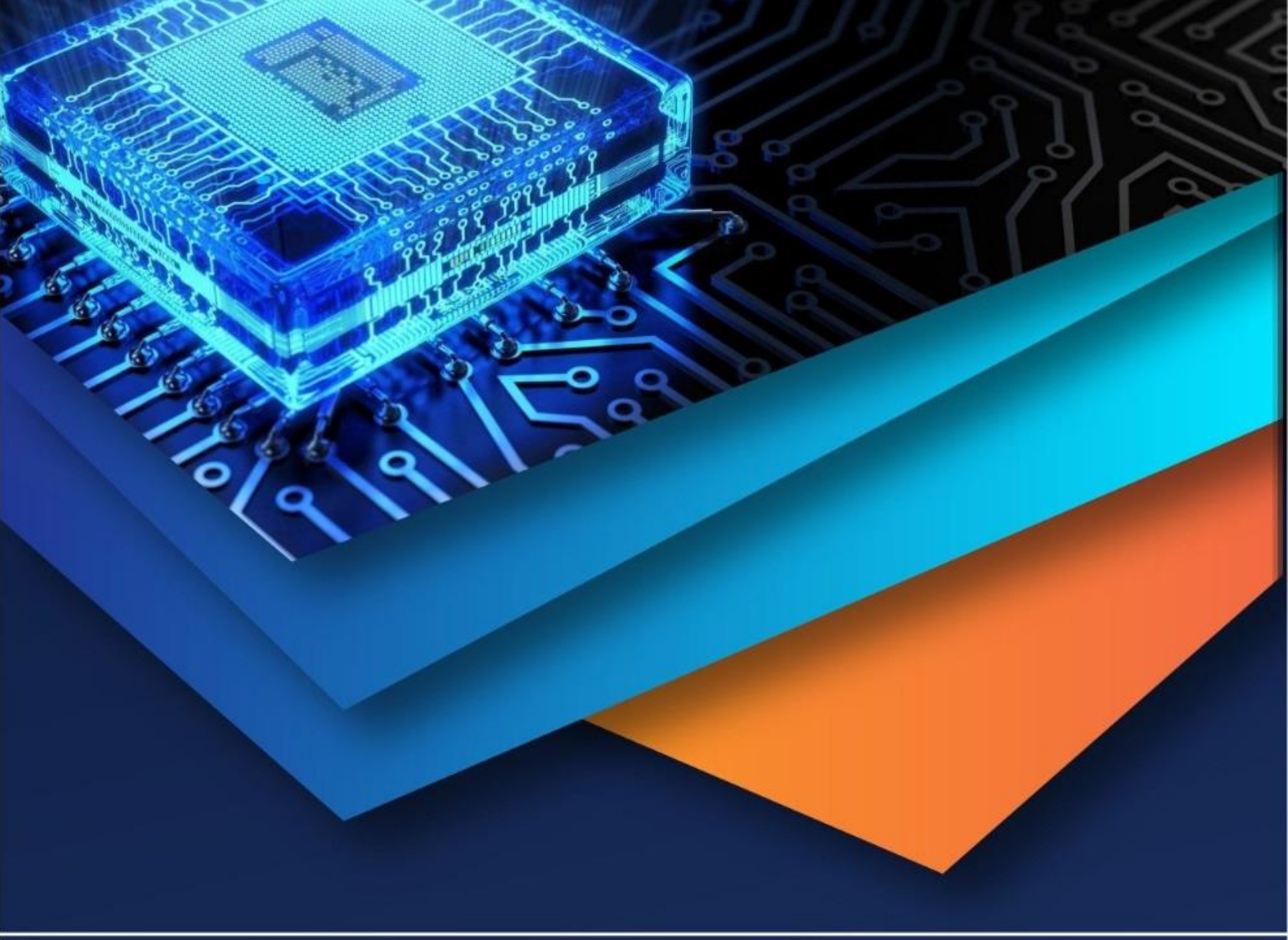

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