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HYDROGEN BONDING EFFECT ON OPTICAL PROPERTIES OF SUPRAMOLECULAR LIQUID CRYSTALS IN UV VISIBLE REGION

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ABSTRACT

This study utilizes spectral analysis techniques and various computational methods to examine the impact of hydrogen bonding on the optical characteristics of supramolecular liquid crystals within the UV-visible spectrum.This investigation involves the computational method of spectral denoising for quantitative analysis of UV-visible spectra, and then spectral analysis is carried out for the measurement of the optical properties of supramolecular liquid crystals. Other computational methods, like the texture absorbance method, are also employed for colorimetric studies of compounds.The investigation will focus on supramolecular systems:p,n- alkoxy benzoic acid: Do-decyl hydroxybenzoate (nOBA:12HB), where n = 7, 8, and 9. The supramolecular system is formed via hydrogen bond development between the OH acceptor of the non-liquid crystalline substance Do-decyl hydroxyl benzoate (12HB) and the COOH donor of the liquid crystal p-n alkoxy benzoic acid. Here, optical properties: such as absorption coefficient, optical energy band gap, Urbach energy, and refractive index of supramolecular liquid crystals are measured for the denoised spectral data. The influence of hydrogen bonding and alkoxy chain length on the optical properties of liquid crystals has been deliberated upon. Further, the attempt at computation studies on the spectral data will yield important insights for subsequent investigations into UV-visible spectral data measurements.

Keywords: Liquid Crystals, HydrogenBond, UV-Visible Spectroscopy, Absorption, Chromaticity.

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INTRODUCTION

Supramolecular hydrogen-bonded liquid crystal systems (SMHBLCs) have gained significant attention in recent studies because of their wide range of applications in science and technology. For modern technologies, it is more and more crucial to produce and use materials with well-defined molecular structures and properties.[1-4] The creation of a hydrogen bond between the compounds in supramolecular hydrogen-bonded liquid crystals results in the insertion of a lateral group on the aromatic ring of the mesogenic portion, which affects the molecular and physical characteristics. Such characteristics are molecular structure, alignment, polarizability, dipole moment, geometry, transitional characteristics, optical properties, etc.[5-8]These characteristics are necessary for the LC materials to perform better in applications. Such applications include the various device fabrications in optics, photonics, and biology. Such devices are information storage systems, displays, optical switches,projectors, phase shifters, photonic crystals, optical fibers, biological sensors, cosmetics, etc.[1-4,9,10]The structural and physical characteristics of liquid crystals in the presence of external fields, such as temperature, electric field, magnetic field, and wavelengths of various spectrum regions, determine all of these uses.[11,12]Therefore, the investigation of the characteristics of supramolecular materials as a function of external fields is important for the applications of liquid crystal devices.Several theoretical works and different experimental methods have been proposed by many authors to study the structural and other physical properties of supramolecular liquid crystals. However, less work is reported on the wavelengths of different spectral regions for supramolecular liquid crystals. Within this framework, a few scientists investigated the possibilities of supramolecular hydrogen-bonded liquid crystal materials in the ultraviolet to visible spectrum.[5,13]In the UV and visible spectra, the spectrally active supramolecular liquid crystals

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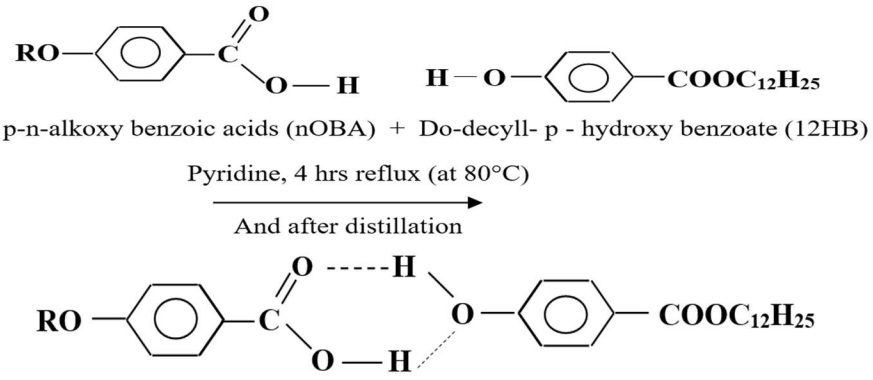
with varying terminal chain lengths and lateral substituents alter the absorption spectra's intensity, wavelength, or shape. The desired electrical or optical properties of materials are influenced by the spectrum absorbencies and their shifts. Investigations into electronic transitions, spectrum absorption, and associated changes in the ultraviolet range will yield important insights for device applications.[14-16]Such organic light emitting devices,solar cells, good absorption, and high UV stability materials are used for sunscreen lotions in cosmetics, etc. Here, supramolecular HBLCs p,n-alkoxy benzoic acids – Do-decyl hydroxyl benzoate are abbreviated as nOBA:12HB (where n = 7, 8, and 9) and are taken for investigation.The COOH groups of p,n-alkoxy benzoic acids (where n = 7, 8, and 9) liquid crystal compounds, and the OH group of Do-decyl hydroxyl benzoate (12HB - non-liquid crystalline chemical) develop a hydrogen bond to form the SMHBLCs. Hydrogen bond formation between the aromatic ring structure of nOBA and hydroxyl benzoate greatly improves the UV absorption performance of SMHBLCs. There is no photodegradation caused by the hydrogen bond formation, which greatly increases photostability.[14,17-19] Therefore, SMHBLCs: nOBA:12HB (n = 7, 8, and 9) with high UV absorption, excellent light stability, etc. may be useful for applications in health care like antibiotics, preservatives, cosmetics, sunscreen lotions, etc., besides optoelectronic devices. Here, the impact of hydrogen bonding on the optical properties of supramolecular liquid crystals in the UV-visible region is investigated using the spectral analysis method and some computational techniques.The quantitative analysis of UV visible spectroscopy encounters challenges related to spectral noise. Before data processing, the spectrum data must be denoised since this spectral noise impairs the precision and accuracy of quantitative spectral outputs.[20,21] Here, some computational techniques of signal denoising methods like median filtering (which is simple and effective) are employed on the UV-visible spectral data of the SMHBLCs. Later, the hydrogen bonding influence on the characteristics of supramolecular liquid crystals in the UV visible region is investigated by measuring the optical properties from the spectral absorbance studies; such properties are absorption coefficient,optical energy band gaps, Urbach energy, refractive index, etc.[22-24]The spectral absorbencies obtained from the UV spectra are supported by the computational study of the texture absorbance method, which gives the colorimetric information of the compounds in terms of computing chromaticity coordinates. The parameter that is measured by these computational methods enhances the precision and accuracy of spectral analysis. The obtained result demonstrates the effect of hydrogen bonds and alkoxy chain length on the optical properties of liquid crystals in the UV region and their applicability in electro-optical and biological contexts.

EXPERIMENTAL

Frinton Laboratory, New Jersey, USA, provided the chemical components do-decyl-p-hydroxy benzoate (12HB) and p,n-alkoxy benzoic acid (nOBA, where n=7,8, and 9). To create the new liquid crystal complexes p-n alkoxy benzoic acid: do-decyl-p-hydroxy benzoate, abbreviated as (nOBA: 12HB, n=7,8, and 9), the nonliquid crystalline compound do-decyl-p-hydroxy benzoate (12HB) is mixed with an equimolar (1:1) ratio of the liquid crystalline compound p-n alkoxy benzoic acids (nOBA).The SMHBLCssynthetic root is provided below:



nOBA: 12HB,Where R= $C_nH_{2n+1,}$ and n = 7,8, and 9.

Scheme-1: Schematic representation for the synthetic root of hydrogen-bonded liquid crystals (nOBA:12HB).

The analysis of synthesized SMHBLCs is carried out using the POM method. With the use of an SD-Tech Polarizing optical microscope that contains a hot stage and an imaging attachment, the phases, and textures of the materials are observed.[25,26]The UV-visible spectra of liquid crystals are obtained using a Japan-manufactured LabIndia analytical UV-visible spectrophotometer type UV-3092. At room temperature, spectral measurements are conducted in the 200–800 nm wavelength range using 1 cm cuvettes containing the sample solution. On the MATLAB platform, computational analysis of pure and hydrogen-bonded liquid crystals has been done.[27,28] The parameters and methods used in this investigation are described in.[22,24,29-31]

RESULTS AND DISCUSSION

The purpose of the current study is to determine how hydrogen bonding affects the optical characteristics of supramolecular liquid crystals in the UV visible range using SMHBLCs (nOBA: 12HB, where n=7,8, and 9). Moreover, to improve the precision and accuracy of quantitative spectrum data before spectral analysis, and to look into their UV stability in light of the absorption wavelength change about alkoxy chain length, the spectral data was denoised using a computational technique of signal denoising called median filtering. This is a simple and effective signal-denoising method, and it was carried out using the computational tool - MATLAB. Fig.-1 illustrates the original and denoised UV-visible absorption spectra of nOBA and nOBA:12HB (where n = 7, 8, and 9). The graph clearly demonstrates the effectiveness of median filtering in reducing noise without compromising important signal characteristics.

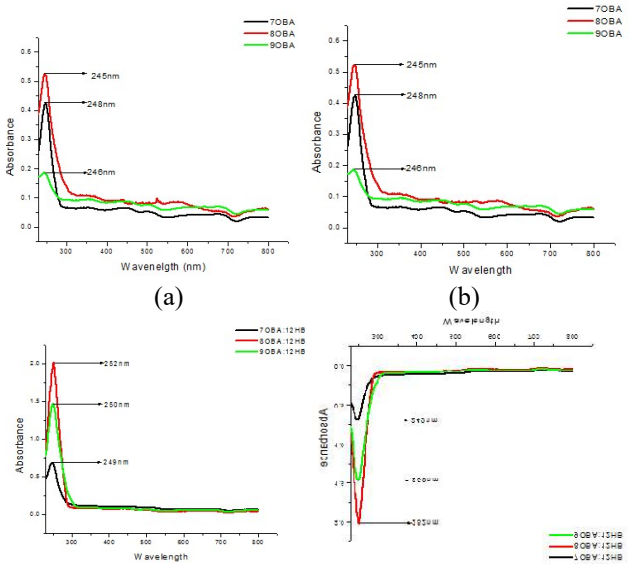


Fig.-1: UV Spectra of nOBA Liquid Crystals (a) Original Spectra; (b) Denoised Spectra; (c)&(d) are Original and Denoised Spectra of nOBA:12HB

The principal absorption bands of pure compounds (nOBA) appear in the wavelength range of 245-248 nm, and for SMHBLCs, they appear in the range of 249-252nm. The primary cause of these spectrum shifts in both pure and SMHBLCs is the $\pi \rightarrow \pi^*$ transitions in the molecule's benzene homologous region. In the visible area of pure and SMHBLCs nOBA:12HB (where n = 7, 8, and 9), there is no maximum absorption detected. The magnitudes of absorbance of SMHBLCs are higher compared to the nOBA. Spectral shifts and their absorbencies are influenced by intermolecular interactions; alkoxy chain length, degree of conjugation, etc.[16, 22,2332,33] The maximum absorption wavelengths and their absorbencies of liquid crystals are given in Table-1.

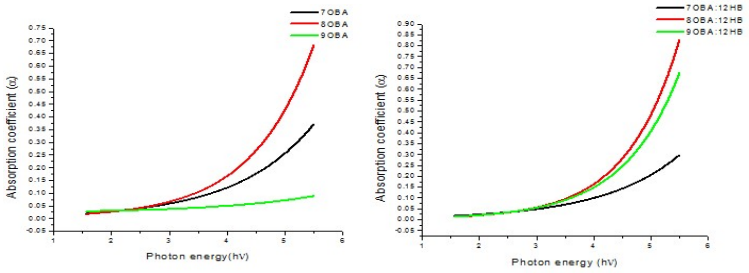
The excited state molecule's energy is decreased by intermolecular interactions between the molecules, increasing the energy needed for the electron to move from the ground state to the excited state. This leads to shifts in the maximum absorption wavelength, mainly redshift, and the absorbance of the samples will increase. Here also, the formation of hydrogen bonds in SMHBLCs leads to the shift of the maximum absorption wavelength towards the longer wavelength (red shift) side of the pure compounds.[22] And the absorbencies of the SMHBLCs are also higher than those of pure compounds. Because of the elongated

molecule size, an increase in absorbance or hyperchromic effect reduces packing density, and thus the overall absorption is increased. However, the absorption wavelength (red shift) and hyperchromic effect of UV stability are higher for 8OBA:12HB compared to 7OBA:12HB and 9OBA:12HB. These effects are due to the molecular size, strength of the hydrogen bond,and odd-even behavior of the liquid crystal. For several molecules molecular and optoelectronic applications, these redshifts and high absorption values in the UV and visible ranges are attractive characteristics.

Table-1: Maximum Wavelength and their corresponding absorbencies of nOBA and nOBA:12HB (n=7,8, and9**)**

Compound	λ_{max}	absorbance
7OBA	248	0.42
8OBA	245	0.522
9OBA	246	0.182
7OBA:12HB	249	0.68267
8OBA:12HB	252	2.0175
9OBA:12HB	250	1.4663

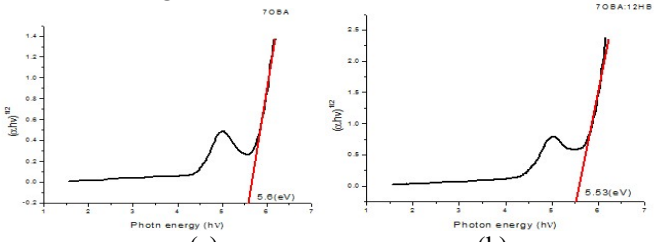
Therefore, the study of these spectroscopic transitions of supramolecular liquid crystals with different chain lengths is very important from an application point of view. Such optical properties are absorption coefficient, optical energy band gap, Urbach energy, refractive index, etc., and are computed from the equations given in.[22, 24, 29-31]The atomic structure, electronic band structure, and electrical characteristics of the material are intimately linked to these optical characteristics. The optical constants of the given nOBA and nOBA:12HB (where n = 7, 8, and 9) are measured from the denoised spectral data. Plots related to optical properties are shown in Fig.-2 to 4, and values are given in Table-2.



(a) (b)

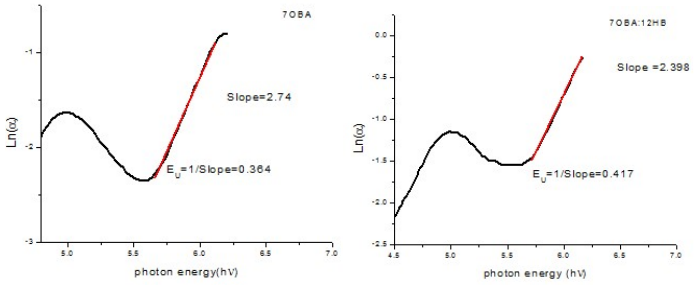
Fig.-2: Absorption Coefficient of the (a) nOBA; (b) nOBA:12HB as a FunctionofPhoton Energy

Figure-2 shows the variation of the absorption coefficient of nOBA, nOBA:12HB as a function of wavelength (photon energy). The magnitude of the absorption coefficient of pure compounds is low compared to SMHBLCs and it shows variations with increasing alkoxy chain length.This indicates that the intermolecular interactions have a profound influence on the absorbance intensity. But for both pure and hydrogen-bonded liquid crystals absorption has its minimum value at low energy states and increases with high optical energy, like the absorption of liquid crystal samples at higher temperatures. HBLC 8OBA:12HB has high values of the absorption coefficient for all wavelengths compared to the remaining. Parameters: optical energy band gap (E_g), Urbach energy(E_U),and refractive index of SMHBLCs are derived from these absorption coefficient values. The values of the optical energy band gap (E_g) for both samples nOBA and nOBA:12HB (where n=7,8and 9) can be derived by extending the linear segments of the $(\alpha h\nu)^{1/2}$ and 'hv' plots, as seen in Fig.-3.



(a) (b)

Fig.-3: Optical Energy Band Gap Measurements from the Linear Fitting of $((\alpha h\nu)^{1/2}$ v/s 'hv') (a) for 70BA; (b) for 7OBA:12HB



(a) (b)

Fig.-4: logarithmic variation of the Absorption Coefficient as a function of Photon Energy ('ln(α)' v/s 'hν')(a) 7OBA; (b) 7OBA:12HB

The slopes of the linear fitting of "ln(α)" and "hν" yielded the values of E_U, and Fig.-4 displays these curves. Table-2 reports the values that were obtained.

As a representative case, the finding of the optical energy band gap and Urbach energy of the 7OBA and 7OBA:12HB liquid crystals are shown in Fig.-3 and 4. A similar procedure was applied for the remaining compounds to calculate the parameters, E_g, and E_U.

Table-2: Optical Properties of nOBA,nOBA:12HB (where n=7,8, and9)

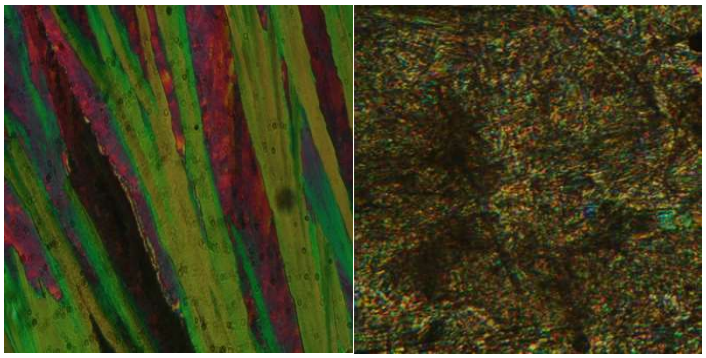
Compound	Optical energy band gap E_g (eV)	Urbach Energy E_U (eV)	Refractive Index
7OBA	5.63	0.364	1.462
8OBA	5.61	0.371	1.484
9OBA	5.55	0.4	1.547
7OBA:12HB	5.53	0.417	1.646
8OBA:12HB	5.57	0.394	1.5
9OBA:12HB	5.47	0.44	1.678

The optical energy band gap and Urbach energy values of nOBA and nOBA:12HB (n=7,8, and 9) determined from the plots displayed in Figs.-3, 4, and 5 are presented in Table-2.Both parameters are important for the characterization of the disorder of the materials, but they are inversely related to each other, which means that a sample having a smaller optical band gap is expected to have a wider band tail. From Table-2, it was observed that the optical band gap of nOBA liquid crystals is high compared to the SMHBLCs and vice versa for Urbach energy. This is due to the intermolecular energy transfer in nOBA: 12HB complexes. Once the donor molecule is excited by absorbing the UV visible wavelength, it transfers part or all of the energy to the acceptor to get excited.The increase in intermolecular energy transfer efficiency reduces the optical band gaps of SMHBLCs and induces considerable structural changes. This leads to the formation of less and more order mesophase**.**Urbach energy values presented in Table-2 prove the presence of disorder in the supramolecular compounds since Urbach energy is associated with micro-structural lattice disorders as well.[30,34] From Table-2, it was also observed that there are certain variations in the optical energy band gaps and Urbach energy with increasing alkoxy chain length.SMHBLCs having odd alkoxy chain lengths 7OBA:12HBand 9OBA:12HB have fewer band gaps and higher Urbach energy compared to the even alkoxy chain length compound 8OBA:12HB.The observed increase and decrease in the band gaps of SMHBLCs 7OBA:12HB, 8OBA:12HB, and 9OBA:12HB describe the strength of the molecular interactions, the odd even behavior of the molecules, and the density of localized states. The refractive index is also another important optical property of a liquid crystal material. Here, the refractive index of hydrogen-bonded liquid crystals is measured from the absorption coefficient and reflectance of the UV-visible spectra. Table-2 shows that the refractive index of pure compounds is lower compared to the SMHBLCs. Complexes 7OBA:12HB and 9OBA:12HB have higher refractive index values compared to 8OBA:12HB. This is due to the strength of the bond formation between the compounds,the alkoxy chain length, and the odd-even behavior of the molecules.[35] 8OBA:12HB with an even number of methylene units has a difference in molecular shape, and structure and exhibits properties different from 7OBA:12HB and 9OBA:12HB (which have an odd number of methylene units). This leads to the variations in the optical properties of 8OBA:12HB from 7OBA:12HB

and 9OBA:12HB. From the above studies, it was observed that supramolecular systems nOBA:12HB (where n = 7, 8, and 9) have low absorbencies from pure compounds (nOBA) in the UV-visible region. Also, changes in the optical parameters of the SMHBLCs in the visible region are very small since the samples were not sensitive to any single wavelength. The same was proven from the textural absorbance studies. For this, the textures of liquid crystals nOBA and nOBA:12HB are recorded from the crystal phase of the sample since the UV experiment was done at room temperature. Measurements of textural absorbance are done in the visible area here. The absorbance of nOBA and nOBA:12HB is carried out on the MATLAB platform using equation (1).[36]Absorbance of the optical texture is

$$A = -log_{10}\left(\frac{I - I_{min}}{I_{max} - I_{min}}\right) \quad (1)$$

Where "A" denotes the texture's absorbance, "I" denotes the texture image captured from the sample's crossing polarization components, and "I_{max}" is the texture captured from the sample's parallel polarization components in the absence of a sample. Fig.-5 displays the textures of the nOBA and nOBA:12HB that were captured at 30^{O}C. Table-3 provides the computed absorbencies for the three picture planes with wavelengths in the visible spectrum.



(a) (b)

Fig.-5: Optical Textures of (a) nOBA; (b) nOBA:12HB at Temperature 30^{o}C

Table-3: Absorbencies of nOBA, nOBA:12HB Liquid Crystals

Compound	Wavelength	Absorbance
nOBA	Red	0.407
	Green	0.194
	Blue	0.284
nOBA:12HB	Red	0.444
	Green	0.353
	Blue	0.365

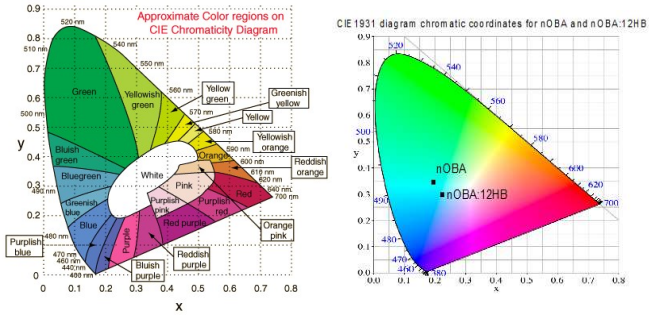
Absorbencies of the nOBA and nOBA samples are given in Table-3. It shows that the absorbencies of nOBA:12HB at three wavelengths in the visible region are high compared to nOBA. Not only from the computed values, but the visual appearance of the liquid crystal textures also tells us that the absorbance of nOBA:12HB is high compared to nOBA. Changes in the molecular alignment due to hydrogen bond formation cause variations in the intensities of transmittance and the absorbencies of the liquid crystals. This was observed pictorially in Fig.-5. The colorimetric study of nOBA and nOBA:12HB was conducted by computing the chromaticity coordinates based on the liquid crystal textures.Chromacity coordinates computed from the equations given in[37,38] and compared with standard chromaticity values given in Table-4 and obtained chromaticity coordinates of the nOBA and nOBA:12HB are given in Table-5.

Table-4: Standard Chromacity coordinates

Co-ordinates	Red	Green	Blue
x	0.5290	0.3050	0.1880
y	0.3930	0.5810	0.1710

Table-5: Chromacity Coordinates (x,y) of the Noba and nOBA:12HB.

Compound	x	y
nOBA	0.2	0.35
nOBA:12HB	0.23	0.3



(a) (b)

Fig.-6: (a) Standard Chromaticity Diagram; (b) Identification of Chromaticity Coordinates from the Textures of nOBA and nOBA:12HB

According to Table-5, it was found that the chromaticity coordinates of nOBA and nOBA:12HB were in closer proximity to green and blue wavelengths.This indicates that the samples are transparent across all wavelengths, as they do not have limited transmission peaks specific to one wavelength. The obtained coordinates of the nOBA and nOBA:12HB are displayed in the chromacity diagram shown in Fig.-6(b). This clearly illustrates that the coordinates of samples occupy the entire chromaticity diagram, indicating a lack of sensitivity to any specific wavelength. This can be observed in the optical textures, which appear with different color combinations. The findings of this work show that the molecular and optical characteristics of nOBA:12HB in the UV-visible range are influenced by hydrogen bond formation in liquid crystals. The influence is varying with increasing alkoxy chain length of the nOBA:12HB (where n = 7, 8, and 9). However, the magnitudes of the variations in the optical prosperities are very small. This is because the liquid crystalline complexes nOBA:12HB (where n=7,8, and 9) are produced by the weak intermolecular interactions between nOBA and 12HB. Compared to 7OBA:12HB and 9OBA:12HB, the supramolecular system 8OBA:12HB exhibits stronger molecular interactions, higher absorbance, and UV stability among the provided nOBA:12HB. Therefore, liquid crystal complexes with high UV stability, high absorption, and a decrease in bandgap are useful for optoelectronic applications, photoelectrochemical hydrogen generation, solar cells, and cosmetics.

CONCLUSION

Using spectral analytic methods and computational approaches, the influence of hydrogen bonding on the optical characteristics of supramolecular liquid crystals in the UV visible range is successfully explored. The weak supramolecular system is formed via hydrogen bond formation between the OH acceptor of the non-liquid crystalline molecule Do-decyl-p-hydroxyl benzoate (12HB) and the COOH donor of the liquid crystal p-n alkoxy benzoic acid. It was explained that the length of the alkoxy chain and hydrogen bonds affect the optical characteristics of liquid crystals. Significant optical properties: high absorption coefficient, UV stability, optical energy band gap, and refractive index of supramolecular liquid crystal 8OBA:12HB improve the material performance for applications. The UV-visible spectral data readings can be objectively and quantitatively analyzed through computational studies such as spectral denoising and texture absorbance research.

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CONFLICT OF INTERESTS

The authors declare that there is no conflict of interest.

AUTHOR CONTRIBUTIONS

All the authors contributed significantly to this manuscript, participated in reviewing/editing, and approved the final draft for publication. The research profile of the authors can be verified from their ORCID IDs, given below:

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