



Synthesis and Evaluation of Novel Fluorescent Dyes using Microwave Irradiation

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Available online at: www.isca.in

Received 29th September 2012, revised 5th October 2012, accepted 6th November 2012

Abstract

Some new fluorescent dyes were prepared using microwave irradiation. Acetyl coumarine were synthesized via reaction of ethyl acetate with salicylaldehyde. The resultant coumarine under go further reaction with different aldehydes to produce a new series of coumarines derivatives which based on chalcone moiety. The structures of the synthesized dyes have been investigated using element analysis, IR –spectra, ¹H- NMR, and mass spectra. The using of the synthesized fluorescent dyes in textile printing of polyester and polyamide fabrics was investigated.

Keywords: Microwave irradiation, coumarine, chalcone, textile printing, polyester, polyamide.

Introduction

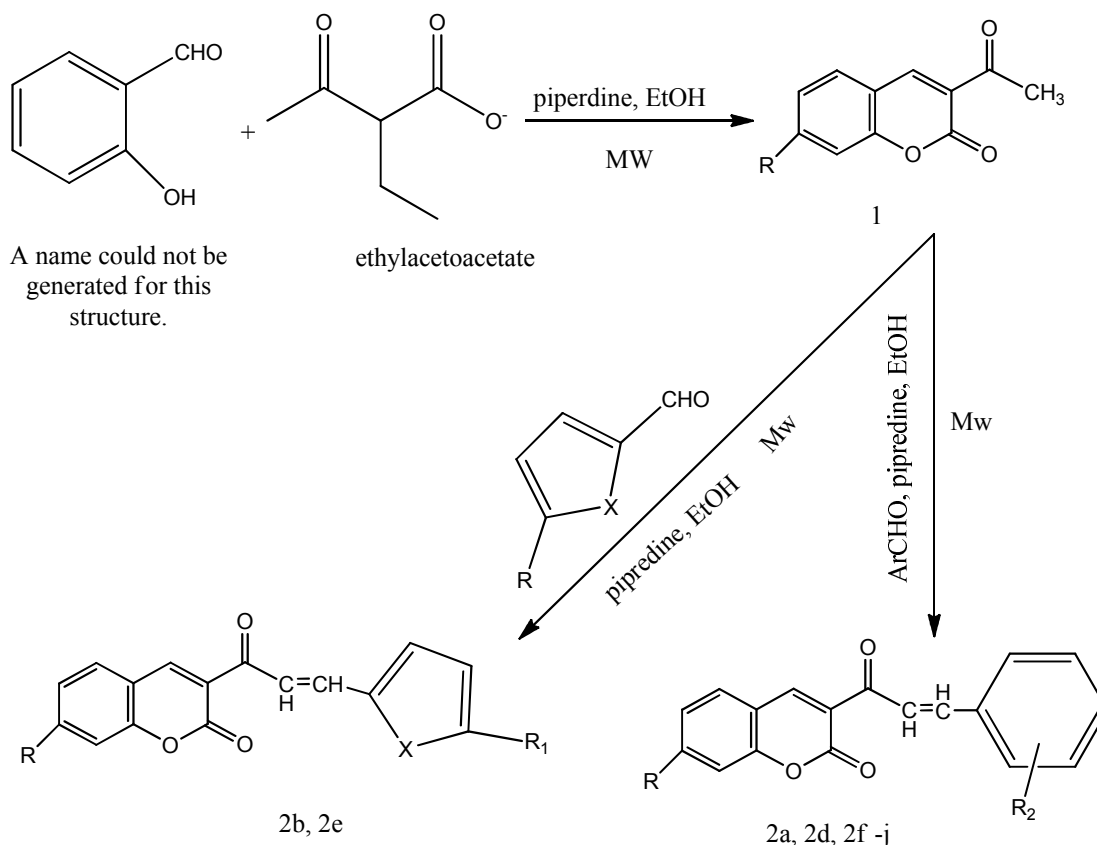
The utilization of green chemistry techniques is dramatically reducing chemical waste and reaction times as has recently been proven in several organic syntheses and chemical transformations. The coumarins are heterocyclic organic compounds, also known as benzo-2-pyrone derivatives and constitute an important group of natural products having varied activities. Coumarin derivatives provide a range of organic materials which owe their commercial exploitation in a broad range of applications to their intense fluorescence. They are of special interest as yellowish-green fluorescent dyes. The most important coumarin fluorescent dyes contain benzothiazole (e.g., coumarin 6, 1a), benzimidazole (e.g., coumarin 7, 1b) or benzoxazole substituent in the 3-position. Initially these dyes were developed for the colouration of synthetic fibres such as polyester, and subsequently their uses have extended to include daylight fluorescent pigments, and functional applications such as dye lasers, solar collector systems, organic light emitting diodes (LED) and numerous biological applications¹⁻⁷. Fluorescent chromophores have been generally known to have planar and rigid π -conjugation systems, and many fluorescent chromophores have rigid ring systems such as coumarin, naphthalimide, perylene and rhodamine 2001). It is a functional group in a molecule which will absorb energy of a specific wavelength and re-emit energy at a different wavelength. The most striking example of fluorescence occur when it absorbed radiation in the ultraviolet region of the spectrum that invisible to human eye and then re-emits the light is in the visible region. The application of fluorescent dyes involves various differentiated fields. In medical diagnostic and biochemical investigations it has developed extremely dynamically. Fluorescent dyes have also been used for the construction of lasers, testing material defects and equipment damage. In textile, fluorescent dyes suitable for synthetic fibers are applied

especially when aesthetic or functional reasons are required. Examples of such dyes are disperse dyes which contain the 7-position of the coumarin ring an electron releasing group such as N, N-diethylamino group and in the 3-position, a heterocyclic electron-acceptor residue. As more fluorescent dyes with different colors are successfully synthesized, the application of fluorescent dyes in textiles is becoming more and more fashionable. Some researchers presented investigations and analysis results of fluorescent disperse dyes used for dyeing polyester fibers to high-visibility colors. Disperse dye as the main colorant for polyester has high performance in hue, brilliant, and color strength⁸. The present study is aimed to synthesized of a series of coumarin-based chromophores containing chalcone moiety using microwave irradiation, where hand, chalcones, the bichromophoric molecules separated by vinyl chains and the carbonyl group, are found to be effective photosensitive materials, and exhibit promising nonlinear optical properties⁹.

Material and Methods

Synthesis of fluorescent dyes using microwave irradiation:

To a mixture of 3-acetyl coumarin (2 mmol) and appropriate aryl and heteroaryl aldehydes (2 mmol) in ethanol (30 mL) in a flask, piperidine (0.3 mL) in ethanol (5 mL) was added drop wise. The mixture was irradiated in a microwave oven at the power of 1500 w for an optimized time and taken out for a few seconds. And at the same time, the mixture was stirred carefully using a bar. The mixture was subjected to microwave irradiation for another optimized time and then taken out for stirring. The operation was repeated several times, and the reaction mixture was monitored by TLC. After the reaction was completed, the reaction mixture was cooled to room temperature. The precipitate was filtrated and recrystallized from EtOH (scheme 1)



Dye no.	R	X	R ₂
2a	H		4- OCH ₃
2b	H	S	
2c	H		2,4 di- OCH ₃
2d	H		2,4 di- NO ₂
2e	H	O	4- OCH ₃
2f	H		2- OH
2g	H		2- OH,-4-N,N-diethyl
2h	4-N,N-diethyl		4-Br
2i	H		3- OCH ₃ ,4- OH
2j	4-N,N-diethyl		4- NO ₂

Scheme-1
 Synthesis of new fluorescent dyes

Printing experiment: Lab scale printing experiments were carried out on polyester and polyamide fabrics using the produced dyes. The printing paste 100 gm. consists of 3gm. of Alko print as synthetic thickener, 3 g. of prepared dye and 96 ml of water. Samples of fabrics were silk screen printed using the above printing paste, the printed fabric was dried and fixed at different temperatures 150, 170, and 190°C, fixation time 2 min. the printed sample were washed twice by cold water, then twice with hot water and finally rinsed with cold water, then air dried.

Measurements: Melting point: Melting points were taken on Electro thermal IA 9000 series digital melting point apparatus.

Element analysis: Elemental analytical data were obtained from the micro analytical unit, National Research Centre, Dokki, Giza, Egypt.

IR spectra: The IR spectra (KBr) were recorded on a Shimadzu CVT-04 spectrophotometer. The infra red of the synthesized dyes was measured using a Perkin Elmer/1650.FT-IR instrument

¹H – NMR spectra: The NMR spectra were recorded on a Varian Mercury VX-300 NMR spectrometer. ¹H spectra were run at 300 MHz in deuterated dimethylsulphoxide (DMSO-*d*₆).

Mass spectra: Mass spectra were measured on a Varian MAT CH-5 spectrometer (70 eV).

Colour strength measurements: The reflectance values of the fabric were measured using a Data Colour SF 600+. Relative colour strengths (K/S values) were determined using the Kubelka-Munk equation¹⁰.

$$K/S = (1-R)^2 / 2R$$

Fastness properties measurements: Fastness to washing, rubbing, light and perspiration were assessed according to standard methods¹¹⁻¹⁴.

Spectrophotometric measurements: The absorbance of the dyes was measured in the ultraviolet-visible region between 300-700 nm by a UNICAM UV spectrophotometer using a 1cm. quartz cell. The dyes were dissolved in absolute ethanol at a concentration of 10⁻⁴ mole/l.

Results and Discussion

Synthesis of new fluorescent dyes: The required 3-acetyl coumarins 1 were prepared from substituted salicylaldehyde with ethyl acetoacetate according to the method reported in literature¹⁵. Coumarin derivatives 2 were readily synthesized by refluxing 3-acetyl coumarins with various aryl or heteroaryl aldehydes in the presence of piperidine in ethanol. The same condensation between 3-acetyl coumarin and aldehydes could be carried out under microwave conditions. It is noteworthy that the reaction which required 3-5 h. in conventional method was completed efficiently, with 78-92% yields, in 2-10 min under microwave conditions (scheme 1). All the synthesized compounds (2a-2k) have been characterized on the basis of their physical data and spectral analysis. These compounds show the band at 1706-1734 cm⁻¹ for typical lactone carbonyl group. All the compounds show the ¹H-NMR signals for different kinds of protons at their respective positions. The values are consistent with their predicted structures, mass spectra (scheme 1).

3-(3-(4-methoxybenzene)prop-2-enoyl)-2H-1-benzopyran-2-one (2a): yellow crystals, m.p. 220-222°C; yield: (90%); IR (KBr), 2958 (CH=CH), 1728(C=O), cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 3.50 (s, 3H, OCH₃), 6.6-7.7. (m, 4H, Ar-H), MS (70 eV): *m/z* = 304 (M⁺, 000). Analysis for C₁₉H₁₄O₄ (305) Calc.: C, 74.50; H, 4.6; O, 20.89. found: C, 74.3; H, 4.13; O, 20.85.

3-(3-(thiophene)prop-2-enoyl)-2H-1-benzopyran-2-one (2b): yellow crystals, m.p. 220-222°C; yield: (90%); IR (KBr), 2936 (CH=CH), 1718(C=O), cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 6.6-7.7. (m, 7H, Ar-H), MS (70 eV): *m/z* = 281 (M⁺, 281). Analysis for C₁₆H₁₀O₃S (282) Calc.: C, 68.07; H, 3.57; S, 11.36. found: C, 68.0; H, 3.53; S, 11.31

3-(3-(2,5 dimethoxybenzene)prop-2-enoyl)-2H-1-benzopyran-2-one (2c): yellow crystals, m.p. 220-222°C; yield: (90%); IR

(KBr), 3075 (CH=CH), 1728(C=O), cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 3.50 (s, 6H, 2OCH₃), 6.6-7.7. (m, 4H, Ar-H), MS (70 eV): *m/z* = 337 (M⁺, 336). Analysis for C₂₀H₁₆O₅ (336) Calc.: C, 71.42; H, 4.79; O, 23.87. found: C, 71.39; H, 4.73; O, 23.55

3-(3-(2,4-dinitrobenzene)prop-2-enoyl)-2H-1-benzopyran-2-one (2d): yellow crystals, m.p. 220-222°C; yield: (90%); IR (KBr), 2928 (CH=CH), 1731(C=O), cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 6.6-7.7. (m, 8H, Ar-H), MS (70 eV): *m/z* = 366 (M⁺, 365). Analysis for C₁₈H₁₀N₂O₇ (366) Calc.: C, 59.02; H, 2.75; N, 7.65; O, 30.58. found: C, 59.0; H, 2.70; N, 7.55; O, 30.50

3-(3-(4-methoxyfuran)prop-2-enoyl)-2H-1-benzopyran-2-one (2e): yellow crystals, m.p. 220-222°C; yield: (90%); IR (KBr), 2917 (CH=CH), 1720(C=O), cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 3.50 (s, 3H, OCH₃), 6.6-7.7. (m, 8H, Ar-H), MS (70 eV): *m/z* = 296 (M⁺, 295). Analysis for C₁₇H₁₂O₅ (296) Calc.: C, 68.92; H, 4.08; O, 27.0; found: C, 68.81; H, 4.0; O, 26.85

3-(3-(2-hydroxybenzene)prop-2-enoyl)-2H-1-benzopyran-2-one (2f): yellow crystals, m.p. 220-222°C; yield: (90%); IR (KBr), 2917 (CH=CH), 1736(C=O), cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 6.6-7.7. (m, 9H, Ar-H), MS (70 eV): *m/z* = 365 (M⁺, 364). Analysis for C₂₂H₂₁NO₄ (365) Calc.: C, 72.71; H, 5.82; N, 2.85; O, 17.61. found: C, 72.69; H, 5.73; N, 2.74; O, 17.57

3-(3-(2-hydroxy-4-N,N-diethylaminobenzene)prop-2-enoyl)-2H-1-benzopyran-2-one (2g): yellow crystals, m.p. 220-222°C; yield: (90%); IR (KBr), 2937 (CH=CH), 1726(C=O), cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 6.6-7.7. (m, 9H, Ar-H), MS (70 eV): *m/z* = 365 (M⁺, 364). Analysis for C₂₂H₂₁NO₄ (365) Calc.: C, 72.71; H, 5.82; N, 2.85; O, 17.61. found: C, 72.50; H, 5.70; N, 2.80; O, 17.53

3-(3-(2-bromoene)prop-2-enoyl)-2H-1-benzopyran-2-one (2h): yellow crystals, m.p. 220-222°C; yield: (90%); IR (KBr), 2937 (CH=CH), 1726(C=O), cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 6.6-7.7. (m, 9H, Ar-H), MS (70 eV): *m/z* = 355 (M⁺, 354). Analysis for C₁₈H₁₁BrO₃(355) Calc.: C, 60.87; H, 3.12; Br, 21.82; O, 13.51. found: C, 60.80; H, 3.09; NBr, 21.75; O, 13.48

3-(3-(3-methoxy-4-hydroxybenzene)prop-2-enoyl)-2H-1-benzopyran-2-one (2i): yellow crystals, m.p. 220-222°C; yield: (90%); IR (KBr), 2937 (CH=CH), 1726(C=O), cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 3.50 (s, 3H, OCH₃), 6.6-7.7. (m, 9H, Ar-H), MS (70 eV): *m/z* = 322 (M⁺, 321). Analysis for C₁₉H₁₄O₅ (322) Calc.: C, 70.80; H, 5.14; O, 24.82. found: C, 70.84; H, 5.10; O, 24.77

7-(diethylamino)-3-(3-(4-nitrophenyl)acryloyl)-2H-chromen-2-one (2j): yellow crystals, m.p. 220-222°C; yield: (90%); IR (KBr), 2937 (CH=CH), 1726(C=O), cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 3.50 (s, 3H, OCH₃), 6.6-7.7. (m, 9H, Ar-H), MS (70 eV): *m/z* = 437 (M⁺, 436). Analysis for C₂₂H₂₀N₂O₅(437) Calc.: C, 67.34; H, 5.14; N, 7.14; O, 20.39 found: C, 67.29; H, 5.11; N, 7.10

Absorption and fluorescence spectra: The molecular structures of the coumarinyl-based organic molecules investigated are shown in figure 1-10. These molecules consist of a typical A- π -D structure, where 3-carbonylcoumarinyl, vinyl and (thiophen-2-yl) (or aryl, substituted aryl, 5-substituted hetero aryl) groups are employed as acceptor (A), π -conjugated center (π) and donor (D) moieties, respectively. As seen from figure 1-10, structural modification occurs only in one terminal moiety. Such a modification could be expected to result in notable changes in the π -conjugated length and red-shifts in the absorption and emission spectra.

UV-vis absorption spectra of these molecules in diluted ethanol solutions are given in figures 1-10. The results show that the compounds comprise 3-5 bands in the wavelength range from 270 to 600 nm, depending on their molecular structures. There is no reasonable linear absorption in the entire spectral range above 600 nm. The first absorption band of compounds in the region 430-473 nm can be assigned to π - π transition involving the whole electronic system of the compounds with a considerable charge-transfer character originating mainly from the aryl or hearty moieties and pointing towards the coumarin ring which is characterized by a high electron-accepting character.

Fastness properties and color strength: The fastness properties and color strength values of silk screen printed polyester and polyamide fabrics using the newly synthesized

fluorescence dyes (2a-j) are represented by tables 1 and 2. The data shows that excellent rubbing, washing and perspiration fastness. The excellent rubbing fastness results indicate deeper penetration of dyes in the fabric. The light fastness results range from good to very good.

The molecular size of dye molecule plays an important role in the rate of movement of the dye out of the fabric during washing so, it is seen from table 1, 2 that the result of washing fastness of polyester and polyamide printed fabrics including alteration and staining on cotton fabric, indicate values 4-5 i.e. from very good to excellent.

Color strength: The effect of the screen printing dye fixation conditions (fixation temperature 150, 170, and 190°C and fixation time 2 min.) on the color strength (K/S) value using the synthesized dyes 2a-j to print polyester and polyamide fabrics are shown by figure 11 and 12. It can be seen that, the increase in the temperature of fixation from 150°C to 190°C leads to increase in the color strength over all the fabrics or the dyes. It can be seen also that, the dye 2j possesses highest color strength value and this may be attributed to the molecular structure of the dye which contains a nitro group (electron withdrawing group) attached to the aryl moiety and N, N-diethyl group attached to the coumarin moiety, which result in notable changes in the π -conjugated length and red-shifts in the absorption and emission spectra.

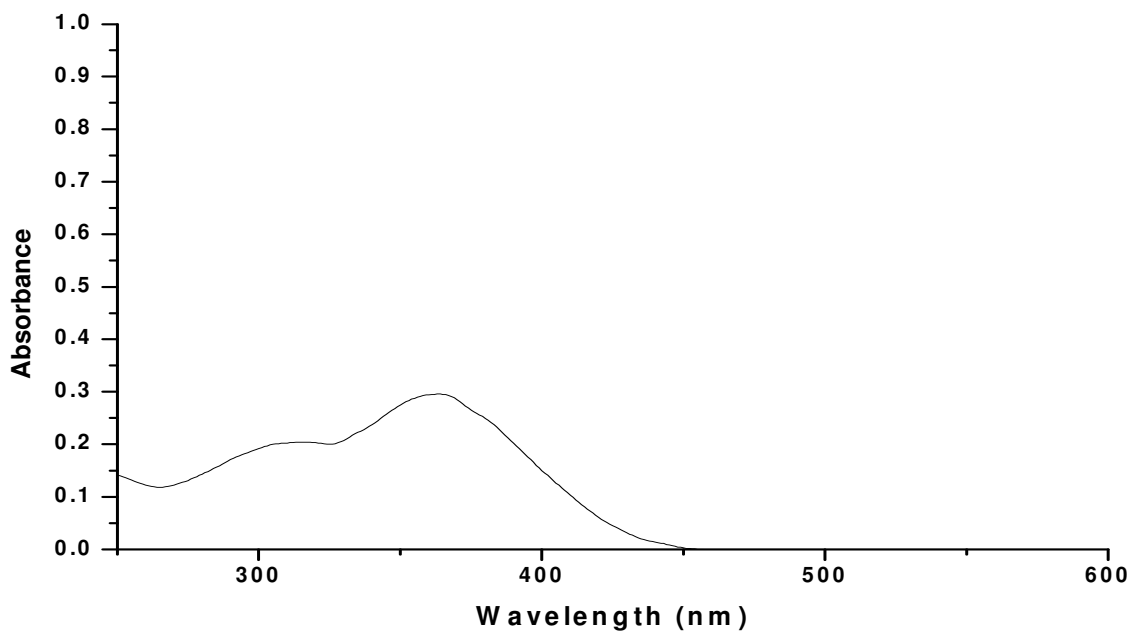


Figure-1
UV-vis absorption spectra of (2a), in dilute ethanol solution ($C=1 \times 10^{-6}$ M)

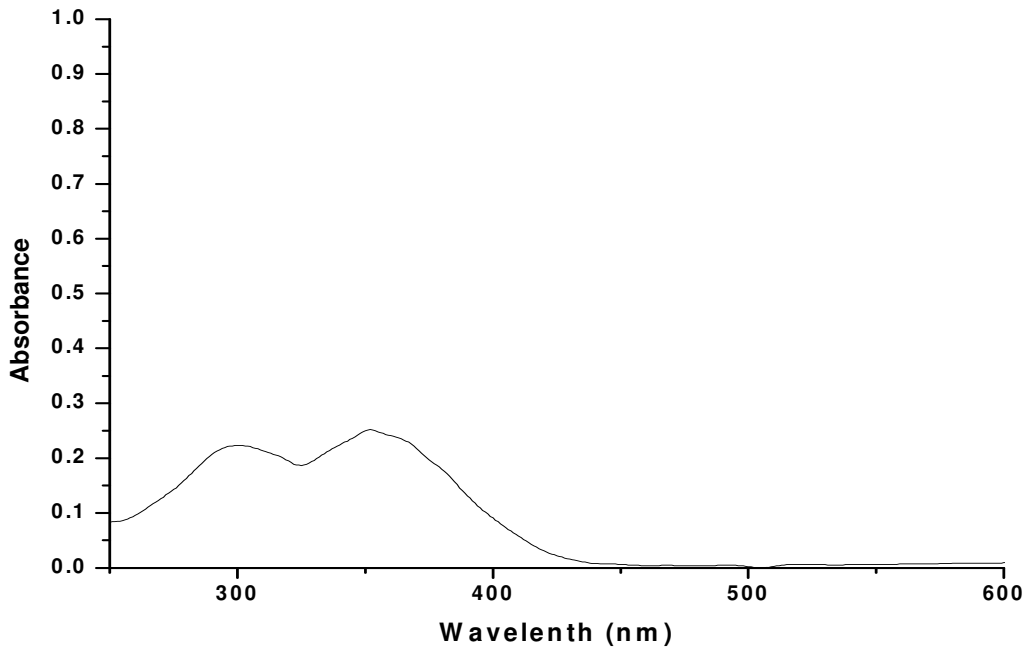


Figure-2
UV-vis absorption spectra of (2b), in dilute ethanol solution ($C=1 \times 10^{-6}$ M)

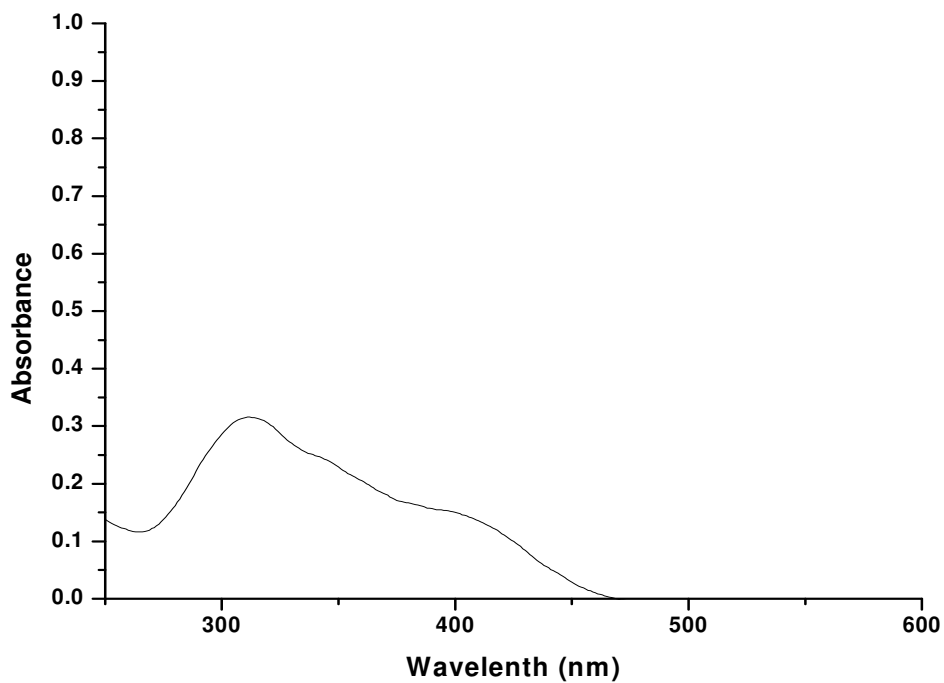


Figure-3
UV-vis absorption spectra of (2c), in dilute ethanol solutions ($C=1 \times 10^{-6}$ M)

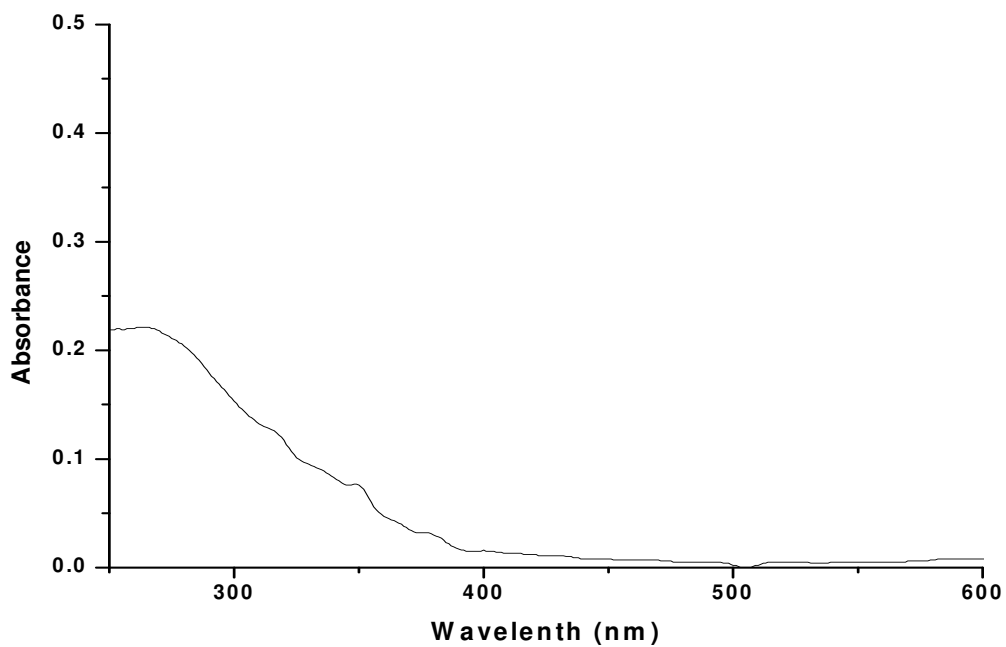


Figure-4
UV-vis absorption spectra of (2d), in dilute ethanol solution ($C=1 \times 10^{-6}$ M)

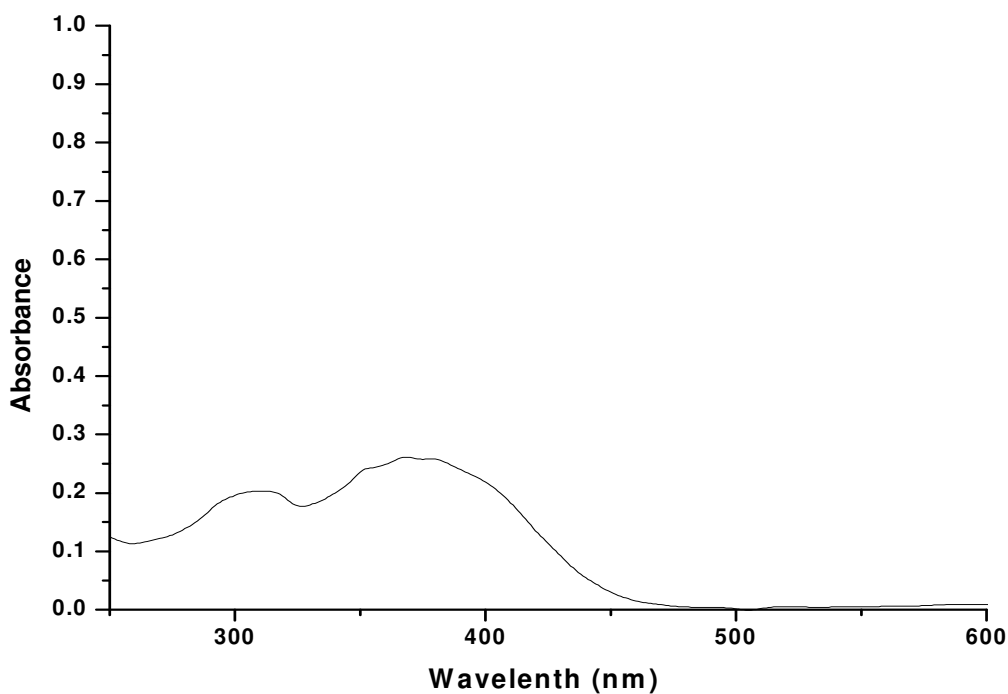


Figure-5
UV-vis absorption spectra of (2e), in dilute ethanol solution ($C=1 \times 10^{-6}$ M)

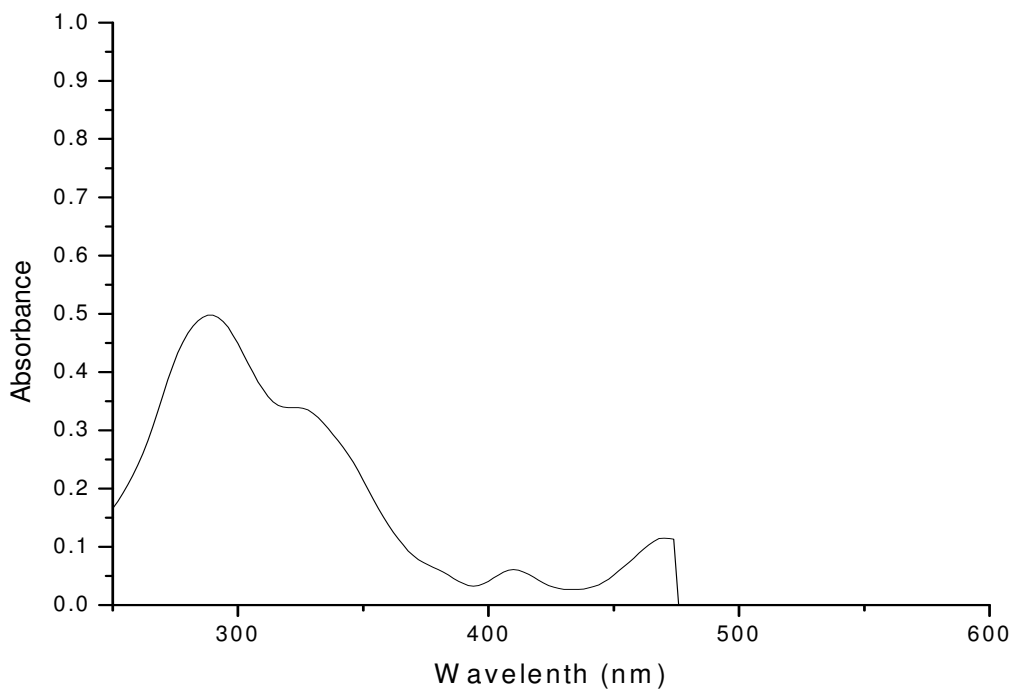


Figure-6
UV-vis absorption spectra of (2f), in dilute ethanol solution ($C=1 \times 10^{-6}$ M)

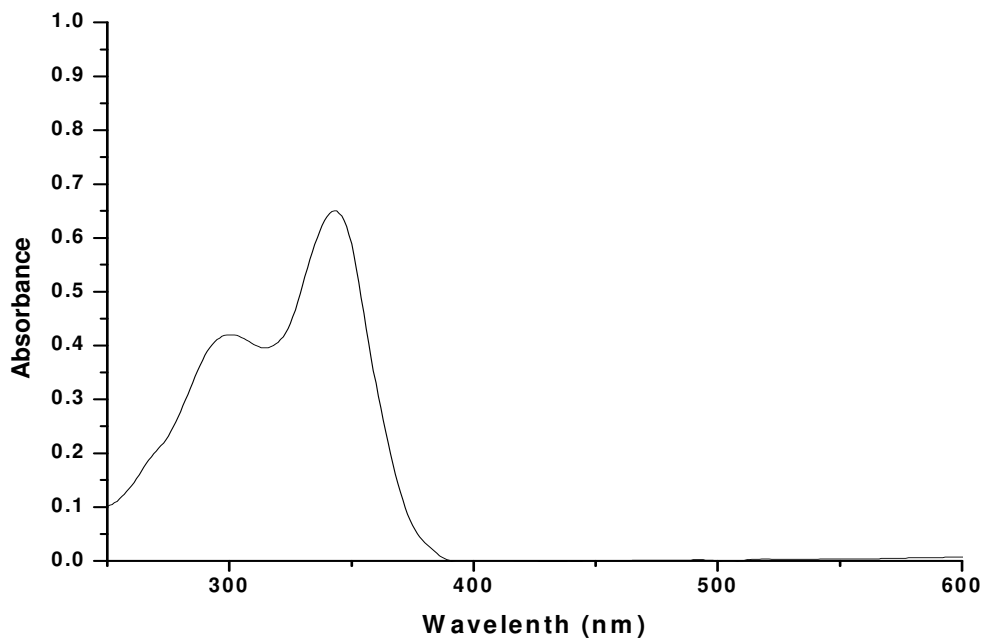


Figure-7
UV-vis absorption spectra of (2g), in dilute ethanol solution ($C=1 \times 10^{-6}$ M)

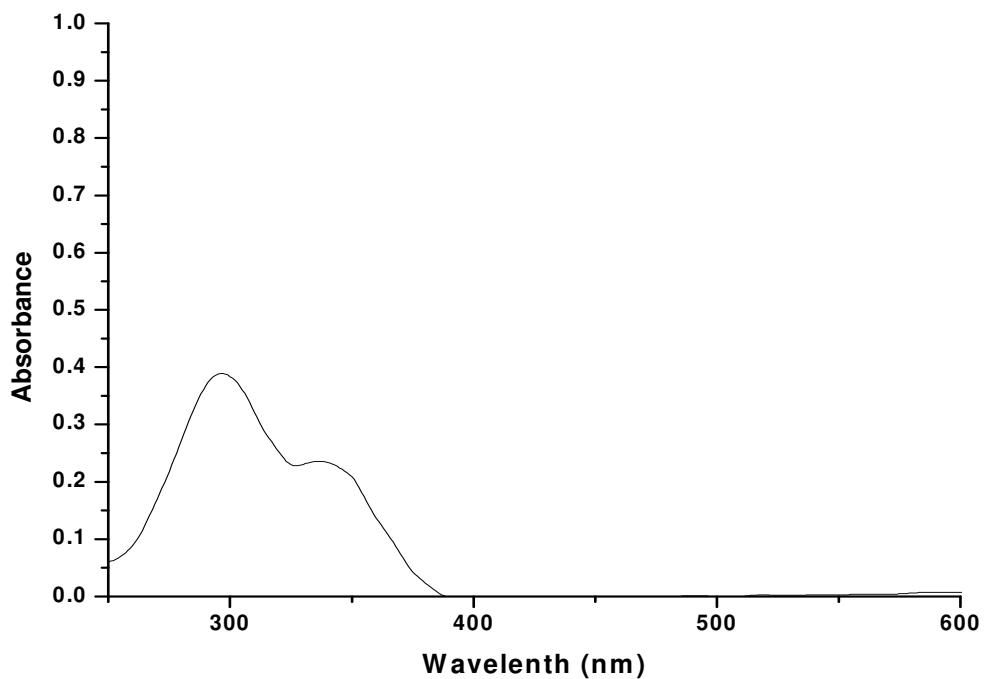


Figure-8
UV-vis absorption spectra of (2h), in dilute ethanol solution ($C=1 \times 10^{-6}$ M)

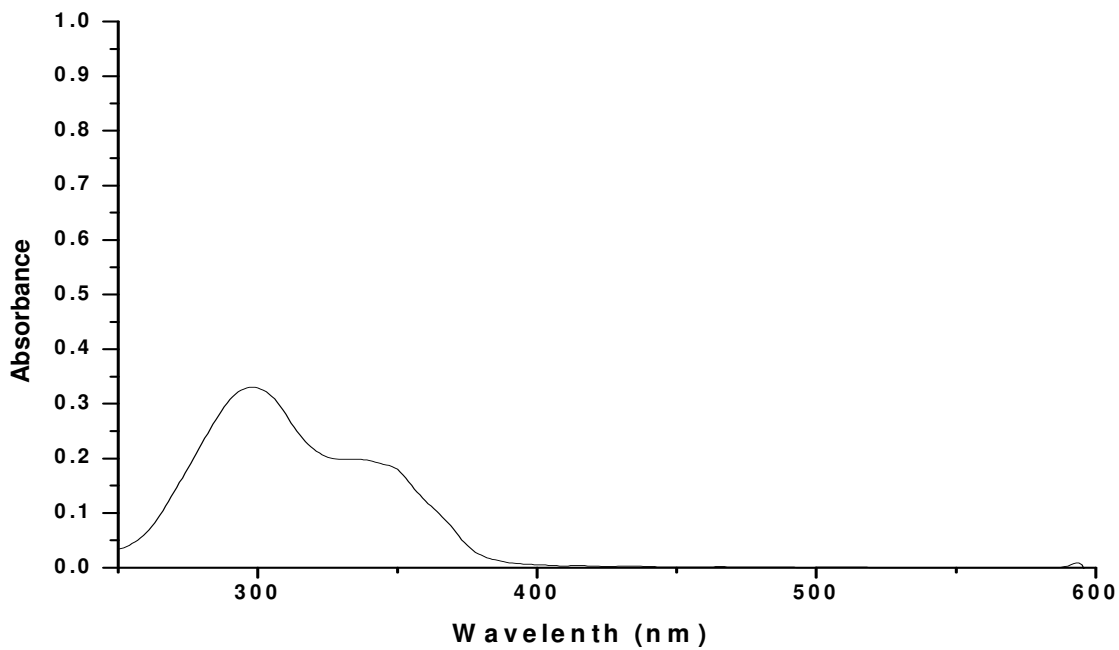


Figure-9
UV-vis absorption spectra of (2i), in dilute ethanol solution ($C=1 \times 10^{-6}$ M)

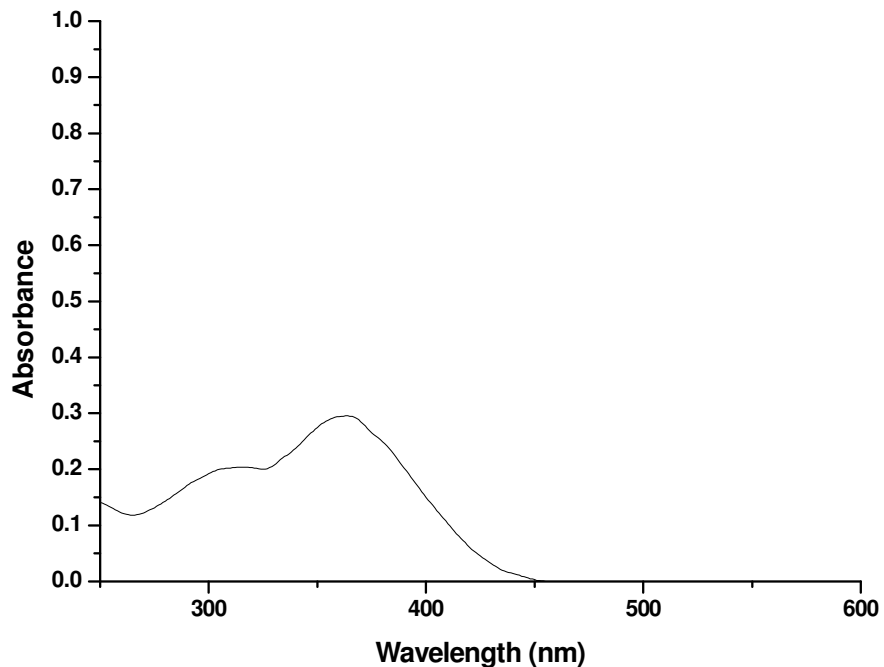


Figure-10
UV-vis absorption spectra of (2j), in dilute ethanol solution ($C=1 \times 10^{-6}$ M)

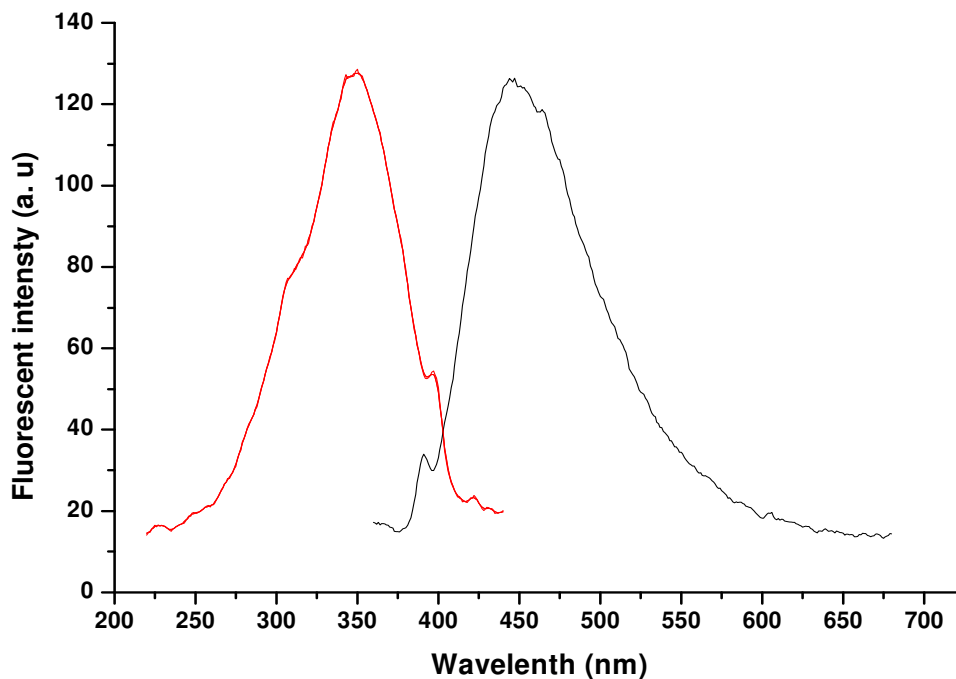


Figure-11
Excitation and emission spectra of (2a), in dilute ethanol solution at room temperature
($C=1 \times 10^{-6}$ M, $\lambda_{ex}=350$ nm, $\lambda_{em}=450$ nm)

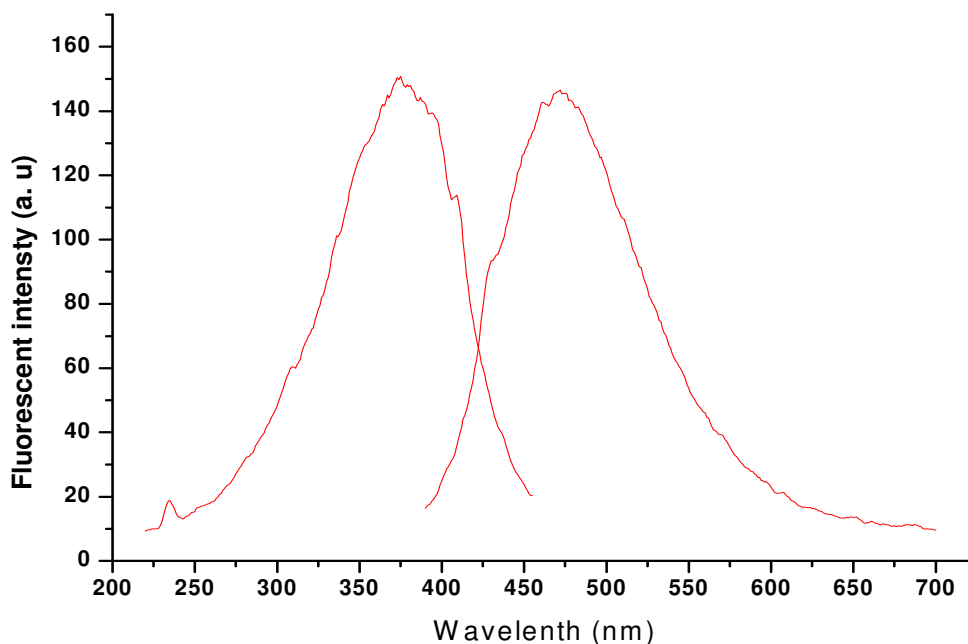


Figure-12
Excitation and emission spectra of (2b), in dilute ethanol solution at room temperature
($C=1 \times 10^{-6}$ M , $\lambda_{ex}=380$ nm , $\lambda_{em}=465$ nm)

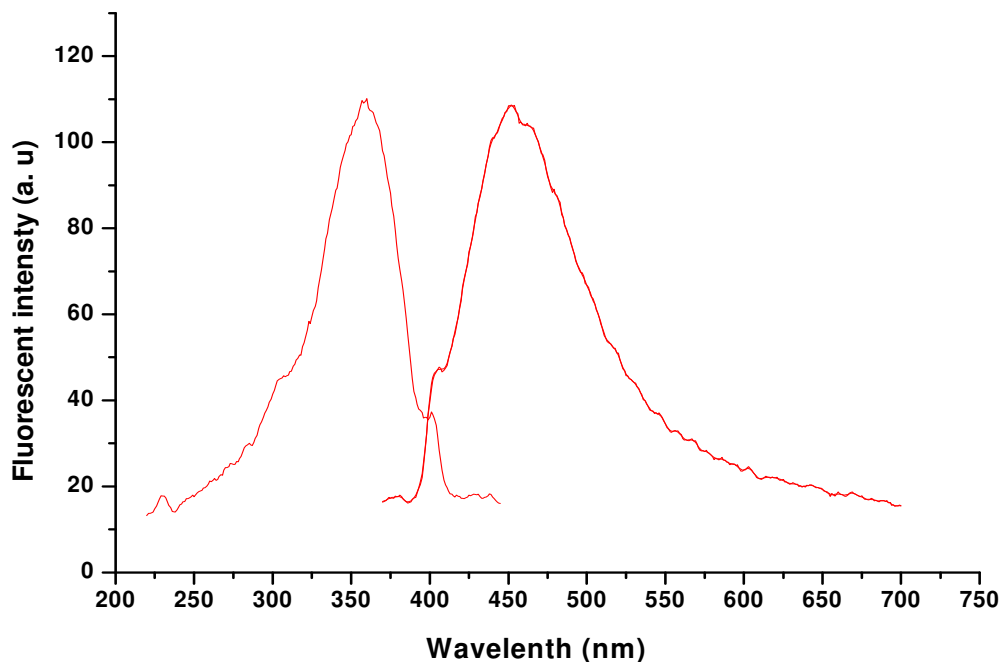


Figure-13
Excitation and emission spectra of (2c), in dilute ethanol solution at room temperature
($C=1 \times 10^{-6}$ M , $\lambda_{ex}=360$ nm , $\lambda_{em}=455$ nm)

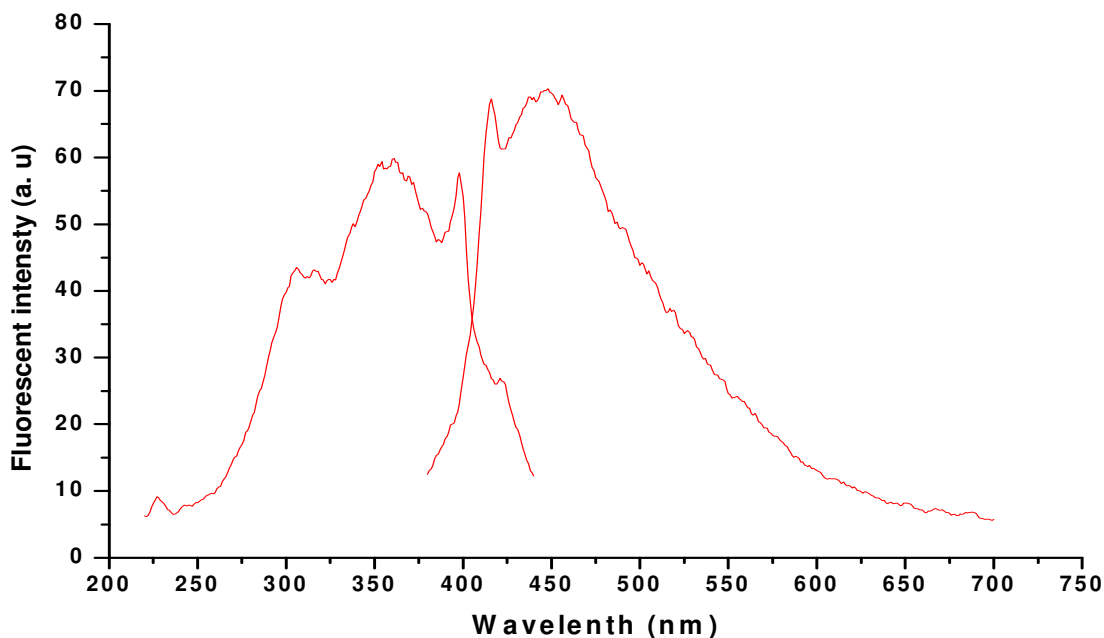


Figure-14
Excitation and emission spectra of (2d), in dilute ethanol solution at room temperature
($C=1 \times 10^{-6}$ M, $\lambda_{ex}=370$ nm, $\lambda_{em}=440$ nm)

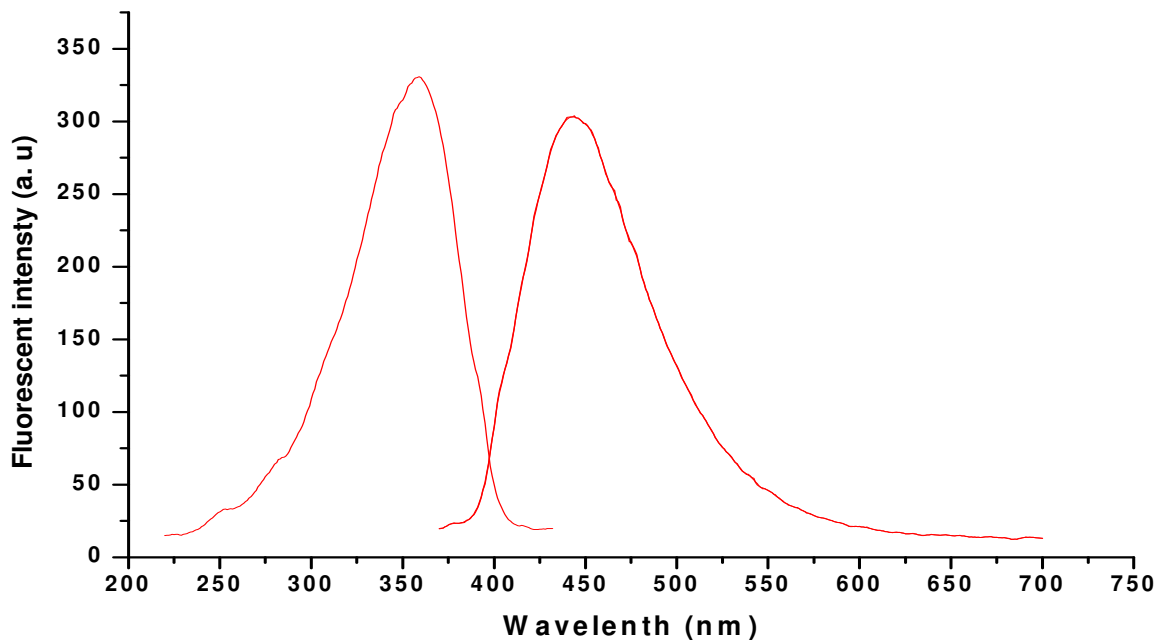


Figure-15
Excitation and emission spectra of (2e), in dilute ethanol solution at room temperature
($C=1 \times 10^{-6}$ M, $\lambda_{ex}=350$ nm, $\lambda_{em}=440$ nm)

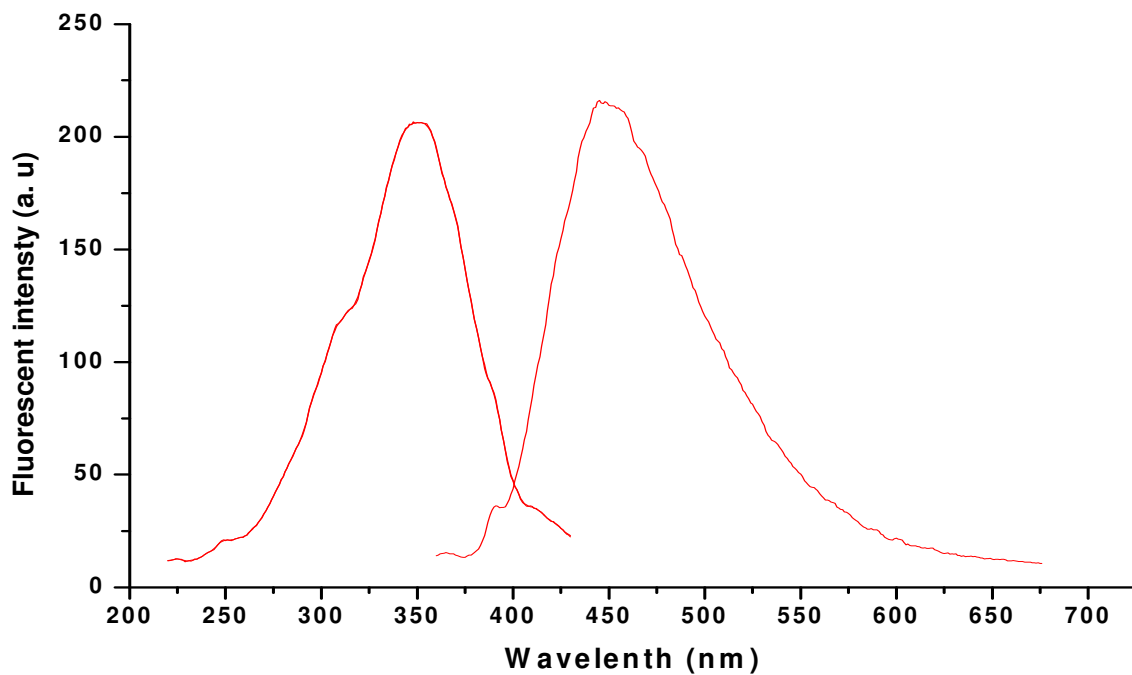


Figure-16
Excitation and emission spectra of (2f), in dilute ethanol solution at room temperature
($C=1 \times 10^{-6}$ M, $\lambda_{ex}=360$ nm, $\lambda_{em}=450$ nm)

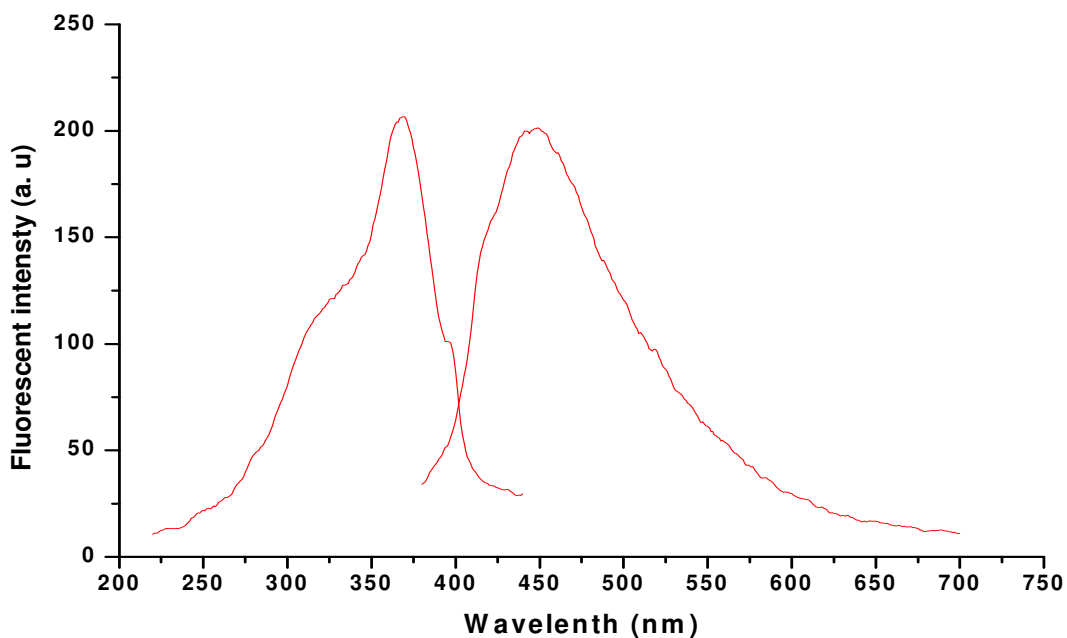


Figure-17
Excitation and emission spectra of (2g), in dilute ethanol solution at room temperature
($C=1 \times 10^{-6}$ M, $\lambda_{ex}=370$ nm, $\lambda_{em}=450$ nm)

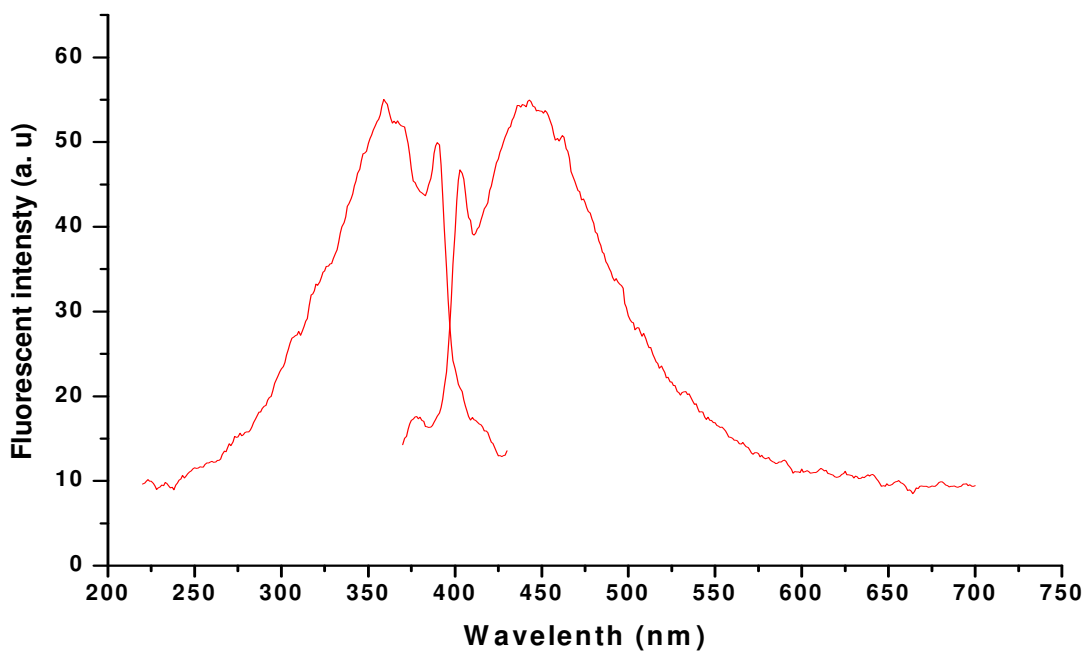


Figure-18
Excitation and emission spectra of (2h), in dilute ethanol solution at room temperature
($C=1 \times 10^{-6}$ M, $\lambda_{ex}=360$ nm, $\lambda_{em}=440$ nm)

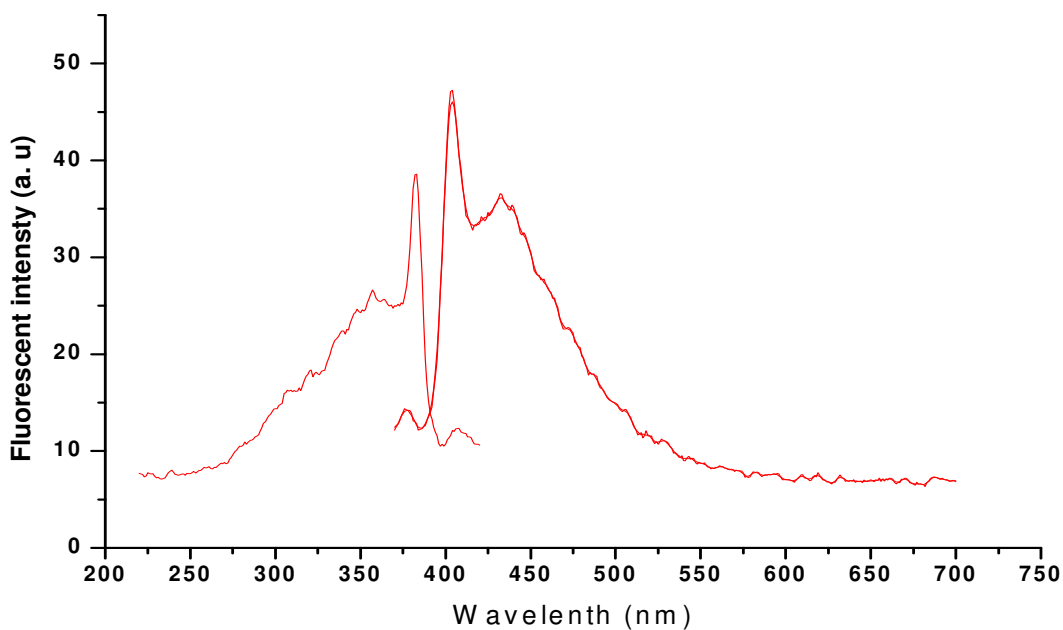


Figure-19
Excitation and emission spectra of (2i), in dilute ethanol solution at room temperature
($C=1 \times 10^{-6}$ M, $\lambda_{ex}=370$ nm, $\lambda_{em}=430$ nm)

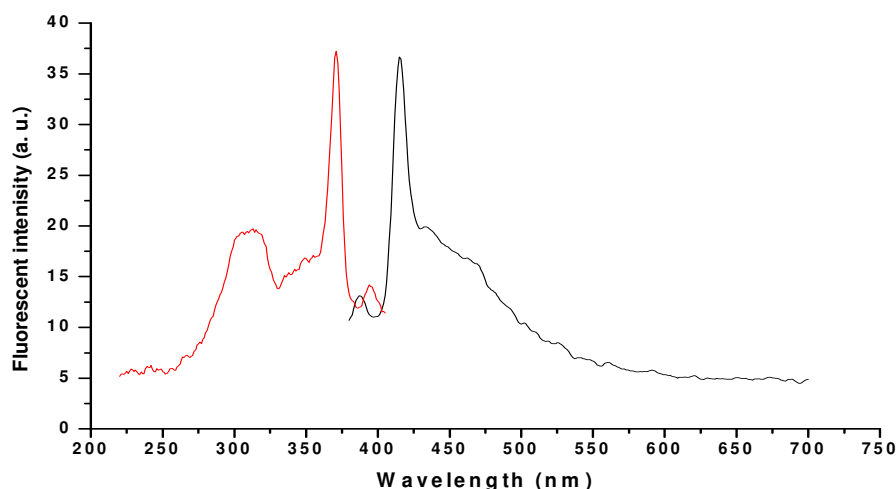


Figure-20
Excitation and emission spectra of (2j), in dilute ethanol solution at room temperature
($C=1 \times 10^{-6}$ M, $\lambda_{ex}=370$ nm, $\lambda_{em}=415$ nm)

Table - 1
Fastness properties and color strength of screen printed *polyester fabric using the newly synthesized dyes (2a-k)

Dye No.	K/S	Rubbing		washing		perspiration				Light fastness
		Dry	Wet	Alt.	St.	Alkali		Acid		
						Alt.	St.	Alt.	St.	
2a	15.1	4-5	4	4-5	5	4-5	5	5	5	3-4
2b	15.3	5	4-5	5-4	5	5	5	5	5	5
2c	13.4	5	4-5	5	5	4-5	5	5	5	4
2d	10.1	4	4	5	5	5	5	5	5	4-5
2e	17.3	5	4-5	5	5	5	5	5	5	5-6
2f	16.2	5	5	5	5	5	5	5	5	5
2g	15.54	4	3-4	4	5	4-5	5	5	5	4-5
2h	7.9	4	3-4	4-5	5	5	5	5	5	6
2i	7.9	4	4-5	5	5	5	5	5	5	5-6
2j	5.7	4	4	4	4-5	5	5	5	5	5

*Fixation temperature 190°C, Fixation time 2 min.

Table - 2
Fastness properties and color strength of screen printed *polyamide fabric using the newly synthesized dyes (2a-k)

Dye No.	K/S	Rubbing		washing		perspiration				Light fastness
		Dry	Wet	Alt.	St.	Alkali		Acid		
						Alt.	St.	Alt.	St.	
2a	18.8	4-5	4	4-5	5	4-5	5	5	5	3-4
2b	19.2	4	4-5	5-4	5	5	5	5	5	5
2c	16.8	4	4-5	5	5	4-5	5	5	5	4
2d	13.22	4	4	5	5	5	5	5	5	4-5
2e	21.91	4	4-5	5	5	5	5	5	5	5-6
2f	21	4	5	5	5	5	5	5	5	5
2g	20.2	3-4	3	4	5	4-5	5	5	5	4-5
2h	12.9	4	3-4	4-5	5	5	5	5	5	6
2i	12.11	4	3-4	5	5	5	5	5	5	5-6
2j	11.65	4	3-4	4	4-5	5	5	5	5	6

*Fixation temperature 190°C, Fixation time 2 min.

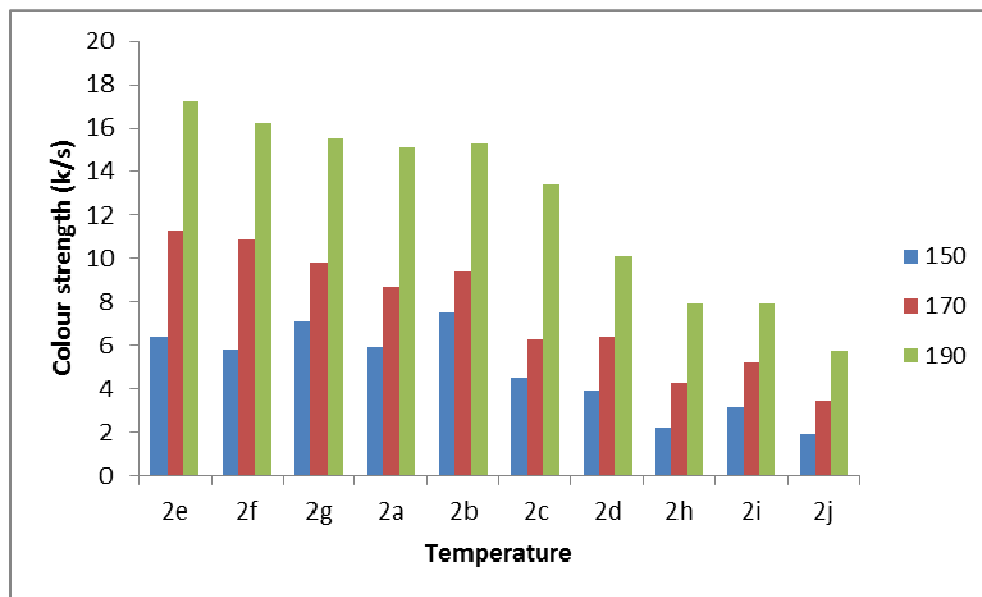


Figure-21

Effect of silk screen printing temperature on the color strength of screen printed polyester fabric using dyes 2a-j, Fixation time 2 min

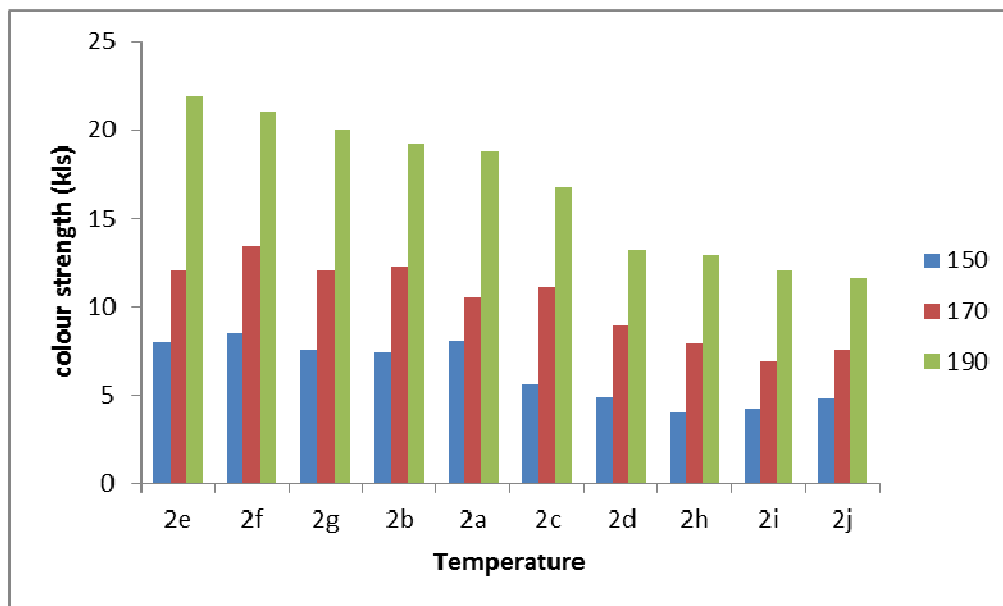


Figure-22

Effect of silk screen printing temperature on the color strength of screen printed polyamide fabric using dyes 2a-j, Fixation time 2 min

Conclusion

The synthesis and characterization for coumarin-based chromophores containing chalcone moiety are presented in this paper. The absorption and emission spectra of selected coumarin derivatives were studied in solution. It was found that the absorption and emission spectra show red-shift according to the strength of the electron-donating moieties and conjugation length. Replacement of carbazolyl donor with a hetaryl or

substituted hetaryl group in the coumarinyl-based chromophores resulted in a strong bathochromic shift. The prepared coumarinyl-based chromophores in the printing of polyester fabric using silk screen printing technique. These coumarinyl-based chromophores could be potential optical materials for some fields, such as OLED materials, two-photon absorption materials, as well as fluorescent probes in biological applications.

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