



Synthesis of biscoumarin bifunctional reactive fluorescent whitening agents and their application on nylon-6 fabric

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Received 25 November 2020; revised received and accepted 12 April 2021

Two new bifunctional reactive fluorescent whitening agents based on joining two coumarin moieties together through triazinylstilbene fluorescent moiety have been successfully synthesized, applied on nylon-6 fabrics and then characterized by NMR and elemental analysis. Spectral properties, such as absorption and emission spectra in solution and on fabrics are investigated. The application of all the prepared compounds as fluorescence [fluorescence whitening agents (FWAs)] and their whiteness as well as light fastness properties on nylon-6 fabric have been compared with monofunctional agent. Despite the fact that the FWAs, containing either mono or bis methyl coumarin derivatives, show better whiteness properties than the FWA containing phenyl coumarin derivative, the later compound shows higher light fastness (excellent).

Keywords: Coumarin, Fluorescent whitening agents, Stilbene, Whiteness properties, Nylon-6 fabric

1 Introduction

Fluorescent brightening agents (colorless dyes) are characterized by their ability to increase fabric brightness and reduce its yellowness, by absorbing ultra-violet radiation present in the light source and re-emitting it as visible blue light¹. The optical brighteners are built up from aromatic or heteroaromatic structures². Mostly they are derivatives of stilbene, coumarin, benzothiazole, pyrazolines, imidazolines, diazoles, triazoles and benzoxazolines²⁻⁴.

Triazinylstilbene fluorescent brighteners are considered to be the most widely used in the textile industry especially for cotton, viscose, linen and polyamides^{2,5}. However, the stilbenes have low photo resistance, which may cause fluorescence loss, yellowing of the substrate in addition to increasing rate of dye fading⁵. Many attempts have been made to increase the triazinylstilbene photostability, by studying the influence of functional groups present^{1,6}. The heteroaromatic types like coumarins are attractive molecules due to their photostability, biological activities, extended spectral range and high emission quantum yields⁷⁻⁹. The presence of an electron-donating group at position 7 exhibits strong fluorescence. Amino coumarin derivatives considered as an important chromophoric system for different kind of applications such as fluorescent dyes for

synthetic polyamide fabrics², solar energy collectors^{10,11}, fluorescent probes^{12,13}, and laser dyes for the blue-green region¹⁴. Their photophysical properties are influenced significantly by the type and position of the substituents on the coumarin moiety^{9,15}. Recently, the synthesis of biscoumarins has been advanced for multiple biological activities and optical properties^{16,17}, through Knoevenagel–Michael reactions of 4-hydroxy coumarin with different aldehydes¹⁸. Moreover, the synthesis of symmetrical 3,3'-phenylene biscoumarin dyes have been investigated, to achieve high optical properties, these biscoumarin dyes could be applied as fluorescent probes, in a wide range of solvents¹⁶. Also, some biscoumarins could possess lasing properties¹⁹. The linking of two coumarin moieties extends the absorption and emission wavelength ranges, by increasing/expanding the electron conjugated system¹⁶.

The main objective of the present study is to introduce two coumarin moieties as functional group in triazinylstilbene moiety, in order to perform two new biscoumarin bis(monochlorotriazine) bifunctional reactive fluorescent whitening agents (FWA). The compounds have two 7-amino coumarin derivatives (substituted at 3 or 4 position). To the best of our knowledge, there is no study having the merit of joining two coumarin moieties using triazinylstilbene fluorescent moiety. They behave like bifunctional reactive colorless dyes due to the

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presence of bis(monochlorotriazine) reactive groups, their application properties are examined on nylon fabric and compared with the mono (coumarin) mono-functional reactive FWA.

2 Materials and Methods

Methyl chloroformate (98%), 3-amino phenol, ethyl acetoacetate, 4,4'-diaminostilbene-2,2'-disulphonic sodium salt (96%), 7-amino-3-phenyl-2*H*-chromen-2-one were purchased from Sigma Aldrich (Germany) and used directly without any further purification. All solvents used (ethanol, dimethylformamide and acetone) were of spectroscopic grade. 100% nylon-6 fabric was supplied by El-Mahalla El-Kobra Company, Egypt. Thin-layer chromatography (TLC) was performed by a Kieselgel 60 F254 (Merck, Darmstadt, Germany), for observation of reaction progress and the purity of the prepared intermediates and synthesized FW agents.

¹HNMR spectra were recorded on a JEOL 500 MHz spectrometer (Japan) using TMS as an internal standard, and the chemical shift values are expressed in δ ppm and *J* values given in Hz. The UV/Vis spectra were measured on a Shimadzu UV-2401PC UV/Vis spectrophotometer (Shimadzu, Japan). The steady-state fluorescence spectra were measured using JASCO FP-8300 spectrophotometer. Instrumental whiteness index ((WI)₃₁₃) values were recorded using UltraScan PRO spectrophotometer (Hunter Lab, USA) under illuminant D65, 10° standard observer.

2.1 Synthesis of 7-Amino-4-Methyl-2*H*-Chromen-2-One

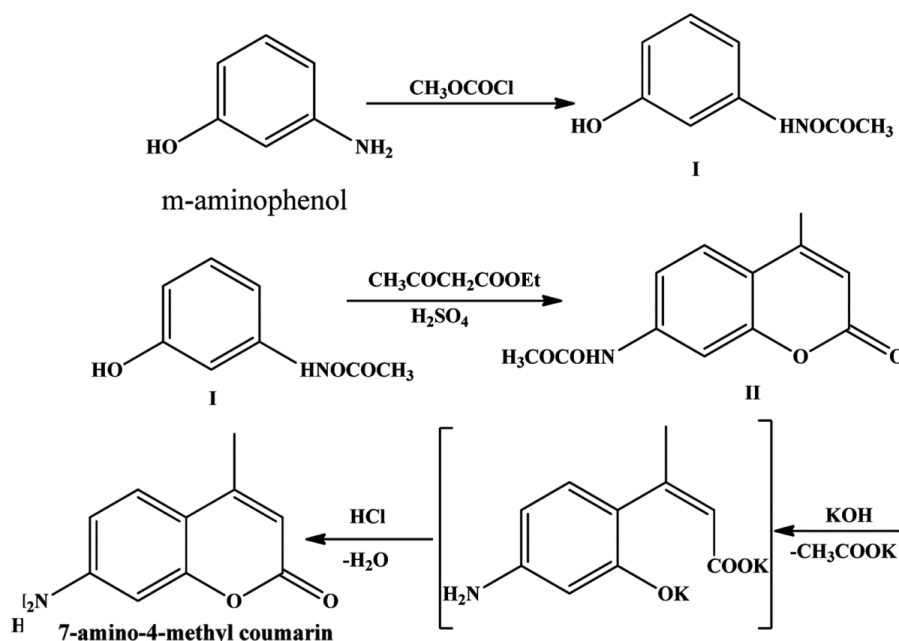
The starting intermediate, 7-amino-4-methyl-2*H*-chromen-2-one, can be synthesized by using two steps²⁰⁻²² (Scheme 1):

2.1.1 First Step— Synthesis of *M*-(*N*-Methoxycarbonylamino) Phenol Intermediate

Methyl chloroformate (18 mL, 234 mmol) was added dropwise during 30 min to a suspension of 22g (202 mmol) of *m*-aminophenol, 25g KHCO₃, 150 mL of ethylacetate and 10 mL distilled water at 10-15 °C. The mixture was stirred for further 1h, then 50 mL of distilled water was added to the mixture and stirred for 3h at room temperature (25 °C). The mixture was then transferred to a separating funnel, aqueous layer was removed and then organic layer was washed with water, 1M H₂SO₄, and brine solution. Solvent was evaporated by Rota evaporator, and the precipitate obtained was recrystallized from toluene. Yield 30.5 g (90%), and mp 98-99 °C were reported²⁰.

2.1.2 Second Step— Synthesis of 7-Amino-4-Methyl-2*H*-Chromen-2-One Intermediate

A suspension of 23g (137 mmol) *m*-(*N*-methoxycarbonylamino) phenol and 25 ml of β -ketoester was added portion wise to 60 mL of concentrated H₂SO₄. The mixture was stirred for 2h at room temperature (25 °C), then 400 mL of ice water was added slowly to the mixture and the mixture was stirred till crystal appear. The precipitate was filtered and washed by distilled water.



Scheme 1— Synthetic routes for the preparation of 7-amino-4-methyl coumarin

Then the precipitate was added to 60 mL of 45% KOH and the mixture was heated to 90 °C for 25 min till a clear solution is obtained. The mixture was then cooled down and diluted with 150 mL H₂O. Finally, the solution was filtrated to remove any insoluble impurities, the filtrated solution was carefully acidified by diluted HCl (1N) to pH 5-6 and then stirred for 10 min. The pH was again increased to 8 using 0.1N NaOH. The mixture was stirred for further 30 min, filtrated, and the precipitate obtained was washed by distilled water. Finally, 7-amino-4-methyl-2H-chromen-2-one, was recrystallized from ethanol (yield 85 %, mp 224-226 °C), which is in agreement with the reported results²¹.

2.2 Monofunctional FWA 1

Monofunctional FWA 1 based on methyl coumarin was prepared (Scheme 2). A suspension of cyanuric chloride (1.94 g, 95% purity, 0.01 mol) in acetone (25 mL) and crushed ice (15-20 g) was freshly prepared, to which an aqueous solution of sulphanilic acid (1.92 g, 90% purity, 0.01 mole) was added at pH 6-7 and over 20 min at 0-5 °C. The reaction mixture was stirred for 4h, while the pH was adjusted at 4-5 using 2M aqueous solution of sodium carbonate. The reaction was controlled by thin layer chromatography (TLC). After the first condensation reaction was completed, 7-amino-4-methyl-2H-chromen-2-one (1.75 g, 0.01 mole) dissolved in a least amount of acetone was added to the reaction mixture over 45 min. The reaction temperature was then raised to 35-40 °C, adjusting the pH at 5-5.5, and controlling the reaction process by TLC. After the reaction was completed, the precipitated compound was filtered off

and washed severally with acetone, then dried at room temperature (25 °C) under vacuum (yield 92 %). The purity of the produced compound (C₁₉H₁₃N₅O₅SNaCl) was determined by elemental analysis, and following results are obtained:

Compound name—Sodium 4-(4-chloro-6-(4-methyl-2-oxo-2H-chromen-7-ylamino)-1,3,5-triazin-2-ylamino) benzenesulfonate

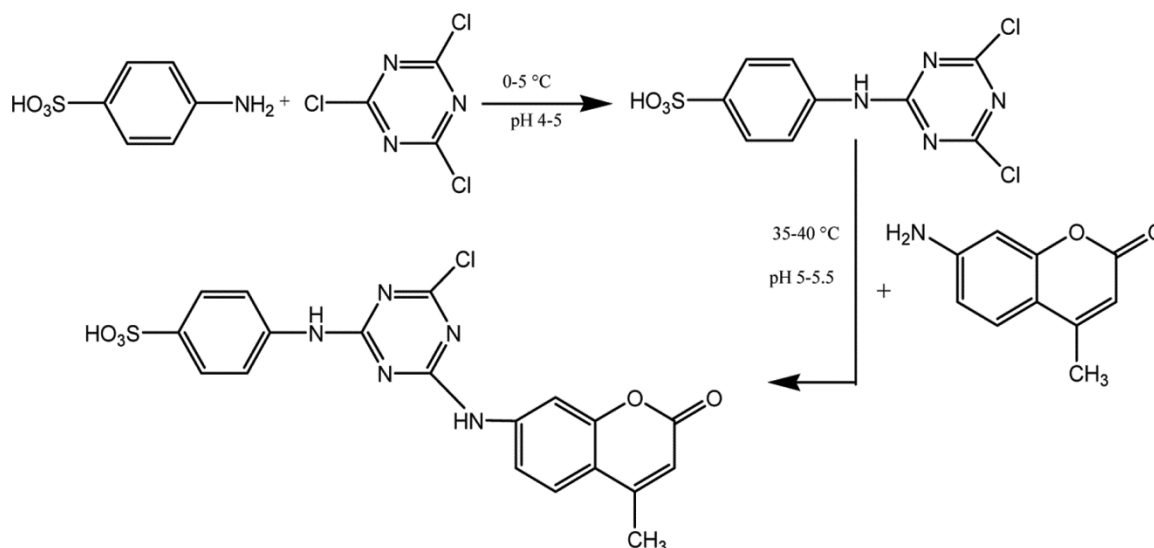
Calc. (%)—C 47.36, H 2.72, N 14.53, S 6.66.

Found (%)—C 47.2, H 2.41, N 14.7, S 6.8.

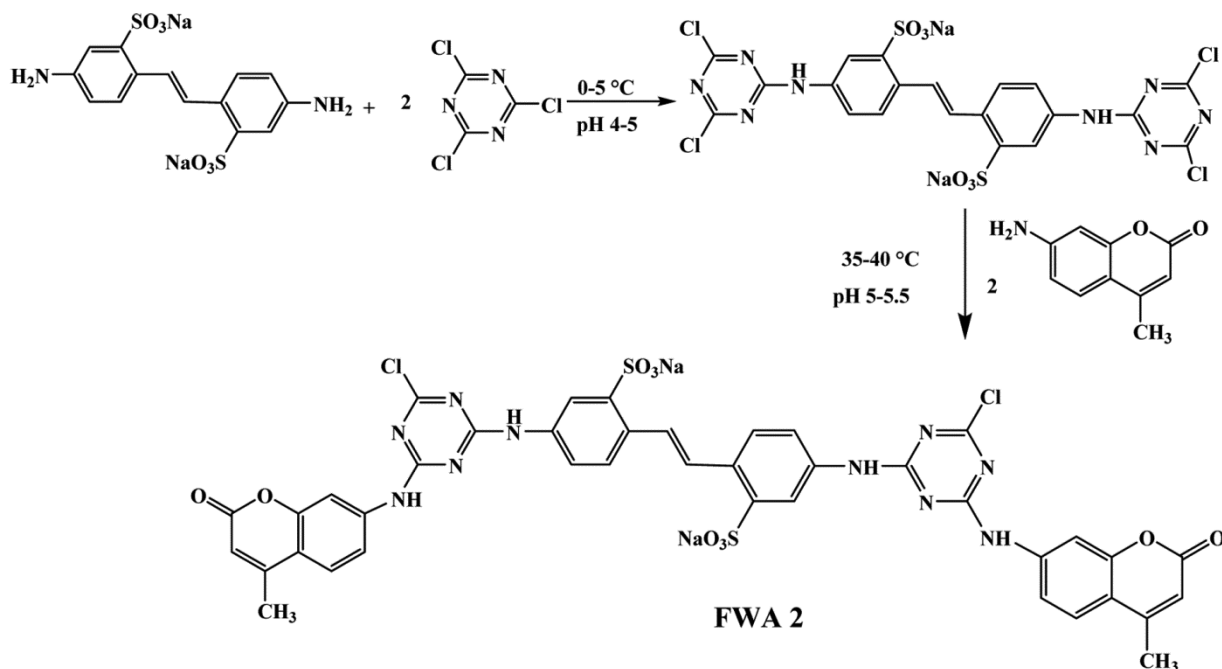
¹H-NMR (400 MHz, DMSO-d₆, δ, ppm) —2.25 ppm (s, 3H, CH₃), 6.2-7.65 ppm (m, 8H, CH aromatic), 10.44 ppm (s, 1H, NH proton) and 10.64 ppm (s, 1H, NH proton).

2.3 Bifunctional FWA 2

Bifunctional FWA 2 based on methyl coumarin was prepared (Scheme 3). A solution of 4,4'-Diamino-2,2'-stilbenedisulfonic acid (4.03 g, 92% purity, 0.01 mole) was adjusted at pH 6-7, then add dropwise over 20 min to a freshly prepared suspension of cyanuric chloride (3.88 g, 95 % purity, 0.02 mole) in acetone (50 mL) and crushed ice (50 g). The reaction mixture was stirred at 0-5 °C for 4h while controlling the pH at 4-5 using 2M aqueous solution of sodium carbonate and TLC. After the first condensation reaction was completed, 7-amino-4-methyl-2H-chromen-2-one (3.5 g, 0.02 mole) dissolved in a least amount of acetone was added to the reaction mixture in over 45 min using ratio of 2:1 with the amine. The temperature was maintained at 35-40 °C while adjusting the pH at 5-5.5, the second condensation reaction was also controlled by TLC. After the reaction was completed, the precipitated compound was filtered off and washed severally with acetone,



Scheme 2— Synthesis of monofunctional FWA 1 based on methyl coumarin



Scheme 3— Synthesis of bifunctional FWA 2 based on methyl coumarin

then dried at room temperature (25 °C) under vacuum (yield 90 %).

Compound name—sodium 5-((4-chloro-6-((4-methyl-2-oxo-2H-chromen-7-yl) amino)-1,3,5-triazin-2-yl)amino)-2-(4-((4-chloro-6-((4-methyl-2-oxo-2H-chromen-7-yl)amino)-1,3,5-triazin-2-yl) amino)-2-sulfonatostyryl)benzenesulfonate

Calc. (%) — C 48.64, H 2.65, N 14.18, S 6.49.

Found (%) — C 48.87, H 2.55, N 14.1, S 6.39.

¹H-NMR (400 MHz, DMSO-d₆, δ, ppm) — 2.30 ppm (s, 6H, CH₃), 6.2-8.22 ppm (m, 14H, CH aromatic), 10.41 ppm (s, 2H, 2NH proton) and 10.58 ppm (s, 2H, 2NH proton).

2.4 Bifunctional FWA 3

Bifunctional FWA 3 based on phenyl coumarin was prepared (Scheme 4). The FWA 3 was prepared using the method as described for FWA 2, except the use of 7-amino-3-phenyl-2H-chromen-2-one instead of 7-amino-4-methyl-2H-chromen-2-one in the second condensation reaction (yield 80 %).

The purity of the produced compound (C₅₀H₂₈N₁₀O₁₀S₂Na₂Cl₂) was determined by elemental analysis, and following results are obtained:

Compound name—Sodium 5-((4-chloro-6-((2-oxo-3-phenyl-2H-chromen-7-yl)amino)-1,3,5-triazin-2-yl) amino)-2-(4-((4-chloro-6-((2-oxo-3-phenyl-2H-chromen-7-yl)amino)-1,3,5-triazin-2-yl)amino)-2-sulfonato-styryl)benzenesulfonate

Calc. (%) — C 54.11, H 2.54, N 12.62, S 5.78.

Found (%) — C 54.2, H 2.5, N 12.7, S 5.8.

¹H-NMR (400 MHz, DMSO-d₆, δ, ppm) —7.3-8.19 ppm (m, 24H, CH aromatic), 10.42 ppm (s, 2H, 2NH proton) and 10.65 ppm (s, 2H, 2NH proton).

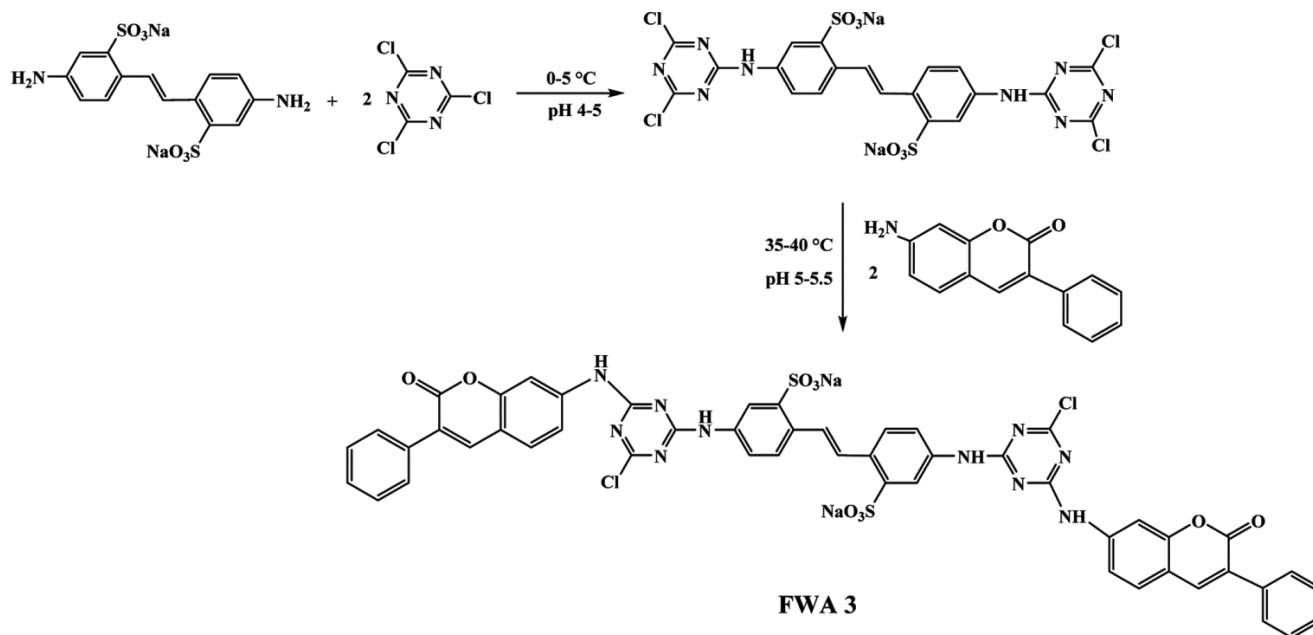
2.5 Application on Nylon 6 Fabrics

Nylon 6 fabric was treated using FWA 1, 2 and 3 in an Ahiba dyeing machine at a liquor-to-material ratio of 40:1. The treatment bath was prepared at room temperature (25 °C) in a range of concentrations (0.05, 0.1, 0.2, 0.3, 0.4 and 0.5% owf) for FWA 1-3 and the pH was adjusted at 5 using diluted acetic acid solution. Treatment was started at 40 °C, then the temperature was raised to 100 °C over 60 min. The treatment was further continued for another 60 min at the desired temperature. After treatment, all the treated samples were rinsed with water and air dried.

2.6 Measurements

2.6.1 Dye Exhaustion

The uptake of FWA by nylon 6 fabric was measured by sampling the treatment bath before and after treatment. The FWA concentration (g/L) of the treatment bath, diluted with 50 % aqueous DMF, was measured on a Shimadzu UV-2401PC UV/VIS spectrophotometer at λ_{max} using a calibration curve; previously obtained using known FWA concentrations in 50% aqueous DMF. The percentage of treatment bath exhaustion (%E) was calculated using the following equation²³:



Scheme 4— Synthesis of bifunctional FWA 3 based on phenyl coumarin

$$\%E = \left[1 - \left(\frac{C_2}{C_1} \right) \right] \times 100 \quad \dots (1)$$

where C_1 and C_2 are the concentrations of FWA in the treatment bath before and after application respectively.

2.6.2 FWA Fixation

The FWA fixation ratio ($\%F$), the percentage of exhausted FWA chemically bound on the fibre, was measured by refluxing the treated samples in 50% aqueous DMF (liquor ratio 20:1) for 10 min to extract the unfixated FWA. This procedure was repeated until the extract was clear. The concentration of the extract was then measured spectrophotometrically at (λ_{\max}) and the FWA fixation ratio was calculated using the following equation²³:

$$\%F = \frac{(C_1 - C_2 - C_3)}{(C_1 - C_2)} \times 100 \quad \dots (2)$$

where C_3 is the concentration of extracted FWA.

From the treatment bath exhaustion (E) and FWA fixation ratio (F), the total FWA fixation (T), which is the percentage of FWA chemically bound relative to the total amount of FWA used, was calculated for all treatments using the following equation²³.

$$\%T = \frac{(\%E \times \%F)}{100} \quad \dots (3)$$

2.6.3 Fluorescence Properties

The UV/vis absorption spectra were recorded at room temperature (25°C) in DMF using a Shimadzu UV2401PC UV-visible spectrophotometer with 1 cm path length quartz cuvettes. The solutions of prepared compounds were prepared by dissolving 10 μ mole of FWA in DMF and used directly for the measurement of absorption spectra.

The steady-state fluorescence spectra were measured using JASCO FP-8300 spectrophotometer on solution as well as on the treated fibers. The fluorescence emission spectra were recorded by excitation at absorption maxima, and the excitation spectra were recorded at fluorescence emission maxima. The fluorescence quantum yields (qF) were measured using a quinine sulphate (qF 0.54 in 0.5 mol/L H_2SO_4) standard²⁴.

2.6.4 Fastness Properties

The treated fabrics containing 0.1 and 0.4 % owf FWAs 1, 2 and 3 were tested for light fastness property using a xenon arc lamp in accordance with ISO 105-B02²⁵.

3 Results and Discussion

3.1 Synthesis of 7-Amino-4-Methyl-2h-Chromen-2-One

7-amino-4-methyl-2H-chromen-2-one has been prepared as a key intermediate by multistep reaction as previously reported²⁰⁻²². The synthesis was based on the *N*-substitution reaction of 3-aminophenol, masking the amino group using methyl chloroformate, which gives a

high yield of methoxycarbonylamino phenol. This step has been followed by the condensation reaction of methoxycarbonylamino phenol with ethylacetoacetate, forming 7-methoxycarbonylamino-4-methyl coumarin, as shown in Scheme 1. By the hydrolysis of the methoxycarbonylamino coumarin derivatives in strong alkaline medium, 7-amino-4-methyl-2*H*-chromen-2-one (7-amino-4-methyl coumarin AMC) is obtained in good yield (> 80%).

3.2 Synthesis of FWAs

The synthesis of FWAs 1-3 is basically based on electrophilic substitution reaction of the reactive chlorine atoms in cyanuric chloride with different amino derivatives, such as coumarin and stilbene. The first condensation reaction is carried out at 0-5° C due to the high reactivity of the first substituted chlorine atom in cyanuric chloride and the pH of the reaction mixture is controlled at 5 using aqueous solution of sodium carbonate to neutralize the HCl liberating. The second substitution of the second chlorine atom is carried out at 30-35°C and 5-5.5 pH.

3.3 Characterization of FWAs

The ¹HNMR data for FWAs 1-3 has been measured in DMSO-*d*⁶, which shows a significant band for all expected protons. ¹HNMR shows a single beak at 2.25 ppm, corresponding to the methyl proton localized on coumarin ring in FWA 1. In the case of FWA 2, the methyl proton is appeared at 2.30 ppm, which represents 6 protons for the symmetrical two methyl groups. The aromatic protons in all prepared compounds are overlapped, thus showing multiplet beaks appeared in the range 6.2-8.22 ppm. The NH protons appear as singlet in the range 10.42-10.64 ppm.

3.4 FWAs Application on Nylon-6

The application of the prepared FWA's has been examined on nylon 6 fabric at pH 5 and at different FWA concentrations (0.05, 0.1, 0.2, 0.3, 0.4 and 0.5% owf). The results are given in Figs 1(a) and (b). It can be seen that the highest exhaustion and the total fixation are achieved in the following order:

FWA 2 > FWA 3 > FWA 1.

As a result of having two reactive monochlorotriazine groups in each of FWAs 2 and 3, this increases their probability of reacting with the reacting sites of the fabric as compared to FWA 1 which has one monochlorotriazine reactive group. In general, the extent of exhaustion and fixation yield of the three compounds decrease with increasing concentration. This is due to the fact that, at higher concentrations the

probability of aggregation increases, resulting in lowering their penetration to the fabric. Additionally, the number of available reacting sites on the fibre decreases, resulting in a lower extent of exhaustion and fixation yield of the compounds.

3.5 Fluorescence Properties

The absorption spectra of the prepared FWAs 2 and 3 show a significant HOMO-LUMO sharp absorption band without any vibronic beaks in the UV region in the range from 314nm to 372 nm, which could be assign to the π - π^* transition [Figs 2 (a)-(c)]. FWA 1 shows a shoulder vibronic band in absorption spectra [Fig. 2 (a)]. The emission spectra and λ_F for the solution of prepared FWAs 1-3 and for the treated fibre are summarized in Table 1. A significant bathochromic shift in absorption spectra is achieved according to the following order:

FWA 3 > FWA 2 > FWA 1

The emission spectra of the prepared FWAs show only significant fluorescence band in the range

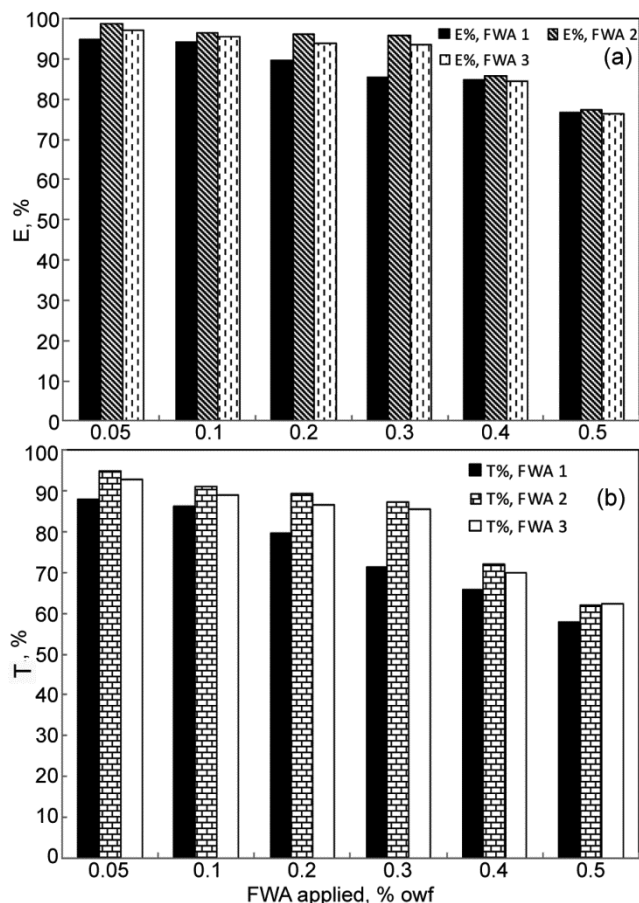


Fig. 1 — (a) Exhaustion (E) and (b) total fixation (T) of FWAs 1, 2 and 3 at different concentrations on nylon 6 fabric

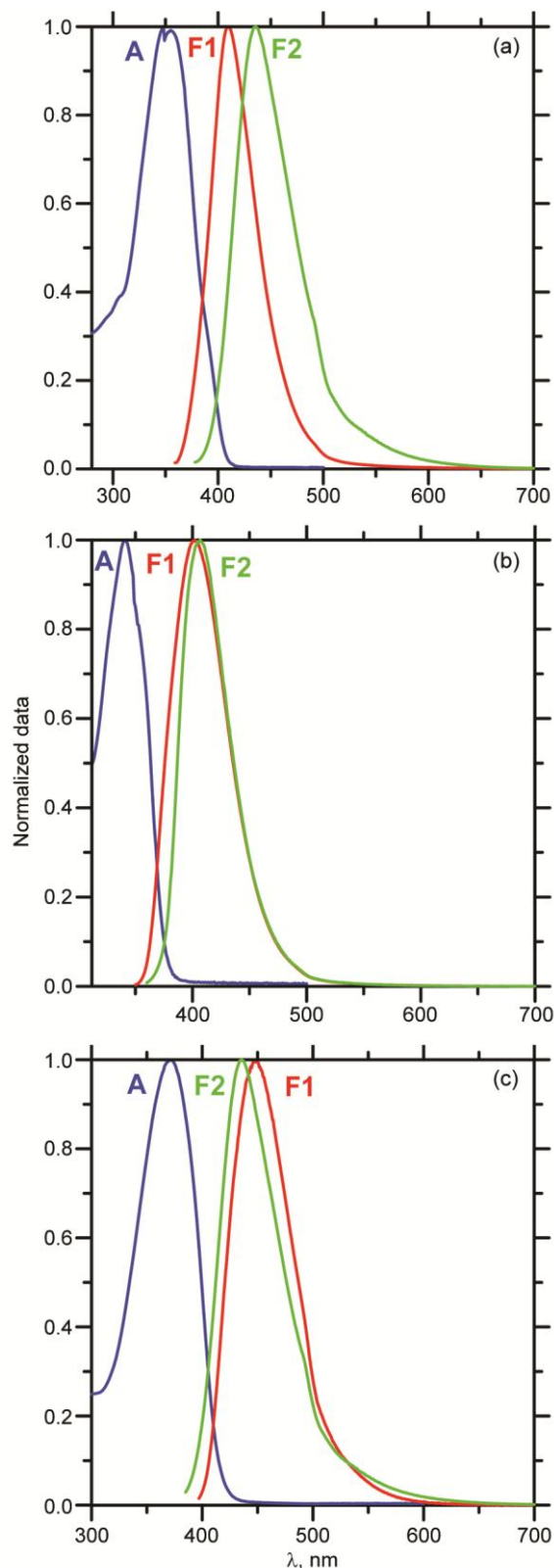


Fig. 2—Absorption (A) and fluorescence (F1) spectra of (a) FWA 1, (b) FWA 2 and (c) FWA 3. (F2)- Fluorescence spectra on the fibre (0.05 % shade)

403-449 nm in solution. There is a slight bathofluoric shift obtained on the treated fabric as compared to that with the emission in solution, while in the case of FWA 3, a significant hypsofluoric effect is observed in the emission for the fabric than in solution by 14 nm. These findings may be due to the prolonged π -conjugated system in the case of FWA 3 due to the terminal phenyl ring connected in position 3 on coumarin ring. The highest fluorescence quantum yield is achieved according to the following order:

$$\text{FWA 1} > \text{FWA 3} > \text{FWA 2}$$

3.6 Whiteness Properties

The whiteness index (WI) of the treated fabrics is measured with different concentrations of FWAs 1-3 and the results are given in Table 2. The results clearly show that the three compounds attain higher degree of WI for all shades studied as compared to the untreated fabric, except in FWA 3 at 0.5 % shade. The highest degree of WI is achieved according to the following order:

$$\text{FWA 2} > \text{FWA 1} > \text{FWA 3}$$

This may be due to the fact that the presence of the phenyl group may cause slight yellowness especially at high concentration (0.5% shade) in FWA 3, which ascribes to the highly conjugation as compared with FWAs 1 and 2.

3.7 Light Fastness Properties

The light fastness properties of the FWAs 1-3 on nylon fabric using 0.1 and 0.4 % owf concentrations are studied and the results are given in Table 3. The results show that the FWA 3 achieves excellent light

Table 1—Absorption (λ_A) and fluorescence (λ_F) maxima (nm) and fluorescence quantum yields (qF) of all compounds in dimethyl formamide (DMF)

Compound	λ_A , nm	λ_F (solution) nm	λ_F (fibre) nm	qF
FWA1, mono (MeQ)	314	403	407	0.6
FWA2 bis (MeQ)	348	410	430	0.1
FWA3 bis (PhQ)	372	449	435	0.15

Table 2 — Whiteness index (WI) of the treated fabrics at different concentrations of FWAs 1-3

FWAs	Whiteness index on nylon-6						
	0.00 %	0.05 % owf	0.1 % owf	0.2 % owf	0.3 % owf	0.4 % owf	0.5 % owf
1	49.66	68.79	76.10	79.88	82.25	84.67	83.84
2	49.66	74.40	80.34	82.41	84.75	86.61	84.28
3	49.66	64.13	61.50	56.16	53.6	50.26	47.03

Table 3—Light fastness of treated fabrics at different concentrations of FWAs 1-3

FWA	Conc., % owf	Light fastness
1	0.1	3-4
	0.4	3-4
2	0.1	4
	0.4	4
3	0.1	6-7
	0.4	6-7

fastness on nylon fabric, while FWAs 1 and 2 achieve good light fastness.

4 Conclusion

4.1 The presence of two reactive bis(monochlorotriazine) groups in FWA 2 and 3 as compared to the presence of one monochlorotriazine reactive group in FWA 1, results in higher substantivity, exhaustion and fixation efficiency of FWA 2 and 3 as compared to FWA 1.

4.2 By virtue of the three FWA structures, FWA 1 and 2 contain methyl coumarin derivative and secure the highest degrees of WI as compared to FWA 3 that contains phenyl coumarin derivative, over all the shades studied.

4.3 By studying the fluorescence properties, the highest fluorescence quantum yield is achieved in following order: FWA 1 (0.6) > FWA 3 (0.15) > FWA 2 (0.1).

4.4 The FWA 3 obtains higher light fastness as compared to FWA 1 and 2.

4.5 FWA 2 [bis(methyl coumarin)] is found the best for application on nylon-6 fabric because this compound achieves good light fastness, the highest WI, and the highest total fixation values, as compared to the other two compounds FWA 1 (mono methyl coumarin) and FWA 3 (bisphenyl coumarin).

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