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Novel Simple Cyanine, Carbocyanine, and Dicarbo-cyanine Dyes: Synthesis, Characterization and Application on Polyester Fabric

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Abstract

Novel simple cyanine dyes, carbocyanine dyes and dicarbo-cyanine dyes derived from the nucleus 6-amino-3-methyl-1,2 diaza 4,13 dihydronaphthacene 5, 7, 12 trione (2) were prepared. The electronic visible absorption spectra of all the novel synthesized cyanine dyes were investigated in 95 % ethanol to evaluate their photosensitization characters and uses these dyes as photographic sensitizers in industry. The antimicrobial activity evaluation of mono, tri and pentamethine cyanine dyes against some bacterial and fungi strains(Escherichia coli, Staphylococcus aureus, Aspergillus lavus and Candida albicans) was tested. The antimicrobial activity of the dyes usually increases when they give higher inhibition zone diameter against the tested bacterial and fungi strains. The dyeing process and fastness properties of the new synthesized cyanine dyes were examined on polyester fabric. Polyester is the hydrophobic fibres and usually dyed with cyanine dyes due to their high tinctorial strength and good fastness properties. The structural characterization of dyes carried out by elemental analysis, visible, mass spectroscopy, ¹H NMR and IR spectra.

Keywords: synthesis, cyanine dyes, visible spectra, antimicrobial activity, polyester fabric, methine cyanine dyes.

1. Introduction

Cyanine dyes (Shindy, 2017; Li et al., 1998; Yadav, 2005; Kabatc et al., 2012; Miki et al., 2017; Park et al., 2013; Keisar et al., 2017; Upadhyayula et al., 2015; Antonious, 1997; Sener et al., 2018; Hilal et al., 2007; Xiang et al., 2008; Ferreira et al., 2015; Li et al., 2012; Wada et al., 2015; Wang et al., 2017; He et al., 2017; Li et al., 2017) are important class of organic heterocyclic dyes. This is due to the excellent photochemical and photophysical properties, extraordinary applications and uses of these dyes in a diverse and a board area, such as biochemistry, engineering, physics, biotechnology, biology, pharmacology and medicine. These dyes can be used in photography, analytical reagents over a wide pH media, in high energy laser and digital image storage, as indicator for solvent polarity, in biomedical and biological use as molecular probes and as fluorescent dyes for DNA visualization assays. In addition cyanine dyes (Sun et al., 2013; Sha et al., 2018; Christenson et al., 2014) used in high technique such as in optical recording materials. On the other side, cyanine dyes possess a wide range of bioactivities including antimicrobial efficiency, antibacterial agents, antioxidants, anticancer and antitumer agents (Power et al., 2009; Gomaa, 2014; Shindy et al., 2016; Fayez, 2009; Fayez et al., 2015; Badran et al., 2007; Mohareb et al., 2007; Vicini et al., 2002; Mishra et al., 2019; Yong et al., 2009; Rathish et al., 2012). The work in this paper aimed to synthesize novel methine cyanine dyes covers monomethine, trimethine and

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pentamethine cyanine dyes to study their electronic transitions through investigating their visible absorption spectra in ethanolic solution to evaluate their photosensitization properties to be used as photographic sensitization in industry. The antimicrobial activities of monomethine, trimethine and pentamethine cyanine dyes were evaluated versus many species of both bacteria and fungi strains and showed promising results. The present study is an attempt to synthesis of novel methine cyanine dyes using for dyeing of polyester fabric to obtain dyed polyester fabric have good fastness properties. The polyester fabrics give high affinity for some prepared dyes.

2. Materials and methods

2.1. General

All melting points of the synthesized cyanine dyes measured by using Electrothermal 15v, 45w 1 A9100 melting point apparatus (Faculty of Science, Aswan University, Aswan, Egypt) and are uncorrected. Elemental analysis carried out at the Microanalytical Center by an automatic analyzer (Vario EL III Germany) (Cairo-Uni – versity). The IR (KBr) spectra measured with a FT-IR (4100 Jasco, Japan) (Cairo University). ¹H NMR spectra accomplished with Varian Gemini-300 MHz NMR spectrometer (Cairo University). The electronic absorption spectra carried out on visible spectrophotometer spectra 24 RS Labomed, INC. (Faculty of Science, Aswan University, Egypt). Mass spectroscopy recorded on Mass 1: GC2010 Shimadzu Spectrometer (Cairo University). Dyeing process examined on Infra dyeing machine in National Research Centre (Dokki, Giza, Egypt).

2.2. Synthesis

2.2.1. Synthesis of 6-amino-3-methyl-1,2 diaza 4,13 – dihydronaphthacene 5,7,12 trione (2)

This compound was synthesized according to reference described earlier (El-Kanzy et al., 2007).

2.2.2. Synthesis of 6-amino 2,3 dimethyl 1,2 diaza 4,13-dihydronaphthacene 5, 7, 12 trione-2 ium iodide (3)

A pure sample of compound (2) was suspended in excess of ethyl (methyl) iodide and heated in a sealed tube at 140°C for 2 hrs. The sealed tube was cooled, opened and the product (3) was collected, washed with ether and crystallized from ethanol to give dark brown crystals (Table 1).

Table 1. Characterization of compounds (3–8c)

Comp. No.	Nature of products			Mol. Formula, (M. Wt)	Elemental analysis, % Calculated (Found)		
	Colour	Yield %	M.P., °C		C	H	N
3	Dark brown	90	160	C ₁₈ H ₁₆ N ₃ O ₃ I (449)	48.10 (48.25)	3.56 (3.50)	9.35 (9.15)
4a	Brown	70	295	C ₂₅ H ₂₃ N ₄ O ₃ I (554)	54.15 (54.00)	4.15 (4.13)	10.10 (10.20)
4b	Deep red	80	260	C ₂₉ H ₂₅ N ₄ O ₃ I (604)	57.61 (57.68)	4.13 (4.22)	9.27 (9.40)
4c	Brownish red	75	240	C ₂₉ H ₂₅ N ₄ O ₃ I (604)	57.61 (57.80)	4.13 (4.16)	9.27 (9.33)
5	Brown	70	210	C ₂₃ H ₂₆ N ₃ O ₅ I (551)	50.09 (50.11)	4.71 (4.53)	7.62 (7.47)
6a	Red	65	280	C ₂₇ H ₂₅ N ₄ O ₃ I (580)	55.86 (55.66)	4.31 (4.27)	9.65 (9.83)
6b	Violet	70	290	C ₃₁ H ₂₇ N ₄ O ₃ I (630)	59.04 (59.23)	4.28 (4.26)	8.88 (8.73)
6c	Brownish red	67	260	C ₂₇ H ₂₅ N ₄ O ₃ I (580)	55.86 (55.93)	4.31 (4.09)	9.65 (9.55)
7	Brown	80	200	C ₂₁ H ₁₈ N ₃ O ₄ I (503)	50.09 (50.33)	3.57 (3.66)	8.34 (8.22)
8a	Deep red	77	260	C ₂₉ H ₂₇ N ₄ O ₃ I (606)	57.42 (57.44)	4.45 (4.60)	9.24 (9.35)

8b	Deep violet	68	285	C ₃₃ H ₂₉ N ₄ O ₃ I (656)	60.36 (60.50)	4.42 (4.33)	8.53 (8.41)
8c	Brownish red	75	250	C ₂₉ H ₂₇ N ₄ O ₃ I (606)	57.42 (57.30)	4.45 (4.40)	9.24 (9.20)

2.2.3. Synthesis of monomethine cyanine dyes (4a-c)

A mixture of compound (3) (0.01mole) and iodoethane quaternary salts of either (pyridine, quinoline or isoquinoline) (0.01 mole) was refluxed for 7-8 hrs in ethanol (40 ml) containing piperidine (2-6 drops), filtered hot, concentrated and acidified with acetic acid. The precipitated products after dilution with water and ice filtered off and crystallized from ethanol. The data were recorded in [Table 1](#).

2.2.4. Synthesis of intermediate compound (5)

1:1 molar ratios of the quaternary salt (3) and triethylorthoformate were refluxed in ethanol (30 ml) containing piperidine (3-6 drops) for 4 hrs., filtered hot to remove unreacted materials, concentrated to one half its initial volume, cooled, acidified, and precipitated by cold water. The brown precipitate was filtered, washed with water, dried and crystallized from ether. The data are registered in [Table 1](#).

2.2.5. Synthesis of trimethine cyanine dyes (6a-c).

A mixture of the intermediate compound (5) (0.01 mol) and N-ethyl (2-picolinium, quinaldinium,4-picolinium) iodide quaternary salts (0.01 mol) heated under reflux in ethanol (35 ml) containing piperidine (3-9 drops) for 8hrs., filtered hot, concentrated, cooled, acidified with acid and precipitated by ice. The precipitates were filtered off, dried and crystallized from absolute ethanol. The data were tabulated in [Table 1](#).

2.2.6. Synthesis of intermediate compound (7)

This intermediate compound (7) was prepared by refluxing compound (5) (0.01 mol) with acetaldehyde (0.01 mol) in ethanol (30 ml) and piperidine (2-6 drops) for 2-4 hrs., filtered hot, concentrated, cooled then precipitated by ice and water. The brown intermediate compound was crystallized from ethanol. The results are reported in [Table 1](#).

2.2.7. Synthesis of pentamethine cyanine dyes (8a-c)

Equimolar ratios of the previously synthesized intermediate compound (7) (0.01mol) and N-ethyl (2-picolinium, quinaldinium, 4-picolinium) iodide quaternary salts (0.01) were dissolved in ethanol (40 ml) and piperidine (3-7 drops). The reaction mixture was refluxed for 9 hrs., filtered hot, concentrated, cooled , acidified with acid and precipitated by ice. The precipitates were filtered off, dried and crystallized from absolute ethanol. The data were tabulated in [Table 1](#).

2.3. Antimicrobial studies

Antimicrobial activity of the tested samples (4a, 4b, 4c, 6a, 6b, 6c, 8a, 8b and 8c) was determined using a modified Kirby-Bauer disc diffusion method. Briefly, 100µl of the test bacteria/fungi were grown in 10 ml of fresh media until they reach a count of approximately 10⁸ cells/ml for bacteria or 10⁵ cells/ml for fungi.100µl of microbial suspension was spread on agar plates corresponding to the broth in which they were aintained. Isolated colonies of each organism that might be playing a pathogenic role should be selected from primary agar plates and tested for susceptbility by disc diffusion method. Disc diffusion method for filamentous fungi used to evaluate the susceptibilities of filamentous fungi to antifungal agent. Samples were dissolved in DMSO to give a final concentration (1 mg/ml). The agar used is Mueller-Hinton agar that is rigorously tested for composition and pH. Further the depth of the agar in the plate is a factor to be consider in the disc diffusion method. This method is well documented and standared zones of inhibition have been determined for susceptible and resistant values. Blank paper disks with a diameter of 8.0 mm were impregnated 10µ of tested concentration of the stock solutions. The biological activity for each substance was tested on surface – seeded nutrient agar medium with the prepared susceptible disc. Bacterial strains and the biological effect are shown in [Table 4](#).

2.4. Fabric

Polyester fabric, bleached and mill-scoured were supplied by the company called El-Mahalla El-Kobra, Egypt. The fabrics always scoured at temperature 50 °C for 30 min, L:R(1:50), 2 g/L of Na₂CO₃ and 2 g /L of nonionic detergent solution (Hostapal; Clariant, Swiss). Then dried at room temperature after rinsed with cold water.

Dyeing Method

Dyeing process carried out by using distilled water while dyeing experiments required two step. Firstly, Polyester fabric dyed take place by using the prepared disperse dyes at pH 5 by adding acetic acid, liquor ratio at 1:50. Dye bath contains Matexil DA-N which (supplied by ICI Company, UK) as dispersing (1 ml/L) and 1 % of dye . The temperature start at 40 °C, then raised to 130 °C for about 60 min. using cold water in washing and reduction cleaning mad by both hydrosulphite (2g/L), sodium hydroxide (2g/L) at 60 oC for about 10 min. Then , treated the samples by acetic acid (1ml/L), cooled water and dried (Tarulata et al., 2011; Tarek et al., 2015).

2.5. Color Measurements of the dyed samples

Color Strength

Hunter Lab ultra Scan® PRO spectrophotometer use to determined the colorimetric analysis of dyes while the Kubelka Munk equation (Kubelka et al., 1931) used to determine colour strength value (K/S) as follow:

$$K/S = \frac{(1-R)^2}{2R} \quad (1)$$

Where,

R = decimal fraction of the reflection of the dyed fabric,

K = absorption coefficient, and S = scattering coefficient

Fastness testing

The dyed samples subjected to washing, rubbing, light, sublimation test and perspiration according to the standard ISO methods, like ISO 105-X12 (1987), ISO 105-C04 (1989), ISO105-E04 (1989), ISO 105-B02 (1988) respectively.

3. Results and discussion

3.1. Synthesis

The polyheterocyclic starting compound namely 6-amino 2,3 dimethyl 1,2 diaza 4,13-dihydronaphthacene-5,7,12 trione-2 ium iodide (3) was synthesized by quaternization of compound (2) using an excess of iodomethane. The reaction of the compound (3) with an iodoethane quaternary salts of either pyridine, quinoline or isoquinoline in equimolar ratios in ethanol catalyzed by piperidine gave the 6-amino 2 methyl 1,2 diaza 4,13-dihydronaphthacene-5,7,12 trione 3[4(1)]monomethine cyanine (4a-c) Scheme (1), Table 1.

Treating on the newly synthesized monomethine dyes (4a-c) by conc. H₂SO₄ resulted in liberating iodine vapor on heating. This can be attributed to the liberation of hydrogen iodide and hydrogen molecule.

The reaction of the quaternary salt (3) with triethylorthoformate in equimolar ratio and in ethanol containing few mls of piperidine as a basic catalyst gave the corresponding intermediate compound (5) (Scheme 1).

The intermediate compound (5) reacted with equimolar ratios of N-ethyl(2-picolinium, quinaldinium, 4-picolinium) iodide quaternary salts in ethanol and piperidine to give 6-amino 2 methyl 1,2 diaza 4,13-dihydronaphthacene-5,7,12 trione 3[2(4)] trimethine cyanine dyes(6a-c) (Scheme 1). Treatment on the previous trimethine cyanine dyes (6a-c) by conc. H₂SO₄ liberating iodine vapor. This back to elimination two molecules of ethanol and hydrogen iodide molecule.

Finally, reaction of equimolar ratio of intermediate compound (5) with acetaldehyde in ethanol and piperidine yielded compound (7). The later compound (7) undergoes condensation reaction with equimolar ratios of N-ethyl(2-picolinium, quinaldinium, 4-picolinium) iodide quaternary salts in ethanol and piperidine to give 6-amino 2 methyl 1,2 diaza 4,13-dihydronaphthacene-5,7,12 trione 3[2(4)]petamethine cyanine dyes (8a-c). (Scheme 1). The structure of the dyes were identified by elemental analysis, visible spectra Tables 1, 2, IR, ¹H NMR and Mass spectroscopic (Table 3).

3.2. Spectral behavior in 95 % ethanol solution:

Dyes (4a-c), (6a-c), and (8a-c) are highly coloured compounds in their ethanolic solution ranging from deep red to deep violet. The electronic absorption spectrum features (λ_{\max} and ϵ_{\max} values) of the newly synthesized mono cyanine dyes (4a-c), tri cyanine dyes (6a-c) and penta methine cyanine dyes (8a-c) were examined in 95 % ethanol solution and the results are summarized in Table 2.

The Uv-Visible absorption spectrum of the monomethine cyanine dyes (4a-c) in 95 % ethanol shows bands in the visible region 405-520 nm. These bands underwent displacements to give red(or blue) shifts with increasing (or decreasing) depending upon the nature of the quaternary residue(A), extension of π -delocalization and their linkage position. So, the dye (4a), A=1-ethyl pyridinium-4-yl salt showed λ_{\max} at 405,445 nm. Substitution A = 1-ethyl pyridinium-4-yl salt in dye (4a) by A = 1-ethyl quinolinium-4-yl salt in dye (4b) caused a bathochromic shift of λ_{\max} = 75 nm, so compound (4b), exhibited λ_{\max} = 520 nm. This back to the more extensive π -delocalization and conjugation within the extra phenyl ring in quinolinium ring in dye (4b) (Table 2) (Ahmed et al., 2018; Gomaa et al., 2012; Shindy et al., 2019; Shindy, 2018; Shindy et al., 2017; Shindy et al., 2015; Soriano et al., 2015; Shindy et al., 2018).

It is also interesting to note that the visible absorption maximum of dye(4b), A = 1-ethyl quinolinium-4-yl salt is red – shifted to dye (4c), A = 2-ethyl isoquinolinium-1-yl so dye (4c) showed λ_{\max} = 500 nm. This is due to the increasing π -delocalization within 4-yl salt in dye (4b) if compared with 1-yl salt in dye (4c) (Table 2).

Moreover, the Uv-visible absorption spectrum of the trimethine cyanine dyes (6a-c) discloses bands in the visible region 360-600 nm. Their intensity and positions are influenced by the nature of the heterocyclic quaternary residue (A), extension of π -delocalization and their linkage position. So, the absorption spectra of dye 6a, A = 1-ethyl pyridinium -2-yl salt showed λ_{\max} = 360, 505 nm. Substituting of A = 1-ethyl pyridinium-2-yl salt in dye (6a) by A = 1-ethyl quinolinium -2-yl salt in dye (6b) resulted in red- shifted of λ_{\max} = 10 nm with increasing the number of absorption bands, (6b λ_{\max} = 370, 450, 505 and 600 nm). This is due to the decreasing π -delocalization in dye (6a) than analogous (6b).

Additionally, changing the linkage position from 2-yl salt in trimethine cyanine dye (6a), A= 1-ethyl pyridinium -2- yl salt to 4-yl salt in trimethine cyanine dye (6c), A = 1-ethyl pyridinium 4-yl salt showed a remarkable bathochromic shift of λ_{\max} = 10 nm if compared with compound (6a), (6c, λ_{\max} = 365, 415,515 nm). This illustrated according to the increasing of the extension conjugation of 4-linkage pyridine moiety in dye (6c) better than 2-linkage analogous in dye (6a) (Table 2).

Finally, the Uv-visible absorption spectrum of pentamethine cyanine dyes (8a-c) displays bands in the visible region 530-690 nm. Their molar absorptivity of bands and positions are effected by the nature of quaternary salt residue (A) and their linkage position. So, substituting A = 1-ethyl pyridinium-2-yl salt in pentamethine cyanine dye (8a) by A = 1-ethyl quinolinium-2-yl salt in pentamethine cyanine dye (8b) causes red shifted by 30 nm and increasing the number of bands. This back to the increasing π -delocalization and conjugation in quinaldinium dye (8b) compared to α -picolinium dye (8a) (Table 2). Changing the linkage position from 2-yl salt in pentamethine dye (8a) to 4-yl salt in pentamethine dye (8c) produced red shifted by 10 nm. This can be explained in the light of increasing conjugation in the γ -picolinium dye (8c) (Table 2).

Comparing the Uv-visible absorption spectrum of trimethine cyanine dyes(6a-c) with those of the pentamethine cyanine dyes (8a-c) showed that the latter dyes are red shifted dyes compared with the former dyes. This is due to the increasing number of methine groups in pentamethine cyanine dyes which increasing conjugation. It is also notice that trimethine cyanine dyes (6a-c) showed absorption bands and their molar extinction coefficients, their positions of these bands are highly effected if compared with monomethine cyanine dyes (4a-c) (Table 2).

Table 2. The electronic absorption spectra of new synthesized cyanine dyes (4a-c), (6a-c) and (8a-c) in 95 % EtOH

Compound	λ_{\max} , nm (ϵ_{\max} mol ⁻¹ , cm ⁻¹)		
	4a	4b	4c
Monomethine cyanine dyes (4a-c)	405(19500) 445(15000)Sh.	520(17500)	500(22000)
Trimethine cyanine dyes (6a-c)	6a 360(22000) 505(20000)	6b 370(22000) 450(14000) 505(12000)Sh. 600(12500)	6c 365(22500) 415(16000) 515(20000)
Pentamethine cyanine dyes (8a-c)	8a 530(20000) 670(5500)Sh.	8b 560(14500) 670(2500)Sh. 690(2500).	8c 450(19500) 680(11000)

Table 3. IR and ¹H NMR (Mass) spectral data of the prepared compounds

Comp. No.	IR Spectrum (KBr, cm ⁻¹)	¹ H NMR Spectrum (DMSO, δ); & (Mass data)
3	1387 (C-N), 1451 (C=N), 1593 (C=C), 1652 (quinone ring), 2940 (quaternary salt), 3434 (NH ₂).	0.82-1.08 (S, 3H, CH ₃ , N-methyl iodide), 2.00-2.50 (S, 2H, NH ₂), 3.17 (S, 2H, CH ₂), 3.50 (S, 3H, CH ₃), 4.10 (S, 1H, quinone), 7.40-8.40 (m, 5H, 4Ar-H+NH).
4b	1369 (C-N), 1451 (C=N) 1666 (quinone ring), 2924 (quaternary salt), 3423(NH ₂).	0.80-2.00 (m, 9H, CH ₃ ,CH ₃ of N-quinolinium, CH ₂ , CH), 2.50 (S, 2H, NH ₂), 3.31 (S, 2H, CH ₂ of N-quinolinium), 7.00-8.50 (12H, 10Ar-H+NH+ = CH).
5	1263 (C-O ether), 1379 (C-N), 1360-1447 (C=N), 1590 (C=C), 1625 (quinone ring) 2936 (quaternary salt), 3424 (NH ₂).	1.15-1.20 (t, 6H, 2 CH ₃ of diethoxyethyl), 1.25-1.90 (m, 4H, CH ₃ of methyl iodide+ CH of diethoxyethyl), 2.50 (S, 2H, NH ₂), 2.70 (S, 2H, CH ₂), 3.00 (S, 4H, 2CH ₂ of diethoxy ethyl), 3.40 (m, 2H, CH ₂ of diethoxyethyl), 3.90-4.00 (S, 1H, quinone), 6.90-8.60 (m, 5H, 4Ar-H +NH). M+: 551
6a	1375 (C-N), 1492 (C=N), 1622 (C=C), 2920, 2858 (quaternary salt), 3432(NH ₂).	0.80-2.00 (m, 9H, CH ₃ ,CH ₃ of N-pyridinium, CH ₂ , CH), 2.50 (S, 2H, NH ₂), 3.43 (S, 2H, CH ₂ of N-pyridinium), 7.20-8.50 (m, 12H, 8Ar-H+NH+ = CH).
7,8b	1371, 1339(C-N), 1441, 1443 (C=N), 1630, 1621 (C=C), 1763(C=O) aldehydic for compound (7). 2925, 2924 (quaternary salt), 3424, 3422 (NH ₂).	(M+1): 504 For compound (7) (8b): 1.17-2.20 (m, 9H, CH ₃ , CH ₃ of N-quinolinium, CH ₂ , CH), 2.50 (S, 2H, NH ₂), 3.33 (S, 2H, CH ₂ of N-quinolinium), 7.45-8.32 (m, 16H, 10Ar-H+NH+ = CH).

3.3. Antimicrobial activity

The antibacterial activities and antifungal activities of all the newly synthesized quinone cyanine dyes 4a, 4b, 4c, 6a, 6b, 6c, 8a, 8b, and 8c were tested against some bacterial strains (*Escherichia coli* and *Staphylococcus aureus*) and fungi strains (*Aspergillus flavus* and *Candida albicans*) (Table 4). When a filter paper disc impregnated with a tested chemical is placed on agar the chemical will diffuse from the disc into the agar. This diffusion will place the chemical in the agar only around the disc. The solubility of the chemical and its molecular size will determine the size of the area of chemical infiltration around the disc. If an organism is placed on the agar it will not grow in the area around the disc if it is susceptible to the chemical. This area of no growth around the disc is known as a [Zone of inhibition] or [Clear zone]. The data obtained are expressed as size (mm) of inhibition zone. Diameter of the inhibition zone were high (22-18 mm), moderate (17-12 mm), low (11-1 mm), no response(-) (Ballatore et al., 2012; King et al., 2010; El-Mashad et al., 2012; Mohamed et al., 2014). Studying the antimicrobial (antibacterial, antifungal) activity evaluation against some bacterial and fungi strains have a great practical value in cyanine dyes because of the uses and applications of these dyes as bactericidal and fungicidal in pharmaceutical industry. In this study, all the newly synthesized dyes are biologically active against bacteria this is due to the presence of quinone ring with its high potency. Dyes (4a-c) possess highest potency as antimicrobial activity if compared with the others showed moderate to low (Table 4). Generally, decreasing carbon content of the tested dyes (4a-c) if compared with (6a-c, 8a-c) increased the activity against bacterial strains. This could be correlated to the hydrophilic and hydrophobic structural equilibria of the tested dyes. Comparing the antibacterial activity of the monomethine cyanine dye (4a), trimethine cyanine dye (6a) and pentamethine cyanine dye (8a) by their analogous (4b), (6b) and (8b) resulted in that the latter dyes (4b, 6b, 8b) have higher inhibition zone diameter than the former dyes (4a, 6a, 8a). This is due to the increasing π -delocalization and conjugation in the quinoline ring system in (4b, 6b, 8b). Moreover, the antimicrobial activity of the trimethine cyanine dye (6c) and pentamethine cyanine dye (8c) showed higher inhibition zone diameter than analogous (6a) and (8a). This attributed to the extension of π -delocalization in the former dyes (6c, 8c) which contains γ -picoline. Conversely, all the prepared cyanine dyes are biologically inactive against fungi except (4b) showed moderate fungicidal activity only on *Candida albicans* (Table 4). From Table 4 we can conclude that: all the tested dyes can be used as antibacterial agents against bacterial strains (*Escherichia coli* and *Staphylococcus aureus*) while dye (4b) only can be used as fungicidal against (*Candida albicans*). Finally: the antimicrobial activity of dyes depending upon the type of bacterial and fungi strains, number of methine groups, type of quaternary salt residue (A) and linkage position.

3.4. Color strength

ΔE , b, a, L and color strength K/S values of the polyester fabrics dyed by using disperse dyes are recorded in Table 5. The dyeing process carried out at pH 4.5, L. R 1:50, at 130 °C, and 1 % (w.o.f) for 60 min. The CIE (L^* , a^* , b^*) system was employed to determine the color coordinates, where the value of (b^*) range from yellow (positive) to blue (negative), (a^*) range from green (negative) to red (positive), and (L^*) denote to lightness or darkness (0-100). From Table 5 we conclude that the dyed polyester fabric with high L^* value (62.35-75.58), a^* value (2.75-8.17), and b^* value (1.75-16.43) for dyed the polyester fabric. The value of K/S refer to the amount of dye which absorbed on the the fiber. Table 5 shows that dyes 4b and 8b exhibit the highest value of K/S and the polyester fabrics give high affinity for some prepared dyes.

Table 4. Biological activity of some newly synthesized compounds

Sample		Inhibition zone diameter (mm/mg sample)			
		<i>Escherichia coli</i> (G ⁻)	<i>Staphylococcus aureus</i> (G ⁺)	<i>Aspergillus flavus</i> (Fungus)	<i>Candida albicans</i> (Fungus)
Control: DMSO		0.0	0.0	0.0	0.0
Standar	Ampicillin antibacterial agent	22	18	--	--
	Amphotericin B antifungal agent	--	--	16	19
4a		14	18	0.0	0.0
4b		16	17	0.0	12
4c		14	18	0.0	0.0
6a		10	11	0.0	0.0
6b		13	14	0.0	0.0
6c		12	13	0.0	0.0
8a		12	13	0.0	0.0
8b		14	16	0.0	0.0
8c		13	16	0.0	0.0

Table 5. Color strength K/S, L, a, b, ΔE value of polyester fabrics dyed by disperse dyes

Samples	K/S	L	a	b	ΔE
4 b	2.63	63.40	3.71	7.91	21.53
6 a	1.84	75.58	6.87	9.19	14.03
6 b	2.12	71.45	8.17	16.43	22.01
6 c	2.00	72.50	6.49	9.59	15.91
8 a	1.96	71.58	5.12	8.48	15.35
8 b	2.37	62.35	2.75	1.75	20.84

Table 6. Fastness properties of polyester fabrics dyed by disperse dyes

Dyes	Washing fastness			Rubbing fastness		Perspiration fastness						Sublimation		Light fastness	
	Alt.	St.*	St.**	Dry	Wet	Acidic			Alkaline			210	180		
						Alt.	St.*	St.**	Alt.	St.*	St.**				
4b	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	3-4
6a	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4
6b	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4	4-5	4-5	4
6c	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4	4-5	4-5	4
8a	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4
8b	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	3-4

St.* Staining on cotton

St.** Staining on wool

Alt. Alteration in colour

3.5. Fastness properties

Most polyester fabric which dyed by using quinone cyanine dyes were examined towards light, sublimation, rubbing, perspiration and washing and showed promising results. The results are recorded in Table 6. From Table 6 we could conclude that the light fastness properties were good (3-4) while (washing, fastness, dry and wet crocking, both acidic and alkaline perspiration) test gave a very good (4-5).

4. Conclusion

From all the previous discussed results we summarized that:

1. The (bathochromic, hypsochromic) shifts, number of bands and intensity of bands for the new prepared quinone cyanine dyes depending upon:

a- The nature of quaternary salt residue(pyridinium dye < quinolinium dye in monomethine cyanine dyes, picolinium dyes < quinaldinium dyes in the trimethine and pentamethine cyanine dyes).

b- Linkage position(isoquinolinium dye < quinolinium dye in monomethine cyanine dyes , α -picolinium dyes < γ – picolinium dyes in trimethine and pentameth -ine cyanine dyes).

c- The number of methine units in the order of: (pentamethine cyanine dyes > trimethine cyanine dyes > monomethine cyanine dyes).

2. The intensity of the colours of (mono, tri, and penta)methine cyanine dyes (4a-c, 6a-c, 8a-c) are discussed according to the presence of two mesomeric structures (A) and (B) producing a delocalized positive charges over the conjugated system of the quinone cyanine dyes. Scheme (2).

3. The quinone dyes (4a-c, 6a-c, 8a-c) can be used:

a- As photographic sensitizers in industry due to their spectral properties.

b- As antimicrobial agents against some(fungal and bacterial) strains due to their biological activity and promising results. Antimicrobial activity depending upon: i-Types of dyes [penta, tri, mono], ii – Nature of quaternary salt residue (A) [quinolinium, pyridinium, quinaldinium and/or picolinium salt residue], iii-Linkage positions [γ -picolinium, α -picolinium, isoquinolinium and/or quinolinium] and kinds of bacterial and fungi strains.

4. The dyeing process and fastness properties of the synthesized quinone cyanine dyes were examined on polyester fabric. The polyester fabrics give high affinity for some prepared quinone dyes.

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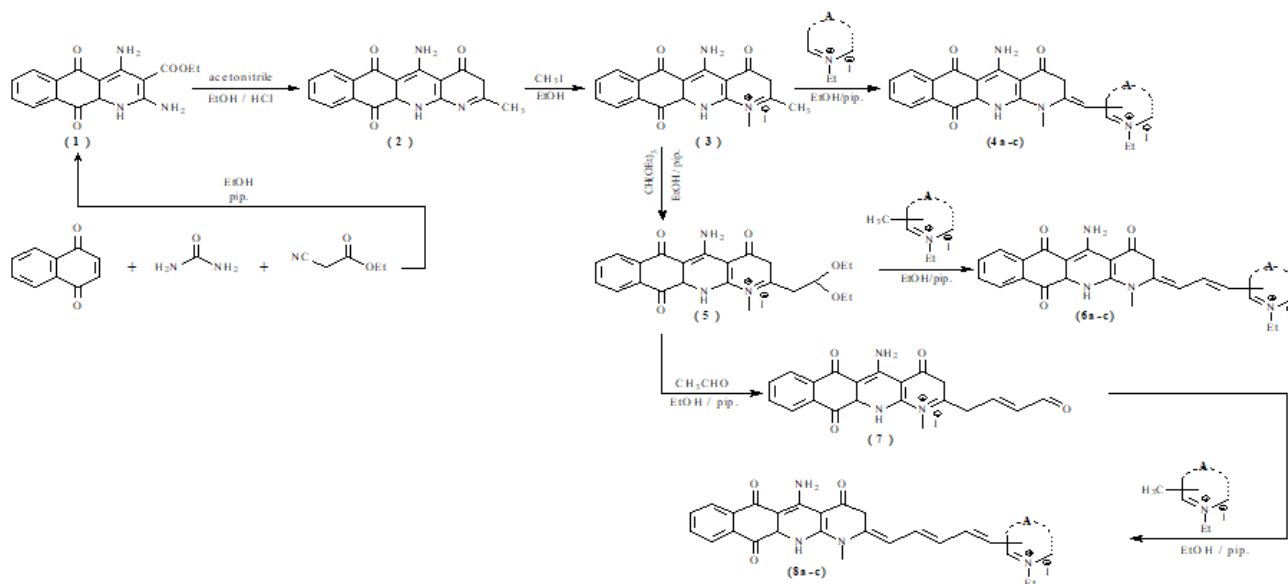
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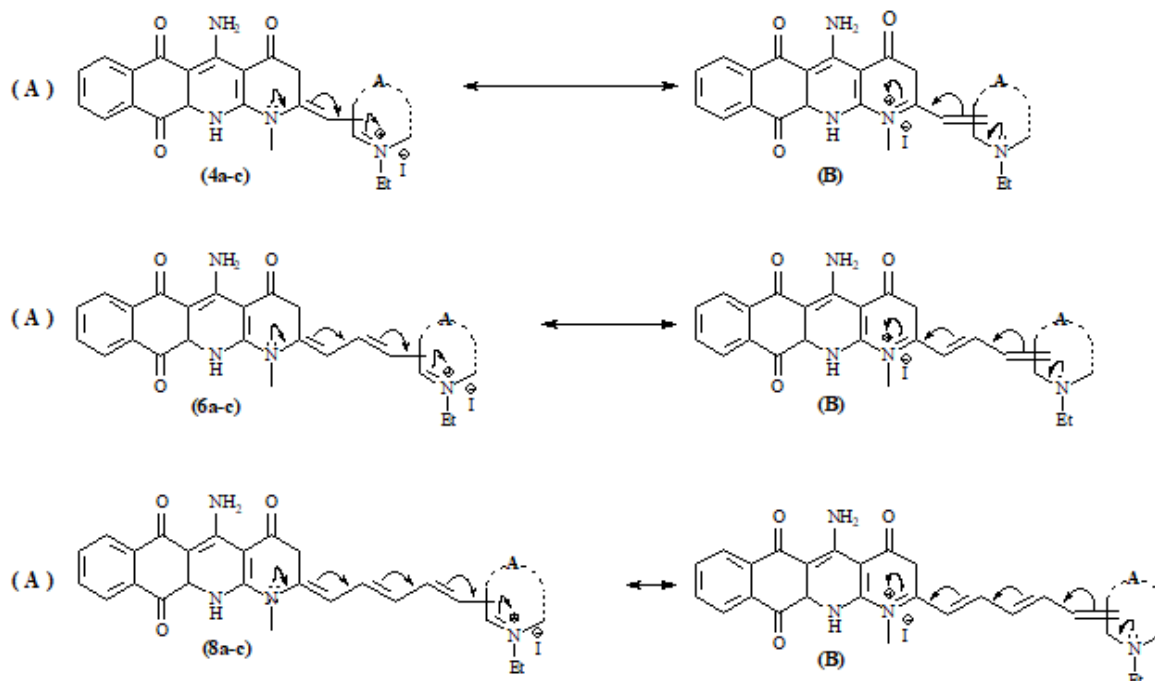
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Appendix

(4a-c): A = 1-ethyl pyridinium-4-yl salt (a),
 A = 1-ethyl quinolinium-4-yl salt (b),
 A = 2-ethyl isoquinolinium-1-yl-salt (c).
(6a-c)& (8a-c): A = 1-ethyl pyridinium-2-yl salt (a),
 A = 1-ethyl quinolinium-2-yl salt (b),
 A = 1-ethyl pyridinium-4-yl-salt (c).



Scheme (1). Synthesis strategy of the prepared compounds (3), (4a-c), (5), (6a-c), (7) and (8a-c)



Scheme (2). Colour intensity and the electronic charge transfer pathways illustration of the synthesized monomethine cyanine dyes (4a-c), trimethine cyanine dyes (6a-c) and pentamethine cyanine dyes (8a-c)