Evaluation of colouration properties of newly synthesized curcumin derivatives

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Received 26 February 2018
Accepted 25 July 2018

Egyptian Pharmaceutical Journal 2018, 17:155–162

Background and objective
Turmeric is a widely known spice that comes from the roots of the plant.
Materials and methods
Phenylpyridazine derivatives based on curcumin (Turmeric, *Curcuma longa*) were synthesized via diazotization of curcumin with appropriate aryl amine derivatives such as p-nitro aniline, p-amino benzoic acid and m-amino benzoic acid. The used fabrics were bleached poplin cotton fabric, mill scoured natural silk fabric and mill scoured pure wool fabric. Effects of mordant and fastness properties for treated fabric were evaluated.
Results and conclusion
The structure of the obtained diazonyl derivatives has been confirmed from their spectroscopic data (ultraviolet, IR and 1H-NMR). Comparable novel curcumin derivatives with curcumin have been investigated with and/or without simultaneous mordanting for natural fibres (cotton, silk and wool). A variety of colours have been obtained by using different mordants. The fastness properties of the dyed fabrics to washing and light fastness were improved.

Keywords:
curcumin, diazotization, modified natural dyes, mordant, natural fabrics, new phenylpyridazine derivatives, textile colouration, ultraviolet

Egypt Pharmaceut J 17:155–162

1687-4315

Introduction

Turmeric is a widely known spice that comes from the roots of the plant, *Curcuma longa*. Physical and classification name is listed in Table 1. The root consists of central rhizome with numerous short ‘fingers’ branching from it. The root’s colour is brownish-yellow but may be lighter or darker, according to plant origin. The flesh inside the root is yellow to orange-yellow [1–8].

A mixture of three polyphenol pigments synthesized in plant’s rhizomes produced turmeric’s yellow hue. These pigments’ collection is called curcumin and consists of curcumin (the dominant pigment), methoxy curcumin and bis-methoxy curcumin.

Curcumin is a polyphenolic compound [1,7-bis-(4-hydroxyl-3-methoxy phenyl)-1,6-heptadiene-3,5-dione] that can exist at least in two tautomeric forms. It undergoes rapid degradation by hydrolysis in alkaline conditions or upon exposure to ultraviolet (UV) radiation [3,4,9].

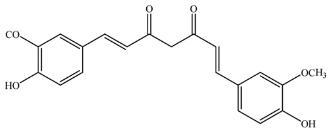
The present investigation is focused on the synthetic, structural and application aspects of monoazo compound derived from diazotized aromatic primary amines and active methylene coupling components such as β-dicarbonyl compound and their metal complexes [10–13]. Thus, curcumin derivatives have been evaluated for a comparative study of their colouration properties to that of curcumin itself when used as a natural dye for natural fibres (cotton, wool and silk).

Materials and methods

Natural colouring matter (natural dye)

Curcumin (Turmeric, *C. longa*), as dry *C. longa* powder, was purchased from local markets in Jazan. It was used after extraction of its colour in water or ethanol as follows: in water, by boiling 20% curcumin powder, leave overnight, and then filter. The filtrate was used as the dyeing solution, or 100 g of *C. longa* powder was extracted using 600-ml absolute ethanol. The solution was soaked for 2 days in a refrigerator and then filtered. The filtrate (dye extract) was used for preparation of curcumin derivatives.

Table 1 Physical and classification name of turmeric

Common name	Turmeric, Olena	
English name	Indian Saffron	
Scientific name	*Curcuma longa* L, Curcumin	
Family	Zingiberaceae	
Group	Monocot	
Dye class	Pigment	
Part used	Root, Rhizome	
Chemical structure	Polyphenol Curcuma	
CI	75 300, natural yellow 3	

Fabrics

(1) Desized, kier boiled, and bleached poplin cotton fabric (165 g/m^2, 40 yarn/cm warp and 36 yarn/cm weft) used in this study has been produced by Misr/Helwan Co. for Spinning and Weaving, Cairo, Egypt.
(2) Mill scoured natural silk fabric (140 g/m^2, 32 yarn/cm warp and 30 yarn/cm weft) was supplied by Hussein M. El-Khatib Sons Co., Suhag, Egypt.
(3) Mill scoured pure wool fabric (220 g/m^2, 24 yarn/cm warp and 22 yarn/cm weft) was supplied by Misr Co. for Spinning and Weaving, Mehalla El-Kubra, Egypt.

Mordant

The mordant used, comprising copper sulphate, ferric chloride, and potassium dichromate, was a laboratory grade chemical.

Chemicals

Chemicals used were of laboratory grade. The purity of the newly synthesized compounds was based on TLC analysis. *p*-Nitroaniline, *p*-aminobenzoic acid, *m*-aminobenzoic acid, sodium nitrite, HCl, sodium acetate anhydrous and ethanol were used.

Synthesis and methods

Amine hydrochloride salt solution of the appropriate aryl amine as 2 mmole *p*-nitroaniline, *p*-aminobenzoic acid or *m*-aminobenzoic acid was added to 5-ml conc. HCl in ice bath at 0–5°C for 10 min; after that, sodium nitrite solution [0.145 g, (2.1 mmole) in 5 ml of water] was added drop wise with stirring to the prepared amine hydrochloride salt solution in 20–25 min at 0°C.

Sodium acetate anhydrous (5 g) in 100 ml ethanol and curcumin (0.67 g, 2 mmol) were added to a well with cold and stirred solution of amine hydrochloride salt at 0–5°C. Stirring was continued for additional 2 h. Then the solution was left overnight in the refrigerator. Cold water (250 ml) was added, and the solid formed was collected by filtration and crystallized from the appropriate solvent to get compounds 3a-c (see Scheme 1).

3a: (60% yield), (Ethanol) [14], chemical formula: $C_{28}H_{26}N_3O_8$ (532.53), elemental analysis: C: 63.27%; H: 4.81%; N: 7.71%; O: 24.22%, UV at λ_{max}: 410 nm. I.R. (ν/cm^{-1}): ^{1}H-NMR (DMSO-d$_6$): δ/ppm=, M.S.: (E.I.) m/z%=530 (M$^+$, 100%)
3b: (62% yield), m.p. (Ethanol)=157–9°C, chemical formula for $C_{28}H_{24}N_2O_8$ (516.51), elemental analysis: C: 65.53%; H: 5.12%; N: 5.27%; O: 24.08%, UV at λ_{max}: 475 nm. I.R. (ν/cm^{-1}): 3420–3358 cm^{-1} (broad OH) and 1735 cm^{-1} (CO). ^{1}H-NMR (DMSO-d$_6$): δ/ppm=4.15 (2s, 6H, 2 OCH_3), 5.06 (2bs, 2H, OH exchangeable with D_2O), 3.12, 3.72 (2s, 2H, 2 CH), 7.07-6.93 (m, 10H, Ar-H), 6.56, 6.34 (2s, CH=CH), and 10.34 (bs, H, OH, exchangeable with D_2O). M.S.: (E.I.) m/z%=515 (M$^+$, 56%), 65 (100).
3c: (55% yield), m.p (Ethanol)=230–2°C, Analytical Calculated for $C_{28}H_{24}N_2O_8$ (516.51), C: 65.33%; H: 5.12%; N: 5.35%; and O: 24.2%; UV λ_{max}: 460 nm. I.R. (ν/cm^{-1}): 3420–3358 cm^{-1} (OH) and 1735 cm^{-1} (CO). ^{1}H-NMR (DMSO-d$_6$): δ/ppm=4.15 (2s, 6H, 2 OCH_3), 5.06 (2s, 2H, OH exchangeable with D_2O), 3.12, 3.62 (2s, 2H, 2 CH), 7.07-6.93 (m, 10H, Ar-H), 6.56, 6.34 (2s, CH=CH), and 10.34 (bs, H, OH, exchangeable with D_2O), M.S.: (E.I.) m/z%=515 (M$^+$, 56%) 65 (100).

Dyeing of cotton, silk and wool fabrics

Dye bath was made by using 2% dye solution, and the mordant was used simultaneously in the dyeing bath with a concentration of 5%. The dye bath with a fabric to liquor ratio of 1 : 20 was maintained at 50°C for 60 min. The dyed samples were taken out and quizzed gently and then dried.

Washing

The dyed fabrics were washed as follows: rinsing thoroughly with cold water, then treatment at 45°C with a solution containing 2 g/l of nonionic wetting agent for 15 min, and after that, rinsing with cold water and then air drying.

Analysis and measurements

All melting points are uncorrected and determined by the open capillary method using Gallen Kamp melting point apparatus. Dye absorbance properties in aqueous solutions were measured using 6800 UV/Vis spectrophotometer from JENWAY, Faculty of Science and Arts-Samtah, Jazan University. IR Spectra (KBr, cm^{-1}) have been obtained using Perkin Elmer 580 Spectrophotometer, Faculty of Science and Arts-Samtah, Jazan University. 1H-NMR was carried on JNM, FTNR-EX 270, run 1H-NMR 270 MHz, in DMSO-d6 using TMS as a standard, Cairo University. Mass Spectra recorded using Varian Mat 112 Spectrophotometer.

Colour measurements [15–20]

Measurements of the colour strength for dyed fabrics (expressed as K/S) have been done at the wavelength of maximum absorbance (l=410, 475 and 460 nm) using the Hunter Lab Ultra-Scan Pro, at the National Research Centre, Egypt. K/S value of the fabrics was evaluated by reflectance technique according to the Kubalka–Munk equation as follows:

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

where K is the absorption coefficient, S is the scattering coefficient, R_o is the reflectance of uncoloured (white) sample and R is the reflectance of coloured sample.

Fastness properties

The colour fastness to washing was determined according to the AATCC Test method 61–2007 using Laudner-Ometer. Colour fastness to light was determined according to AATCC test method (16A–2004). The evaluation was established using the blue scale as reference of colour change [21–25].

Results and discussion

Diazotization is the reaction of a primary aromatic amine with a nitrosating agent, such as sodium nitrite or to a lesser extent with nitrosylsulfuric acid $NOSO_4H$, nitrous gases or organic nitrites in an aqueous acidic solution at a temperature between 0 and 5°C, converting the amine to its diazonium salt:

$$Ar{-}NH_2 + 2\ HY + Na{-}NO_2 \longrightarrow Ar{-}\overset{\oplus}{N}{\equiv}N\ \overset{\ominus}{Y} + Na{-}Y + H_2O$$

The active methylenic carbon in these compounds can act as nucleophilic centre for the electrophilic attack. The reaction with ArN^{+2} can be represented as Scheme 1 and the resultant product is capable of keto-enol as well as azo-hydrazone (Scheme 1).

The diazenyl derivatives based on curcumin were prepared by the diazotization of curcumin with appropriate aryl amine derivative such as *p*-nitroaniline, *p*-aminobenzoic acid and *m*-aminobenzoic acid, correspondingly. The structures and the characterization data for these compounds are also summarized in Scheme 1 and the experimental part individually.

Colouration properties

To start with the evaluation of curcumin as a natural dye, cotton, silk and wool fabric samples were dyed with curcumin water extract solution in presence or absence of mordant using simultaneous mordanting method. Another series of samples was prepared via dyeing of natural fabrics (cotton, silk or wool) with the prepared curcumin derivatives in 50% ethanol/water

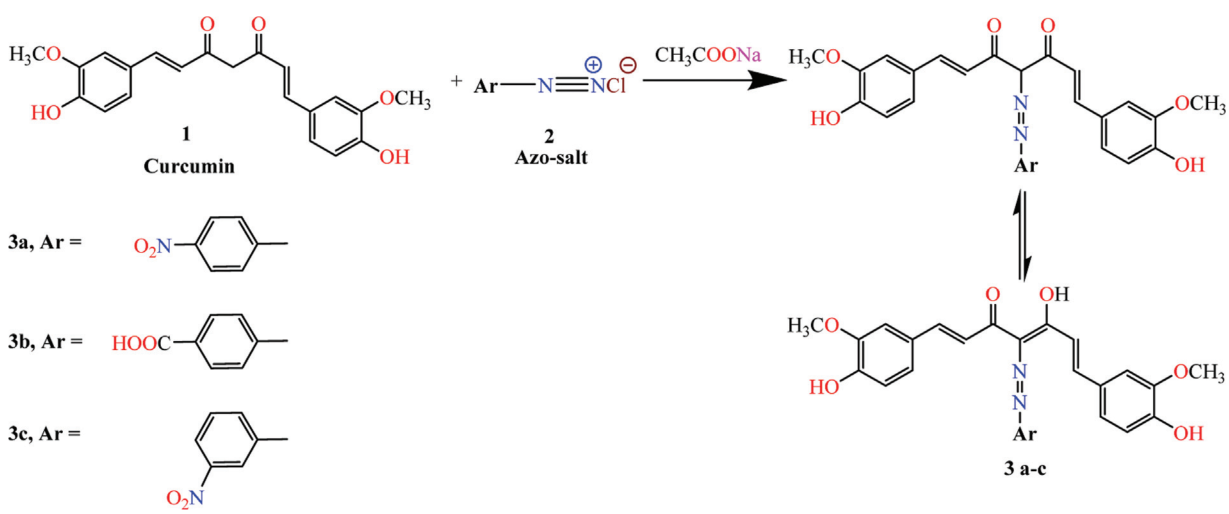
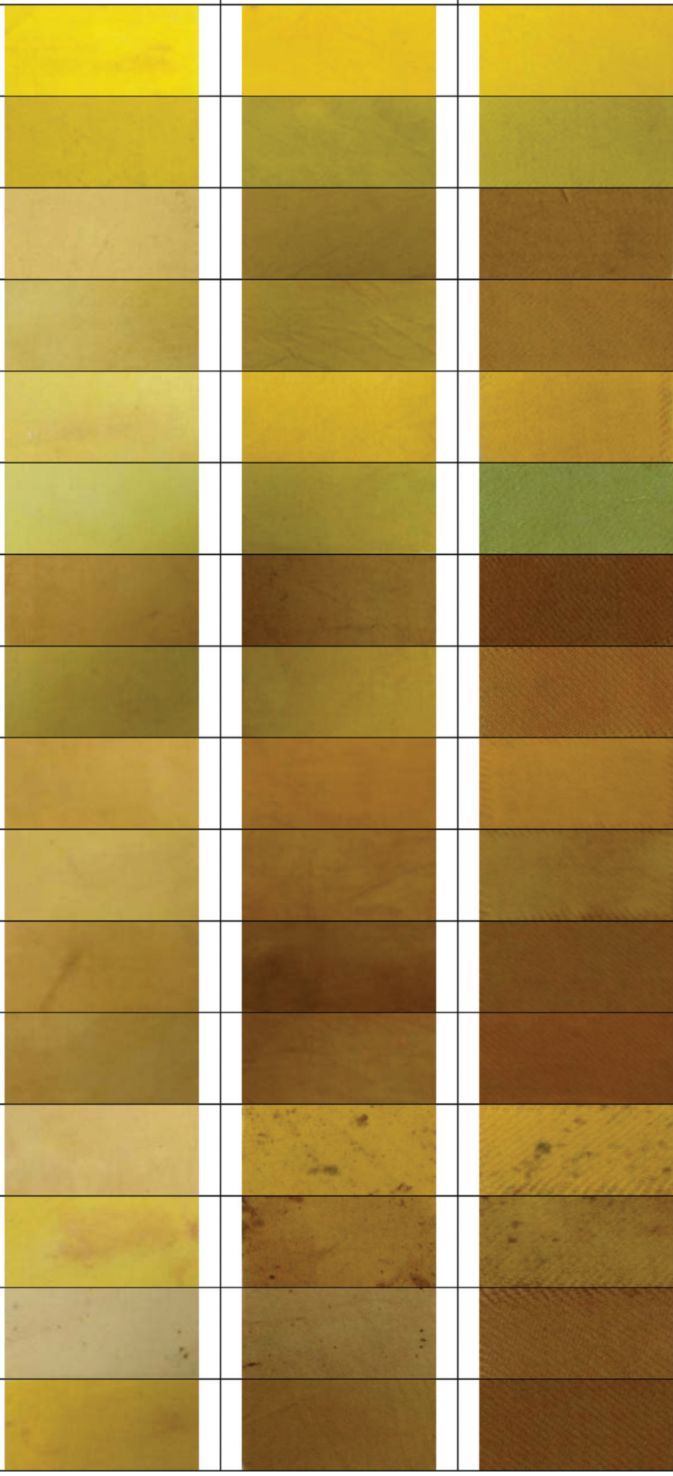


Table 2 Effect of mordant on the colour of fabrics dyed using curcumin and its derivative

Dye used	Mordant used	Cotton	Silk	Wool
Curcumin	Without mordant			
	Copper sulphate			
	Ferric chloride			
	Potassium dichromate			
Curcumin derivative (with *p*-nitro aniline)	Without mordant			
	Copper sulphate			
	Ferric chloride			
	Potassium dichromate			
Curcumin derivative (with p-amino benzoic acid)	Without mordant			
	Copper sulphate			
	Ferric chloride			
	Potassium dichromate			
Curcumin derivative (with m-amino benzoic acid)	Without mordant			
	Copper sulphate			
	Ferric chloride			
	Potassium dichromate			



mixture with or without simultaneous mordanting (Table 2). The results obtained are represented in Table 2.

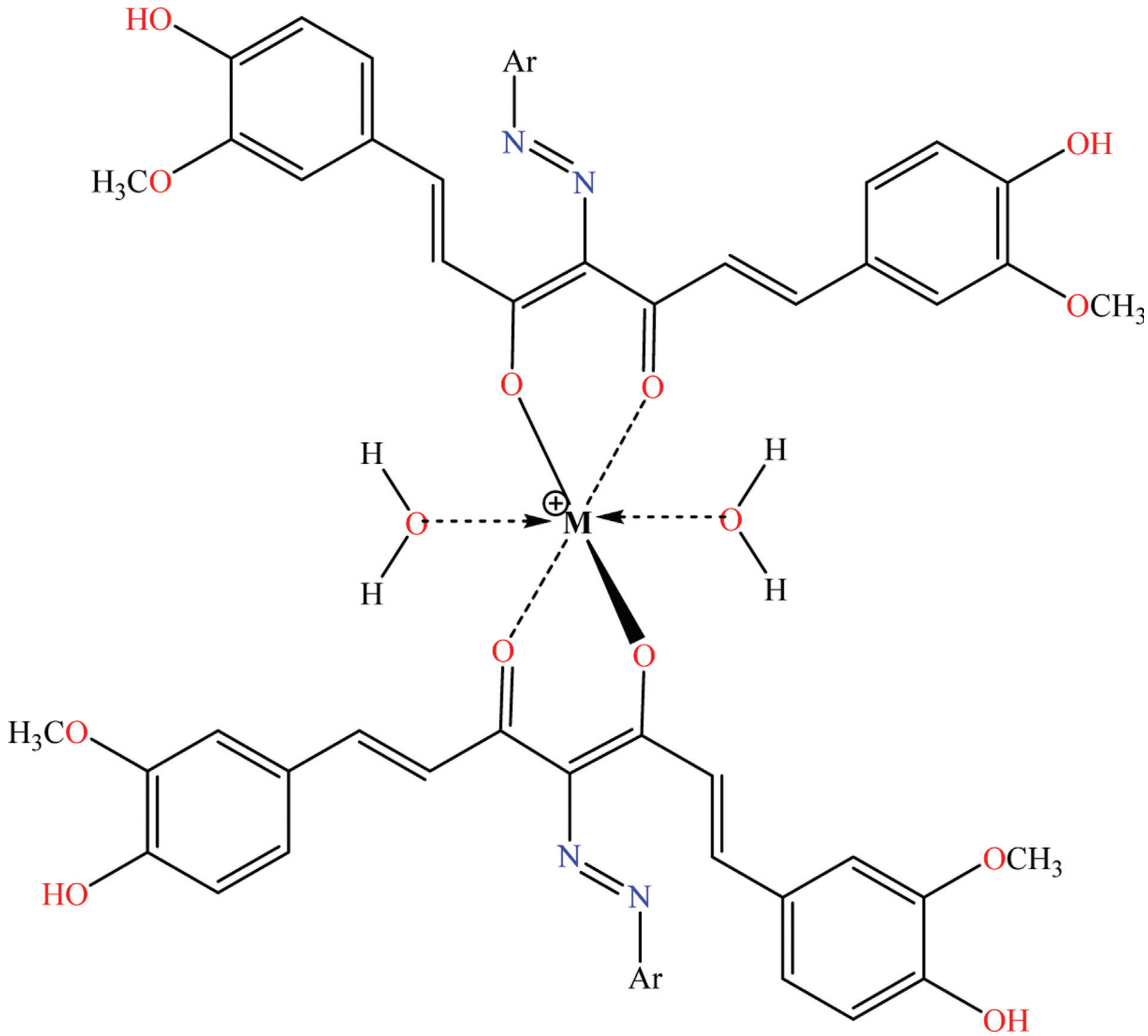
The absorbance spectra of dye solutions have been also studied to investigate the effect of the coupling reaction on absorbance wavelength of the dye chromophore. The data show that the absorbance spectrum of curcumin (λ_{max} 425 nm) is affected by the substituent in the aryl amine used. A hypsochromic shift is observed in case of coupling with *p*-nitroaniline where the spectral peak is shifted to 410 nm. However, in case of coupling with aminobenzoic acids, a pathochromic shift is observed, and the dye spectral peak obtained was 475 and 460 nm for products obtained upon coupling with *p*-amino benzoic acid and *m*-amino benzoic acid, respectively.

Table 2 shows that the colour strength of the dyed samples (K/S) depends on the following: (a) coupling agent used, (b) mordant used, and (c) fabric used. The data show that in most cases, regardless of the coupling agent and/or mordant used, the K/S obtained for dyed protein fabrics (i.e. silk or wool) is higher than their corresponding samples of cotton. These results (i.e. the higher K/S in case of protein fabrics) may be explained as follows: in case of protein fabrics, three types of bonds are formed namely: hydrogen bond, ionic bond and covalent bond, and in case of cellulosic (cotton) fabrics, the curcumin derivatives could form only two types of bonds that are hydrogen bond and covalent bond [22].

Effect of mordantMordants are usually added on using natural dye to bind the dye better to the fabric, keep natural dye from fading, improve colour fastness properties and/or deepen or dull the colour to obtain a full range of colours [17,21,23,26]. A variety of colours may be obtained for the same dye by using different mordants, as it is clear from Table 2. These colour changes may be attributed to the complex formation between the dye and a mordant.

In literature Moustafa *et al.* [5], the complex formation between curcumin and Cu(II) ion as an example of metallic mordant have been investigated via

Figure 1



The formed complex with metallic mordant (M).

Table 3 Fastness properties of fabrics dyed using curcumin and its derivative in absence or presence of different mordants

	Fabric used	Mordant used	K/S	Washing fastness		Light fastness
				Colour change	Staining on cotton	
Curcumin	Cotton	Without mordant	2.65	3	3–4	3
		Copper sulphate	3.2	3–4	4	4
		Ferric chloride	2.41	3–4	3–4	3
		Potassium dichromate	2.78	4	4	4
	Silk	Without mordant	2.96	4	4	4
		Copper sulphate	3.11	4	4	5
		Ferric chloride	2.86	4	4	4
		Potassium dichromate	2.58	4	4	5
	Wool	Without mordant	2.70	4	4	4
		Copper sulphate	3.8	3–4	3–4	4
		Ferric chloride	3.12	4	4	3
		Potassium dichromate	3.19	4–5	4–5	4
Curcumin derivative with *p*-nitroaniline	Cotton	Without mordant	2.14	4	4	3
		Copper sulphate	3.2	4	4–5	4
		Ferric chloride	2.32	3–4	4	3–4
		Potassium dichromate	2.67	4–5	4–5	4–5
	Silk	Without mordant	2.42	3–4	3–4	4
		Copper sulphate	3.10	4–5	4–5	4–5
		Ferric chloride	2.65	4	4–5	4
		Potassium dichromate	3.12	4–5	4–5	4–5
	Wool	Without mordant	3.17	4	4	4
		Copper sulphate	3.88	4	4	4
		Ferric chloride	3.83	3–4	4	4
		Potassium dichromate	4.05	4	4	4
Curcumin derivative with *p*-amino benzoic acid	Cotton	Without mordant	4.78	4	4	3–4
		Copper sulphate	4.56	4–5	4–5	4
		Ferric chloride	4.50	4–5	4–5	3
		Potassium dichromate	4.34	4–5	4–5	4
	Silk	Without mordant	5.8	4–5	4–5	4
		Copper sulphate	4.22	4–5	4–5	4
		Ferric chloride	5.01	4–5	4–5	4–5
		Potassium dichromate	5.63	4–5	4–5	4–5
	Wool	Without mordant	5.54	4	4	4
		Copper sulphate	5.00	4–5	4	4–5
		Ferric chloride	4.89	3–4	4	4–5
		Potassium dichromate	4.77	4–5	4–5	4
Curcumin derivative with *m*-amino benzoic acid	Cotton	Without mordant	2.78	4	4	4
		Copper sulphate	3.56	4–5	4–5	4
		Ferric chloride	2.98	4	4	3–4
		Potassium dichromate	3.14	4–5	4–5	4
	Silk	Without mordant	4.22	4	4	4
		Copper sulphate	4.67	4–5	4–5	4
		Ferric chloride	3.74	4	4	4
			4.63	4	4	4–5

(*Continued*)

Table3 (*Continued*)

	Fabric used	Mordant used	K/S	Washing fastness: Colour change	Washing fastness: Staining on cotton	Light fastness
		Potassium dichromate				
	Wool	Without mordant	4.45	4	4	4
		Copper sulphate	4.70	4–5	4	4–5
		Ferric chloride	3.20	4	4	3–4
		Potassium dichromate	4.65	4–5	4–5	4–5

spectrophotometric methods. A significant hypsochromic shift in the absorbance wavelength has been observed. Other metallic mordant ions are also able to form such complexes. These complexes are able to be directly applied for textile dyeing giving a variety of colours. The complex formed in case of metallic mordant (M) may be represented as presented in Fig. 1.

The data also obtained show that the presence of mordant causes an increase in the colour strength (K/S) of the dyed fabric samples, and an increase in the K/S in the presence of mordants may be owing to the fixation of the colour by the mordant.

Fastness properties

The fastness properties of the dyed fabric samples to washing and to light for curcumin and the prepared curcumin derivatives are shown in Table 3. The data show that the washing fastness properties of these samples are nearly comparable regardless of the following:

(1) The nature of curcumin derivative used.
(2) The fabric used, that is, cotton, silk, or wool.
(3) The type of mordant used.

The data in Table 3 also show that the washing fastness properties range from 4 to 4–5. The light fastness for the dyed fabrics is improved by using mordant and/or by using curcumin derivatives. The light fastness properties obtained upon using curcumin itself as a natural dye ranges from 3 to 4, whereas upon using curcumin derivatives, the fastness properties ranges from 3 to 5. The improvements of the light fastness properties owing to the presence of mordant or by using curcumin derivatives reflect the increase in stability of the dye molecules owing to complex formation with the metallic mordant and/or via coupling with aryl amines.

Conclusion

Phenylpyridazine derivatives based on curcumin have been prepared via diazotization with aryl amines such as *p*-nitroaniline, *p*-aminobenzoic acid and *m*-aminobenzoic acid. A hypsochromic shift was observed for compound 3a, whereas a pathochromic shift was observed for compounds 3b and 3c. A variety of colours have been obtained by using different mordants. The colour strength of the dyed samples (K/S) depends on the type of coupling agent used, type of mordant, and type of dyed fabric. The fastness properties of the dyed fabrics to washing were comparable. However, the light fastness for the dyed fabrics is improved by using mordant and/or by using curcumin derivatives as modified natural dyes.

Financial support and sponsorship

Nil

Conflicts of interest

There are no conflicts of interest.

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