Research Article

Preparation of nano disperse dye based on benzopyran in one pot reaction using microwave irradiation and its appliance in textile printing

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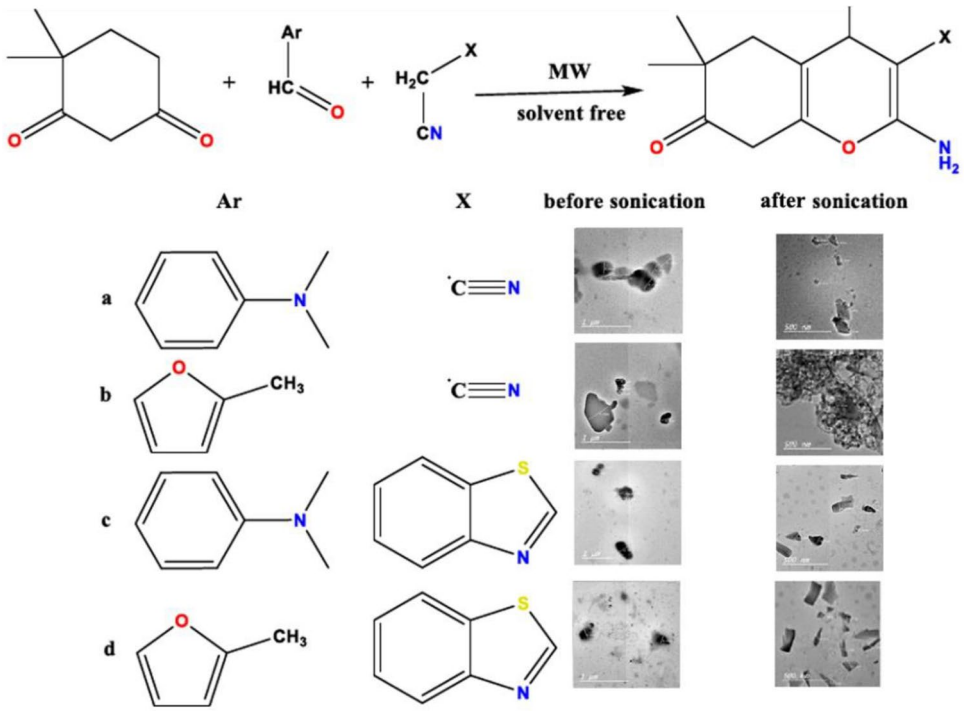
Abstract
This paper aims to synthesis some novel of benzopyran derivatives using high efficient, solvent free, excellent yield, one pot multicomponent reaction via microwave technique. The prepared compounds are subjected to ultra-sonication to reduce their particle size to nano size and used as a pigment in printing of polyester/cotton, polyester and linen fabrics the fastness properties of the printed fabrics are very good. The structures of prepared pigment are confirmed using (element analysis, IR, H-NMR and MS). The size of prepared and treated dyes are identify by TEM.

Graphic abstract Graphic abstract explain the suggested scheme for synthesise the hyper branched compound I with TEM image for the different resulting compounds with different moieties before and after sonication.



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1 Introduction

Benzopyran compounds are belongs according to its structure to natural product which have a lot of interesting biological and pharmaceutical application [21, 38]. 4-H-chromene derivatives received a considerable attention in the last decades because it act as starting compounds for anticancer, anticoagulant, and biodegradable agrochemicals. Only few research articles reported it as a pigment without further application [13, 33].

There are a lot of reported methods for preparation of 4-H-chromene (benzopyran) derivatives via multi-component reaction including of catalyst such as acetic acid, and organo-catalyst. These methods leads to low yield, waste time [14, 35, 36, 51].

Disperse dye normally known as an extensively water insoluble dye with an affinity to hydrophobic fibres only. They are usually utilized in the textile industry to impart a colour to synthetic fibers, for example polyester, acrylic and acetate. Before using, disperse dye converted to a low particle size dispersed in water and added to treatment bath during textile coloration process.

Increasing awareness of environmental issues around the world make various treatments such as UV, ultrasonic, gamma radiation and microwave in the light spot for many scientist who are working in the field of textile wet treatments (pre-treatment and dyeing) to improve the quality of the dyed fabrics and reduce environmental pollution in the same time. UV radiation is mainly used in the pretreatment of both the fabric and dye solution separately to lower the dyeing conditions (time and temperature) and enhance the colour strength and fastness properties of treated fabrics [9, 16, 20, 22, 27, 28]. The same results were noticed by using the gamma radiation and plasma on different fabrics [8, 44]. The microwave radiation application in textile takes the attention of researchers as an alternative eco-friendly heating system. Shahid et al. improve the color strength and fastness properties of polyester fabric dyed with disperde Yellow 211 dye under the influence of microwave for 6 min [10, 11, 15, 23]. Limited study has been performed on the effect of radiation on the synthesis of organic dyes itself. The synthesis of benzopyran is obtained from different kinds of starting materials using basic catalysts [52] or by condensation reactions [30, 40, 50]. Most of these methods has limitations such as long time of reaction, consume an expensive strongly basic materials, tedious work-up and low products. Chaturvedi et al. [19] employ the microwave radiation in the synthesis of benzopyrans from substituted acetophenones and keto compounds mediated by Triton-B under solvent-free conditions and mild condition in one pot reaction given excellent yield.

Introduction of various radiations, for example, ultraviolet [41] and microwave [32] treatment which can impart better color fastness properties to the modified fabrics [12, 17, 43]. Some researchers have found that, microwave treatment improve the extraction of color from their natural source, which offers deep shading to the fabrics [32]. Microwave can equitably and effectively enter into a medium and consistently heat the substance [31, 34]. During microwave treatment, polar groups of the medium are excited which results into heat creation. Past investigations have demonstrated that, microwave treatment didn't change the micro fibrils of cellulose [49] and the physical properties for example, wrinkling and tensile strength were highly improved.

Ultrasound (US) improve a wide range of both chemical and physical processes; mostly because of the phenomenon known as cavitation inside the liquid medium that is the development and breakdown of tiny bubbles which can create a hot spots [14], for example restricted high temperature and pressure, shock waves and extremely shear forces equipped for breaking bonds. Many researchers have been investigate this method in the textile coloration as it is a noteworthy wet process, which required high energy, water and releases huge effluent to the environment. In addition any enhancements in coloration processes are generally ascribed to the cavitation phenomena [35, 51] and, as an outcome, other mechanical and chemical impacts are as listed below:

- Decreasing any agglomerations with high molecular mass;
- Remove any dissolve/entrap air from cavities;
- Accelerate the diffusion rate of dye;
- Powerful agitation of the liquid;
- Breaking down the diffusion layer interfaces;
- Producing a free radicals.

This work combine the using of microwave radiation in the preparation of some novel heterocyclic compounds based on 4-Hchromene derivatives using in one pot multicomponent reaction then the produced disperse dyes were subjected to sonication to reduce its particle size and be applied as nano-pigment in printing of linen, polyester and cotton/polyester fabrics.

The aim of this work is to print nature fibre as linen and cotton/polyester blend using the solvent free nano disperse dye prepared with high colour strength and good to excellent fastness properties via pigment printing

technique. The advantages of both economic and environmental, points of view using of solvent free technique and green tool as microwave are very interesting. So this article report preparation of some novel heterocyclic compounds based on 4-Hchromene derivatives using eco-friendly techniques (solvent free one pot multi-component reaction and microwave heating). The prepared compounds are sonicated to reduce its particle size and used as nano-pigment in printing of linen, polyester and cotton/polyester fabrics.

2 Experimental

2.1 Materials

Linen, cotton/polyester. (40/60) and polyester fabric are supplied by a private sector company. Malononitrile, 2-(benzo[d]thiazol-2-yl)acetonitrile, 2-methyl cyanobenzothiazole, dimedone and benzaldehyde derivatives, tetramethyl silane, sodium dihydrogen phosphate dehydrate, all chemicals were purchased from Merck (Germany) and were used without further purification. Commercial binder, namely EBCAPRINTTB manufactured by Egyptian British Company was used. Commercial synthetic thickener namely Printofix thickener MTB 01 liq. (Clariant Company).

2.2 Synthesis

A mixture of malononitrile, or 2-methyl cyanobenzothiazole 2 (0.01 mmol), dimedone 3 (0.01 mmol) and benzaldehyde derivatives 1 (0.01 mmol) were mixed and subjected to microwave radiation (multimode Milestone MW reactor with a frequency of 2.45 GHz and maximum MW power of 1200 W) for 10 min. The reaction progress was observed using thin layer chromatograph (TLC). After the reaction was completed, the mixture was poured into ice water then filtered (Fig. 1).

2.3 Ultra-sonication of pigment

A 3 g of disperse dyes were suspended in 100 ml water under stirring with ultrasonic stirrer; 80% power and 35 frequency (SONICS & MATERIALS, INC), Model: VCX750, Volts: 230 V AC50l60 HZNOM, USA) for 30 min at room temperature then added to the printing paste according to the following recipe.

Prepared dye	20 g of dye suspend
Urea	2.5 g
Synthetic thickener	50 g
Binder	5 g
Sodium dihydrogen phosphate dehydrate	0.5 g
Distilled water	Y
Total	100

2.4 Characterisation

The melting points for synthesized dyes were investigated using a melting point apparatus. FT-IR, nuclear magnetic resonance, Mass spectra and transmission electron microscope of the untreated and treated samples were recorded using FT-IR spectrometer (JASCO connected with ATR), Varian 400 or Wilmad 270-MHz spectrometer, Varian MAT112 spectrometer and JEOL JEM-1230 equipment respectively. Perkin Elmer double-beam spectrophotometer is used to measure Absorption spectra.

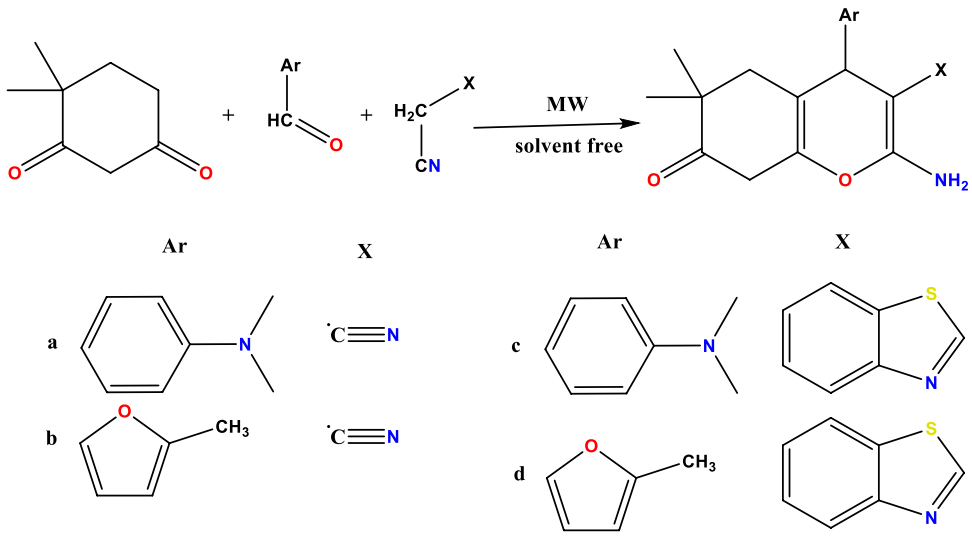

Fig. 1 Scheme for synthesis of hyper branched composite I

Colour strength values of the dyed fabrics was measured using reflection spectroscopy (Hunter Lab UltraScanPRO USA, 2007) at the wavelength of the maximum absorbance and it was expressed as K/S which assessed by applying the Kubelka–Munk equation [6, 7, 22, 24, 25, 45, 46]. Fastness properties of dyed fabric to washing, crocking (dry and wet rubbing), perspiration and light were determined according to standard methods AATCC Test Method 61-2007, AATCC Test Method 8-2007, AATCC Test Method 15-2013, AATCC Test Method (16-2004) respectively. The evaluation established using the blue scale as reference of colour change [1–4, 26, 27, 47, 48].

3 Result and discussion

3.1 Synthesis

The present research deals with a novel synthesis of functionalized heterocyclic compounds using 4-chromene (benzopyran) derivatives through the reaction of malononitrile, or 2-(benzo[d]thiazol-2-yl)acetonitrile (2), dimedone (3) and benzaldehyde (1) derivatives in a microwave (Fig. 1). The benzopyran derivatives were synthesised via Michael adduct, after that, water elimination and dehydrogenation was carried out using microwave irradiation (Fig. 1). The chemical structure of the pigments were confirmed using different techniques; element analysis, IR, ^{1}H-NMR and mass spectral [5, 18, 37, 42]. The result was illustrated in Tables 1, 2 and 3.

3.2 Effect of sonication on the particle size of the pigment

It is clear from TEM images (Fig. 2a–h) that the sonication reduced the particle size over all the compounds **4a**–**d** from micro scale to nano-scale. The results of transmission electron microscope of nano-benzopyran pigment presented a very homogeneous morphology with quite uniform particle size distribution. The particle size diameters obtained were in the range of 25–70 nm. The conversion of several benzopyran dyes into nano-sized organic pigment has been achieved mechanically via miniaturization using ultrasonic processor for 30 min at

Table 1 Physical and analytical data of synthesized dyes **4a–d**

	Molecular formula	M.wt	Colour	Yield %	λ (nm)	m. p
4a	$C_{20}H_{23}N_3O_2$	337	Greenish yellow	97	375	222
4b	$C_{17}H_{20}N_2O_3$	300	Yellow	97	370	219
4c	$C_{26}H_{27}N_3O_2S$	445	Yellow	94	380	230
4d	$C_{23}H_{24}N_2O_3S$	408	Yellow	94	382	232

Table 2 Elemental analysis, mass and IR spectral data of synthesized dyes **4a–d**

Compounds	FT-IR (KBr, cm^{-1})		Element analysis								Mass spectra
			Theoretical (%)				Found (%)				
	NH_2	C=O	C	H	N	O	C	H	N	O	
4a	3450	1650	71.19	6.87	12.45	9.48	71.11	6.77	12.40	9.39	336
4b	3456	1640	67.98	6.71	9.33	15.98	67.88	6.69	9.21	15.94	299
4c	3440	1667	70.08	6.11	9.43	7.18	70.01	6.08	7.11	6.05	444
4d	3400	1670	67.62	5.92	6.86	11.75	67.55	5.90	6.84	11.65	407

Table 3 ^{1}H-NMR spectral data of the synthesized dyes **4a–d**

	^{1}H-NMR
4a	1.25 (6H, s, 2-CH_3), 3.06 (6H, s, 2-CH_3), 3.17 (2H, s, CH_2), 2.29 (2H, s, CH_2), 3.94 (1H, s, CH), 8.51 (2H, s, NH_2), 6.68–6.97 (4H, m, aromatic protons)
4b	1.25 (6H, s, 2-CH_3), 2.3 (3H, s, CH_3), 3.17 (2H, s, CH_2), 2.32 (2H, s, CH_2), 4.17 (1H, s, CH), 8.59 (2H, s, NH_2), 6.14–7.58 (4H, m, aromatic protons)
4c	1.28 (6H, s, 2-CH_3), 3.11 (6H, s, 2-CH_3), 3.19 (2H, s, CH_2), 2.31 (2H, s, CH_2), 3.89 (1H, s, CH), 8.91 (2H, s, NH_2), 6.64–8.18 (4H, m, aromatic protons)
4d	1.27 (6H, s, 2-CH_3), 2.5 (3H, s, CH_3), 3.21 (2H, s, CH_2), 2.21 (2H, s, CH_2), 4.21 (1H, s, CH), 8.89 (2H, s, NH_2), 614–8.12 (6H, m, aromatic protons)

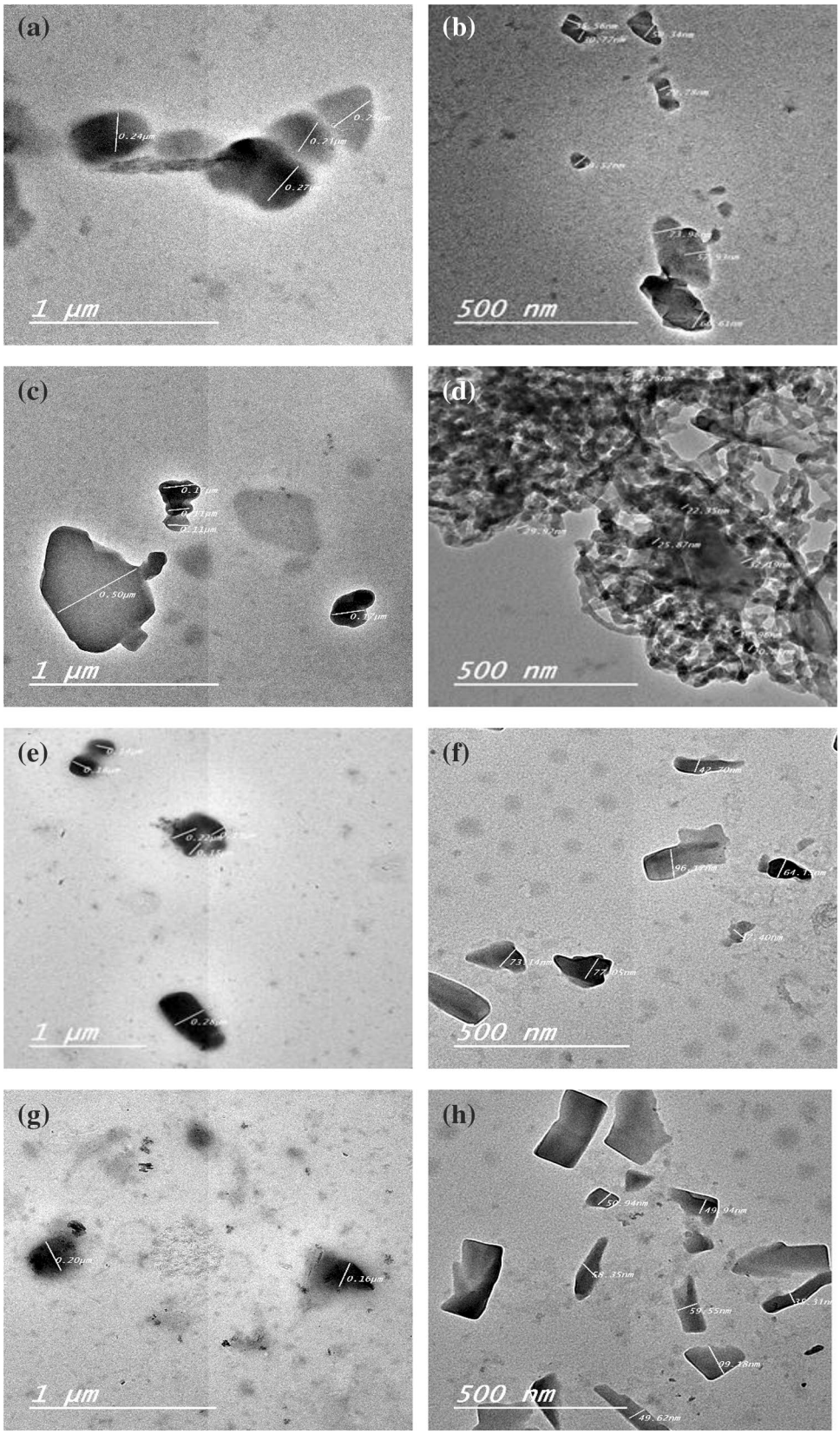

Fig. 2 TEM image 1 compound **a** Compound **4a** before sonication **b** Compound **4a** after sonication **c** Compound **4b** before sonication **d** Compound **4b** after sonication **e** Compound **4c** before sonication **f** Compound **4c** after sonication **g** Compound **4d** before sonication **h** Compound **4d** after sonication

room temperature. This study also investigated the use of the prepared nano-pigment dispersion in printing (linen, polyester and its blend with cotton) with pigment printing paste in presence of binder using thermofixation. The size of nano-pigment obtained and the colour strength values (K/S) are greatly affected by the molecular structure of the prepared benzopyran dyes.

3.3 Colour strength and fastness properties

The colour strength of the pigment printed polyester, linen, and polyester/cotton fabrics using the synthesized pigment (before and after sonication is represented in Tables 4 and 5. It's clear from the tables that all pigment (with normal size) possesses good colour strength value ranging from 8 to 10. Reducing the particle size over all pigments leads to high increasing in K/S values ranging from 12 to 14.7 over all compounds.

To determine the colour parameters and the colour difference of printed fabrics, CIE lab system is used where, (L) values reefer to lightness-darkness values from 100 to 0 representing white to black, (a) value run from negative (green) to positive (red) and (b) values run from negative (blue) to positive (yellow) and the total colour difference is given by ΔE from Tables 4 and 5 it can be noticed that the (L) values decrease in all nano-sized printed samples which indicate that the printed samples (with nano-sized pigment) become more darker than printed samples with micro size pigment. It is expected as by decreasing the size of dye more molecules were capture on the fabric surface and that trend is noticed in several researches (asma, hamad) [29, 39]. From a and b values the colour hue changed to reddish yellow.

Table 4 also represent the colour difference ΔE it is clear that there is a colour difference between the prints using nano-sized pigments and prints with micro size

Table 4 Colour strength and colour parameters of compounds **4a–d**

Pigment	Fabric type	ΔE	b	a	L	k/S	Pigment (**4a**)
4a	Linen	45.67	42.84	−11.66	78.15	6.58	Org.
		47.44	44.8	−10.9	78.82	11.48	Nano.
	Polyester	43.10	40.61	−12.41	80.14	7.95	Org.
		46.01	43.56	−11.95	78.62	12.98	Nano.
	Cotton/polyester	49.28	46.96	−9.61	77.88	7.68	Org.
		50.97	48.45	−9.1	74.85	14.05	Nano.
4b	Linen	86.53	40.5	−0.52	77.93	8.38	Org.
		87.63	46.96	11.67	71.74	12.04	Nano.
	Polyester	79.66	21.12	−0.47	77.59	7.32	Org.
		80.42	35.70	15.47	69.52	11.06	Nano.
	Cotton/polyester	81.85	31.98	−1.66	77.79	8.33	Org.
		84.12	40.61	14.08	69.66	13.94	Nano.
4c	Linen	77.39	16.70	−1.73	77.59	8.91	Org.
		79.09	32.05	−0.26	72.30	14.83	Nano.
	Polyester	80.05	23.89	−1.39	75.66	7.9	Org.
		79.34	18.04	−1.28	77.25	13.16	Nano.
	Cotton/polyester	79.71	16.7	−1.73	71.74	8.73	Org.
		77.39	29.02	1.89	77.18	14.74	Nano.
4d	Linen	86.53	40.5	−0.52	77.93	8.38	Org.
		87.63	46.96	11.67	71.74	12.04	Nano.
	Polyester	79.66	21.12	−0.47	77.59	7.32	Org.
		80.42	35.70	15.47	69.52	11.06	Nano.
	Cotton/polyester	81.85	31.98	−1.66	77.79	8.33	Org.
		84.12	40.61	14.08	69.66	13.94	Nano.

Compound (**4a**): 2-amino-4-(4-(dimethylamino)phenyl)-6,6-dimethyl-7-oxo-5,6,7,8-tetrahydro-4H-chromene-3-carbonitrile

Compound (**4b**) 2-amino-6,6-dimethyl-4-(5-methyl-2,3-dihydrofuran-2-yl)-7-oxo-5,6,7,8-tetrahydro-4H-chromene-3-carbonitrile

Compound (**4c**) 2-amino-3-(benzo[d]thiazol-2-yl)-4-(4-(dimethylamino)phenyl)-6,6-dimethyl-5,6-dihydro-4H-chromen-7(8H)-one

Compound (**4d**) 2-amino-3-(benzo[d]thiazol-2-yl)-6,6-dimethyl-4-(5-methyl-2,3-dihydrofuran-2-yl)-5,6-dihydro-4H-chromen-7(8H)-one

Table 5 Fastness properties of printed samples

	Pigment **4a**	Pigment **4b**	Pigment **4c**	Pigment **4d**
Washing fastness				
Linen				
Alt	4	4	4	4–5
St.	4–5	4	4	4
Polyester/cotton				
Alt.	4–5	4	4	4
St.	4	4–5	4–5	4–5
Polyester				
Alt.	4	4–5	4	4–5
St.	4–5	4–5	4–5	4–5
Rubbing fastness				
Linen				
Alt	4	4	4	4
St.	4	4	4	4
Polyester/cotton				
Alt.	4	4	4	4
St.	4	4	4	4
Polyester				
Alt.	4	4	4	4
St.	4	4	4	4
Perspiration				
Linen				
Alkali				
Alt	4–5	4–5	4–5	4–5
St.	4–5	4–5	4–5	4–5
Acidic				
Alt.	4–5	4–5	4–5	4–5
St.	4–5	4–5	4–5	4–5
Polyester/cotton				
Alkali				
Alt.	4–5	4–5	4–5	4–5
St.	4–5	4–5	4–5	4–5
Acidic				
Alt	4–5	4–5	4–5	4–5
St.	4–5	4–5	4–5	4–5
Polyester				
Alkali				
Alt.	4–5	4–5	4–5	4–5
St.	4–5	4–5	4–5	4–5
Acidic				
Alt.	4–5	4–5	4–5	4–5
St.	4–5	4–5	4–5	4–5
Light fastness				
Linen	5–6	5	5–6	5
Polyester/cotton	5–6	5	5–6	5
Polyester	5–6	5	5–6	5

Alt Alternation, *St* Staining on cotton

pigments where the pigment concentration is constant. All the prints have ranging from good to very good Fatness to rubbing washing, perspiration and light (Table 5).

4 Conclusion

This paper describes the preparation of benzopyran derivatives disperse dye in microwave as an eco-friendly approach and its conversion into nano size using microwave at optimum conditions.

The results obtained show that:

- The particle size diameters obtained were in the range of 25–57 nm for Nanoparticles of the prepared dyes, while that of the original dye particles ranges from 200 to 700 nm.
- The nanostructure of benzopyran derivatives dyes and its application via pigment printing technique bring a series of unique properties: excellent colour fastness, good ecological performance and advanced performance and non-selectivity to various fibers provide a wide market for the product application, and provide a strong technical support to transform the traditional printing and dyeing industry with frontier technology.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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