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A facile synthesis of some biologically active disperse dyes derived from arylazonicotinates and their application on polyester fabrics

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ABSTRACT

A series of 2-hydroxy- and 2-amino-6-substituted-5-arylazonicotinates monoazo disperse dyes **6** and **8a-d** were prepared via condensation of 3-oxo-3-substituted-2-arylhydrazonals **1a-d** with active methylene nitriles **2a**,**b**. A high temperature dyeing method was employed to apply these disperse dyes for polyester fabrics. Fastness properties of the dyed samples were measured. Most of the dyed fabrics tested displayed very good fastness level to washing, perspiration and light. Finally, the biological activities of the prepared dyes against gram positive and gram negative bacteria were evaluated.

1. Introduction

The past four decades have witnessed considerable innovation in the field of azo dye chemistry based on heterocyclic systems and studies in the synthesis of such derivatives have been reported [1]. Heterocyclic based azo dyes are not only important for their excellent properties as disperse dyes for polyester textiles; they have also been utilized in non-textile applications such as photodynamic therapy, lasers, functional dye applications and non-linear optical systems [2]. The majority of disperse dyes belong to azo family. Firstly due to the ease with which an extraordinary number of molecular combinations can be generated by varying the diazo and coupling components; secondly, the dyes enjoy relatively simple manufacturing processes and finally, they provide a very wide color gamut of high color strength [3]. Derivatives of nicotinates, and nicotinamide have a long history of use as heterocyclic components for various disperse dyes [4-6]. Moreover, they have been proved to constitute the active part of several biologically active compounds [7-9]. In view of these findings and in continuation of our previous studies [10], new syntheses of these disperse dyes and their application for dyeing polyester fabrics was discussed. Also, the present study was undertaken to investigate the biological activities of the prepared dyes against Gram positive and Gram negative bacteria.

2. Experimental

2.1. Instrumentation

Melting point was recorded on a Gallenkamp apparatus. IR spectra were recorded using KBr pellets on a Jasco FTIR-6300 FT-IR spectrophotometer. ^{1}H and ^{13}C NMR spectra were recorded on Bruker DPX 400 MHz. Mass spectra were measured on a high resolution GC/MS DFS-Thermo. Micro-analyses were performed on Elementar-Vario Micro cube Analyzer.

2.2. Ethyl 5-[(4-bromophenyl) diazenyl]-2-hydroxyl-6-phenyl nicotinate (6)

Independent mixtures of compounds **1a** (0.01 mol), ethyl cyanoacetate **2a** (0.01 mol), and ammonium acetate (0.5 g) in acetic acid (3 mL) were stirred at reflux for 45 min. The mixtures were cooled and then poured into ice-water. The solids that formed were collected by using filtration and crystallized from dioxane to give compound **6** (Scheme 1). Color: Dark red crystals. Yield: 84%. M.p.: 125-128 °C. FT-IR (KBr, ν, cm^{-1}): 3407 (OH), 1699 (CO). ^{1}H NMR (400 MHz, DMSO-d_6, δ, ppm): 1.33 (t, 3H, *J* = 7.2 Hz, CH_3), 4.34 (q, 2H, *J* = 7.2 Hz, CH_2), 7.35-8.56 (m, 10H, Ar-H and OH), 8.57 (s, 1H, pyridyl-H). ^{13}C NMR (100 MHz, DMSO-d_6, δ, ppm): 166.6 (CO), 162.4, 160.2, 151.7, 137.6, 136.9, 132.9, 131.0, 129.8, 128.8, 128.7, 127.8, 124.7, 105.5, 61.5 (CH_2), 14.6 (CH_3). MS (*m/z*, (%)): 425 ([M$^+$], 100). HRMS: *m/z* (EI) $C_{20}H_{16}BrN_3O_3$; calcd.: 425.0363; found: 425.0363.

2.3. General procedure for the preparation of compounds 8a-d

Independent mixtures of each of compounds **1a-d** (0.01 mol), ethyl cyanoacetate **2a** or **2b** (0.01 mol), and ammonium acetate (2.0 g) in acetic acid (3 mL) were stirred at reflux for 45 min. The mixtures were cooled and then poured into ice-water.

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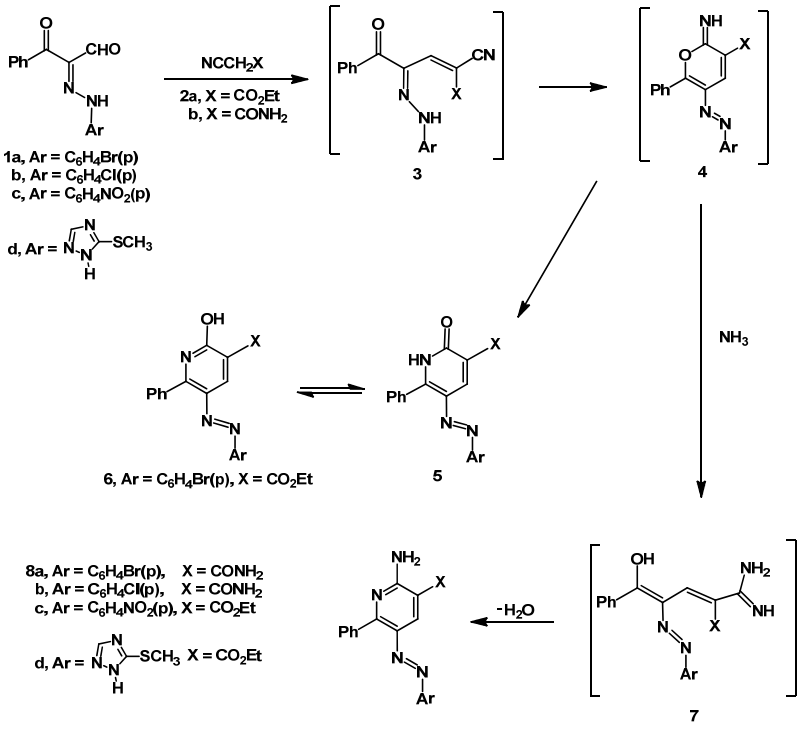


Scheme 1

The solids that formed were collected by using filtration and crystallized from dioxane to give compounds **6a-d** (Scheme 1).

2-Amino-5-[(4-bromophenyl)diazenyl]-6-phenylnicotinamide (**8a**): Color: Light brown crystals. Yield: 86%. M.p.: 185 °C. FT-IR (KBr, ν, cm^{-1}):) 3396, 3188 (NH_2), 1650 (CO). ^1H NMR (400 MHz, DMSO-d_6, δ, ppm): 7.28-7.85 (m, 9H, Ar-H), 8.03 (brs, 2H, NH_2), 8.32 (brs, 2H, NH_2), 8.49 (s, 1H, pyridyl-H). ^{13}C NMR (100 MHz, DMSO-d_6, δ, ppm): 172.8 (CO), 169.8, 171.9, 160.7, 160.4, 151.8, 136.9, 132.9, 131.3, 129.5, 127.9, 124.5, 123.8, 109.6. MS (*m/z*, (%)): 396 ([M+1] $^+$, 100). HRMS: *m/z* (EI) $C_{18}H_{14}BrN_5O$; calcd.: 395.0377; found: 395.0377.

2-Amino-5-[(4-chlorophenyl)diazenyl]-6-phenylnicotinamide (**8b**): Color: Light brown crystals. Yield: 85%. M.p.: 118-122 °C. FT-IR (KBr, ν, cm^{-1}): 3398, 3188 (NH_2), 1685 (CO). ^1H NMR (400 MHz, DMSO-d_6, δ, ppm): 7.26-8.01 (m, 11H, Ar-H and NH_2), 8.32 (brs, 2H, NH_2), 8.48 (s, 1H, pyridyl-H). ^{13}C NMR (100 MHz, DMSO-d_6, δ, ppm): 169.3 (CO), 160.2, 159.9, 151.0, 137.5, 136.4, 134.5, 130.3, 129.5, 129.8, 128.9, 128.4, 127.4, 118.9. MS (*m/z*, (%)): 350.1 ([M-1] $^+$, 100). HRMS: *m/z* (EI) $C_{18}H_{14}ClN_5O$; calcd.: 351.0881; found: 351.0881.

Ethyl-2amino-5-[(4-nitrophenyl) diazenyl]-6-phenyl nicotinate (**8c**): Color: Red brick crystals. Yield: 87%. M.p.: 175 °C. FT-IR (KBr, ν, cm^{-1}): 3406, 3278 (NH_2), 1697 (CO) 1515, 1384 (NO_2). ^1H NMR (400 MHz, DMSO-d_6, δ, ppm): 1.35 (t, 3H, *J* = 7.2 Hz, CH_3), 4.36 (q, 2H, *J* = 7.2 Hz, CH_2), 7.51-7.52 (m, 3H, Ar-H), 7.77-7.80 (m, 2H, Ar-H), 7.83 (d, 2H, *J* = 8.8 Hz, Ar-H), 8.25 (brs, 2H, NH_2), 8.34 (d, 2H, *J* = 8.8 Hz, Ar-H), 8.60 (s, 1H, pyridyl-H). ^{13}C NMR (100 MHz, DMSO-d_6, δ, ppm): 166.0 (CO), 163.0, 160.2, 155.6, 147.5, 136.9, 136.8, 131.0, 129.5, 127.5, 127.4, 125.0, 123.1, 105.3, 61.2 (CH_2), 14.1 (CH_3). MS (*m/z*, (%)): 390 ([M-1] $^+$, 100). HRMS: *m/z* (EI) $C_{20}H_{17}N_5O_4$; calcd.: 391.1273; found: 391.1273.

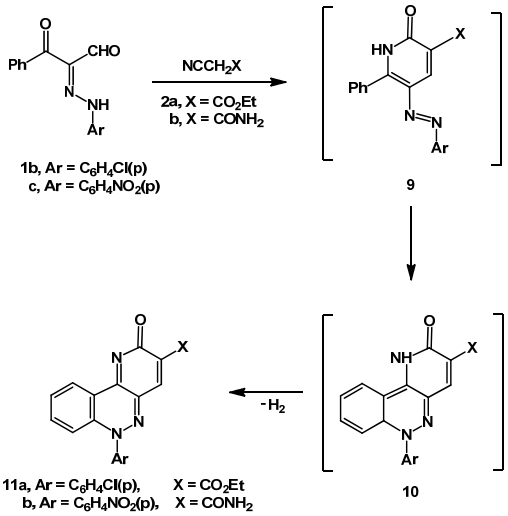
Ethyl 2-amino-5-[(5-(methylthio)-1H-1,2,4-triazole-3-yl) diazenyl]-6-phenylnicotinate (**8d**): Color: Yellow crystals. Yield: 89%. M.p.: 231-234 °C. FT-IR (KBr, ν, cm^{-1}): 3421, 3278 (NH_2), 3147 (NH), 1701(CO). ^1H NMR (400 MHz, DMSO-d_6, δ, ppm): 1.36 (t, 3H, *J* = 7.2 Hz, CH_3), 2.62 (s, 3H, S-CH_3), 4.36 (q, 2H, *J* = 7.2 Hz, CH_2), 7.47-7.50 (m, 3H, Ar-H), 7.85-7.87 (m, 2H, Ar-H), 8.18 (brs, 2H, NH_2), 8.57 (s, 1H, pyridyl-H), 14.54 (brs, 1H, NH). ^{13}C NMR (100 MHz, DMSO-d_6, δ, ppm): 162.0, 160.0, 137.1, 136.8, 131.2, 129.5, 127.5, 127.3, 105.2, 61.1 (CH_2), 14.1 (CH_3), 14.0 (CH_3). MS (*m/z*, (%)): 383.1 ([M] $^+$, 100). HRMS: *m/z* (EI) $C_{17}H_{17}N_7O_2S$; calcd.: 383.1158; found: 383.1158.

2.4. General procedure for the preparation of compounds **11a,b**

Independent mixtures of compounds **1b** or **1c** (0.01 mol), ethyl cyanoacetate **2a** or **2b** (0.01 mol), and ammonium acetate (0.5 g) in acetic acid (3 mL) were stirred at reflux for 120 min. The mixtures were cooled and then poured into ice-water. The solids that formed were collected by using filtration and crystallized from dioxane to give compounds **11a**,**b** (Scheme 2).

Ethyl-6-(4-chlorophenyl)-2-oxo-2,6-dihydropyrido[3,2,c] cinnoline-3-carboxylate (**11a**): Color: Dark brown. Yield: 83%. M.p.: 143-148 °C. FT-IR (KBr, ν, cm^{-1}): 1710, 1633(CO). ^1H NMR (400 MHz, DMSO-d_6, δ, ppm): 1.36 (t, 3H, *J* = 7.2 Hz, CH_3), 4.36 (q, 2H, *J* = 7.2 Hz, CH_2), 7.40-8.03 (m, 8H, Ar-H), 8.57 (s, 1H, pyridyl-H). ^{13}C NMR (100 MHz, DMSO-d_6, δ, ppm): 182.6, 166.1 (CO), 161.9 159.7, 150.9, 137.1, 136.4, 134.8, 130.9, 129.4, 129.3, 127.5, 127.3, 123.9, 105.0, 61.1 (CH_2), 14.1 (CH_3). MS (*m/z*, (%)): 379 ([M] $^+$, 100). HRMS: *m/z* (EI) $C_{20}H_{14}ClN_3O_3$; calcd.: 379.0719; found: 379.0719.

6-(4-Nitrophenyl)-2-oxo-2,6-dihydropyrido[3,2,c]cinnoline-3-carboxamide (**11b**): Color: Dark orange crystals. Yield: 82%. M.p.: 195 °C. FT-IR (KBr, ν, cm^{-1}): 3423, 3365 (NH_2), 1654, 1639(CO), 1518, 1336 (NO_2). ^1H NMR (400 MHz, DMSO-d_6, δ, ppm): 7.50-7.90 (m, 6H, Ar-H and NH_2), 8.28 (d, 2H, *J* = 9.6 Hz, Ar-H), 8.37 (d, 2H, *J* = 9.2 Hz, Ar-H), 8.55 (s, 1H, pyridyl-H). ^{13}C NMR (100 MHz, DMSO-d_6, δ, ppm): 189.4, 169.2 (CO), 147.5, 145.9, 142.2, 140.1, 136.8, 136.4, 132.6, 130.9, 130.2, 128.1, 127.5, 125.1, 122.9, 116.7, 109.3, 82.7. MS (*m/z*, (%)): 361.1 ([M] $^+$, 100). HRMS: *m/z* (EI) $C_{18}H_{11}N_5O_4$; calcd.: 361.0804; found: 361.0804.



Scheme 2

2.5. High temperature dyeing method (HT)

2.5.1. Materials

Scoured and bleached polyester 100% was used. The fabric was treated before dyeing with a solution containing non-ionic detergent (5 g/L) and sodium carbonate (2 g/L) in a ratio of 50:1 at 60 °C for 30 min, then thoroughly washed with water and air dried at room temperature.

2.5.2. Dyeing

The dye baths were prepared from the dye (1% and 3% weight of fabrics) to a final liquor of 40:1 (*w*:*w*). The pH value of the bath was adjusted to 4-5 with acetic acid (10%) in the presence of a 1:1 ratio of the dispersing agent (Sodium lignin sulphonate). The polyester fabrics, previously wetted, were placed into the liquor at 25-30 °C. The temperature was raised to 130 °C at the rate of 2 °C/min, and dyeing continued for 60 min. After cooling, the dyed fabrics were then washed and dried [11].

2.6. Fastness test

The dyed fabrics were tested, employing ISO standard methods [12]. Wash and perspiration fastness tests were carried out in accordance with ISO:105-C04:1989(E):Test 4 and ISO: 105-E04:1994. Daylight fastness was carried out in accordance with ISO: 105-B01:1994.

2.7. Antimicrobial activities test

The antimicrobial activities of four disperse dyes were tested against five different microbial cultures using the agar-well diffusion technique [13]. Pure cultures of *Staphylococcus auerus* and *Bacillus subtilus* (Gram positive bacteria), *Escherichia coli, Klebsiellae pneumoniae* and *Pseudomonas aeruginosa* (Gram negative bacteria) were employed in the test. An aliquot of each bacterial strain (0.1 mL) was inoculated and spread on nutrient agar (NA). The inoculated plates were supplied with 100 µL of each of the tested disperse dyes with concentrations of 1, 2, and 3 mg/mL. The dyes were placed in 4 mm wells produced by sterile cork borer. The NA plates were incubated at 37 °C for 24 h. The zones of inhibition around the wells were determined and the averages based on three replicas were recorded.

3. Results and discussion

Recently we have reported the synthesis of 2-hydroxy- and 2-amino-6-substituted-5-arylazonicotinates dyes [14-16]. Herein, we report a new strategy for the preparation of these dyes in better yields by condensing of 3-oxo-3-substituted-2-arylhydrazonals with active methylene nitriles. We observed that reaction of compound **1a-d** with active methylene, **2a**,**b**, in the presence of ammonium acetate and a few drops of acetic acid leads to ethyl 5-[(4-bromophenyl) diazenyl]-2- hydroxyl-6- phenylnicotinate, **6** (Scheme 1). It is believed that the pathways for these processes involve initial reaction of **1a-d** with active methylene ethyl cyanoacetate, **2a**, to yield the hydrazono-enone, **3**, that then cyclizes to generate the pyran-imine, **4**. In the absence of ammonium ion, compound **4** undergoes a Dimroth type rearrangement to yield ethyl 5-[(4-bromophenyl) diazenyl]-2-hydroxyl-6-phenylnicotinate monoazo disperse dye, **6**. In contrast, when the condensation reaction of compound **1a-d** with active methylene, **2a**,**b**, in presence of ammonium acetate, ethyl 2-amino-6-substituted-5-arylazonicotinate monoazo disperse, **8a-c**, are produced. It is believed that the pathways for these processes involve initial reaction of compound **1a-d** with active methylene, **2a**,**b**, to yield the hydrazono-enone, **3**. Compound **3** cyclizes to generate the pyran-imine, **4**, that participates in ring opening to yield amidine, **7** which cyclizes followed by water elimination to yield compound **8a-d** (Scheme 1).

In contrast, arylhydrazonals, **1b**,**c**, react with active methylene nitriles to afford the novel ethyl-6-(4-chlorophenyl)-2-oxo-2,6-dihydropyrido[3,2,c]cinnoline-3-carboxylate, **11a**, or 6-(4-nitrophenyl)-2-oxo-2,6-dihydropyrido[3,2,c]cinnoline-3-carboxamide, **11b**. This substance is believed to be formed via a 6 π-electrocyclization reaction of the initially formed arylazonicotinate, **9**, that generates the tricyclic intermediate product **10**, which then aromatizes to produce the cinnoline derivatives **11a**,**b** (Scheme 2).

In continuation of our research the disperse dyes **6** and **8a-d** were applied to polyester fabrics at dye shades 1 and 3% using high temperature high pressure dyeing method (HT) at 130 °C. Color shades were obtained, varying from very dark brown to orange-brownish hue. The dyeing on the polyester fabrics was evaluated in terms of their fastness properties (e.g., fastnesses to washing, perspiration, and light). The optical measurements and fastness properties data for the dyed fabrics are listed in Tables 1-4.

Table 1. Optical measurements of the synthesized arylazonicotinates disperse dyes on the polyester fabrics at 1% shade [a].

Dye No	*K/S* at (λ_{max})	*L**	*a**	*b**	*C**	*h*
6	10.88 (365)	76.59	10.19	54.21	55.16	79.35
8a	12.90 (385)	73.08	16.36	60.9	63.06	74.96
8b	20.98 (370)	64.44	23.94	64.06	68.39	69.51
8c	22.19 (415)	58.69	26.13	64.38	69.48	67.91
8d	10.48 (370)	78.38	11.18	72.41	73.27	81.23

[a] K/S = Amount of dye absorbed on the surface of the fabrics; L* = Lightness; C* = Chroma (The brightness or dullness of colour on polyester fabrics); a * = Whose value represents the degree of redness (positive) and greenness (negative); b* = Whose value represents the degree of yellowness (positive) and blueness (negative); h = Hue.

Table 2. Optical measurements of the synthesized arylazonicotinates disperse dyes on the polyester fabrics at 3% shade [a].

Dye No	*K/S* at (λ_{max})	*L**	*a**	*b**	*C**	*h*
6	20.42 (365)	66.47	19.13	59.46	62.46	72.17
8a	21.59 (385)	64.53	22.4	64.63	68.4	70.89
8b	23.94 (370)	59.63	23.95	61.48	65.98	68.71
8c	28.08 (410)	45.85	24.81	50.01	55.82	63.62
8d	15.68 (370)	74.79	15.52	75.18	76.77	78.33

[a] K/S = Amount of dye absorbed on the surface of the fabrics; L* = Lightness; C* = Chroma (The brightness or dullness of colour on polyester fabrics); a * = Whose value represents the degree of redness (positive) and greenness (negative); b* = Whose value represents the degree of yellowness (positive) and blueness (negative); h = Hue.

Table 3. Fastness properties of monoazo disperse dyes on polyester fabrics at 1% shade [a].

Dye	Color shade on polyester	Washing fastness		Perspiration fastness				Light fastness
				Alkaline		Acidic		
		Alt	SC	Alt	SC	Alt	SC	
6	Dark brown	5	4	5	5	5	5	8
8a	Light brown	5	4-5	4-5	5	5	5	8
8b	brown	5	4-5	5	5	5	5	7
8c	Very dark brown	5	5	5	5	5	5	8
8d	Orange - brownish	4-5	3	4	4-5	4-5	4-5	4-5

[a] Alt = alteration; SC = staining on cotton.

Table 4. Fastness properties of monoazo disperse dyes on polyester fabrics at 3% shade [a].

Dye	Color shade on polyester	Washing fastness		Perspiration fastness				Light fastness
				Alkaline		Acidic		
		Alt	SC	Alt	SC	Alt	SC	
6	Dark brown	5	4	5	5	5	5	7-8
8a	Light brown	5	4	4-5	5	4-5	5	7-8
8b	brown	5	4	5	5	5	5	6-7
8c	Very dark brown	5	4	5	5	5	5	7-8
8d	Orange - brownish	4	2-3	4	4-5	4-5	4-5	4-5

[a] Alt = alteration; SC = staining on cotton.

The color of dyeing on polyester fabrics is expressed in terms of CIELAB values, and the following CIELAB coordinates were measured as: lightness (*L**); chroma (*C**); hue angle (*h*) from 0 to 360 °; *a**, whose value represents the degree of redness (positive) and greenness (negative); and *b**, whose value represents the degree of yellowness (positive) and blueness (negative). A reflectance spectrophotometer was used for the colorimetric measurements of the dyed samples. The *K/S* values (where K is the absorbance coefficient and S is the scattering coefficient) given by the reflectance spectrometer were calculated at λ_{max} (wavelength of maximum absorption) and were directly correlated with the dye concentration on the dye substrate according to the Kubelka-Munk equation. Tables 1 and 2, show the relationship between shade and *K/S* as a parameter for dye concentration, the K/S increase with increasing shades. These results in this study support the findings of the other researchers who showed the linear relationship between shade and *K/S*.

The physical data for the dyed fabrics, given in (Tables 3 and 4), shows that these disperse dyes displayed very good washing and perspiration and light fastness with respect to most of the tested dyes , except **8d** which showed good results**.** It is clear from the data obtained from *p*-NO_2, *p*-Cl and *p*-Br of disperse dyes **8c**, **8b** and **8a**, the color with *p*-NO_2 very dark brown while that of *p*-Cl brown and *p*-Br light brown. This is consisting with the effect of electronegativity where NO_2 more effect than Cl and Br. This may be due that nitro group is more electron withdrawing with two electrons through adding extra resonance forms delocalizing negative charge on nitro group. Similarly, electronegativity of Cl more than that of Br. Thus we can conclude that as electronegativity of substituted atom/group decrease more light color is observed.

The antimicrobial activities of the synthesized dyes were screened against selected bacteria by the agar well diffusion method and their inhibition zones diameters, given in (Table 5), reveal that six of the tested dyes showed positive antimicrobial activities against at least two of the tested microorganisms. Three dyes, **8a**, **8c** and **11b** showed strong activities, while three dyes **6**, **8b** and **11a** showed moderate activities with significant inhibition zones >10 mm, against *Staphylococcus auerus* and *Bacillus subtilus* (Gram positive bacteria). It is of value to mention here that dye **8d** has showed no antimicrobials activities against all of the tested microorganisms, and all the dyes have no antimicrobials activities against the *Escherichia coli* and *Pseudomonas aeruginosa* (Gram negative bacteria).

The results obtained for the synthesized disperse dyes in this study support the findings of the other researchers [17] who showed that arylazonicotinates cores have various biological activities. Arylazonicotinates were proved to possess biological effects, including antimicrobial activities [18]. Therefore, the new synthesized classes showed promising results for possessing the potentials to be utilized for medicinal purposes.

4. Conclusion

In summary, a series of arylazonicotinates disperse dyes were synthesized via condensation of 3-oxo-3-substituted-2-arylhydrazonals with active methylenes. The dyed polyester fabrics, those displays very dark brown to orange-brownish hues, exhibited very good washing, perspiration and light fastness and moderate fastness properties for dye **8d**.

Table 5. Diameter of the zones of inhibition of the tested disperse dyes against microorganisms [a].

Dye No	Conc. % of Dye	Inhibition zone diameter (Nearest mm)				
		Gram positive bacteria		Gram negative bacteria		
		S. aureus	*B. subtilus*	*E. coli*	*K. pneumoniae*	*P. Aeruginosa*
6	1 mg/mL	-	-	-	-	-
8a		-	15	-	-	-
8b		-	-	-	17	-
8c		-	10	-	10	-
8d		-	-	-	-	-
11a		-	-	-	-	-
11b		10	10	-	10	-
6	2 mg/mL	14	14	-	14	-
8a		18	21	-	-	-
8b		-	-	-	17	-
8c		10	15	-	14	-
8d		-	-	-	-	-
11a		-	-	-	-	-
11b		15	15	-	12	-
6	3 mg/mL	20	21	-	21	-
8a		30	24	-	-	-
8b		-	17	-	17	-
8c		15	19	-	16	-
8d		-	-	-	-	-
11a		-	17	-	14	-
11b		20	21	-	15	-

[a] (-) No inhibition.

Finally, the biological activity of the synthesized dyes against Gram positive and Gram negative bacteria were discussed.

References

[1]. Annen, O.; Egli, R.; Henzi, B.; Jakob, H.; Matzinger, P. *Rev. Prog. Color.* **1987**, *17*, 72-85.
[2]. Singh, K.; Singh, S.; Taylor, J. A. *Dyes Pigments* **2002**, *54*, 189-200.
[3]. Gharanjig, K.; Arami, M.; Bahrami, H.; Movassagh, B.; Mahmoodi, N. M.; Rouhani, S. *Dyes Pigments* **2008**, *76*, 684-689.
[4]. El-Kashouti, M. A.; El Molla, M. M.; Elsayad, H. S.; Ahmed, K. A.; Helal, M. H.; Elgemeie, G. H. *Pigm. Resin. Technol.* **2008**, *37(2)*, 80-86.
[5]. Rasheeda, P.; Shahnaz, P.; Ausaf; A.; Khan, K. M. *Nat. Prod. Res.* **2007**, *21*, 7-12.
[6]. Khan, M. K.; Abdul, M. J.; Shahnaz, P.; Rasheeda, P.; Moazzam, H. S. *J. Chem. Soc. Pakistan* **2010**, *32*, 505-510
[7]. Kim, K. R.; Rhee, S. D.; Kim, H. Y.; Jung, W. H.; Yang, S. D.; Kim, S. S.; Ahn, J. H.; Cheon, H. *Eur. J. Pharm.* **2005**, *518(1)*, 63-70.
[8]. Lin, J. G.; Qiu, L.; Cheng, W.; Luo, S. N.; Wang, K.; Meng, Q. J. *Inorg. Chem. Commun.* **2010**, *13(7)*, 855-858.
[9]. Mohanta, G. P.; Manna, P. K.; Manavalan, R. *Ind. J. Pharm. Sci.* **2002**, *64(5)*, 489-491.
[10]. El-Apasery, M. A.; Al-Mousawi, S. M.; Mahmoud, H.; Elnagdi, M. H. *Int. Res. J. Pure Appl. Chem.* **2011**, *1*, 69-93.
[11]. Maradiya, H. R. *Turk. J. Chem*. **2001**, *25*, 441-450.
[12]. Chrysler, L. P. Methods of Test for Color Fastness of Textiles and Leather, 7th edition, Bradford, 1990, 89-94.
[13]. Dhingra, V.; Pakki, M.; Srinivas, R.; Narasu, L. *Current Sci.* **2000**, *78(6)*, 708-713.
[14]. Al-Mousawi, S. M.; El-Apasery, M. A. *Molecules* **2012**, *17*, 6547-6556.
[15]. Al-Mousawi, S. M.; Moustafa, M. S.; Abdelshafy, I. A.; Elnagdi, M. H. *Tetrahedron Lett.* **2011**, *52*, 202-204.
[16]. Zaky, O. S.; Moustafa, M. S.; Selim, M. A.; El-Maghraby, A. M.; Elnagdi, M. H. *Molecules* **2012**, *17(5)*, 5924-5934.
[17]. Alnajjar, A.; Alsaiedi, M.; El-Apasery, M. A. *Eur. J. Chem.* **2013**, *4(1)*, 53-57.
[18]. Al-Mousawi, S. M.; El-Apasery, M. A.; Mahmoud, H. M. *Molecules* **2012**, *17*, 11495-11506.