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SYNTHESIS OF SOME NEW DISPERSE DYES FOR DYEING POLYESTER FIBRE

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خلاصة ـ بتفاعل ٢ ـ فينيل ـ ٤ ـ (٢ ـ كينوليل) ـ ٢ ـ أكسازولين ـ ٥ ـ أون مع الامينات الثنائية فى ظروف مختلفة أمكن الحصول على المركبات رقم ٢، ٣ ، ٣ ، ٤ ، ٥ ، ٧ ، ٩ بالترتيب . وقد تم ادخال مجموعة الازو على المركب رقم ٤ وازدواجه مع بيتا ـ نافثول أعطى صبغة الازو رقم ٦ ، وكل هذه المركبات أستخدمت كصبغات لتعطى ألوانا ذات حساسية ولمعان عاليين لالياف البولى استر بمقارنتها بالياف البولى أميد وثلاثى الاسيتات ـ وقد أعطت ألياف البولى استر المصبوغة درجات ثبات عالية للغسيل والاحتكاك والضوء .

Abstract

Reaction of 2-phenyl-4-(2-quinolyl)-2-oxazolin-5-one 1 with diamines in different conditions gave compounds 2,3,4,5,7 and 9, respectively. Diazotisation and coupling of compound 4 gave compound 6. All these compounds were used as dyes whose gives a high substantivet and high flourescence colours to polyester fibres, compared with polyamid and triacetate fibres. The dyed polyester samples give a good fastnesses to washing, rubbing and light.

Introduction

Recently, Hassan et al.(1) have reported that oxazolone ring system may be transformed into imidazolone ring upon treatment of the oxazolone with aromatic amines in the presence of anhydrous sodium acetate. However, Yoosif et al.(2) has shown that when 2-phenyl-4-(2-quinolyl)-2-oxazolin-5-one 1 was directly heated with aniline at 150°C, anilinolysis product was the sole product, and no imidazolone derivatives being obtained. In the present work, it is aimed to report our observati in the exploration of this reaction with aromatic amines, diamines and hydrazine hydrate.

2-Phenyl-4-(2-quinolyl)-2-oxazolin-5-one **1**, on condensation with o-phenylene diamine in ethanol gave an open-chained product o-amino-phenyl-carboxamido- -phenylcarboxamido-B-quinoline acrylic acid **2**,(1,3). However, the compound 1 condensed with o-phenylene diamine in acetic acid containing sodium acetate to give benzimidazolo-2-quinolinophenyl-carboxamidomethane **3**,(1), which was obtained also by the refluxing the acrylic acid derivative 2 in acetic acid containing sodium acetate Schem

On the other hand, when 1 was directly heated with o-phenylene diamine in the presence of sodium acetate at 150°C in an oil bath, gave the 1-(2-aminophenyl)-2-phenyl-4-(2-quinolyl)-2-imidazolin-5-one 4, in good yield.

The structure of 4 was supported by i.r., elemental analysis and by the formation of the diazonium salt derivative which undergoes coupling with B-naphthol to give the azodye 6.

Compound 4, underwent cyclization upon refluxing in acetic acid containing sodium acetate to give 4-(2-quinolyl)-imidazolo[5,1a]benzimidazole 5(1,4).

When p-phenylene diamine was refluxed with I in ethanol, gave the bis acrylic acid derivatives 7,(5) in good yield, which undergoes cyclization by refluxing in acetic acid and sodium acetate to give the bis-[2-phenyl-2-imidazolin-5-one 4-(2-quinolyl)]benzene 8.

Treatment of 1 with hydrazine hydrate in ethanol gave the open chained hydrazide acrylic acid derivatives 9. The later compound did not cyclized to imidazolone derivative 10 when refluxed in acetic acid containing sodium acetate.

Colour measurement programe:

Colour measurement data (lightness (L), psychometric chroma (C), and hue angle (H) in opponent colour scale and tristimulus value (X,Y,Z) of the different dyed samples were measured and evaluated by Lab Scan Spectrocolorimeter. Instrument model 5100 produced by Hunter Associates Laboratory, INC. 11495 sunset Hills Road Reston Virginia 22090 which measure the values of :

1- CIE Tristilulus X,Y,Z.

2- CIE Chromaticity Coordinatesn Y,x,y.

3- L,a,b opponent colour scales

4- L, C, H colour system (computerised)

According to CIE(6) system tristimulus values (X,Y,Z) are automatically measured as integrated values:

$$X = \int_{400}^{700} R.E.\bar{x}.$$

$$Y = \int_{400}^{700} R.E.\bar{y}$$

$$Z = \int_{400}^{700} R.E.\bar{z}.$$

where R = reflectance % of sample at $\lambda_{400} - \lambda_{700}$

E = light source energy by microwatt at $\lambda_{400} - \lambda_{700}$

x = extinction coefficient (sinsitivity) of human eye retina to the red region of the visible spectrum $\lambda_{400} - \lambda_{700}$.

y = extinction coefficient to the yellow region of spectrum $\lambda_{400} - \lambda_{700}$.

z = extinction coefficient to the blue region of spectrum $\lambda_{400} - \lambda_{700}$

Lightness (L), psychometric chroma (C), and hue angle (H) are autometically measured according to the more advanced LCH system$^{(7,8)}$ drived from the previous tristimulus values as follows:

$$L^* = 116\ (Y/Y_o)^{1/3} - 16$$

$$C^*_{ab} = (a)^2 + (b)^2.$$

$$H^* = \text{arc Tan } b/a$$

$$\text{where } a = 500\ (\frac{X}{X_o})^{1/3} - (\frac{Y}{Y_o})^{1/3}$$

and it represents the location or the intersection of the colour of the dyed sample on the red-green axis of the colour solid.

$$b = 200\ (\frac{Y}{Y_o})^{1/3} - (\frac{Z}{Z_o})^{1/3}$$

and it represents the location or the intersection of the colour on the yellow-blue axis of the colour solid.

L^* = degree of lightness of colour

C^* = psychometric chroma or saturation or purity of the colour.

H^* = hue angle in opponent colour scale

X_o, Y_o, Z_o equal to tristimulus values of absolute white scale according to CIE system where:

$$X_o = 98.04\ ,\quad Y_o = 100\ ,\quad Z_o = 118.103$$

From table (1) we can see that, the compound $\underset{\sim}{1}$ is a redish brown dye for polyester fibers, due to the presence of the chromophoric groups $>C=C<$, $>C=O$, $-N=C<$ and groups respectively.

On treating compound $\underset{\sim}{1}$ with diamines gave compound $\underset{\sim}{2}$ which its reddish and yellowish colours are less than compound $\underset{\sim}{1}$ but its brightness is increased than compound $\underset{\sim}{1}$, this may be due to the decreasing of the number of chromophoric groups and the effect of the hypsochromic group.

On the other hand cyclisation of compound $\underset{\sim}{2}$ gave compound $\underset{\sim}{3}$ which its reddish and yellowish colours increased but its brightness dicreased, this may be due to the increase of conjugation effect.

Table (1): Colour measurements

Item	Colour solid	a	b	L	C	H
1	red orange	54.07	64.26	57.66	84	49.22
2	orange red	51.28	63.41	58.92	81.55	51.03
3	orange scalet red	55.95	64.94	54.99	85.72	49.25
4	red	56.57	64.76	51.93	85.90	48.86
6	green	-24.65	-32.49	77.97	40.78	52.81
7	red	56.57	65.06	55.32	86.02	48.99
8	orange red	55.95	64.95	54.99	85.72	49.25
9	brown	36.82	45.35	69.58	58.45	50.88

Other treatment of compound 1 with diamines gave compound 4 which has a reddish and yellowish colours more than compound 1 but the brightness decreased this may be due to the hydrogen bonding which occures between the amino group in the benzene ring and the carbonyl group in the imidazolone derivative.

Diazotisation and coupling of compound 4 gave the green dye 6, this may be due to the effect of bathochromic group (B-naphthol).

Treating of compound 1 with paradiamines gave compound 7 which has more reddish and yellowish colours and less brightness than compound 1 this may be due to the increasing of hydrogen bonding in this compound, cyclisation of compound 7 gave compound 8 which gave less reddish and yellowish colours and less brightness, this may be due to the presence of more conjugated douple bond.

Experimental:

Preparation of 4-quinoline-(4H)-2-oxazolin-5-one 1:

Quinoline-N-oxide (1.45 g, 0.01 mole) was added to a solution of hippuric acid (1.789, 0.01 mole) and fused sodium acetate (1 gm) in acetic anhydride (50 ml) at 5°C. The reaction mixture was left for 1 hr. and the precepitated solid collected by filteration and recrystallized from acetic acid to give 1 as red orange crystalls, m.p. 240°C(2).

Preparation of o-aminophenylcarboxamido of -phenylcarboxamido-B-quinoline-acrylic acid 2:

Refluxing (0.01 mole, 2.88 gm) of 4-quinoline (4H)-2-oxazolin-5-one (I) with (0.1 mole gm) of o-phenylene diamine in 100 ml ethanol on a water bath for 2 hours, the reaction mixture cooled and the collected precipitate recrystallized from ethanol to give red orange crystals of 2, m.p. 228°C and yield (-g, 75%), ir. spectra 3300 cm^{-1} for (NH) -C- and

1720 cm^{-1}.

Chemical analysis	C%	H%	N%
Calcd.	72.72	5.05	14.15
Found	72.71	5.03	14.18

Preparation of benzimidazolo-2-quinolino-phenylcarboxamido-methane 3.

(0.01 mole, 2.88 gm) of 4-quinoline-2-oxazolin-5-one 1 was refluxed with (0.01 mole, 1.08 gm) of o-phenylene diamine in 50 ml of acetic acid containing one gm of freshly fused sodium acetate on a water bath for two hours. The collected precipitate recrystallized from acetic acid to give scarlet red orange crystalles of 3 with m.p. 223°C, yield 65% and ir. (3300 cm^{-1}) for NH, 1720 cm^{-1} for $\overset{O}{\text{C}}$—, and 1620 cm^{-1} for C=N.

Chemical analysis

	C%	H%	N%
Calcd.	76.19	4.76	14.81
Found	76.15	4.73	14.79

Preparation of N-O-amino phenyl-2-phenyl-4-quinoline-2-imidazolin-5-one 4.

Fusion of 0.01 mole, 2.88 gm) of the oxazolone 1 with (0.01 mole, 1.08 gm) of o-phenylene diamine in the presence of freshly fused sodium acetate at 140°C. Or an oil bath for two hours, the reaction mixture cooled and the collected precipitate washed with ethanol and recrystallized from ethanol to give red crystalles of the imidazolone derivative 4 with m.p. 300°C, and yield 72% ir. (3300 cm^{-1} for NH), (1620 cm^{-1} for C=N) and (1710 cm^{-1} for -CON-).

Chemical Analysis:

	C%	H%	N%
Calcd.	76.19	4.76	14.81
Found	76.19	4.75	14.80

Preparation of 4-(2-quinolyl)-imidazolo 5,1-a benzimidazole 5:

A solution of o-amino phenyl-2-phenyl-4-quinoline-2-imidazoline-5-one 0.01 mole, 3.78 gm was refluxed in 50 ml glacial acetic acid containing 1 gm freshly fused sodium acetate on a hot plate for two hours. The reaction mixture was cooled and the collected precipitate recrystallized from ethanol to give colourless crystalles of imidazolo benzimidazole derivative 5, with m.p. 135°C and yield 66%, ir (3300 cm^{-1} for NH and 1620 cm^{-1} for C=N).

Chemical analysis:

	C%	H%	N%
Calcd.	80.00	4.41	15.55
Found	79.98	4.39	15.53

n.m.r (7.3 o for (15 phenyl protons) (CD_3Cl) and at 5.4 for (S-NH protons).

Cyclisation of o-aminophenylcarboxamido-B-quinoline acrylic acid:

Refluxing (0.01 mole, 3.96 gm) of o-aminophenylcarboxamido-B-quinoline-arylacrylic acid 2 in 50 ml acetic acid containing 1 gm of freshly fused sodium acetate, the reaction mixture cooled and the collected precipitate recrystallized from acetic acid to give the scarlet red crystalls of 3.

Preparation of o-B-naphthyl-azo phenyl-4-quinoline-2-phenyl-2-imidazolin-5-one 6:

(0.01 mole, 3.78 gm) of o-aminophenyl imidazolone derivatives 4 was added to 10 ml of conc. HCl and 10 ml of water than diazotized with (0.01 mole, 0.69 gm) of sodium nitrite solution. The diazonium salt was coupled with (0.01 mole, 1.44 gm) of B-naphthol at 0.0°C, and the reaction mixture was left for two hours at 0°C, collected precipitate was recrystallized from ethanol to give the green crystalles of the azodye 6 with m.p. 130°C, and yield 67%, ir. 3300 cm^{-1} for (OH), and 1620 cm^{-1} for (C=N), 3400 cm^{-1} for (OH), and 1710 cm^{-1} for (CON<).

Chemical analysis:

	C%	H%	N%
Calcd.	77.28	3.41	13.26
Found	77.28	3.39	13.25

Preparation of bis(p-phenylene carboxamido of -phenylcarboxamido-B-quinaline-acrylic acid 7:

(0.01 mole, 5.76 gm) of 2-phenyl-4-quinoline-2-oxazolin-5-one 1 and (0.6 mole, 1.08 gm) of p-phenylene diamine were refluxed in 50 ml ethanol on a water bath for two hours. The reaction mixture was cooled and the collected precipitate recrystallized from ethanol go give the red crystalles of 7 with m.p. 225°C, and yield -9,92%) ir. 3300 cm^{-1} for (NH) and 1700 cm^{-1} for -CONH-.

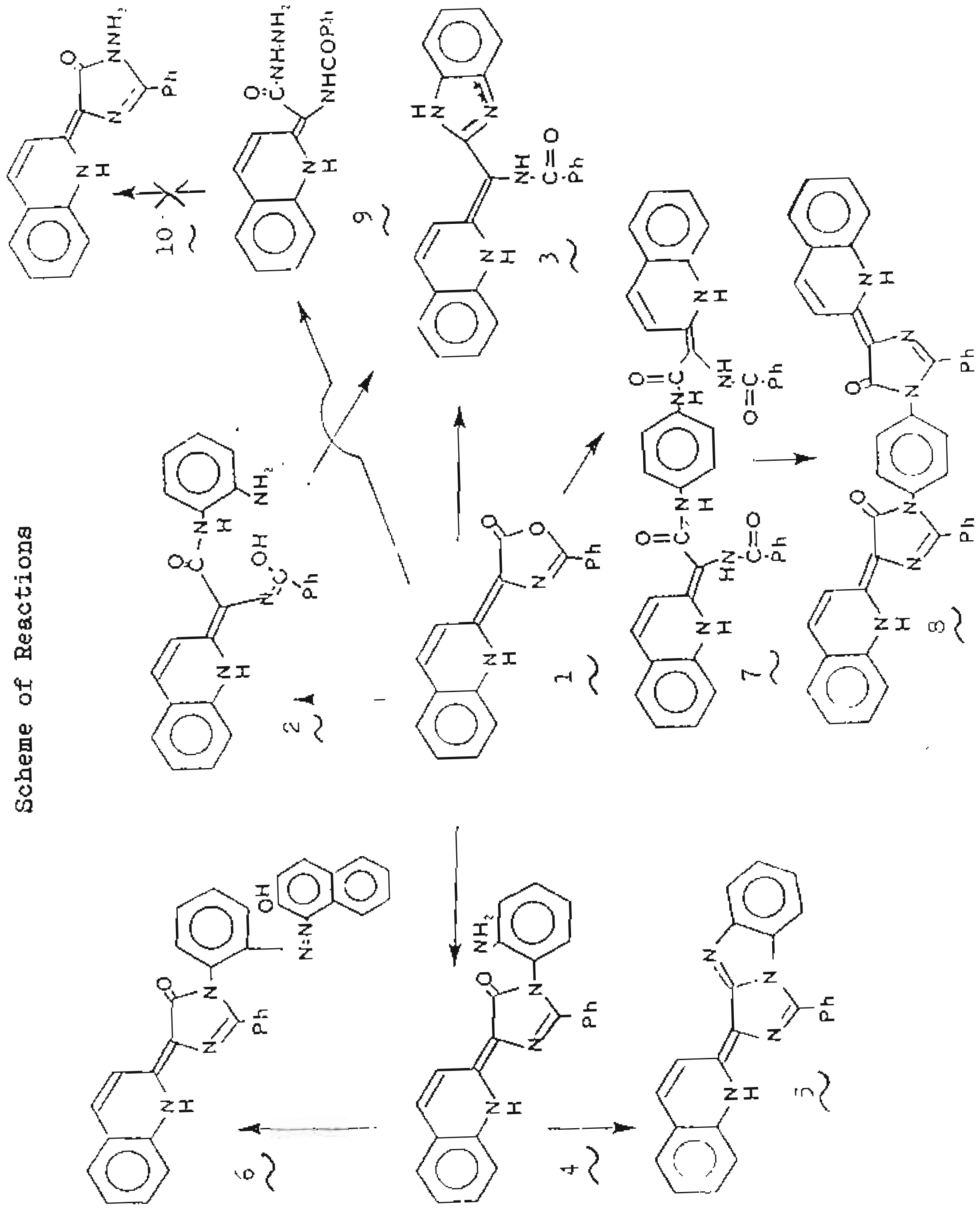
Chemical Analysis

	C%	H%	N%
Calcd.	73.68	4.68	12.28
Found	73.65	4.68	12.27

Preparation of (p-phenylene bis 4-quinoline-2-imidazolin-5-one 8:

Refluxing of (0.02 mole, 5.76 gm) of 2-oxazolin-5-one 1 with (0.01 mole, 1.08 gm) of p-phenylene diamine in 50 ml acetic acid containing 1 gm of freshly fused sodium acetate on a hot plate for two hours, the reaction mixture cooled and the collected precipitate recrystallized from ethanol to give the orange red crystalles of the bis imidazolone

Scheme of Reactions



8 with m.p. 167°C, and yield 87%, ir. 3300 cm^{-1} for (NH), 1620 cm^{-1} for C=N, and 1720 cm^{-1} for (CO-N-).

Chemical analysis

	C%	H%	N%
Calcd.	77.78	4.32	12.95
Found	77.76	4.31	12.93

Preparation of acrylic acid hydrazide 9:

(0.01 mole, 2.88 gm) of the oxazolone 1 was treated with (0.01 mole, 2 ml) of hydrazine hydrate 80% under reflux in 100 ml ethanol for fwo hours, the reaction mixture cooled, and the collected precipitate recrystallized from ethanol to give the brown crystalles of 9 with m.p. 105°C, ir. spectra, 3300 cm^{-1} for NH, 1710 cm^{-1} for -C-NH-).

Chemical analysis

	C%	H%	N%
Calcd.	67.50	5.00	17.50
Found	67.48	4.99	17.52

All these compounds were used as dyes whose gives a high substantivety and high flourescence colours (as yellow, orange, red, paje and green) to polyester fibres, compared with polyamide and triacetate fibres. The dyed polyester samples give a good fastnesses to washing, rubbing and light.

General remarks for the dyeing and fastnesses of the dyed polyester fibres:

1- Polyester (Terylene); polyamide (Nylon 66) and triacete yarns were used to assess the dyeing behaviour of dyes.

2- Fastness to washing was assessed using the automatic Lander-O-Meter machine under the following conditions:

Washing solution	5g/l soap solution 2g/l soda ash
Washing temperature	95°C
Time of washing	30 min.
pH of solution	10.5
Liquor ratio	1 : 50

The specimen was then removed and rinsed with water at 35°C until the rinse showed no alkalinity with phenolphthalein, squeezed and air-dried. The effect on the colour was expressed and defined by reference to the international geometric grey scale.

3- Fastness to rubbing: fastness to rubbing was carried out, using crockmeter of Atlas electronic type, according to the A.A.T.C.C 8-1961 standard methods of testing.

4- The fastness to light was carried out using the xenon weathering tester model 60 XH machine under the following conditions:

Relative humidity 35%
Black panel temperature 65°C,
Time of exposure 40 hr.,

The samples are viewed in a north day light and taken degrees in comparison with the relative blue scale (Standard methods of testing A.A.T.C.C. 16E-1964 T).

Dyeing of polyester fibres with arylazo of barbituric and thiobarbituric dyes were carried out by the carrier process. In dyeing at the boil (100°C), carrier was used to enable intensified absorption of disperse dyestuffs on polyester fibres. Dyeing with carriers was found to be particularly suitable for arylazo dyes. Dispersing the arylazo dyes with dispersing agent (Ekaline F), the polyester fibres were treated by:

6 c.c./l	Carrier (Levagol PT-Bayer)
pH	5-6 with acetic acid
Temperature	60°C
Time	15 minutes

Fifteen minutes after the arylazo dyes (2%) were added, the bath was brought to the boil (100°C) within 40 minutes, then dyeing was continued at boil for 120 minutes. Rinsing with cold water, then washing with:

2 c.c./l	Caustic soda 38 38 Be'
1 g/l	Soap
Temperature	95°C
Time	15 minutes

Followed by another cold water rinse.

All the dyeing processes and the fastness properties made in the laboratories of El-Nasr Weaving, Spinning and Dyeing Co., Mehalla Kobra

Table 2

Fastness Properties of

2-phenyl-4-(2-quinolyl)-2-oxazolin-5-one (2-10) dyes

Dye No	Fastness to		
	Light	Washing	Rubbing
2	6	4	4
3	5-6	4	4
4	6	4-5	4
5	6-7	4-5	4-5
6	5-6	4	4
7	6-7	4	4
8	4-5	5	4-5
9	6-7	4	4
10	6	4-5	4

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