Mansoura University

Mansoura Journal Of Chemistry

Synthesis of Some Novel Azo Coumarin Dyes Containing Benzothiazole Moiety.

A. A. Fadda[1], H.A. Etman[1], K.O. Rashid[1].

[1]*Chemistry Department, Faculty of Science, Mansoura University, Mansoura 35516, Egypt.*

Received 14 September 2014; accepted 28 September 2014

keywords
Benzothiazol acetonitrile; 2H-chromen; coumarin azodye

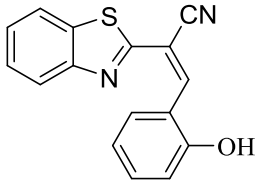
Abstract Azo dyes are extremely important in variety of industries for different technical purposes. Hence, a series of azo dyes 2-(benzo[d]thiazol-2-yl)-3-(2-hydroxy-5-((arylazo)phenyl)acrylonitrile **2a-h** were synthesized via diazotization of substituted aniline derivatives followed by azo coupling with 2-(benzo[d]thiazol-2-yl)-3-(2-hydroxyphenyl)acrylonitrile (**1**). Refluxing of compounds **2a-h** in boiling ethanol containing catalytic amount of triethylamine for long time afforded 3-(benzo[d]thiazol-2-yl)-6-arylazo-2H-chromen-2-one **4a-h**. The chemical structures of all synthesized compounds were confirmed using analytical data and spectroscopic technique which include IR, Mass spectra, ^{1}H- NMR.

Introduction

Over the years, azo compounds constitute one of the largest classes of industrially synthesized organic compounds, potent in drug and cosmetics **(**Rathod., 2013 and Marmion., 1999). Of all classes of dyestuffs, azo dyes have attained the widest range of usage because variations in chemical structures are readily achievable and methods of application are generally not complex (Chudgar, et al., 2003).In fact, 60-70% of all dyes stuff in use and production fall in this group (Carliell , et al.,1995). According to a statistical data survey, one million tons of such dyes are produced annually worldwide (Stolz, 2001). It can simply be defined as any class of artificial dyes that contains the azo group (-N=N-). When describing a dye molecule, nucleophiles are referred to as *auxochromes*, while the aromatic groups are called *chromophores*. Together, the dye molecule is often described as a *chromogen* (Al-Rubaie & Mhessn., 2012). Synthesis of most azo dyes involves diazotization of a primary aromatic amine, followed by coupling with one or more nucleophiles. Amino-and hydroxy-groups are commonly used coupling components (Heinrich,1991). The traditional application field of the synthetic azo dyes still remains the textile industry, and the finishing of fibrous materials. The emergence of diverse classes of synthetic dyes including azo dyes occurred due to constant effort to find specific dye for application in diverse materials of industrial importance, which include, but not limited to textile fabric (Elisangela, et al., 2009) leather, aluminium sheet, ink-jet printer, paper, electro-optical devices (Chakraborty & Saha, 2010). They are among the compounds, which are suitable for biocidal treatment of textile materials because some of them exhibit biological activity, as a result of the presence of some bioactive templates that form a definite type of bonding with the molecules of the fibrous material (Simu, et al.,2010).Azo

compounds are known for their medicinal importance and are well recognized for their use as antineoplastics (Child, et al.,1977) antidiabetics (Garg & Praksh,1972) antiseptics (Browing, et al.,1926), antibacterial (Khalid, et al.2008;Pagga & Brown,1986), antitumor (Thoraya, et al., 2008). They are known to be involved in a number of biological reactions such as inhibition of DNA, RNA and protein synthesis, carcinogenesis and nitrogen fixation (Park, et al., 2007; Rajendra & Madhu, 1998). Furthermore, azo dye compounds also have a lot of applications in industry and photodynamic therapy as well as photosensitive species in photographic or electro photographic systems and are dominant organic photoconductive materials (Thoraya , et al., 2008; Shridhari, et al., 2011). Azo compounds are important structures in the medicinal and pharmaceutical fields (Chandravadivelu & Senniappan, 2011) and it has been suggested that the azoimine linkage might be responsible for the biological activities displayed by some reported Schiff bases (Patel, 2012; Chopde, et al., 2010). In addition, Evans blue and Congo Red are azo dyes being studied as HIV inhibitors of viral replications. This effect is believed to be caused by binding of azo dyes to both protease and reverse transcriptase of this virus (Swati, et al., 2011). The existence of an azo moiety in different types of compounds has caused them to show antibacterial and pesticidal activities. In the recent times, exploration of azo dye as antimicrobial agents has received considerable attention (Shridhari, et al., 2011; Patel, 2012; Avci, et al., 2012; Gopalakrishnan). In the light of variety of diverse applications of azo dyes, it is conceivable to develop synthesis of phenolic azo dyes and their derivatives in order to unfold many more potentials of such compounds.

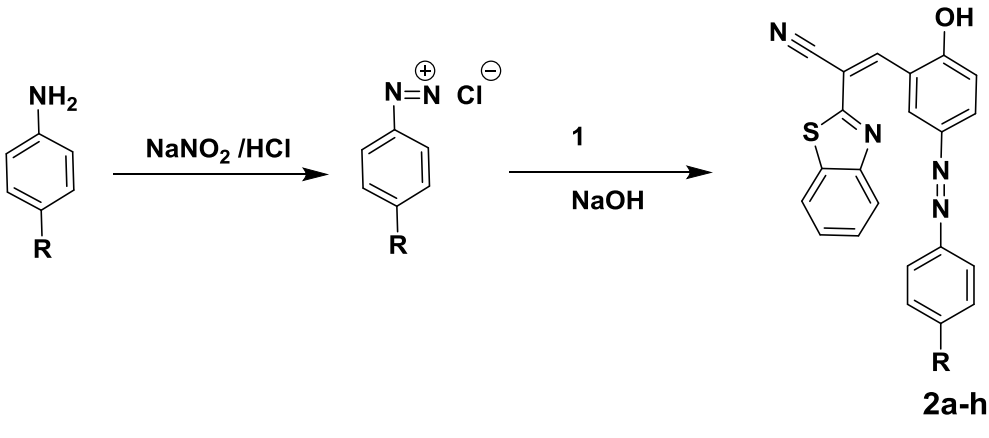
Results and Discussion



1

The title compound 2-(benzo[d]thiazol-2-yl)-3-(2-hydroxyphenyl) acrylonitrile (1) was prepared according to the reported procedure in literature (Masanori, et al., 1995).

Compound **1** act as a versatile material for synthesis of new 3-(5-(arylazo)-2-hydroxyphenyl)-2-(benzo[d]thiazol-2-yl)acrylonitrile **(2a-h)** azodyes. The formation of compounds **2a-h** involved two steps reactions. The first step involved the diazotization of para-substituted aniline to form a reactive intermediate, aryl diazonium chloride while the second step involved the formation of coupling of aryl diazonium chloride with **1** para position of phenolic OH giving 3-(5-(arylazo)-2-hydroxyphenyl)-2-(benzo[d]thiazol-2-yl)acrylonitrile **(2a-h) (Scheme 1).**

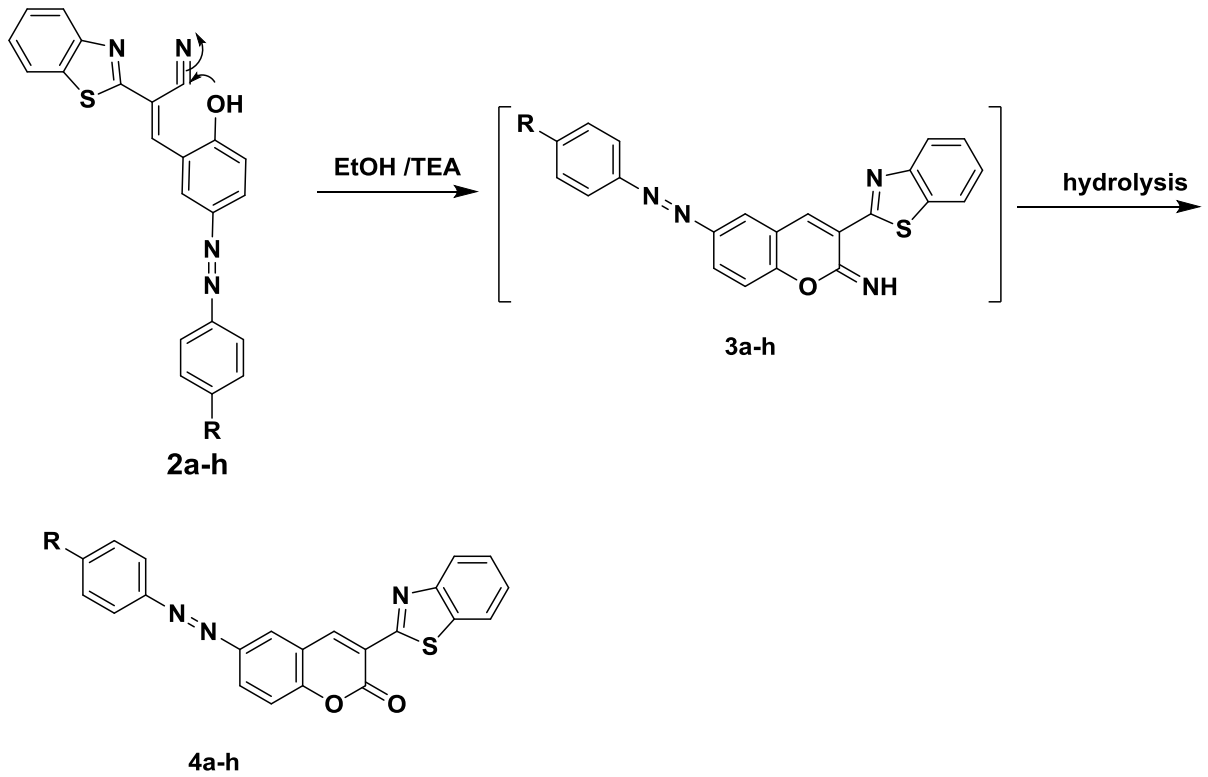


2a: R=H
2b: R =Cl
2c: R = Br
2d: R =NO_2
2e: R = COOH
2f: R = OCH_3
2g: R = SO_3H
2h: R = CH_3

Scheme 1: Synthesis of 3-(5-(arylazo)-2-hydroxyphenyl)-2-(benzo[d]thiazol-2-yl)acrylonitrile (2a-h)

The structures assessment were based on their spectroscopic data. The infrared spectra showed broad absorption bands in the region 3300-3450 cm^{-1} due to phenolic OH group, and bends in the region 1050-1250 cm^{-1} due to C_O group. The characteristic band of cyano group were obtained for stretching at 2100-2250 cm^{-1}, in addition to characteristic band from 1580-1600 cm^{-1} due to azo group. The general aromatic stretching bands were observed at 3100-3000 cm^{-1}. The characteristic bands for halogen groups like chlorine and bromine were found at 740-700 cm^{-1} & 600-500 cm^{-1}. In addition, characteristic stretching frequencies of 1, 4-Disubstituted phenyl ring were found at 800-850 cm^{-1}.

^{1}H NMR spectra of compounds **2a-h** gave a singlet signal at δ 9.30-10.00 ppm (D_2O exchangeable) attractable to phenolic OH group, singlet signal at of δ 8.40-8.60 ppm due to vinylic CH proton, in addition to multiplet signal in region δ 7.0-8.2 ppm due to aromatic protons. The mass spectra gave supporting evidence for the suggested structure by giving the correct molecular ion peak of the proposed structures **2a-h.**Boiling compounds **2a-h** in absolute ethanol containing catalytic amounts of triethylamine for long time afforded 3-(benzo[d]thiazol-2-yl)-6-arylazo-2H-chromen-2-one (**4a-h**) *via* interamolecular cyclization to form the intermediate 2-imino-chromene derivatives **3a-h** which gave on hydrolysis the compounds **4a-h** as shown in scheme 2.



Scheme 2: Synthesis of 3-(benzo[d]thiazol-2-yl)-6-arylazo-2H-chromen-2-one 4a-h.

The formation of 3-(benzo[d]thiazol-2-yl)-6-arylazo-2H-chromen-2-one (**4a-h)** was elucidated based on their analytical and spectroscopic data. Infra-red spectra showed absence of peaks in region of 2100-2250 cm^{-1} due to cyano group, also absence of broad peak in region of 3300-3400 cm^{-1} due to phenolic OH group, This confirm that hydroxyl group and cyano group were involved in cyclization reaction. In ^1H NMR the singlet signal of OH in the region 9.5-10.0 ppm was disappeared. This singlet OH was involved in the cyclization reaction.

Experimental

Melting points were recorded on Gallenkamp electric melting point apparatus (Electronic Melting Point Apparatus, Great Britain, London) and are uncorrected. Precoated Merck silica gel 60F-254 plates were used for thin-layer chromatography (TLC) and the spots were detected under UV light (254 nm). The infrared spectra were obtained from potassium bromide triturate containing 0.5% of the product on Pye Unicam SP 1000 IR spectrophotometer (Thermoelectron Co. Egelsbach, Germany. The ^1H NMR spectra were determined on Varian Gemini 400 MHz (Varian Co., Fort Collins, USA), Deuterated DMSO-d_6 was used as a solvent, tetramethylsilane (TMS) was used as an internal standard and chemical shifts were measured in δ ppm. Mass spectra were determined on a GC-MS.QP-100 EX Shimadzu (Japan). Elemental analyses were recorded on Perkin-Elmer 2400 Elemental analyzer at the Micro-analytical Centre at Cairo University, Cairo, Egypt.

General procedure: Synthesis of 3-(5-(arylazo)-2-hydroxyphenyl)-2-(benzo[d]thiazol-2-yl)acrylonitrile (2a-h)

A well stirred solution of aryl aimne (0.1 mol) in 2N hydrochloric acid (125 ml) was cooled in an ice-bath and diazotized with 0.1N sodium nitrite solution (100 ml). The mixture was stirred at 0-5°C for 1 h. The above cold diazonium solution was added dropwise to a well stirred cold solution of **1** in sodium hydroxide solution (5%, 30 ml). The reaction mixture was stirred for 2-3 h until coupling was complete. The solid precipitate was filtered, washed with water, dried and crystallized from absolute ethanol to give **2a-h.**

2-(benzo[d]thiazol-2-yl)-3-(2-hydroxy-5-(phenyldiazenyl) phenyl)acrylonitrile (2a) .

Light Brown crystal; yield (90%); mp > 350°C. IR υ (KBr) cm^{-1}: 3337(OH), 2215 (CN) and 1589 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= δ 6.90-8.20 (m, 12H, aromatic protons), 8.50 (s, 1H, CH), 9.50 (s, 1H, OH (D_2O exchangeable). MS:(*m/z, %*): 382 , 4.3% , Anal. Calcd. For $C_{22}H_{14}N_4OS$: C, 69.09; H, 3.69; N, 14.65; S, 8.38 %; Found C, 69.11; H, 3.65; N, 14.66; S, 8.34 %.

2-(benzo[d]thiazol-2-yl)-3-(5-((4-chlorophenyl)diazenyl)-2-hydroxyphenyl)acrylonitrile (2b) .

Brown crystal; yield (90%); mp =155°C. IR υ (KBr) cm^{-1}: 3338(OH), 2181 (CN) and 1587 (-N=N-).^{1}H NMR (DMSO-d_6): δ ppm= δ 7.00-8.20 (m, 11H, aromatic protons), 8.53 (s, 1H, CH), 9.56 (s, 1H, OH (D_2O exchangeable). MS :(*m/z, %*): 416, 3.4%, Anal. Calcd. For $C_{22}H_{13}ClN_4OS$: C, 63.39; H, 3.14; Cl, 8.50; N, 13.44; S, 7.69 %; Found C, 63.35; H, 3.12; Cl, 8.51; N, 13.47; S, 7.72 %.

2-(benzo[d]thiazol-2-yl)-3-(5-((4-bromophenyl)diazenyl)-2-hydroxyphenyl)acrylonitrile (2c) .

Brown crystal; yield (90%); mp =130°C. IR υ (KBr) cm^{-1}: 3336 OH) and 2187 (CN) and 1593 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= δ 7.00-8.20 (m, 11H, aromatic protons), 8.57 (s, 1H, CH), 9.50 (s, 1H, OH (D_2O exchangeable). MS:(*m/z, %*): 462 (M^+ +2) , 2.1% , 460 (M^+) , 2.9 % Anal. Calcd. For $C_{22}H_{13}BrN_4OS$: C, 57.28; H, 2.84; Br, 17.32; N, 12.14; S, 6.95 %; Found C, 57.30; H, 2.86; Br, 17.37; N, 12.12; S, 6.90%.

2-(benzo[d]thiazol-2-yl)-3-(2-hydroxy-5-((4-nitrophenyl)diazenyl)phenyl)acrylonitrile (2d)

Brown crystal; yield (90%); mp =290°C. IR υ (KBr) cm^{-1}: 3370 (OH), 2174 (CN) and 1594 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm=δ 7.00-8.20 (m, 11H, aromatic protons), 8.59 (s, 1H, CH), 9.64 (s, 1H, OH (D_2O exchangeable). MS:(*m/z, %*): 427 , 1.6% , Anal. Calcd. For $C_{22}H_{13}N_5O_3S$: C, 61.82; H, 3.07; N, 16.38; S, 7.50 %; Found C, 61.79; H, 3.05; N, 16.36; S, 7.55 %.

4-((3-(2-(benzo[d]thiazol-2-yl)-2-cyanovinyl)-4-hydroxyphenyl)diazenyl)benzoic acid (2e) .

Dark Brown crystal; yield (90%); mp =228°C. IR υ (KBr) cm^{-1}: 3448 (OH), 2500-3060 (OH of COOH), 2203 (CN), 1694 (C=O) and 1586 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= δ 7.00-8.20 (m, 11H, aromatic protons), 8.55 (s, 1H, CH), 9.64 (s, 1H, OH (D_2O exchangeable) 11.69 (s, 1H, COOH). MS (*m/z, %*): 426, 25.7%, Anal. Calcd. For $C_{23}H_{14}N_4O_3S$: C, 64.78; H, 3.31; N, 13.14; S, 7.52 %; Found C, 64.81; H, 3.30; N, 13.17; S, 7.50 %.

2-(benzo[d]thiazol-2-yl)-3-(2-hydroxy-5-((4-methoxyphenyl)diazenyl)phenyl)acrylonitrile (2f) .

Dark Brown crystal; yield (90%); mp =100°C. IR υ (KBr) cm^{-1}: 3415(OH), 2214 (CN) and 1582 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= 3.97 (s, 3H, CH_3) δ 7.00-8.20 (m, 11H, aromatic protons), 8.51 (s, 1H, CH), 9.62 (s, 1H, OH (D_2O exchangeable), MS (*m/z, %*): 412 , 24.0% , Anal. Calcd. For $C_{23}H_{16}N_4O_2S$: C, 66.98; H, 3.91; N, 13.58; S, 7.77 %; Found C, 66.99; H, 3.89; N, 13.54; S, 7.73%.

4-((3-(2-(benzo[d]thiazol-2-yl)-2-cyanovinyl)-4-hydroxyphenyl)diazenyl)benzenesulfonic acid (2g) .

Light Brown crystal; yield (90%); mp =215°C. IR υ (KBr) cm^{-1}: 3357(OH), 2216 (CN) and 1582 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm=7.10 (s, 1H, OH of SO_3H) δ 7.00-8.20 (m, 11H, aromatic protons), 8.59 (s, 1H, CH). MS (*m/z, %*): 462, 20.3%, Anal. Calcd. For $C_{22}H_{14}N_4O_4S_2$: C, 57.13; H, 3.05; N, 12.11; S, 13.86 %; Found C, 57.15; H, 3.03; N, 12.14; S, 13.84 %.

2-(benzo[d]thiazol-2-yl)-3-(2-hydroxy-5-(p-tolyldiazenyl)phenyl)acrylonitrile(2h) .

Light Brown crystal; yield (90%); mp =123 °C. IR υ (KBr) cm^{-1}: 3429(O-H) and 2213 (CN). ^{1}H NMR (DMSO-d_6): δ ppm= 2.30 (s, 3H, CH_3) δ 7.00-8.20 (m, 11H, aromatic protons), 8.54 (s, 1H, CH), 9.52 (s, 1H, OH (D_2O exchangeable) , MS (*m/z, %*): 396 , 39.3% , Anal. Calcd. For $C_{23}H_{16}N_4OS$: C, 69.68; H,

4.07; N, 14.13; S, 8.09%; Found C, 69.65; H, 4.05; N, 14.10; S, 8.07 %.

General procedure for synthesis 3-(benzo[d]thiazol-2-yl)-6-arylazo-2H-chromen-2-one 4a-h.

Boiling compound **2a-h** (0.01ml) in absolute ethanol (10 ml), containing a triethylamine (4drops) was heated under reflux for 14 - 24 hours (TLC controlled). Pour the reaction mixture on ice water acidified by conc. HCl. The precipitate that formed filtered off, recrystallized from absolute ethanol to give compounds **4a-h.**

3-(benzo[d]thiazol-2-yl)-6-(phenylazo)-2H-chromen-2-one (4a)

Brown crystal; yield (77 %); mp > 300˚C. IR υ (KBr) cm^{-1}: 1712(C=O) and 1588 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= δ 6.90-8.20 (m, 13H, aromatic protons and C_4-H pyran). MS (*m/z, %*): 383, 43%, Anal. Calcd. For $C_{22}H_{13}N_3O_2S$: C, 68.92; H, 3.42; N, 10.96; S, 8.36 %; Found C, 68.89; H, 3.40; N, 10.92; S, 8.35 %.

3-(benzo[d]thiazol-2-yl)-6-((4-chlorophenyl)diazenyl)-2H-chromen-2-one (4b) .

Brown crystal; yield (65 %); mp =127˚C. IR υ (KBr) cm^{-1}: 1710 (C=O) and 1591 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= δ 6.90-8.20 (m, 12H, aromatic protons and C_4-H pyran). MS (*m/z, %*): 417, 14.4%, Anal. Calcd. For $C_{22}H_{12}ClN_3O_2S$: C, 63.24; H, 2.89; Cl, 8.48; N, 10.06; S, 7.67 %; Found C, 63.20; H, 2.85; Cl, 8.46; N, 10.03 S, 7.65 %.

3-(benzo[d]thiazol-2-yl)-6-((4-bromophenyl)diazenyl)-2H-chromen-2-one (4c) .

Brown crystal; yield (90%); mp =170˚C. IR υ (KBr) cm^{-1}: 1710 (C=O) and 1591 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= δ 6.90-8.20 (m, 12H, aromatic protons and C_4-H pyran). MS (*m/z, %*): M^++2 460, 20.3%, Anal. Calcd. For $C_{22}H_{12}BrN_3O_2S$: C, 57.16; H, 2.62; Br, 17.28; N, 9.09; S, 6.93%; Found C, 57.13; H, 2.64; Br, 17.27; N, 9.07; S, 6.95 %.

3-(benzo[d]thiazol-2-yl)-6-((4-nitrophenyl)diazenyl)-2H-chromen-2-one (4d) .

Brown crystal; yield (90%); mp >300˚C. IR υ (KBr) cm^{-1}: IR υ (KBr) cm^{-1}: 1719 (C=O) and 1594 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= δ 6.90-8.30 (m, 12H, aromatic protons and C_4-H pyran). MS (*m/z, %*): 428, 17%, Anal. Calcd. For $C_{22}H_{12}N_4O_4S$: C, 61.68; H, 2.82; N, 13.08; S, 7.48 %; Found C, 61.65; H, 2.85; N, 14.97; S, 7.46 %.

4-((3-(benzo[d]thiazol-2-yl)-2-oxo-2H-chromen-6-yl)diazenyl)benzoic acid (4e) .

Brown crystal; yield (90%); mp =260˚C. IR υ (KBr) cm^{-1}: 2500-3200 (br) OH of COOH, 1718, 1689 (two C=O) 1591 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= δ 6.90-8.20 (m, 12H, aromatic protons and C_4-H pyran), 11.42 (s, 1H, OH of COOH). MS (*m/z, %*): 429, 100%, Anal. Calcd. For $C_{23}H_{13}N_3O_4S$: C, 64.63; H, 3.07; N, 9.83; S, 7.50 %; Found C, 64.65; H, 3.06; N, 9.84; S, 7.53 %.

3-(benzo[d]thiazol-2-yl)-6-((4-methoxyphenyl)diazenyl)-2H-chromen-2-one (4f) .

Brown crystal; yield (90%); mp >300˚C. IR υ (KBr) cm^{-1}: 1709 (C=O) and 1582 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= 4.12 (s, 3H, CH_3), 6.90-8.20 (m, 12H, aromatic protons and C_4-H pyran). MS (*m/z, %*): 413, 63.1%, Anal. Calcd. For $C_{23}H_{15}N_3O_3S$: C, 66.82; H, 3.66; N, 10.16, S, 7.75 %; Found C, 66.85; H, 3.64; N, 10.14, S, 7.79 %.

4-((3-(benzo[d]thiazol-2-yl)-2-oxo-2H-chromen-6-yl)diazenyl)benzenesulfonic acid (4g).

Brown crystal; yield (90%); mp =223˚C. IR υ (KBr) cm^{-1}: 1720 (C=O) and 1586 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= 7.10 (s, 1H, OH of SO_3H) δ 6.90-8.20 (m, 12H, aromatic protons and C_4-H pyran). MS (*m/z, %*): 463, 77%, Anal. Calcd. For $C_{22}H_{13}N_3O_5S_2$: C, 57.01; H, 2.83; N, 9.07; S, 13.83%; Found C, 57.04; H, 2.85; N, 9.05; S, 13.85 %.

3-(benzo[d]thiazol-2-yl)-6-(p-tolylazo)-2H-chromen-2- one (4 h) .

Light Brown crystal; yield (90%); mp > 350 ˚C. IR υ (KBr) cm^{-1}: 1711 (C=O) and 1589 (-N=N-). ^{1}H NMR (DMSO-d_6): δ ppm= 2.28 (s,

3H, CH_3), 6.90-8.20 (m, 12H, aromatic protons and C_4-H pyran). MS (*m/z, %*): 397, 13.1% , Anal. Calcd. For $C_{23}H_{15}N_3O_2S$: C, 69.51; H, 3.80; N, 10.57; S, 8.07 %; Found C, 69.54; H, 3.82; N, 10.53; S, 8.00 %.

References

Al-Rubaie LAR, Mhessn RJ. Synthesis and characterization of azo dye para red and new derivatives. *E-J. Chem*. **(2012)**, *1*,465-70.*Physical Review & Research International*. **(2013)**, *40*, 28-41.

Avci GA, Ozkinali S, Ozluk A, Avci E, Kocaokutgen H. Antimicrobial activities, absorption characteristics and tautomeric structures of o,o'-hydroxyazo dyes containing an acryloyloxy group and their chromium complexes. *Hacettepe J. Biol.Chem*.**(2012)**, *2*,119-26.

Browing CH, Cohen JB, Ellingworth S, Gulbransen R. The antiseptic properties of the amino derivatives of styryl and anil quinoline. *Journal Storage*. **(1926)**, *100*,293-25.

Carliell CM, Barclay SJ, Buckley CA. Microbial decolourization of a reactive azo dye under anaerobic conditions. *Water SA*. **(1995)**, *1*, 61–69.

Chakraborty A, Saha PK, Datta C. Synthesis and application of azo-naphthol dyes on wool, silk and nylon fabric. *7Th Int. Conf. TEXSCI Sept. Liberec, Czech.Rep*. **(2010)**, *6*, 8-15.

Chandravadivelu G, Senniappan P. In-vitro antimicrobial activity of novel derivative of azo dye from cyano ester. *Int. J. Res. Pharm. Chem*. **(2011)**, *4*,1082-086.

Child RG, Wilkinson RG, Tomcu-Fucik A. Effect of substrate orientation of the adhesion of polymer joints. *Chem. Abstr*. **(1977)**, *87*, 6031-6038.

Chopde HN, Meshram JS, Pagadala R, Mungole AJ. Synthesis, characterization and antibacterial activity of some novel azo-azoimine dyes of 6-bromo-2-naphthol. *Int. J.Chem. Tech. Res*. **(2010)**, *3*, 1823-830.

Chudgar RJ, Oakes J, Kirk-Othmer Encycl. Molecular interactions of azo dyes with nonionic and anionic surfactants .*Chem. Tech***. (2003),***116*, 1199-208.

Elisangela F, Andrea Z, Fabio DG, Cristiano RM, Regina DL, Artur CP. Biodegradation of textile azo dyes by a facultative Staphylococcus arlettae strain VN-11 using asequential microaerophilic/aerobic process. *Int. Biodeter. Biodegrad*. **(2009)**, *63*,*280*-88.

Garg HG, Praksh C. Preparation of 4-arylazo-3,5-disubstituted-(2H)-1,2,6-thiadiazine1,1-dioxides. *J. Med. Chem*. **(1972)**, *4*,435-36.

Gopalakrishnan S, Nevaditha NT, Mythili CV. Antibacterial activity of azo compounds Synthesized from the natural renewable source. *cardanol. J. Chem. Pharm. Res*.**(2011)**, *4*,490-97.

Heinrich Z, Color chemistry: Syntheses, properties and applications of organic dyes and pigments, *VCH*. **(1991)**, 496.

Khalid A, Arshad M, Crowley DE. Accelerated decolorization of structurally different azo dyes by newly isolated bacterial strains. *Appl. Microbiol. Biotech*. **(2008)**,*78*,361-69.

Marmion DM. Hand book of colorant. Colorants in use today. *Wiley New York*. **(1999)**, 23-26.

Masanori, S; Asae, N; Mino, S; Hideki, O; Yoko, S; Sanae, Y; Mayumi, N; Kimio, O; Tadamasa, D; Osamu, Diels–Alder reaction of benzylidene(cyano)methyl-1,3-benzoxa/thiazoles as stable 1-azabuta-1,3-dienes . T. *J. Chem. Soc*. **(1995)**, *1*, 1759-1779.

Pagga U, Brown D. The degradation of dyestuffs in aerobic biodegradation tests. *Chemosphere*. **(1986)**, *15*,*479*-91.

Pandey A, Singh P, Iyengar L. Bacterial decolorization and degradation of azo dyes.*Int. Biodeter. Biodegrad*. **(2007)**, *59*,73–84.

Park Ch, Lim J, Lee Y, Lee B, Kim S, Lee J, Kim S. Optimization and morphology for decolorization of reactive black 5 by Funalia trogii. *Enzyme Microb. Tech*.**(2007)**,*40*,1758-764.

Patel PS. Studies on synthesis and dyeing performance of disperse azo dyes based on Schiff base of ninhydrin and 3-amino phenol. *Arch. Appl. Sci. Res*. **(2012)**, *2*,846-51.

Rajendra GM, Madhu SV, Naveen KS. Voltammetric investigations of the reduction of direct orange-31 a bisazo dye. *Croatica Chemica Acta*. **(1998)**, *3*,715-26.

Rathod KM, Thakre NS. Synthesis and antimicrobial activity of azo compounds containing m-cresol moiety. *Chem. Sci. Trans*. **(2013)**, *1*, 25-28.

Shridhari AH, Keshavayya H, Hoskeri HJ, Ali RAS. Synthesis of some novel bis 1,3,4-oxadiazole fused azo dye derivatives as potent antimicrobial agents. *Int. Res. J. Pure Appl. Chem*. **(2011)**, *3*, 119-29.

Simu GM, Dragomirescu A, Grad ME, Savoiubalint G, Andoni M, Bals G. Azo compounds with antimicrobial activity. *14Th Int. Electron. Conf. Syn. Org. Chem.ECSOC*. **(2010)**, *14*,1–30.

Stolz A. Basic and applied aspects in the microbial degradation of azo dyes. *Appl.Microbiol. Biotech*. **(2001)**, *56*, 69-80.

Swati G, Romila K, Sharma IK, Verma PS. Synthesis, characterization and antimicrobial screening of some azo compounds. *Int. J. Appl. Biol. Pharm. Tech*. **(2011)**, *2,332*-38.

Thoraya A, Farghaly, Abdallah ZA. Synthesis, azo-hydrazone tautomerism and antitumor screening of N-(3-ethoxycarbonyl-4,5,6,7-tetrahydro-benzo[b]thien-2-yl)-2-arylhydrazono-3-oxobutanamide derivatives. *Arkivoc*. **(2008)**, *17,295-305*.

تشيد بعض مركبات الازوكومارين المحتوية على نواة البنزو ثيازول

أ.د. أحمد علي فضة[١] ، أ.د. حسن على عتمان[١] ، كيوان عمر رشيد[١]

[١] قسم الكيمياء ، كلية العلوم ، جامعة المنصورة ، المنصورة ٣٥٥١٦ ، مصر

الأصباغ ايزو هي في غاية الأهمية في مجموعة متنوعة من الصناعات لأغراض فنية مختلفة. وبالتالي فان سلسلة من الأصباغ ايزو 2- (بنزو [د] ثيازول-2-يل) -٣- (٢-هيدروكسي-٥ - ((اريل ايزو) فينيل) تم تصنيعه الأكريلونيتريل h-a٢ عبر تديز للمشتقات الأنيلين استبداله تليها الآزو اقتران مع ٢- (بنزو [د] ثيازول-2-يل) -٣- (٢-هايدروكسيد فينايل) الأكريلونيتريل (١). التكثيف الراجع من المركبات h-a٢ في الإيثانول الغليان التي تحتوي على كمية الحفاز ثلاثي الإيثيلامين لفترة طويلة الممنوحة ٣- (بنزو [د] ثيازول-2-يل) -٦-اريل ايزو-٢-H-كرومين-٢-اون h-a٤. تم تأكيد البنية الكيميائية لجميع المركبات المشيد باستخدام بيانات تحليلية وتقنية الطيفية والتي تشمل الأشعة تحت الحمراء، كتلة ألطيف(^{1}H-NMR) .