

## **RESEARCH ARTICLE**

## SYNTHESIS, REACTIVITY AND APPLICATIONS OF ARYL AZOTHIAZOLES DERIVATIVE.

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# Manuscript Info

#### Abstract

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### Key words:-

Aryl azothiazole, Thiourea, Diazonium salts, Heterocyclic compounds and applications.

## A great importance has been paid to study new compounds with new sites of action. In this mini-review we aim to show synthesis, chemical reactions and applications of Aryl azothiazoles derivatives.

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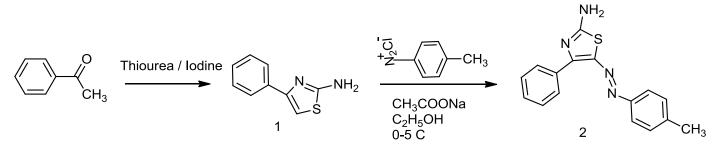
## Introduction:-

Aseries of heterocyclic azo dyes based on thiazolyl derivatives synthesized by a classical method of diazotizingcoupling. Many arylazo derivatives of heterocyclic compounds with thiazolyl group, compounds are very important class of chemical constituents which are having wide applications in different fields such as textile industries for dying the cotton fibers, fabrics, paper industries, paint and coating industries as dyeing agent (Al-Adilee, Abass et al. 2016)hair dyeing and disperse dyes (Shawali and Zayed 2013). A novel calix[4]arene derivative containing benzothiazoleazo groups was synthesized as chromogenic chemosensor which has high selectivity towards Hg<sup>+2</sup>(Bingol, Kocabas et al. 2010). Some arylazo derivatives of heterocyclic compounds used to prepare polyurethanes(Kariduraganavar, Tambe et al. 2011)somearyl azothiazole dyes exist in azo-enol-keto forms in solid state, and in hydrazone-keto forms in solvents.(Yahyazadeh and Yousefi 2014)Some arvlazo based on N-benzyl-Nethyl-aniline with thiazole group Characterized by good optical response(Yazdanbakhsh, Mohammadi et al. 2010)hetaryl-azophenol dyes with thiazole group used as dyes which applied to polyesters and afforded red-orange shades with excellent wash fastness properties. Also use of these azo dyes as an acid-base endpoint indicator(Yousefi, Yahyazadeh et al. 2013)4-(2-thiazolylazo)resorcinol used as ligand and react with Manganese(II), cobalt(II) and nickel(II) acetates to form complexes (Karipcin, Dede et al. 2010)Thiazoles are an important class of heterocyclic compounds, because of their anti-inflammatory, antimicrobial (El-Borai, Rizk et al. 2014). Due to the importance of this class of compounds, we collected methods of preparation, Chemical reactions and applications.

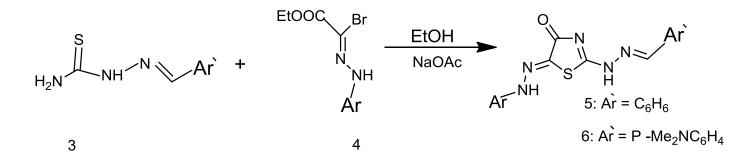
## Aryl azo thiazole derived from thiourea and its derivatives:-

Acetophenone fused with thiourea to obtain 2-amino-4-phenylthiazole **1** which coupled with diazonium salt of 4methylaniline to afford 2-amino-5-(4-methylphenyl)-diazenyl-4- phenyl-1, 3-thiazole **2** (Prajapati and Modi 2011)

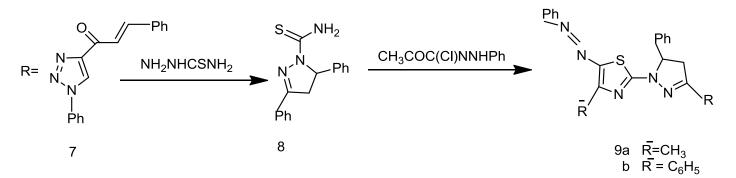
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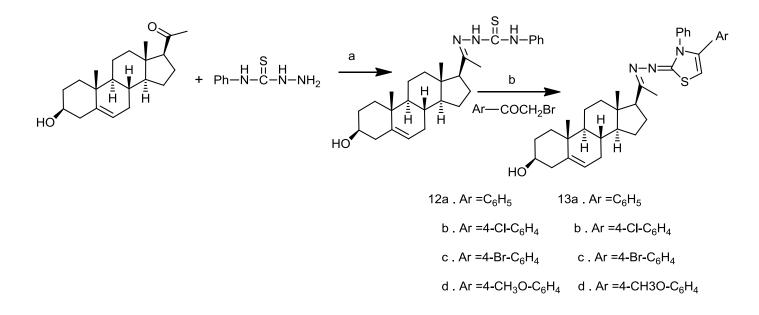
Heating of thiosemicarbazones 3 with ethyl arylhydrazonobromoacetates 4 in ethanol containing anhydrous sodium acetate afforded the corresponding 2-(benzylidenehydrazino)-5-arylazo-thiazolidin-4-one (E. Khalifa, Abdel-Latif et al. 2013)



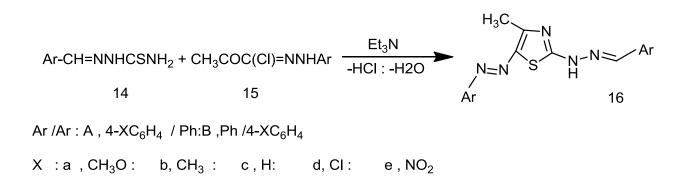
Treatment of 1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)- 3-phenyl-prop-2-en-1-one **7** with thiosemicarbazide in boiling ethanolic sodium hydroxide followed by chloroacetone gave 1-(2-(4,5-dihydro-3-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)-5-phenylpyrazol-1-yl)-4-methylthiazol- 5-yl)-2-phenyldiazene**9a**and 1-(2-(4,5-dihydro-3-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)-5-phenylpyrazol-1-yl)-4-phenylthiazol-5-yl)-2-phenyldiazene**9b**(Abdelhamid, Abdel-Riheem et al. 2012)



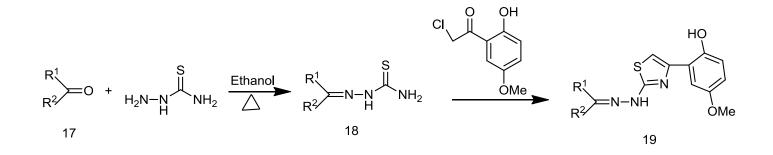
Pregnenolone **10** condensed with 4-phenyl-3-thiosemicarbazide in ethanol followed by phanacylbromide gave thiazolyl hydrazonopregnenolone derivatives **13** (Mohareb, Wardakhan et al. 2012)



Shawali et al. reported that treatment of benzaldehyde thiosemicarbazone **14c** with each of the hydrazonoyl chlorides **15a–e** in ethanol in the presence of triethylamine afforded substituted-benzaldehyde N-(5-arylazo-4-methylthiazol-2-yl)-hydrazones **16** (Shawali and Zayed 2013)

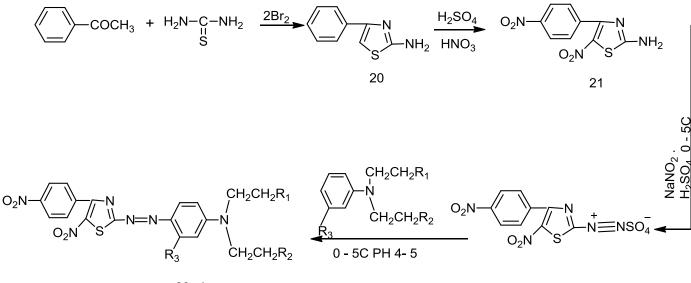


Carbonyl compounds **17a–k** were reflux with thiosemicarbazide in ethanol. The produced thiosemicarbazones **18a–k** condensed with 2-chloro-1-(2-hydroxy-5-methoxyphenyl)-ethanone to give thiazolylhydrazones 19a–k (Maillard, Bertout et al. 2013)



а	: R <sup>1</sup> = Me ,R <sup>2</sup> = Indanyl	f : R <sup>1</sup> = H, R <sup>2</sup> = Imidazo[1,2-a]pyridin-3-yl	
b	: R <sup>1</sup> = H, R <sup>2</sup> = 1,3-Benzodioxol-5-yl	g : R <sup>1</sup> = H, R <sup>2</sup> = Benzofuran-2-yl	
с	: R <sup>1</sup> , R <sup>2</sup> = Cyclohexyl	h : $R^1 = H$ , $R^2 = pyrrol-3-yl$	
d	: R <sup>1</sup> = H, R <sup>2</sup> = Napht-2-yl	i : R <sup>1</sup> = H, R <sup>2</sup> = Fur-2-yl	
е	$: R^1 = H, R^2 = Indol-3-yl$	j : R <sup>1</sup> = H, R <sup>2</sup> = imidazol-5-yl	
k: R <sup>1</sup> = H, R <sup>2</sup> = 3-Methyl-6-phenylimidazo-[2,1-b]thiazol-5-yl			

Thiourea reacted with bromine and acetophenone followed by nitration to get 2-amino-4-(p-nitrophenyl)-5nitrothiazole **21** Then coupled with diazonium salt of N,N-dialkylaniline derivatives to produce **23a-j** (Maradiya 2012)



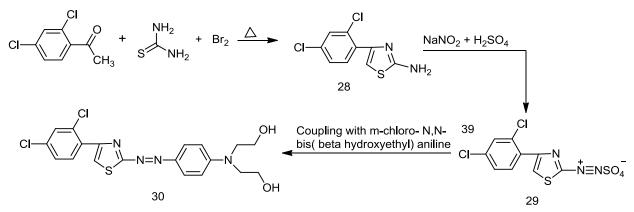
23a-j

a : R <sub>1</sub> =R <sub>2</sub> = OH	R <sub>3</sub> =H	d : R <sub>1</sub> =R <sub>2</sub> = OH	R <sub>3</sub> =CI	g : R <sub>1</sub> =R <sub>2</sub> = CI	R₃=H	
b : R <sub>1</sub> =R <sub>2</sub> = OAc	R <sub>3</sub> =H	e : R <sub>1</sub> =R <sub>2</sub> = OH		$\tilde{h}$ : $R_1 = R_2^2 = CN$		
c : R <sub>1</sub> =R <sub>2</sub> = OH	R <sub>3</sub> =H	$f: R_1 = R_2 = OAc$	$R_3 = CH_3$	$i: R_1 = R_2^2 = CN$		
				$j : R_1 = R_2^- = CN$	R <sub>3</sub> =Cl	2/29
						272J

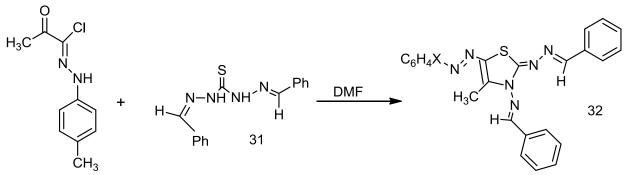
Condensation of bromoacetone 24 with thiosemicarbazone derivatives by reflux in ethanol afforded the corresponding 4-substituted-2- thiazolylhydrazones of ketones 26. By diazocoupling 26 with the antipyrinyl diazonium chloride get the corresponding antipyrinylazo-thiazole dyes 27 )Metwally, Abdel-Galil et al. 2012(

 $Me \xrightarrow{O}_{24} Br + H_2N \xrightarrow{N}_{H} N \xrightarrow{R_1}_{R_2} \underbrace{EtOH}_{S} \xrightarrow{N}_{H} N \xrightarrow{R_1}_{R_2} Ar \xrightarrow{N}_{N} \xrightarrow{N}_{H} \xrightarrow{N}_{R_2} Ar \xrightarrow{N}_{N} \xrightarrow{N}_{H} \xrightarrow{N}_{R_2} Ar \xrightarrow{N}_{R_2} \xrightarrow{$ 

When 2,4-dichloro acetophenone and thiourea reacted with bromine followed by diazotization gave diazonium salt solution **29** which coupled with m-Chloro-N,N-bis( $\beta$ -hydroxyethyl) aniline to afford 2,2-(3-Chloro-4-((4-(2,4-dichlorophenyl)thiazol-2-yldiazenyl)phenylazanediyl)diethanol **30** (Patel, Patel et al. 2014)



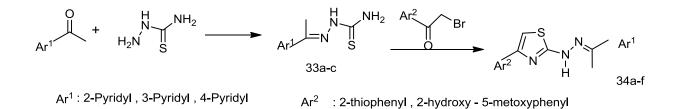
Hydrazonoyl halides reacted with bis-hydrazone **31** afforded Benzylidene-[2-(benzildene-azo)-4-methyl-5-p-tolylazo-thiazol- 3-yl]-amine **32a** (Sayed 2012).



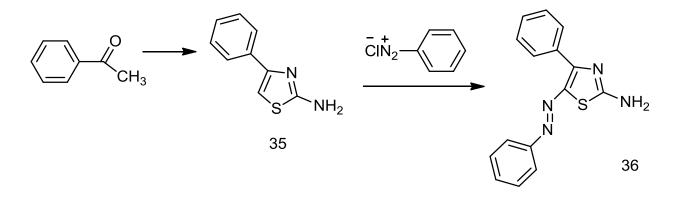
X:a,4-CH<sub>3</sub> c,4-CI

hen acetylpyridines react with thiosemicarbazide produced 1-(1-arylethylidene)- thiosemicarbazide 33a-c

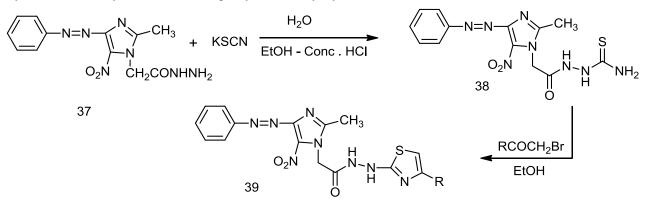
following add 1-(2-thiophenyl)-2-bromoethanone afford N-(1-arylethylidene)-N-(4-arylthiazol-2-yl)-hydrazines **34a–f** (Turan-Zitouni, Kaplancikli et al. 2010)



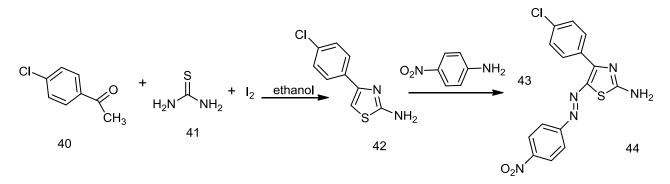
2-Amino-4-phenyl-5-phenylazothizole **36** produced from the reaction of Acetophenone with thiourea to give 2amino-4-phenylthiazole **35** then coupled with diazonium salt of methylaniline (Prajapati and Modi 2010)



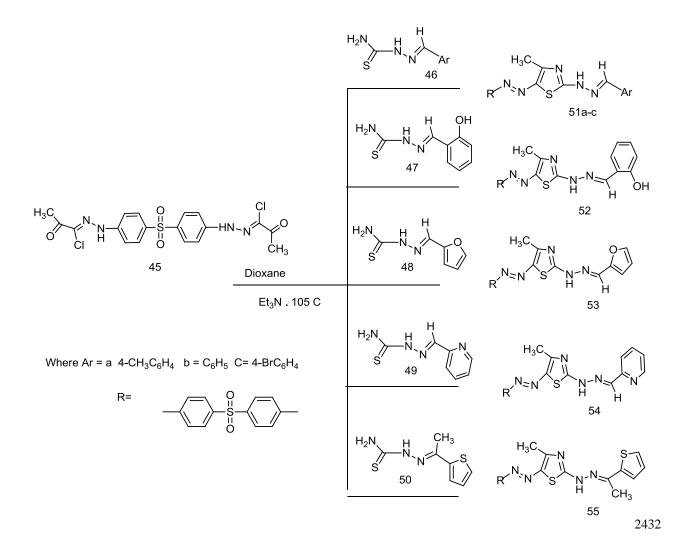
(2-methyl-5-nitro -4-phenyl azo-imidazole-1-yl)-acetic acid hydrazide **37** refluxed with with potassium thiocyanate, concentrated hydrochloric acid and ethyl alcohol to give 2-methyl5-nitro-4-phenyl azo-imidazole-1-yl)-acete thiosemicarbazone **38** which reacted with bromoacetyl derivatives to afford (2-methyl-5-nitro-4-phenylazo-imidazol-1-yl)-aceticacid-N-(4-phenyl-thiazol-2-yl hydrazide **39** [21].



R = phenyl, p-tolyl, p-anisyl, p-hydroxy phenyl, p-nitro phenyl, p-chloro phenyl, p-bromo phenyl, pphenyl sydnonyl , N-p-tolyl sydnonyl and N-p-anisyl sydnonyl Reaction of 4-chloro acetophenone **40** condensed with thiourea **41** gave 4-(4-nitrophenyl) thiazol-2-amine **42** which coupled with diazonium salt of 4-nitro aniline to produce 4-(4-nitrophenyl)thiazol-2-amine **44** (SATBHAIYA and CHOURASIA)

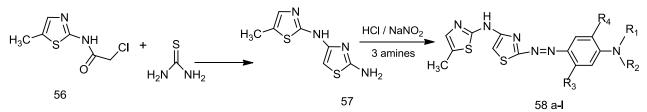


Bis-hydrazonoyl chlorides 45 reacted with thiosemicarbazone derivatives 46 - 50 and triethylamine in dioxane to afford compounds 51 - 55. (Sayed 2015)

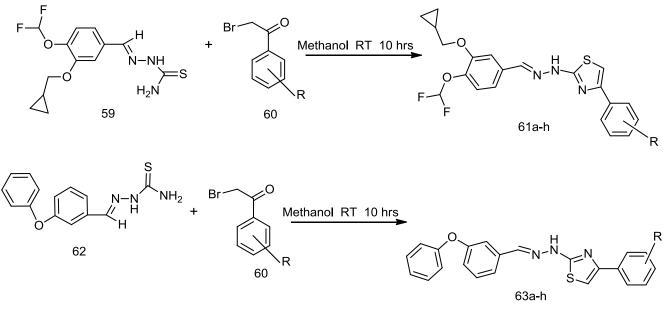


Methyl-1,3-Thiazole-2-yl)-1,3-Thiazole-2-4-diamine 57 which diazotized and coupled with N, N- di methyl aniline derivatives to give corresponding disperse dyes 58 a-l

When 2-chloro-N- (5-methyl-1, 3-Thiazol-2-yl) acetamide 56 refluxed with thiourea in methanol gave N4-(5-(Malik, Patel et al.)

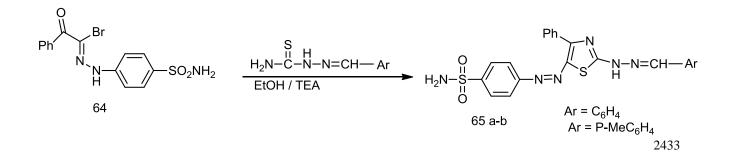


Khushal et al. reported that reaction of (E)-1-Subsituted benzylidene thiosemicarbazide 59, 62 reacted with phenacyl bromide (60) gave (E)-1-(Substituted benzylidene)-2-(4-Substituted phenylthiazol-2-yl) hydrazine 61 a-(Khushal Kapadiya 2015) h , 63 a-h.

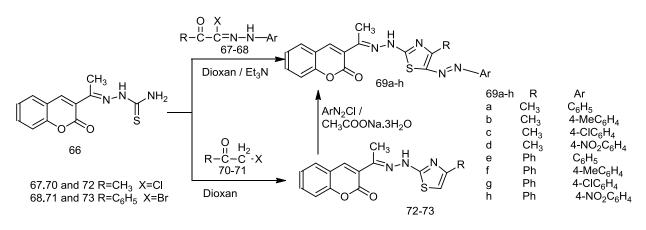


 $R = 4-OCH_3 4-CH_3 4-Br 4-H 3 4-(CI)_2 4-CI 4-NO_2 4-OCHF_2$ 

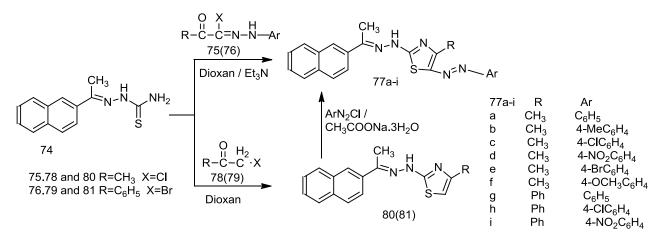
2-(benzylidenehydrazino)-4-phenyl-5-(4-sulfamoyl-phenylazo)-thiazole derivatives 65a -b obtained by refluxed (4-sulfamoyl-phenylhydrazono) phenacyl bromide 64 with thiosemicarbazone derivative in ethanol and triethylamine (Fadda, El-badraw et al. 2016)



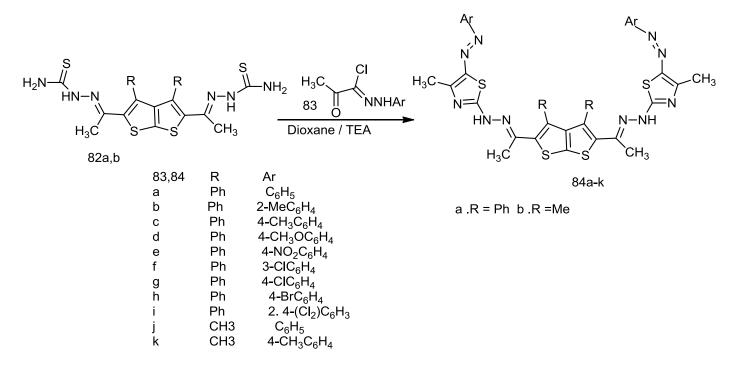
It has been found that,  $2-(1-(2-\infty - 2H-chromen-3-yl)ethylidene)$  thiosemicarbazide **66** reacted with hydrazonoyl halides **67** - **68** gave 3-[1-(4-substituted-5-(aryldiazenyl)thiazol-2 yl)hydrazono)ethyl]-2Hchromen-2-ones**69 a-h**. The thiazole derivatives**72**-**73**were prepared by the reaction of thiosemicarbazide derivatives**66**with chloroacetone**70**or phenacyl bromide**71**in dioxane, then diazotized aniline coupled with thiazole derivatives**72**-**73**, in presence of sodium acetate trihydrate, afford the azo derivatives**69 a-h**(Gomha and Khalil 2012)

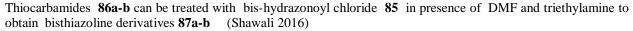


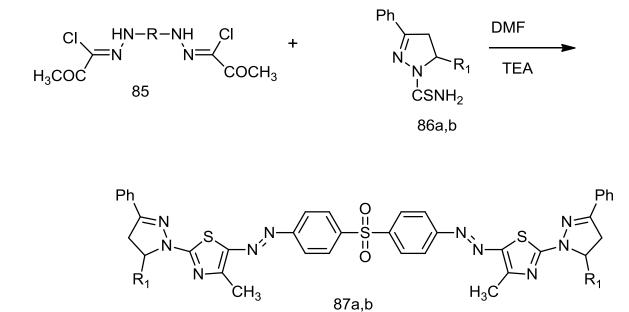
4-substituted-2-(2-(1-(naphthalen-2-yl)ethylidene)hydrazinyl)-5-(aryldiazenyl)thiazole **77a–i** were prepared by reaction of 2-[1-(naphthalen-2-yl)ethylidene]hydrazi necarbothioamide **74** with hydrazonoyl halides **75** and **76** in 1,4-dioxane and presence of triethylamine (TEA), otherwise thiosemicarbazone derivative **74** refluxed with chloroacetone **78** or phenacyl bromide **79** in 1,4-dioxane afford thiazole derivatives **80** and **81**. Azo derivatives **77a–i** prepared by coupling thiazole derivatives **80** and **81** with diazonium salt in the presence of sodium acetate (Gomha and Badrey 2013)



Heating of thiosemicarbazone derivatives **82** with the hydrazonyl halides **83** in 1,4-dioxane and trimethylamine afforded the bisthiazole derivatives **84 a**- $\mathbf{k}$  21 (Gomha, Badrey et al. 2016)



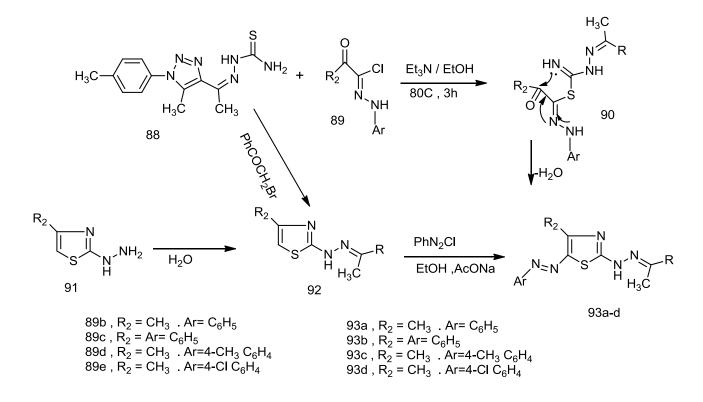




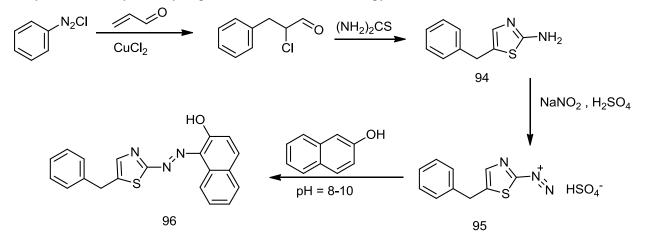
 $R= 1,4-C_6H_4-SO_2-C_6H_4-1,4$   $R_1 = a,Ph : b=2-CIC_6H_4$ 

Refluxing of 2-(1-(5-Methyl-1-(p-tolyl)-1H-1,2,3-triazol-4-yl)ethylidene)hydrazinecarbothioamide **88** with hydrazonyl chloride in ethanolic triethylamine afforded (2-(2-(1-(5-methyl-1-(p-tolyl)-1H-1,2,3-triazol-4-yl)ethylidene)-hydrazinyl)-4-phenyl-5-(phenyldiazenyl)thiazole **93b**, also **88** reacted with bromoacetophenone to

give 2-(2-(1-(5-methyl-1-(p-tolyl)-1H-1,2,3-triazol-4-yl)ethylidene)-hydrazinyl)-4-phenylthiazole **92** which coupled with benzenediazonium chloride to give **93a-d** (Abdelriheem, Mohamed et al. 2017)

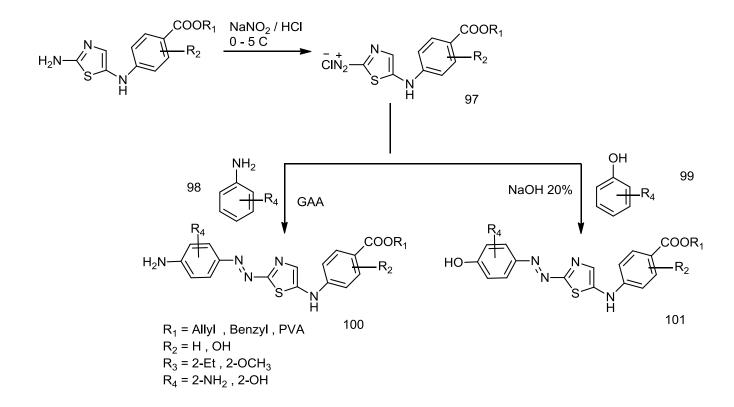


Diazotization of 2-amino-5-benzyl-1,3-thiazole **94** then coupled with naphthalen-2-ol in a basic media gave 1-[(5-benzyl-1,3-thiazol-2-yl)diazenyl]naphthalen-2-ol **96** (BnTAN) (Tupys, Kalembkiewicz et al. 2017)

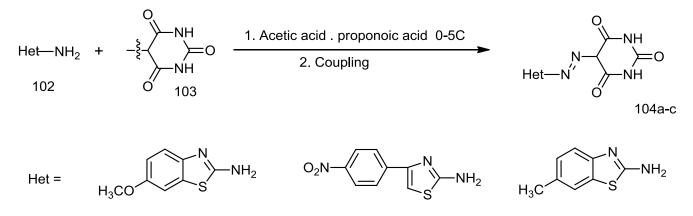


#### Aryl azo thiazole derived from diazonium salt solution:-

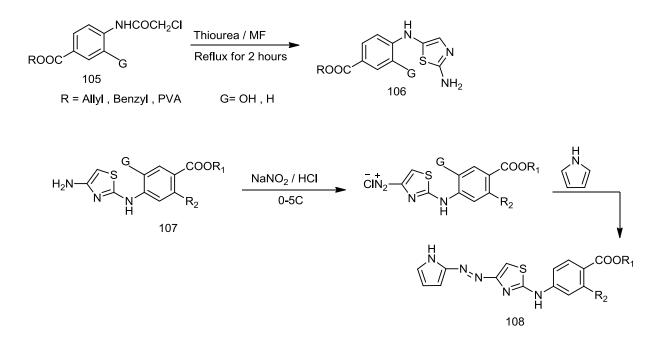
Al-Tamimi et al. reported that substituted thiazole-2-azo 97 coupled with aromatic amines 98 and phenols 99 afforded compound 100-101 (Al-Tamimi 2017)



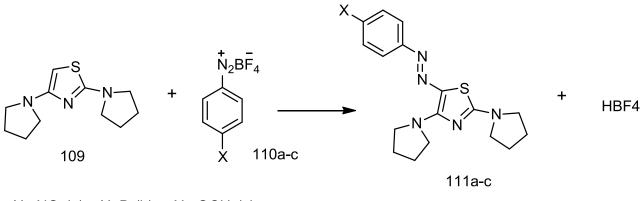
Diazotization of 6-methoxy-1,3-benzothiazol-2-amine , 4-(4-nitrophenyl)-1,3-thiazol-2-amine and 6-methyl-1,3-benzothiazol-2-amine by nitrosyl sulfuric then coupled with barbituric acid **103** gave azo dyes **104a-c** (Harisha, Keshavayya et al. 2017)



Chloroacetyl substituted amides 105 could be refluxed with thiourea to obtain 2-aminothiazol compounds **106.** Aminothiazol compound **107** was diazotized then coupled with pyrrole to give azo compounds **108** (Mahdi 2017)

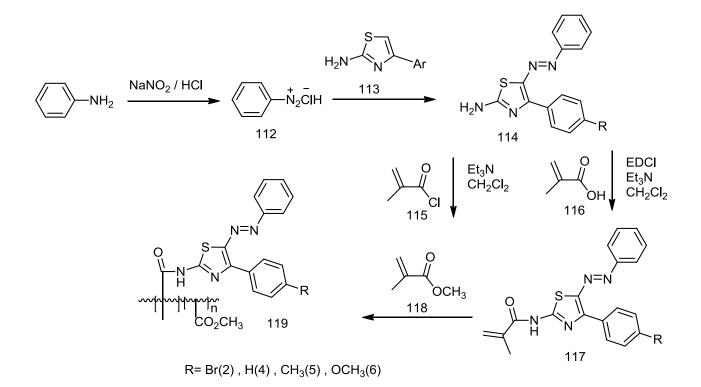


Treatment of 2,4-dipyrrolidinylthiazole **109** with arenediazonium salts **110a-c** afforded 5-(4-Nitrophenyl)diazenyl-2,4-dipyrrolidin-1-yl-1,3-thiazole **111a** and 5-(4-Bromophenyl)diazenyl-2,4-dipyrrolidin-1-yl-1,3-thiazole **111b** and 5-(4-methoxyphenyl)diazenyl-2,4-dipyrrolidin-1-yl-1,3-thiazole **111c** (Boga, Cino et al. 2016)

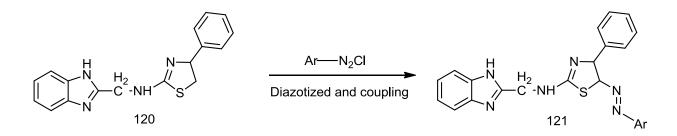


$$X = NO_2(a)$$
,  $X = Br(b)$ ,  $X = OCH_3(c)$ 

Coupling of 4-aryl-1,3-thiazol-2-amine **113** with diazonium salt solution **112** afford 4-(substituted)-5-(phenyldiazenyl)-1,3-thiazol-2-amine **114**, then the product reacted with methacryloyl chloride **115** or condensed with methacrylic acid **116** in the presence of 1-ethyl-3-(3-dimethylaminopropyl)carbodiimide (EDCI) to afford 2-methyl-N-[4-aryl-5-(aryldiazenyl)-1,3-thiazol-2-yl]acrylamides **117**. Copolymers **(119)** produced from the reaction of 2-methyl-N-[4-aryl-5-(aryldiazenyl)-1,3-thiazol-2-yl]acrylamides **117** with methylmethacrylate 118 in anhydrous N,N-dimethylformamide (Derkowska-Zielinska, Skowronski et al. 2016)

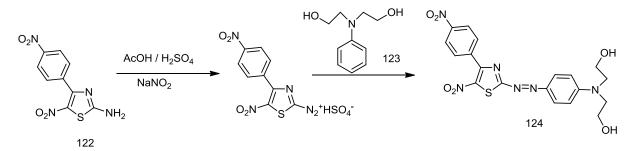


N-((1H-benzo[d]imidazol-2-yl)methyl)-4-phenyl-5-(phenyldiazenyl)- 4,5-dihydrothiazol-2-amine (Dye **121**) could be synthesized by coupling N-((1H-benzo[d]imidazol-2-yl)methyl)-4-phenyl-4,5-dihydrothiazol-2-amine **120** with diazonium salt of arylamine (Chaudhari 2016)

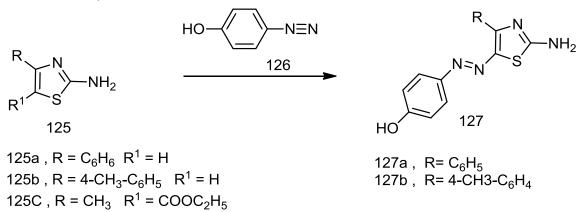


Ar = a=Phenyl , b=4-CloroPhenyl , c=4-BromoPhenyl , d=4-MethoxyPhenyl , e=4-HydroxyPhenyl , f=2,4-DiCloroPhenyl , g=2,4-DiBromoPhenyl , h=4-IsopropylPhenyl

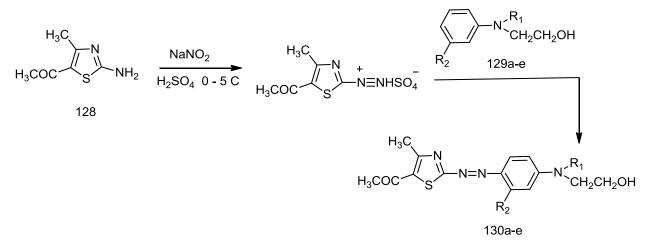
Tasaganva et al. reported that , 5-nitro-4-(4-nitrophenyl) thiazole-2-amine **122** diazotized coupled with N-phenyldiethanol amine **123** to yield chromophore **124** (Tasaganva, Tambe et al. 2011)



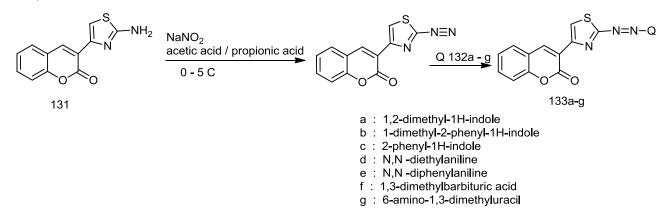
4-(2-amino-4-substituted-thiazol-5-ylazo) phenols (**127a-b**) was prepared from reaction of 4-substituted-thiazol-2-ylamine **125a** or **125b** diazotized and coupled with p-aminophenol diazonium salt **126** (Abdellatif, Abd El Wareth et al. 2015)



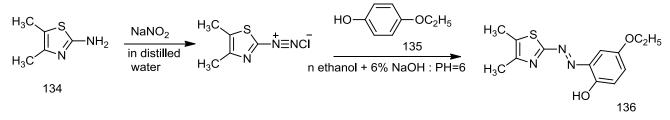
Diazotization of 5-acetyl-2-amino-4-methylthiazole **128** followed by coupling with N-(2-hydroxyethyl)aniline **129ae** afforded azo dye **130a-e** (Maradiya 2010)



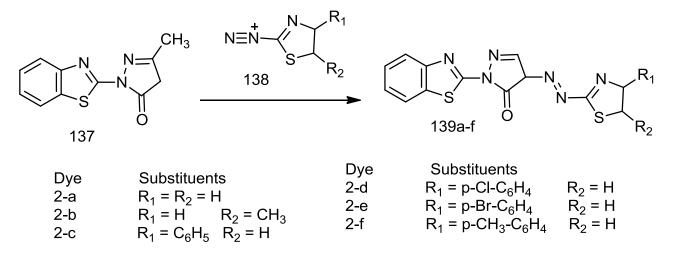
2-amino-4-(coumarin-3-yl)thiazole (**131**) diazotized and coupled with 1,2-dimethyl-1H-indole **132a-g** produced (E)-3-(2-((1,2-dimethyl-1H-indol-3-yl)diazenyl)thiazol-4-yl)-2H-chromen-2-one (**133a-g**) (Özkütük, İpek et al. 2016)



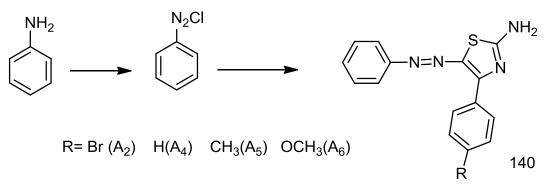
2-Amino - 4,5 -dimethyl thiazole **134** diazotized and coupled with 4-Ethoxy Phenol **135** to yield Ethoxy phenol (DMTAEP) **136** (Al-Adilee, Abass et al. 2016)



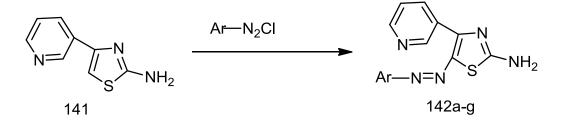
1-(2-benzothiazolyl)-3-methylpyrazol-5-one **137** coupled with heterocyclic amine **138** to afford hetarylazopyrazolone dyes **139-a–f** (Aktan, Ertan et al. 2014)



4-Aryl-5-aryl(hetaryl)diazenyl-1,3-thiazol-2-amines **140** was obtained by diazotization of aromatic amine and coupled with hetarylamine (Derkowska-Zielinska, Skowronski et al. 2015)

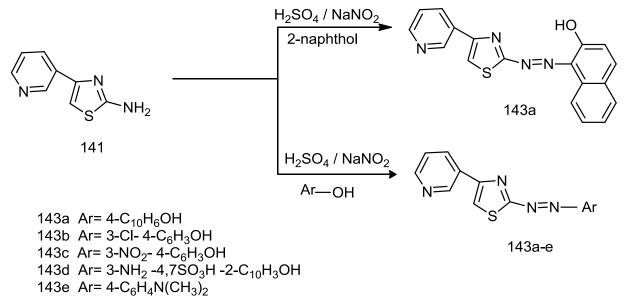


El-Borai reported that 2-amino-4-(pyridin-3-yl) thiazole **141** coupled with diazonium salt of aromatic amines to afford 5-(substituteddiazenyl)-4-(pyridin-3-yl) thiazol-2-amine **142a-g** (El-Borai, Rizk et al. 2014)

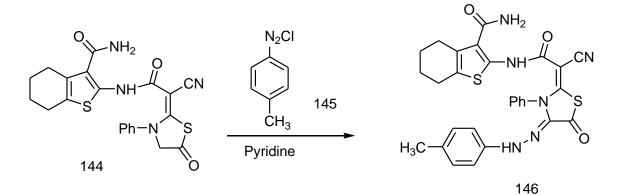


142a Ar= 
$$4-C_6H_4CI$$
142d Ar=  $4-C_5H_4N$ 142b Ar=  $4-C_6H_4OH$ 142e Ar=  $4-C_6H_4COCH_3$ 142c Ar=  $4-C_6H_4OCH_3$ 142f Ar=  $4-C_6H_4COCH=CH-(4-C_6H_4COCH_3)$ 142g Ar=  $C_{10}H_7$ 

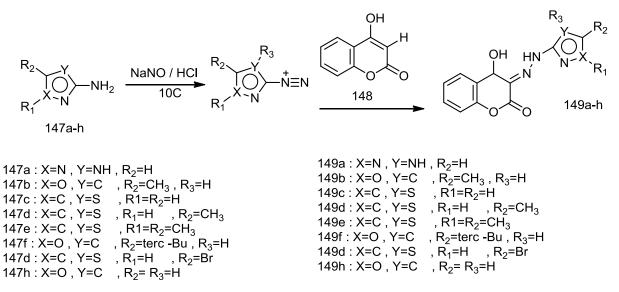
He also reported that 2-amino-4-(pyridin-3-yl) thiazole **141** is diazotized and coupling with aromatic alcohol to obtain 4-pyridyl-2-arylazo thiazoles **143a-e** 



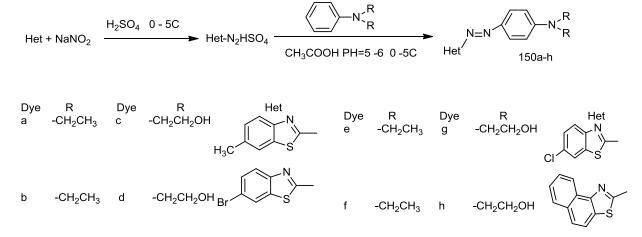
2-[2-cyano-2-(5-oxo-3-phenyl-thiazolidin-2-ylidene)-acetylamino]-4,5,6,7-tetrahydro-benzo[b] thiophene-3carboxamide **144** coupled with diazonium salt of p-tolyldiazonium chloride **145** in pyridine to give 2-[4-(tolylhydrazono)-5-oxothiazolidinone]-cyanoacetamido derivative **146** (Gouda, Berghot et al. 2010)

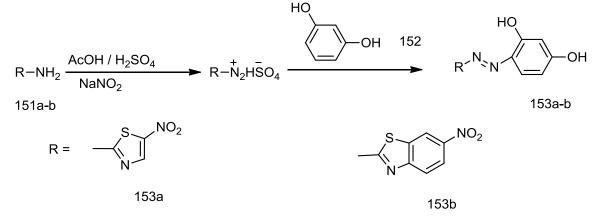


Heterocyclic amines **147a-h** diazotized and coupled with 4-hydroxycoumarin **148** to yield hydrazinylidenechroman-2,4-diones **149a-h** (Jashari, Imeri et al. 2014)

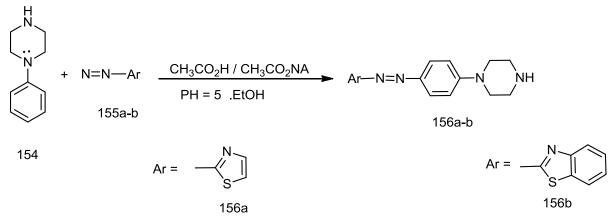


When 2-aminobenzothiazoles diazotized and coupled with N,N-disubstituted anilines gave heterocyclic azo dyes **150a-h** (Moradi Rufchahi, Yousefi et al. 2013)

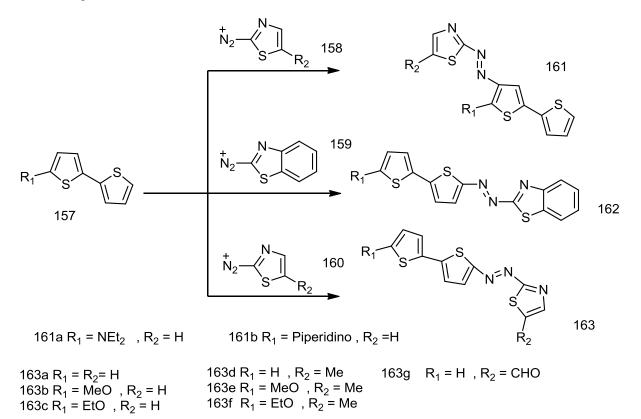




N-phenylpiperazine **154** coupled with diazonium salt of heterocyclic amines **155a-b** yielded triazene dyes **156a-b** (Mohammadi 2014)



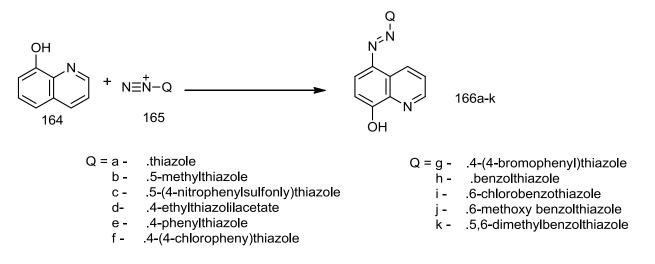
When bithiophenes **157a-e** reacted with diazonium salts of 2-aminothiazole derivatives **158** produced **161a-b**, also when bithiophenes **157a-e** reacted with 2-aminobenzothiazole **159** produced benzothiazolyldiazene 162. bithiophenes **157a-e** reacted with diazonium salts of 2-aminothiazole derivatives **160** to produce **163a-c**, **164a-c**, **165** (Raposo, Castro et al. 2011)



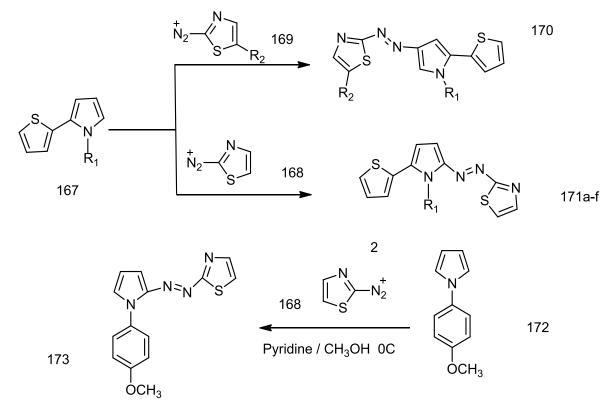
R<sub>1</sub> a =H , b =MeO , c =EtO , d =NEt<sub>2</sub> , e =piperidino R<sub>2</sub> a =H , b =Me , c =CHO

The heteroarylazoquinoline dyes **166a-k** obtained aminothiazole, 2-aminobenzothiazole derivatives **165** 

by coupling 8-hydroxyquinoline **164** with diazotized 2-(Saylam, Seferoğlu et al. 2014)

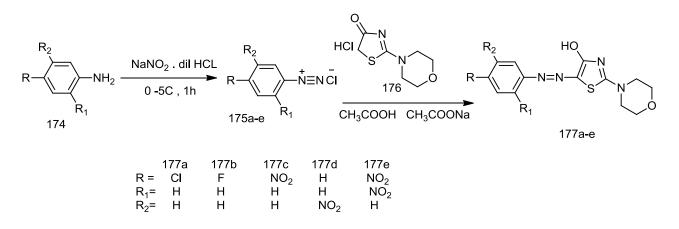


Thienylpyrroles 167 could be coupled with diazonium salts of 2-aminothiazole 168 and 2-amino-5-methylthiazole 169 to give thienylpyrrole azo dyes 171a-f - 170, also diazonium salt of 2-aminothiazole 168 coupled with pyrrole 172 to produce pyrrole azo dyes 173 (Raposo, Fonseca et al. 2011)



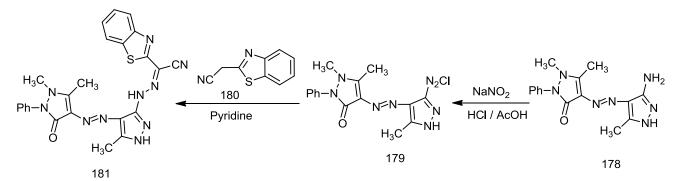
171a R<sub>1</sub> n -Pr 171b R<sub>1</sub> = 4 -MeOC<sub>6</sub>H<sub>4</sub> 171c R<sub>1</sub> = 2,4 -(MeO)<sub>2</sub>C<sub>6</sub>H<sub>3</sub> 171<br/>d $R_1$  = 2,4,5 -(MeO)\_3C\_6H\_2<br/>
171<br/>e $R_1$  = 4 -FC\_6H\_4<br/>
171<br/>f $R_1$  = 4  $BrC_6H_4$ 

Azo dyes **177a-e** afforded by coupling of 2-(morpholin-4-yl)-1,3-thiazol-4(5H)-one hydrochloride **176** with diazonium salt solution of aniline derivatives **175a-e** (Umape, Patil et al. 2013)

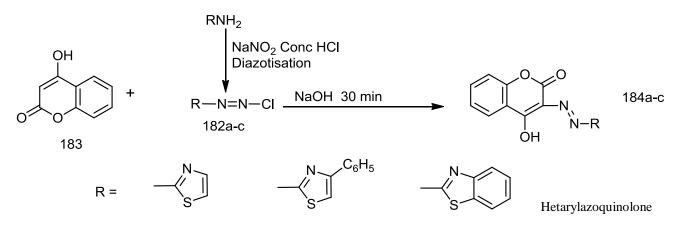


### Aryl azo thiazole derived from benzothiazol and its derivatives:-

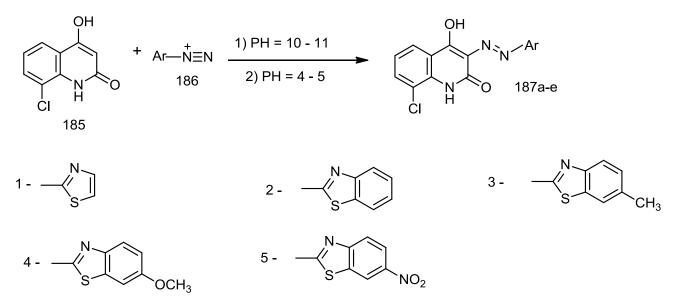
4-((3-amino-5-methyl-1H-pyrazol-4-yl)diazenyl)-1,5-dimethyl-2-phenyl-1H-pyrazol-3(2H)-one **178** is diazotized to give corresponding diazonium salt **179** which coupled with 2-(benzo[d]thiazol-2-yl)acetonitrile **180** in pyridine to give the corresponding hydrazones **181** (Metwally, Gouda et al. 2012)



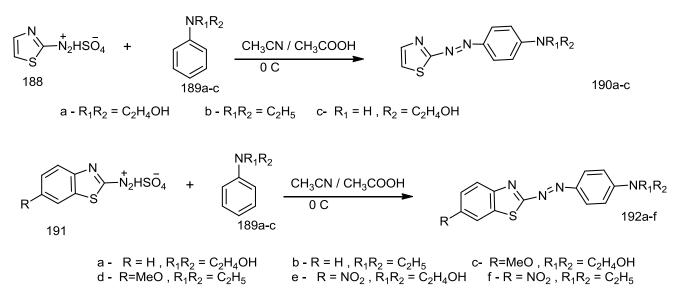
4-hydroxy-3-(heteroaryl-2-yldiazenyl)-2H-chromen-2 -one **184a-c** obtained by coupling 4-hydroxy coumarin (**183**) with diazonium salt of substituted heteryl amine **182a-c** (Sahoo, Kumar Mekap et al. 2015)



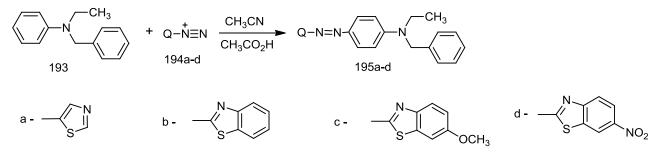
dyes **187a-e** could be constructed through the reaction of 8-chloro-4-hydroxyquinoline-2-(1H)-one **185** with diazonium salt solution of heterocyclic amines 186 (Yahyazadeh and Yousefi 2014)



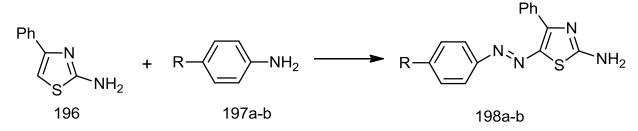
Yazdanbakhsh et al. reported that, diazonium salt solution of thiazolylamines coupled with N-phenyl-2,2-iminodiethanol, N,Ndiethylaniline and 2-anilinoethanol obtained azo dyes **190a-c**, similarly diazonium salt solution of benzo thiazolylamines coupled with N-phenyl-2,2--iminodiethanol, N,Ndiethylaniline and 2anilinoethanol afforded azo dyes **192a-f** (Yazdanbakhsh, Mohammadi et al. 2010)



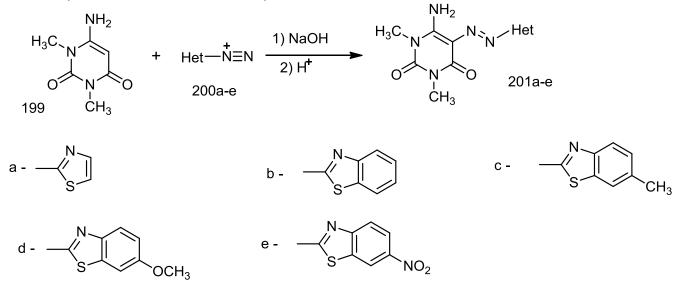
To prepare dyes **195a-d** heterocyclic amines were diazotized then coupled with N-benzyl–N-ethyl–aniline **193** (Yazdanbakhsh, Mohammadi et al. 2010)



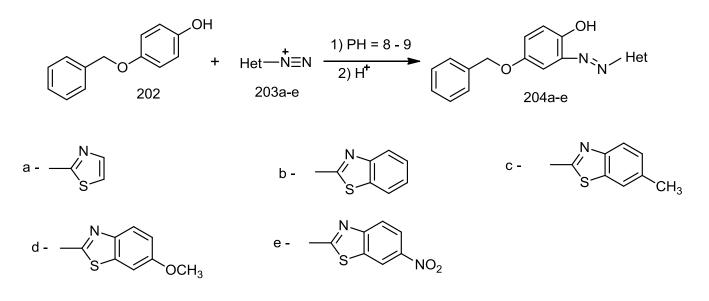
It has been found that, 4-phenyl-2-aminothiazole **196** coupled with diazonium salt solution of p-nitroaniline **197a** or p-methoxyaniline **197b** to afford dyes 5-[2-(4-nitrophenyl)-diazen-1-yl]-4- phenyl-1,3-thiazol-2-amine (**189a**) or dyes 5-[2-(4-methoxyphenyl)-diazen-1-yl]-4-phenyl-1,3-thiazol-2-amine (**189b**) respectively (Yen and Kuo 2012)



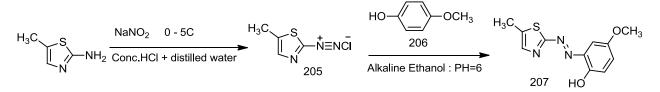
The hetarylazo dyes **201a-e** could be synthesized by coupling of 6-amino-1,3-dimethyluracil **199** with diazotized heterocyclic amines **200a-e** (Yousefi, Yahyazadeh et al. 2012)



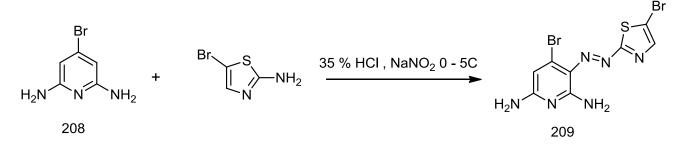
Similarly the hetarylazo dyes **204a-e** obtained by coupling of alkaline solution of 4-benzyloxy phenol **202** with diazotized heterocyclic amines **203a-e** (Yousefi, Yahyazadeh et al. 2013)



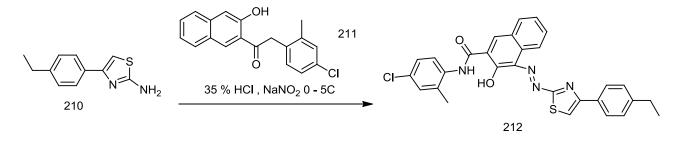
2-[-2-(5-Methyl thiazolyl)azo ]-4-methoxy phenol **207** afforded by coupling 4-methoxy phenol **206** with diazonium chloride solution of 2-amino-5-methyl thiazole **205** (AL-Adilee and Fanfon 2012)



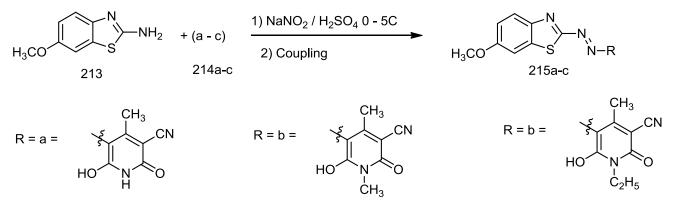
It was reported that , 3-(2-(5-bromothiazol-2-yl) diazenyl)-4-bromopyridine-2,6-diamine **209** obtained by coupling diazonium salt of 5-bromothiazol-2-amine with 4-bromopyridine-2,6-diamine **208** (Savanor Prasanna M. 2013)



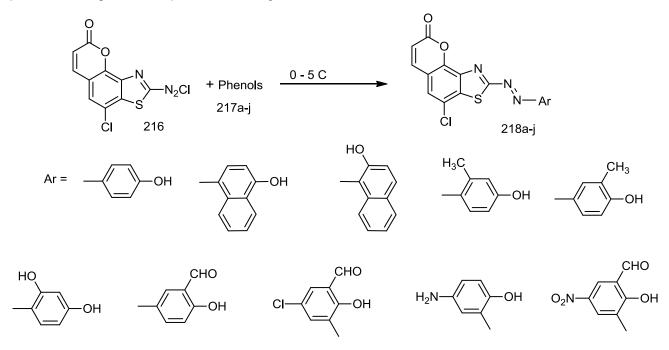
Compound 4-(4-ethylphenyl) thiazol-2-amine **210** was diazotized followed by coupling with N-(4-chloro-2methylphenyl)-3-hydroxynaphthalene-2-carboxamide) **211** to afford N-(4-chloro-2-methyl phenyl)-4-(al 2013)-3hydroxynaphthalene-2-carboxamide **212** (al 2013)



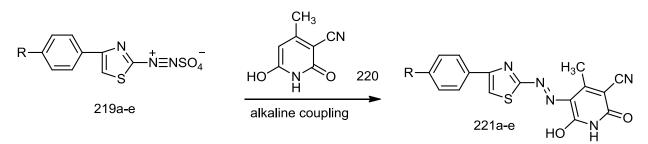
When diazonium salt of 2-Amino6-methoxybenzothiazole **213** reacted with coupler compounds 6-hydroxy 4methyl-2-oxo-1,2-dihydropyridine-3-carbonitrile, 6-hydroxy 4-dimethyl-2-oxo-1,2-dihydropyridine- 3-carbonitrile, 1-ethyl-6-hydroxy-4-methyl-2-oxo-1,2-dihydropyridine-3-carbonitrile **214a–c** gave benzothiazole azo dyes **215a-c** (Harisha S 2015)



2-amino-4-chloro-8Hchromeno-[8,7-d]-1,3-thiazol-8-one **216** diazotized followed by coupling with phenols **217aj** resulted in compound **218a-j** (Thorat, Yamgar et al. 2010)

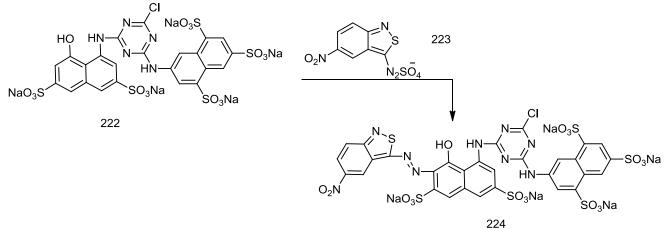


2-amino-4-phenylthiazoles were diazotized by nitrosylsulphuric acid then coupled the diazonium salts **219a-c** with pyridone **220** gave disperse dyes **221a-e** (Franker Amen Imadegbor 2014)

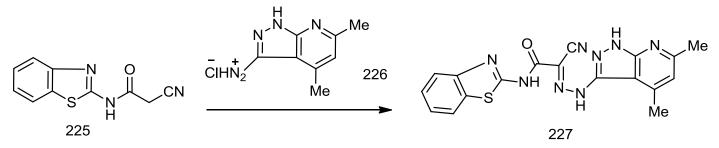


R = 1(a) H, 1(b) CI, 1(c) CH<sub>3</sub>, 1(d) OCH<sub>3</sub>, 1(e) NO<sub>2</sub>

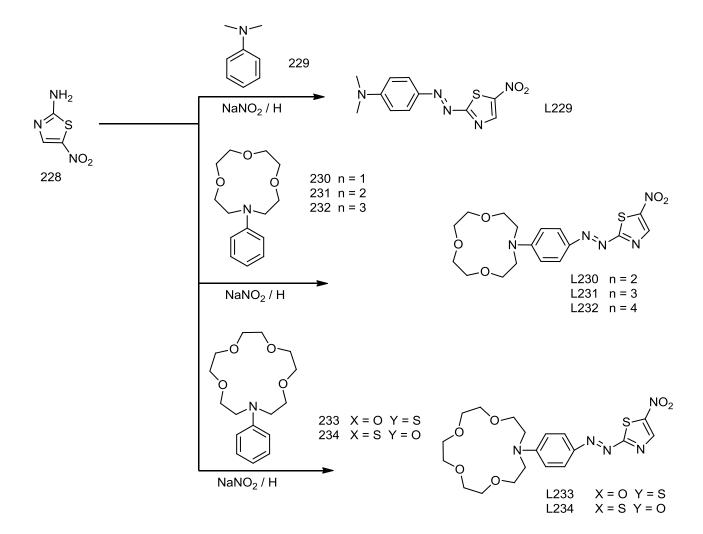
When 3-Amino-5-nitrobenzothiazole 223 diazotized then coupled with the substituted compound containing chlorotriazine 222 afforded pH-sensitive colori-metric dye 224 (Xie, Gao et al. 2014)



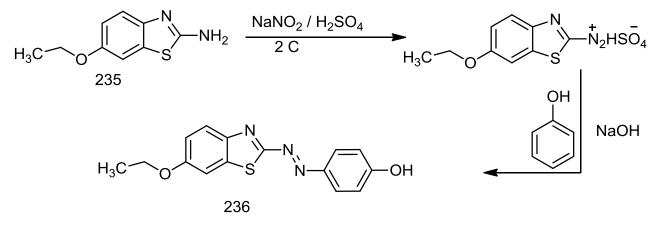
N-(benzothiazol-2-yl)-2-cyanoacetamide **225** coupled with diazotized 3-amino-4,6-dimethyl-2Hpyrazolo[3,4-b]pyridine **226** in pyridine afforded the hydrazono derivative **227** (Bondock, Fadaly et al. 2010)



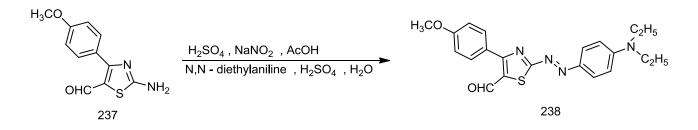
It has been found that, diazonium salt of 2-amino-5-nitrothiazole **228** coupled with N,N-dimethylaniline **229** to yielded **L229**, similarly diazonium salt of 2-amino-5-nitrothiazole **228** coupled with 4-(N-crown)phenyl derivatives to obtain **L230–L234** (Ábalos, Moragues et al. **2012**)



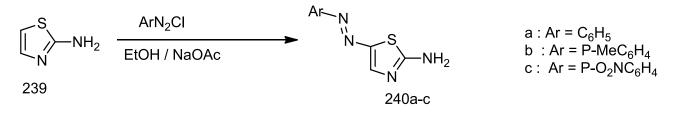
When 6-ethoxy-2-aminobenzothiazole **235** diazotized and coupled with phenol afforded 4-((5-ethoxybenzothiazol-2-yl)diazenyl)phenol **236** (ARWA ALSHARGABI and TAKEUCHI 2013)



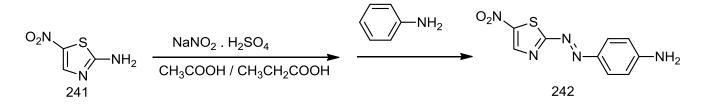
Diazotization of 2-Amino-4-(4-methoxyphenyl)thiazole-5-carbaldehyde **237** then the product coupled with N,Ndiethylaniline to produce azo dye 2-((4-(diethylamino)phenyl)diazenyl)-4-(4-methoxyphenyl)thiazole-5carbaldehyde **238** (El-Shishtawy, Borbone et al. 2013)



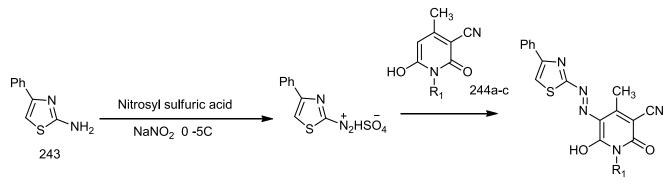
Coupling of 2-aminothiazole **239** with aromatic diazonium salts in ethanol and sodium acetate, afforded the corresponding 2-amino- 5-arylazothiazoles **240a–c** (Khalifa, Abdel-Latif et al. 2015)



When 2-Amino-5-nitrothiazole **241** diazotized and coupled with aniline afford monoazo dye **242** (Kim, Lee et al. 2013)



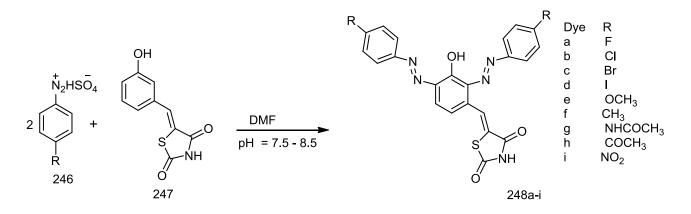
4-Phenyl-2-aminothiazole **243** diazotised with nitrosyl sulfuric acid and coupled with 2-pyridone derivatives **244a–c** to obtain Bishetroaryl Monoazo Dyes **245a–c** (Modi and Patel 2013)



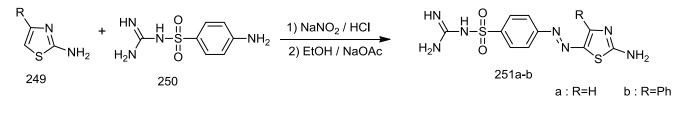
where  $R_1 = (a) - CH_2CH_3$ . (b)  $-CH_2CH_2CH_2CH_3$ . (c)  $-(CH_2CH_2CH_2)OCH_3$ 

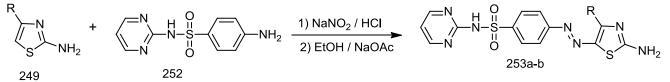
When different aryl diazonium salts **246** coupled with 5-(3-hydroxybenzylidene) thiazolidine-2,4-dione **247** in alkaline medium gave bis-azo dyes **248a** –i (Mohammadi and Safarnejad 2014)

245a-c



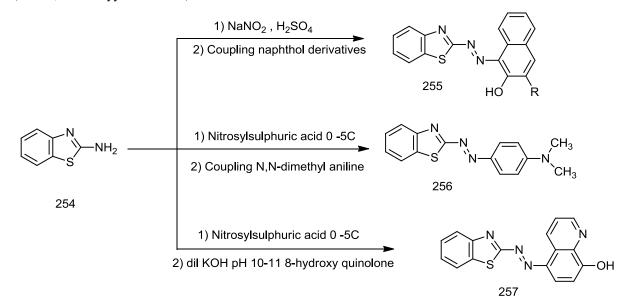
Diazonium chloride derived from sulfaguanidine **250** coupled with 2-aminothiazole derivatives **249** to give 2-Amino-5-substituted thiazole **251a-b.** Coupling of 2-aminothiazole derivatives **249** with the diazonium chloride derived from sulfapyrimidine **252** gave the corresponding 2-amino-5-arylazothiazole derivatives **253a-b** (Gaffer, Fouda et al. **2016**)



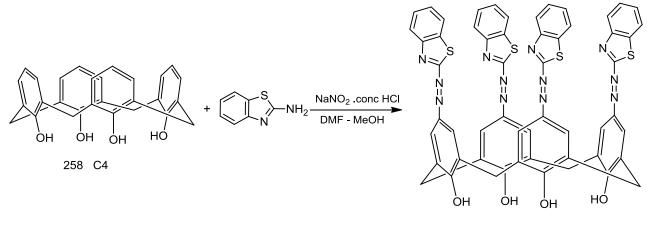


a:R=H b:R=Ph

When 2-amino benzothiazole **254** diazotized and coupled with naphthol derivatives, N, N-dimethyl aniline, 8-hydroxy quinolone and to obtain 2-amino benzothiozole substituted azo dye **255**, **256** and **257** Respectively (Kumar, Keshavayya et al. 2013)

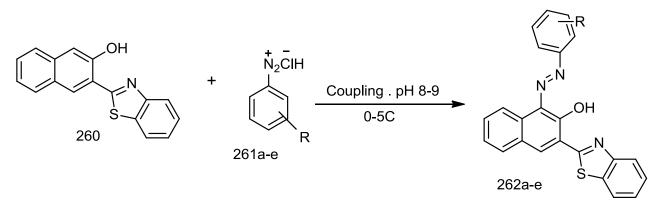


Treatment a solution of benzothiazole diazonium chloride with calix[4]arene **258** C4 produced Chromogenic calixarene, **259** BTC4 (Bingol, Kocabas et al. 2010)



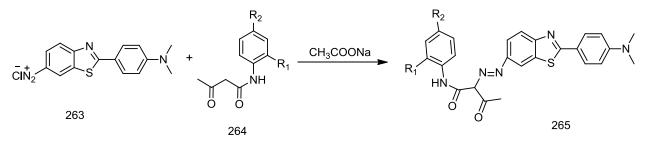
259 BTC4

Diazotized aromatic amine **261a-e** was added to 3-(1,3-benzothiazol-2-yl)naphthalen-2-ol **260** to obtain azo disperse dyes **262a-e** (Satam, Raut et al. 2013)



a:  $R =_4$ -Cl b:  $R =_H$  c:  $R =_4$ -NO<sub>2</sub> d:  $R =_4$ -OCH<sub>3</sub> e:  $R =_2$ -OH, 5-NO<sub>2</sub>

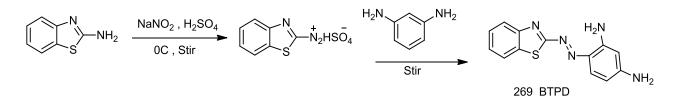
The synthesis of dye **265** was achieved by coupling diazonium salt of 2-(4-(dimethylamino)phenyl) benzo[d]-thiazol-6-amine **263** with acetoacetanilide coupler **264** (Kasture, Sonawane et al. 2010)



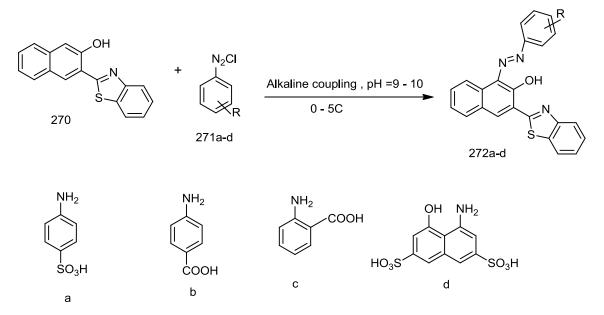
C.I. Disperse Blue **268** prepared by diazotization 3-amino-5-nitro-[2,1]-benzisothiazole **266** which coupled with N-ethyl-N-2-(methoxycarbonyl)ethylaniline **267** (Qian, Wang et al. 2013)



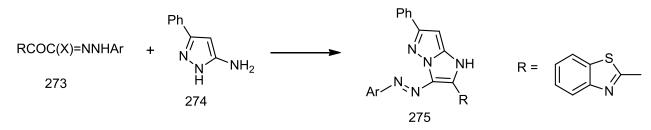
A chromophore molecule 4-[(benzothiazole-2-yl) diazenyl] phenyl-1,3-diamine **269** BTPD prepared by coupling diazonium salt solution of 2-amino benzothiazole with m-phenylenediamine (Qiu, Chen et al. 2014)



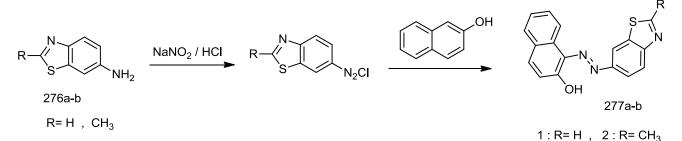
Acid azodyes **272a-d** prepared by coupling diazonium salt of substituted aromatic amines containing acid functionality **271a-d** with 3-(1,3-benzothiazol-2-yl)naphthalen-2-ol **270** (Satam, Raut et al. 2013)



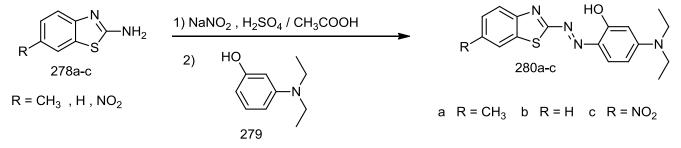
3-Arylazo-2-substituted-1H-imidazo[1,2-b]pyrazoles **275** treated with 5-amino-3-phenyl-pyrazole **274** with 2-oxohydrazonoyl halides **273** in ethanol (Shawali 2010)



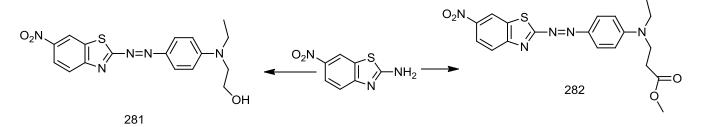
When 6-aminobenzothiazole **276a** and 6-amino-2-methylbenzothiazole **276b** diazotized then coupled with 2naphthol afforded benzothiazolyl azo derivatives **277a-b** (Racané, Mihalić et al. 2013)



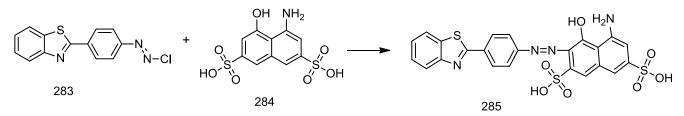
Synthesis of Azo dyes **280a-c** was carried out by diazotization of substituted 2-aminobenzo thiazole **278a-c** then coupled with 3-(Diethylamino)phenol **279** (Tao, Xu et al. 2012)



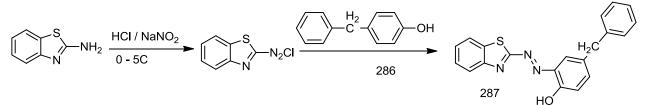
When 2-amino-6-nitrobenzothiazole diazotized followed by coupling with N-Ethyl-N'-phenyl ethanolamine and N-ethyl-N'-2-(methoxycarbonyl)ethylaniline afforded dye **281** and dye **282** respectively (Wang, Wang et al. 2015)



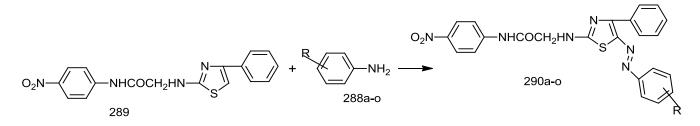
The synthesis of dye 5-Amino-3-(4-Benzthiazol-2-yl-Phenylazo)-4-Hydroxy-Naphthalene-2,7- Disulphonic Acid **285** was achieved by coupling diazonium salt of 4-Benzthiazole-2-yl-phenylamine **283** with H-Acid **284** (D. M. Vashi 2014)



It was reported that , 2 – Amino benzothiazole diazotized and coupled with p-benzyl phenol **286** to afford 2-(Benzothiozolyl azo-4- benzyl phenol **287** (Mehdi, Al-khafagy et al. 2013)

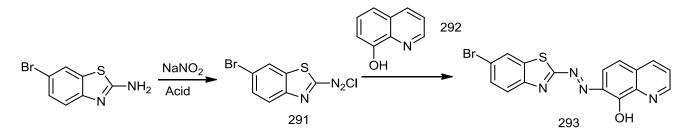


Different aryl amines **288a-o** diazotized and coupled with N-(4-nitrophenyl)-2-[(4-phenyl-1, 3-thiazol-2-yl) amino] acetamide **289** give dye **290a-o** (Zadafiya, Tailor et al. 2013)



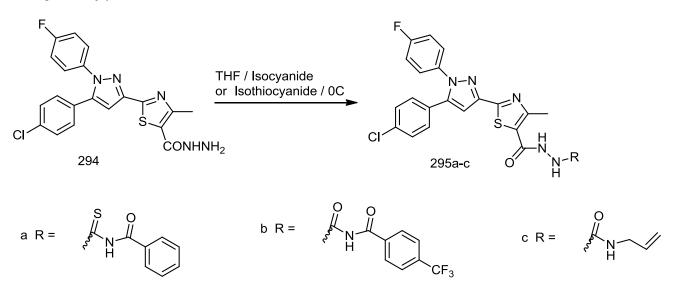
R = H , 4-NO<sub>2</sub> , 3-NO<sub>2</sub> , 2-CN,4NO<sub>2</sub> , 2-OH , 3-OH , 4-OH , 4-CH<sub>3</sub> , 3-CH<sub>3</sub> , 4-CI , 2,4-di NO<sub>2</sub>,6-CI , 2,6-di Br,4-NO<sub>2</sub>

when 8-hydroxyquinoline **292** coupled with diazonium salt of 2-amino-6-bromobenzothiazol **291** produced 7-(6-bromo-2-benzothiazolylazo)-8-hydroxyquinoline **293** (Kasim Hassan Kadhim 2014)

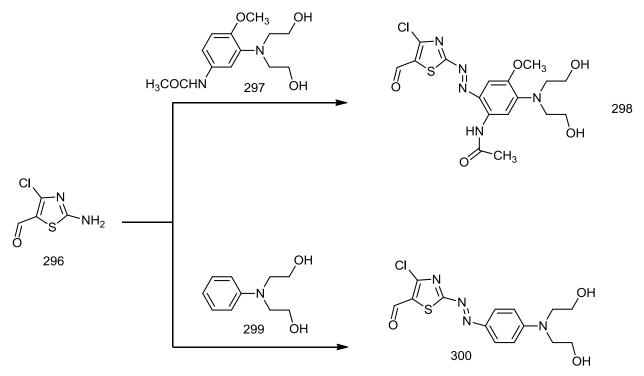


#### Aryl azo thiazole derived from different Heterocyclic Compounds:-

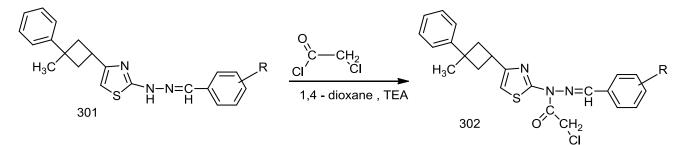
It has been found that, 2-[5-(4-chlorophenyl)-1-(4-fluorophenyl)-1Hpyrazol-3-yl]-4-methyl-thiazole-5-carboxlic acid hydrazide (**294**) was reacted with the substitutional isocyanate (or) isothiocyanate afford **295a-c** (Ragavan, Vijayakumar et al. 2010)



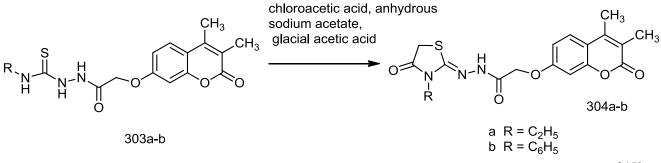
Diazotation of 2-amino-4-chloro-5-formylthiazole **296** followed by coupling with N-phenyl diethanolamine **299** and N-acetamide **297** afforded 2-((4-(bis(2-hydroxyethyl) amino) phenyl) diazenyl)-4-chlorothiazole-5-carbaldehyde **300** and N-(5-(bis (2-hydroxyethyl) amino)-2-((4-chloro-5-formylthiazol-2-yl) diazenyl)-4-methoxyphenyl)acetamide **298** respectively (Borbone, Carella et al. 2011)



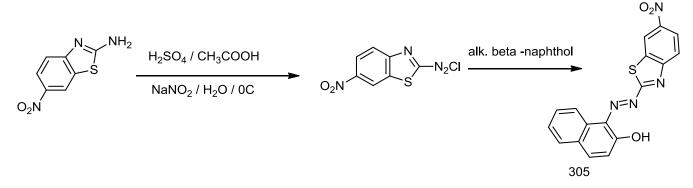
Solution of thiazole derivatives **301** in dioxane reacted with chloroacetyl chloride in the presence of of triethylamine gave N-( substitutional bromobenzylidene)-2-chloro-N-(4-(3-methyl-3-phenylcyclobutyl)-thiazol-2-yl)acetohydrazide **302** (Cukurovali and Yilmaz 2014)



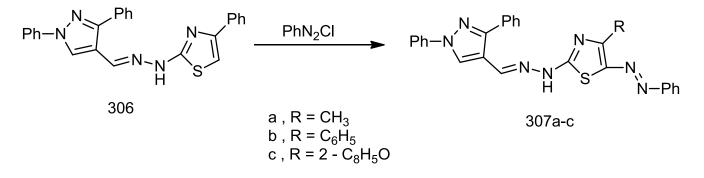
1-(3,4-dimethyl-2-oxo-2H-1-benzopyran-7-yloxy) acetyl-4-substituted thiosemicarbazides **303a-b** reacted with chloroacetic acid, anhydrous sodium acetate in glacial acetic acid gave N<sup>-</sup>-(4-Oxo-3-substituted thiazolidin-2-yl)-2-(3,4-dimethyl-2-oxo-2H-1-benzopyran-7-yloxy) acetohydrazides **304a-b** (El-Ansary, Hussein et al. 2014)



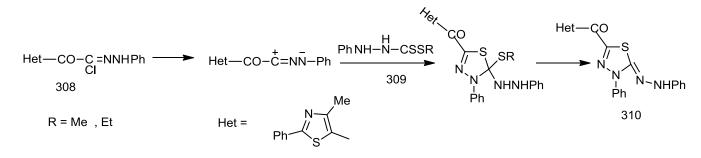
To obtain 1-(6-nitrobenzothiazole-2-ylazo)naphthalene-2-ol **305** added diazonium salt of 2-amino-6-nitrobenzothiazole to alkaline  $\beta$ -naphthol (Misra, Shahid et al. 2010)



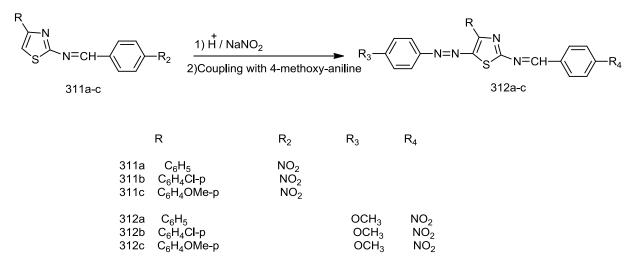
When 1-((1,3-diphenyl-1*H*-pyrazol-4-yl)methylene)-2-(4-phenylthiazol-2-yl)hydrazine **306** reacted with benzenediazonium chloride gave **307a-c** (2013)



Reaction of methyl N-phenyldithiocarbazate **309** with the nitrilimines, generated from the N-aryl C-hetaroylhydrazonoyl chlorides **308** in ethanol in the presence of triethylamine, gave the thiadiazole derivative **310** (Shawali 2014)



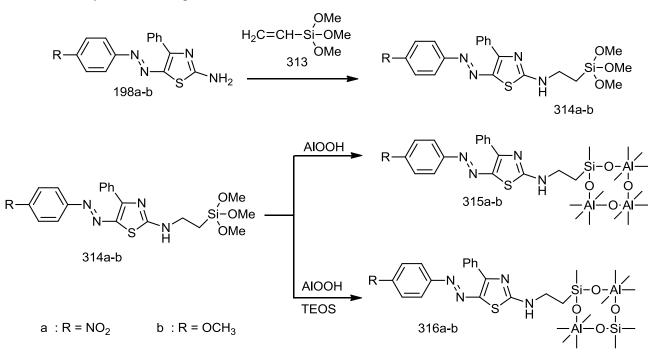
Reactions between 4-methoxy-aniline , as diazo components, and coupling components dyes **311a-c** would lead to obtain dyes **312a-c** (Yen 2012)



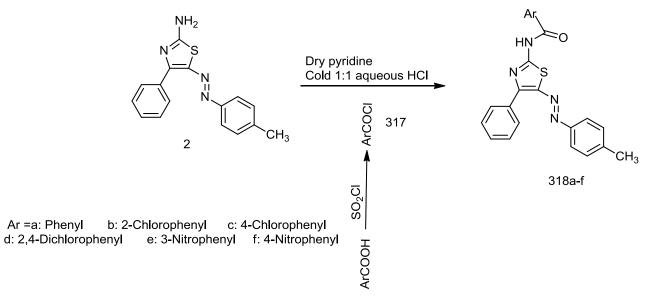
## Chemical reactivity of Aryl azo thiazole:-

### Chemical reactivity of 5-arylazo-thiazole Derivatives:-

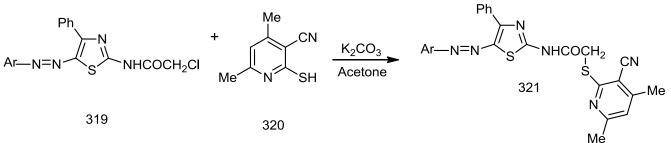
Reaction of dyes 5-[2-(4- substituted phenyl)-diazen-1-yl]-4-phenyl-1,3-thiazol-2-amine **198a-b** with vinyltriethoxysilane **313** afforded 5-[2-(4- substituted phenyl)-diazen-1-yl]-4-phenyl-N-[2-(triethoxysilyl)ethyl]-1,3-thiazol-2-amine, also compounds **314a-b. 314a-b** condensed with boehmite sol AlO(OH) in ethanol and hydrochloric acid yielded **315a-b**, similarly, compound **314a-b** condensed with TEOS and boehmite sol AlO(OH) in ethanol and hydrochloric acid produced **316a-b** (Yen and Kuo 2012)



It has been found that, 2-amino-5-(4-methylphenyl)-diazenyl-4- phenyl-1, 3-thiazole **2** was condensed with appropriate substituted acid chlorides **317** afforded N-[5- (4-Methylphenyl)diazenyl-4-phenyl-1,3-thiazol-2-yl]benzamide derivatives **318** (Prajapati and Modi 2011)



2-(N-chloroacetyl)-5-arylazo-thiazole derivatives **319** refluxed with 4, 6- dimethyl-2-mercaptonicotinonitrile **320** and anhydrous potassium carbonate in acetone to give 2-(3-cyano-4,6-dimethylpyridin-2-ylthio)-N- (5-arylazo-4phenylthiazol-2-yl)acetamides **321** (E. Khalifa, Abdel-Latif et al. 2013)

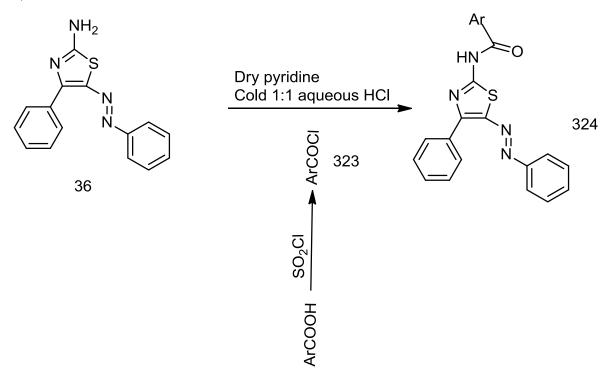


The nicotinonitrile derivative **321** refluxed with solution of sodium ethoxide afforded 3-amino-N-(4-phenyl-5-arylazo-2-thiazolyl)-thieno[2,3-b]pyridine-2-carboxamide dyes **322** 

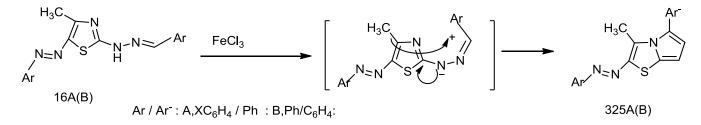
 $\begin{array}{c} Ph \\ Ar-N=N \\ 321 \\ 321 \\ Me \end{array} \xrightarrow{\begin{subarray}{c} Ph \\ S \\ S \\ 321 \\ Me \end{array} \xrightarrow{\begin{subarray}{c} CN \\ NaOEt \\ NaOEt \end{array} \xrightarrow{\begin{subarray}{c} Ph \\ Ar-N=N \\ S \\ 322 \\ 322 \\ Me \end{array} \xrightarrow{\begin{subarray}{c} Ph \\ H_2N \\ H$ 

a : Ar =  $C_6H_5$  b : Ar = p-MeC<sub>6</sub>H<sub>4</sub> c : Ar = p-MeOC<sub>6</sub>H<sub>4</sub> d : Ar = p-NO<sub>2</sub>C<sub>6</sub>H<sub>4</sub> e : Ar = p-BrC<sub>6</sub>H<sub>4</sub>

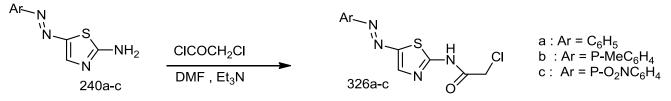
It was reported that, 2-amino-4-phenyl-5-phenylazothizole **36** reacted with substituted aromatic acid chloride **323** in dry pyridine to obtain N-(4-phenyl-5-phenylazo-thiazol-2-yl)-substituted arylamide **324** (Prajapati and Modi 2010).



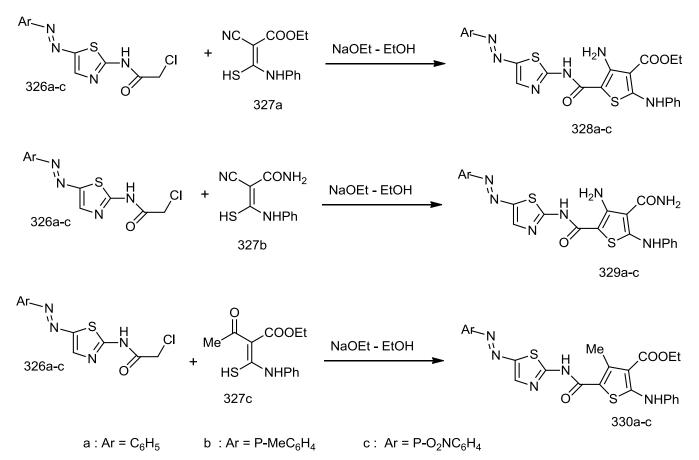
When substituted-benzaldehyde N-(5-arylazo-4-methylthiazol-2-yl)-hydrazones **16** in ethanol refluxed with ferric chloride afforded 3-aryl-5-methyl-6-phenylazo[thiazolo[2,3-c][1,2,4]-triazoles **325** (Shawali and Zayed 2013)



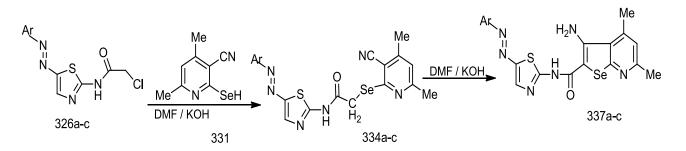
2-Amino-5-arylazothiazoles **240a-c** reacted with chloroacetyl chloride in DMF and triethyl amine to give the corresponding 5- arylazo-2-(N-chloroacetyl)amino-thiazole derivatives **326a-c** (Khalifa, Abdel-Latif et al. 2015)

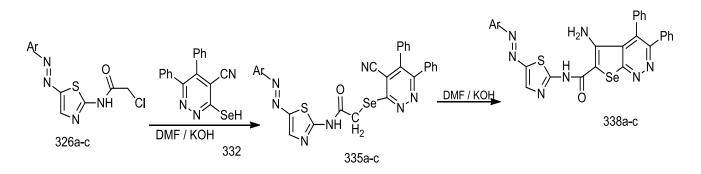


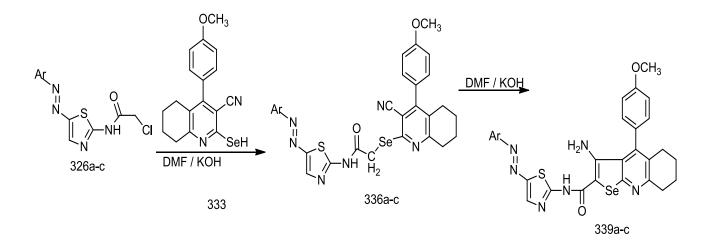
Thiocarbamoyl derivatives **327a-c** undergo condensation reaction with 5-arylazo- 2-(N-chloroacetyl)amino-thiazoles **326a-c** in ethanol and sodium ethoxide afforded the corresponding ethyl 4-amino-5-(5-arylazo-thiazol-2-ylcarbamoyl)-2-phenylamino-thiophene-3-carboxylates **328a-c** , 4-amino-5-(5-arylazo-thiazol-2-ylcarbamoyl)-2-phenylamino-thiophene-3-carboxamides **329a-c** and ethyl 4-methyl-5-(5-arylazo-thiazol-2-ylcarbamoyl)-2-phenylamino-thiophene-3-carboxylates **330a-c** , respectively



when 5-arylazo-2-chloro-N-(thiazol-2-yl)acetamide derivatives **326a-c** reacted with 2-hydroseleno-4,6dimethylpyridine-3-carbonitrile **331**, 3-hydroseleno-5,6- diphenylpyridazine-4-carbonitrile **332** and 5,6,7,8tetrahydro-2-hydroseleno-4-(4'- methoxyphenyl)quinoline-3-carbonitrile **333** in dimethyl formamide with aqueous potassium hydroxide, gave seleno-(pyridine,pyridazine and/or quinoline)carbonitrile derivatives **334a-c**, **335a-c** and **336a-c** respectively. Cyclization happened when the compounds **334a-c**, **335a-c** and **336a-c** reacted with an excess of aqueous potassium hydroxide in (DMF) and afforded 3-amino-4,6-dimethyl-*N*-(5-arylazothiazol-2yl)selenopheno[2,3-b]pyridine-2- carboxamide derivatives **337a-c**, 3-amino-4,6-diphenyl-N-(5-arylazothiazol-2yl)selenopheno[2,3-b]pyridazine-2-carboxamide derivatives **338a-c** and 3-amino-5,6,7,8-tetrahydro-4-(4methoxyphenyl)-*N*-(5-arylazothiazol-2-yl)selenopheno[2,3-b]quinoline-2-carboxamide derivatives **339a-c** (Khalifa, Abdel-Hafez et al. 2015)







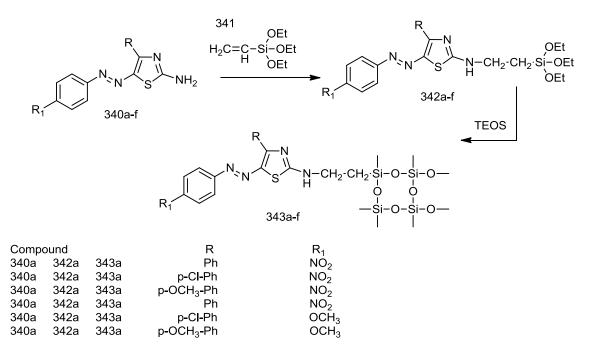


b : Ar= 4-CH<sub>3</sub>-Ph

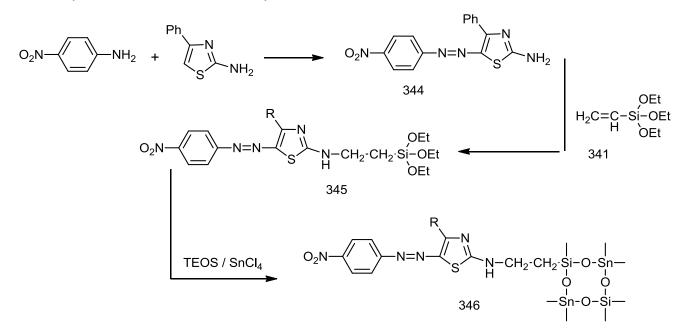
h c:Ar

c : Ar= 4-NO<sub>2</sub>-Ph

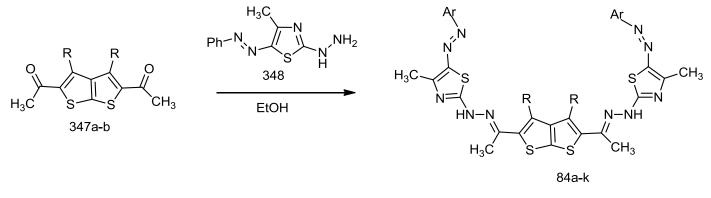
When dyes **340a-f** refluxed with tetrahydrofuran and vinyltriethoxysilane afforded **342a-f** Compounds **343a-f** could be obtained by reaction of **342a-f** with tetraethoxysilane (Yen, Kuo et al. 2012)



Hybrid tin dioxide/silica/thiazole dye materials **346** prepared by heating tetrahydrofuran solutions containing thiazole dyes with tin chloride and tetraethoxysilane (Yen, Kuo et al. 2015)



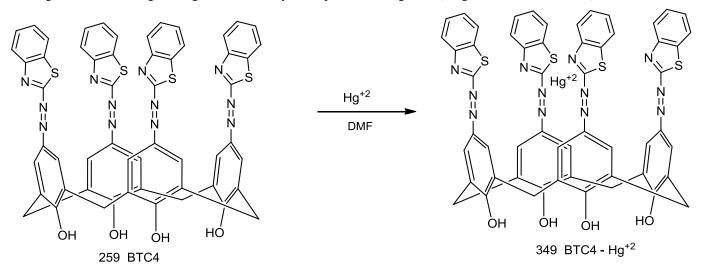
Bisthiazole derivatives **84a-b** prepared by reaction the diacetylthieno[2,3-*b*]thiophene derivative **347a-b** with 2-hydrazino-4-methyl-5-(phenylazo)thiazole **348** (Gomha, Badrey et al. 2016)



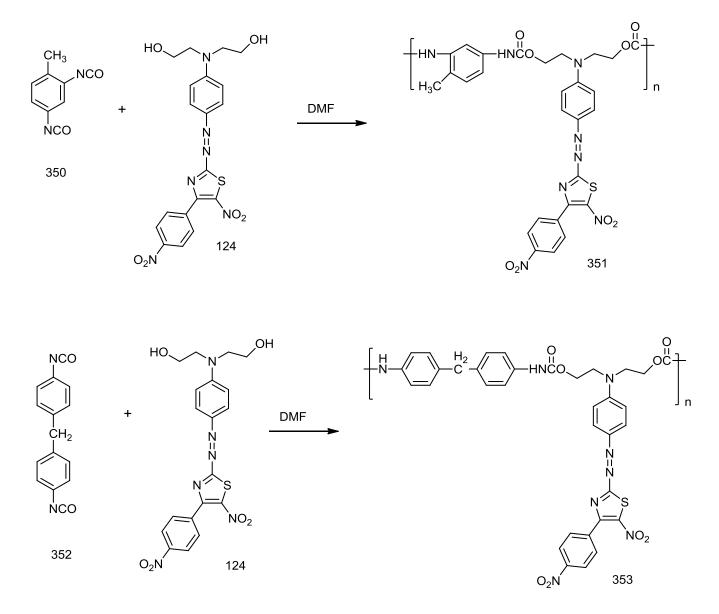
a .R = Ph b .R =Me

## Chemical reactivity of 2-arylazo-thiazole derivatives:-

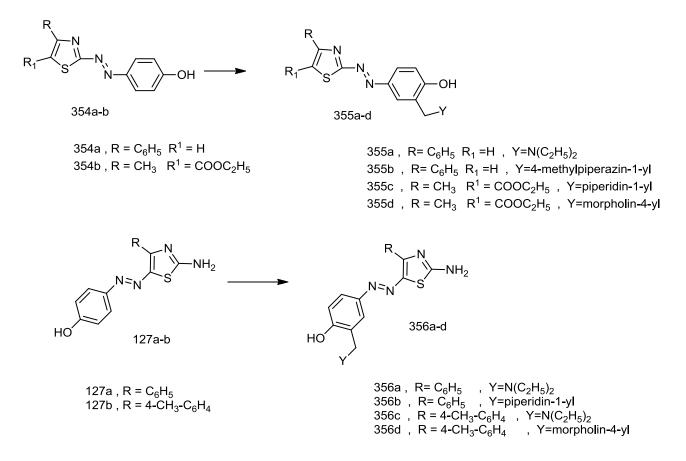
The metal solutions were added into the solution of **259** BTC4 in DMF. The solution of **259** BTC4 showed dramatic changes in color from light orange to reddish only in the presence of  $Hg^{2+}$  (Bingol, Kocabas et al. 2010)



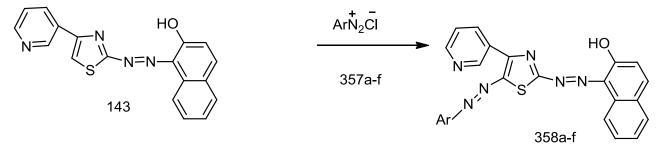
When chromophore **124** reacted with tolylene-2,4-diisocyanate in presence of dry DMF afforded Polyurethane **351** similar procedure for the syntheses of polyurethanes **353** (Tasaganva, Tambe et al. 2011)



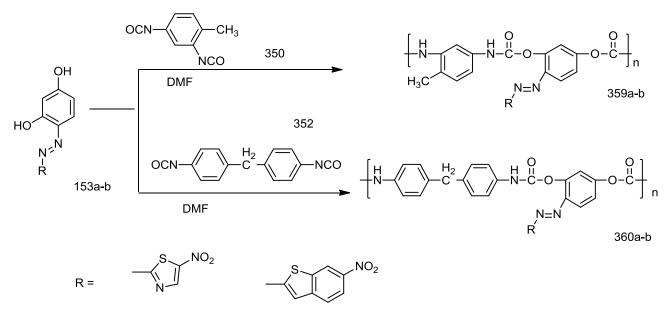
When a Phenolic azo dye **354a-b** added to a mixture of the secondary amine (diethylamine, 4-methylpiperazine, piperidine or morpholine) and formaldehyde gave 4-(thiazol-2-ylazo)-2-substituted-methyl-phenol derivatives (**355a-d**). When a solution of the azo dye 4-(2-amino-4-substituted-thiazol-5-ylazo) phenols (**127a-b**) added to a mixture of the (diethylamine, piperidine, dimethylamine or morpholine) and formaldehyde give 4-(2-amino-4-substituted-thiazol-5-ylazo) phenols (**127a-b**) added to a substituted-thiazol-5-ylazo)-2-substituted-methylphenols **356a-d** (Abdellatif, Abd El Wareth et al. 2015)



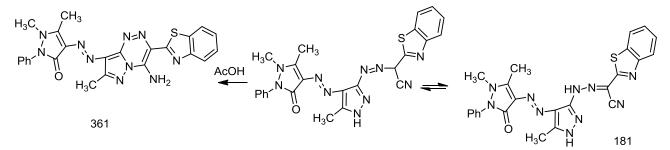
1-[(4-(pyridin-3-yl)thiazol-2-yl)diazenyl]naphthalen-2-ol **143** coupled with some aromatic diazonium salts gave the corresponding 2, 5 diarylazo-thiozoles **358a-f** (El-Borai, Rizk et al. 2014)



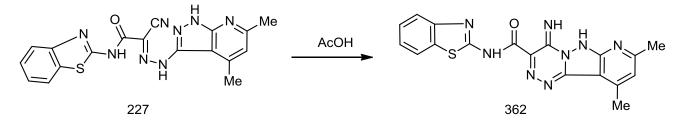
358a Ar=  $4-C_6H_4CH_3$ 358b Ar=  $4-C_6H_4OCH_3$ 358c Ar=  $4-C_6H_4CI$ 358d Ar=  $4-C_6H_4COCH_3$ 358e Ar=  $4-C_6H_4COCH=CH-(4-C_6H_4COCH_3)$ 358f Ar=  $C_{10}H_7$  Chromophores **153a-b** were condensed with tolylene-2,4-diisocyanate (**350**) and 4,4-methylenedi( phenyl isocyanate) (**352**) in a dry DMF solvent to yielded polyurethanes **359a-b** and **360a-b** Respectively (Kariduraganavar, Tambe et al. 2011)



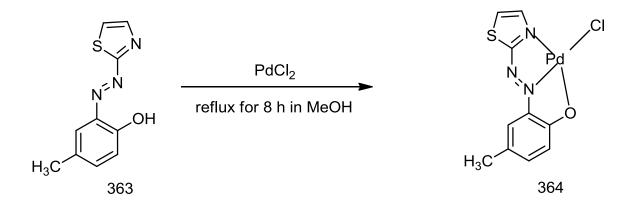
Refluxing of compound **181** in acetic acid afforded the pyrazolotriazines **361** (Metwally, Gouda et al. 2012)



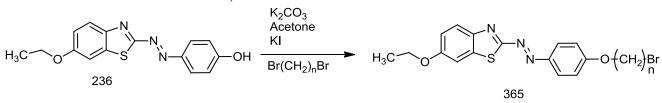
Heating hydrazono derivative **227** in glacial acetic acid obtained the triazine derivative **362** (Bondock, Fadaly et al. 2010)



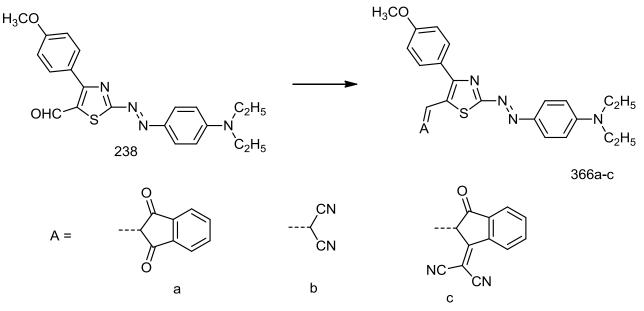
PdCl<sub>2</sub> dissolved in methanol then refluxed with 2-(2-thiazolyl)-4-methylphenol (TAC) **363** afforded [Pd(TAC)Cl] **364** (Biswas, Pramanik et al. 2015)



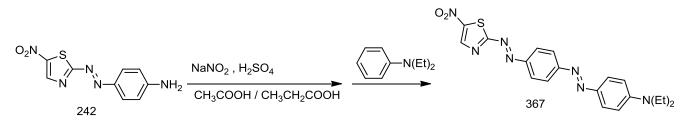
When 4-((5-ethoxybenzothiazol-2-yl)diazenyl)phenol **236** refluxed with potassium carbonate and potassium iodide and dibromoalkane to obtain 4-((6-ethoxybenzothiazol-2-yl)diazenyl) bromo-alkoxyphenyl **365** (ARWA ALSHARGABI and TAKEUCHI 2013)



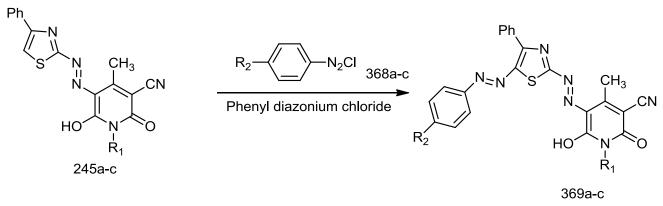
Aldehyde 2-((4-(diethylamino)phenyl)diazenyl)-4-(4-methoxyphenyl)thiazole-5-carbaldehyde **238** undergo Knoevenagel condensation with 1H-indene-1,3(2H)-dione, malononitrile and 2-(3-oxo-2,3-dihydro-1H-inden-1-ylidene)malononitrile in the presence of piperidine/acetic acid catalyst afforded the corresponding NLO chromophores **366a-c** respectively (El-Shishtawy, Borbone et al. 2013)



When monoazo dye **242** diazotized and coupled with N,N-Diethylaniline afford disazo dye **367** (Kim, Lee et al. 2013)

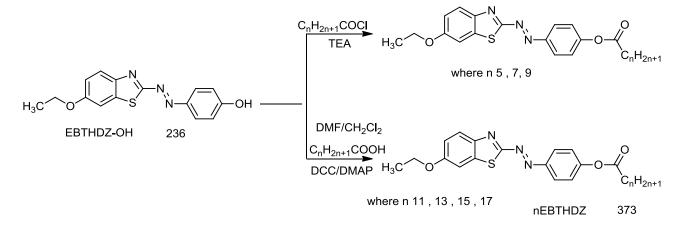


Different aromatic amines **368a-c** diazotized and coupled with Bishetroaryl Monoazo Dyes **245a-c** to afford Bisheteroaryl Bisazo Dyes **369a-c** (Modi and Patel 2013)

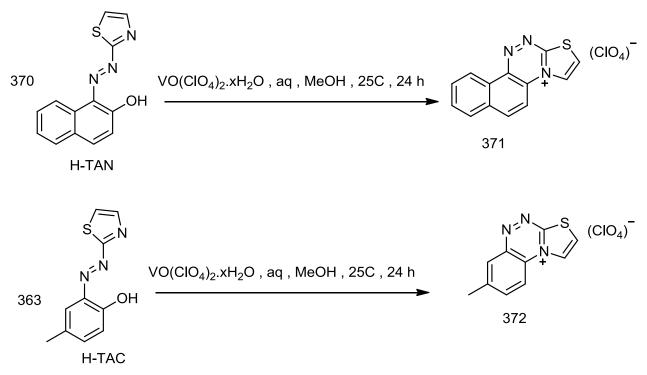


where  $R_1$  = (a) -CH\_2CH\_3  $\ .$  (b) -CH\_2CH\_2CH\_2CH\_3  $\ .$  (c) -(CH\_2CH\_2CH\_2)OCH\_3  $R_2$  = (a) -H ,  $\$  (b) -NO\_2 , (c) -COOH

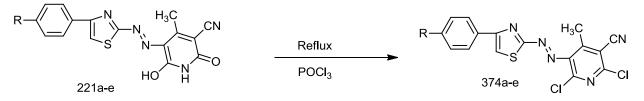
when H-TAN **370** or H-TAC **363** reflux with oxovanadium(IV) perchlorate obtain cyclised triazinium cationic **371 – 372** Respectively (Prasad, Khan et al. 2013)



Yeap et al. reported that, 4-((6-ethoxybenzothiazol-2-yl)azenyl)phenol, EBTHDZ-OH **236** dissolved in THF then reacted with acid chloride  $CnH_{2n+1}COCl$  (where n = 5, 7, and 9) in THF and triethylamine afforded 4-((6-ethoxybenzothiazol-2-yl) diazenyl) phenylalkanoate (nEBTHDZ ) **373** (Yeap, Alshargabi et al. 2012)

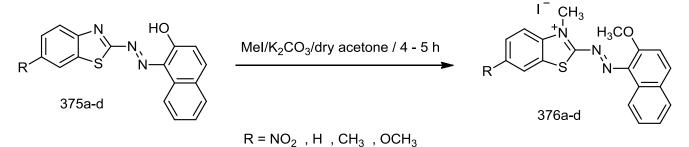


It was reported that, disperse dyes **221a-e** refluxed with phosphorus oxychloride afforded the disperse-reactive dyes **374a-e** (Franker Amen Imadegbor 2014)



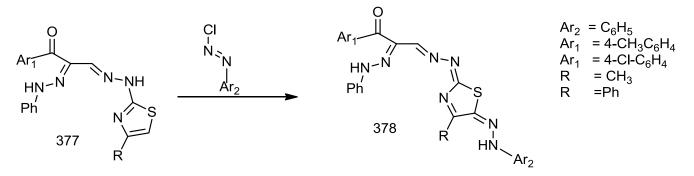
R = 1(a) H, 1(b) CI, 1(c) CH<sub>3</sub>, 1(d) OCH<sub>3</sub>, 1(e) NO<sub>2</sub>

Compound **375a-d** refluxed with acetone, and  $K_2CO_3$  and methyl iodide to give Compound **376a-d** (Misra and Shahid 2010)

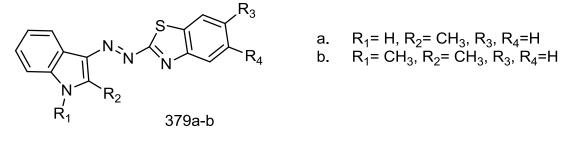


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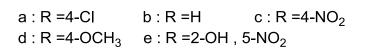
It has been found that, 3-[(4-substituted-thiazol-2-yl)-hydrazono}-1-aryl-2-(aryl-hydrazono) -propan-1-one derivatives **377** reacted with benzendiazonium chloride afforded 3-1-aryl-2-(phenyl-hydrazono)-propan-1-one derivatives **378** (Sayed A. Ahmed 2014)

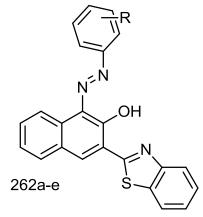


Application of Aryl azo thiazole:-Aryl azo thiazole used as Dyes:benzothiazolylazo indole dyes **379a-b** is stable dye in the solid state. (Seferoğlu, Kaynak et al. 2013)

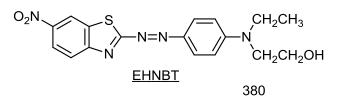


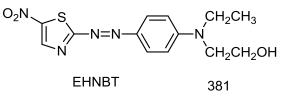
Azo disperse dyes 262a-e showed red shades on polyester fabric. (Satam, Raut et al. 2013)



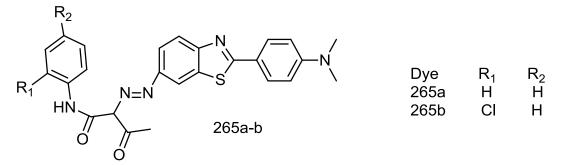


Hetarylazo chromophores **380** - **381** Involved in non-linear optical fluorinated polyimides which exhibited excellent film-forming properties with good optical quality (He, Zhou et al. 2010)

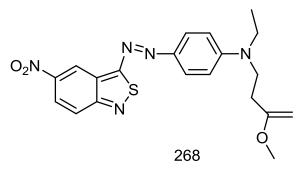




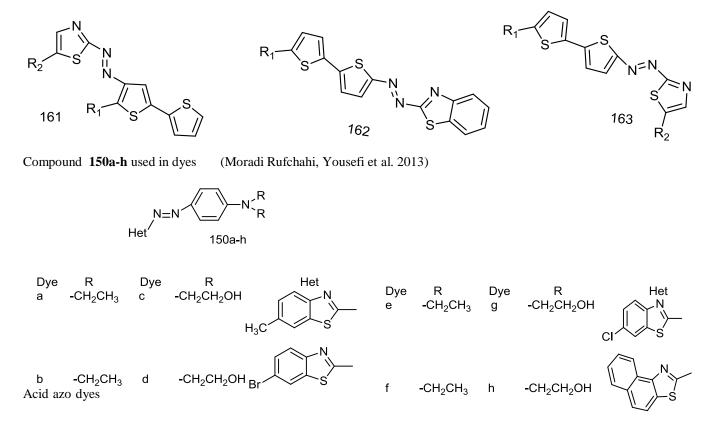
Fluorescent azo dyes **265a-b** exhibited thermal stability, therefore it used for polymer application. (Kasture, Sonawane et al. 2010)



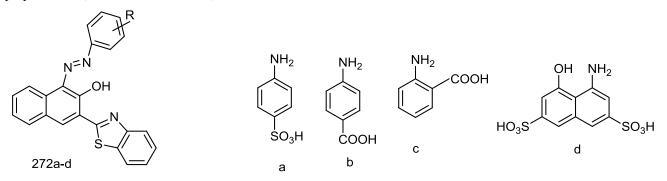
C.I. Disperse Blue 268 is trichromatic blue azo dye and used in cellulose acetate. (Qian, Wang et al. 2013)



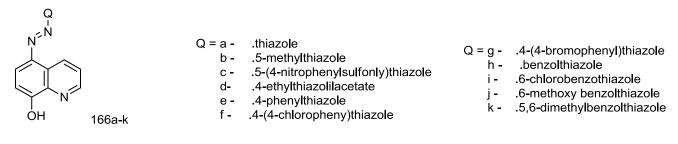
Compounds **161 - 162 - 163** used for dyes with redox properties and exhibited good thermal stability (Raposo, Castro et al. 2011)



272a-**d** is dyes with red color on wool, nylon and silk fabrics but dye **272d** show purple color and has good fastness properties. (Satam, Raut et al. 2013)



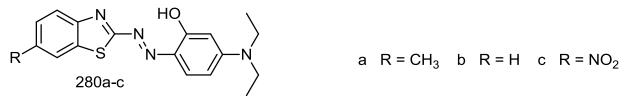
Heteroarylazo-8-hydroxyquinoline 166a-**k** used as dyes that have larger solvatochromic effects than azo-benzene based dyes. (Saylam, Seferoğlu et al. 2014)



6-[(2-hydroxy-1-naphthyl)diazenyl]-2-methylbenzothiazole, **277a** and 6-[(2-hydroxy-1-naphthyl)diazenyl]benzothiazole **277b** used as dyes (Racané, Mihalić et al. 2013)

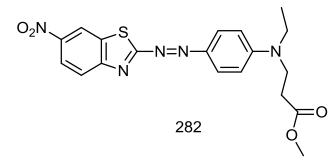


5-(Diethylamino)-2-((6-methylbenzo[d]thiazol-2-yl)diazenyl)phenol **280a** and 2-(Benzo[d]thiazol-2-yldiazenyl)-5-(diethylamino)phenol **280b** and 5-(diethylamino)-2-((6-nitrobenzo[d]thiazol-2-yl)diazenyl) phenol **280c** used as Disperse Red azo dyes (Tao, Xu et al. 2012)



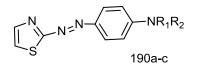
Compound 282 used as dye which exhibit reversible acid-base discoloration

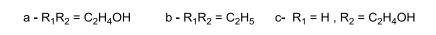
(Wang, Wang et al. 2015)

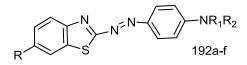


Compounds 190a-c, 192a-f used for dyes

(Yazdanbakhsh, Mohammadi et al. 2010)



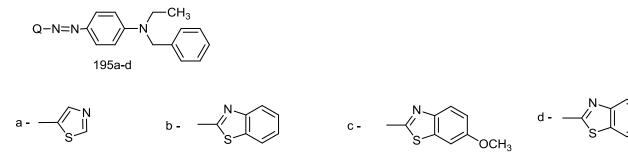




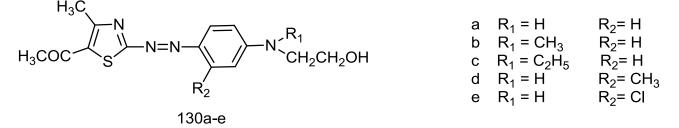
a - R = H ,  $R_1R_2 = C_2H_4OH$ d - R=MeO ,  $R_1R_2 = C_2H_5$ 

 $b - R = H , R_1R_2 = C_2H_5 \qquad c - R = MeO , R_1R_2 = C_2H_4OH \\ e - R = NO_2 , R_1R_2 = C_2H_4OH \qquad f - R = NO_2 , R_1R_2 = C_2H_5$ 

Compounds 195a-d used as azo disperse dyes with metallic luster (Yazdanbakhsh, Mohammadi et al. 2010)

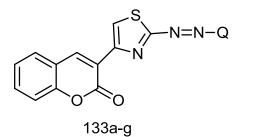


Compounds **130a-e** used as disperse dyes for cellulose triacetate fabric. The dyed fabric show good light fastness (Maradiya 2010)



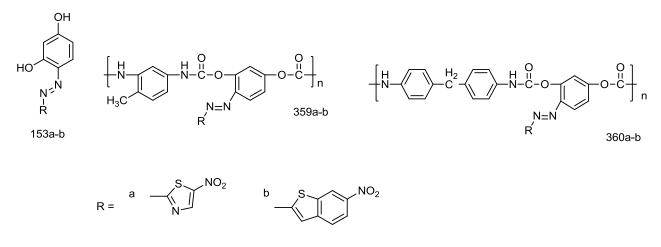
 $NO_2$ 

Compounds **133a-g** used as azo disperse dyes where these dyes are thermal stable up to 269  $^{\circ}$ C (Özkütük, İpek et al. 2016)

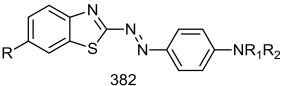


- a : 1,2-dimethyl-1H-indole
- b : 1-dimethyl-2-phenyl-1H-indole
- c : 2-phenyl-1H-indole
- d : N,N -diethylaniline
- e : N,N -diphenylaniline
- f: 1,3-dimethylbarbituric acid
- g: 6-amino-1,3-dimethyluracil

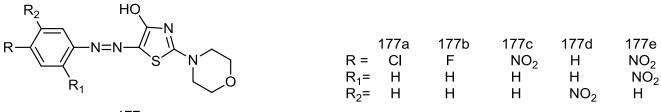
**153a-b** is achromophores which used to produce polyurethanes. These could be easily processed into high quality optical thin films. The polyurethanes show high glass transition temperatures in the range of 140-165 °C (Kariduraganavar, Tambe et al. 2011)



Diazo disperse dyes **382** characterized with bathochromic shift due to extended resonance system (Rauf and Hisaindee 2013)

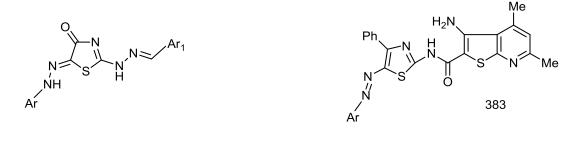


2-morpholin-4-yl-1,3-thiazol-4(5H)-one based azo dyes **177a-e** used as dyes with good thermal stability (Umape, Patil et al. 2013)



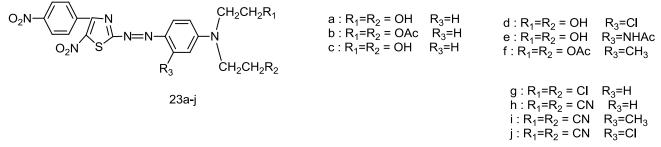
177а-е

Compounds 2-(benzylidenehydrazino)-5-arylazo-thiazolidin-4-one derivatives **5** and **6** and compound 3-amino-N-(4-phenyl-5-arylazo-2-thiazolyl)-thieno[2,3-b]pyridine-2-carboxamide dyes **383** used as disperse dyes on polyester fibers which exhibit very good to excellent washing properties (E. Khalifa, Abdel-Latif et al. 2013)

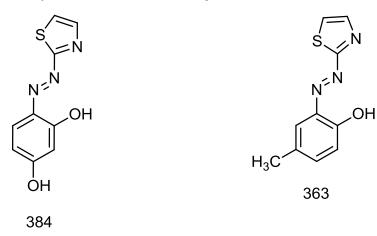


5:  $Ar_1 = C_6H_5$  6:  $Ar_1 = p-MeNC_6H_4$  383:  $Ar_1 = C_6H_5$  -  $p-MeC_6H_4$  -  $p-MeOC_6H_4$  -  $p-NO_2C_6H_4$  -  $p-BrC_6H_4$ 

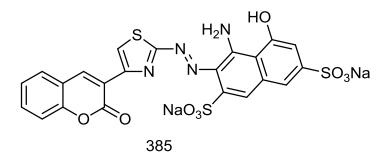
2-[4<sup>-</sup>[N,N-bis(2-hydroxyethyl)amino]phenylazo]-4-(4<sup>-</sup>p-nitro phenyl)-5-nitrothiazole **23** used as dyes when applied on cellulose triacetate fabric , showed good light fastness. (Maradiya 2012)



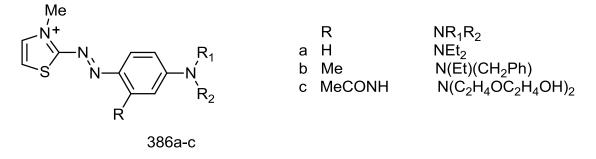
4-(2-thiazolyazo)resorcinol (TAR) **384** and 2-(2-thiazolyazo)-p-cresol (TAC) **363** used as dyes and have high fluorescence intensity in the cyclodextrins solution (Rajendiran, Venkatesh et al. 2014)



Compound Sodium 4-amino-5-hydroxy-3-((4-(coumarin-3-yl)thiazol-2-yl)diazen-yl)naphthalene-2,7-disulfonate **385** used for dyeing of wool and silk fabrics. The dyed fabric show good light fastness (Mohamed, Bashandy et al. 2014)

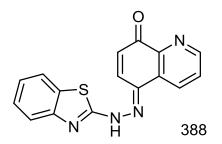


2-[2-[4-(diethylamino)phenyl]diazenyl]-3-methyl-thiazolium **386a-c** used as dyes (Abbott, Batchelor et al. 2013)

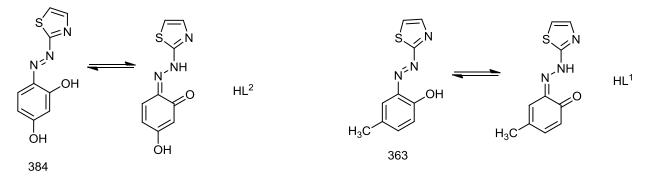


## Aryl azo thiazole used as ligand

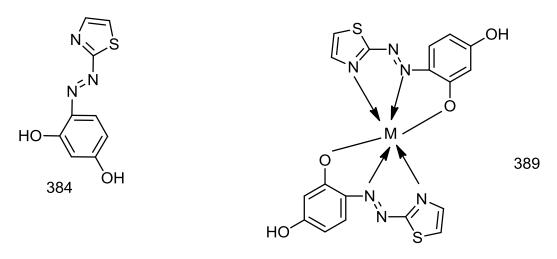
5-(2-benzothiazolylazo)-8-hydroxyquinolene **388** is aligand which used for determination of cobalt in biological,water, soil and pharmaceutical preparation samples (Amin 2014)



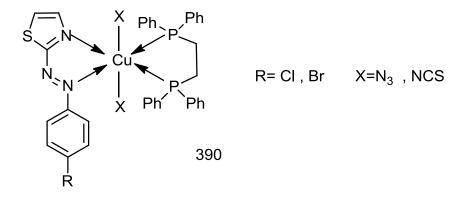
2-(2-thiazolylazo)-p-cresol **363** and 4-(2-thiazolylazo)-resorcinol **384** reacted with  $Zn^{2+}$  to afforded complex the complex exhibit enhancement of fluorescence intensity (Hens, Mondal et al. 2015)



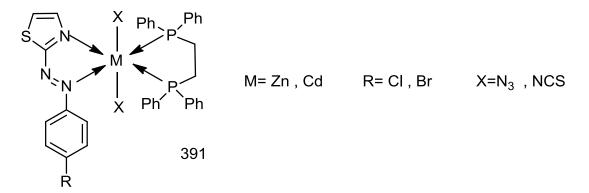
4-(2-thiazolylazo)resorcinol **384** used as ligand and reacted with Manganese(II), cobalt(II) and nickel(II) acetates to form complexes **389**. Mn(II) complex of 4-(2-thiazolylazo)resorcinol used as catalyst for the disproportionation of  $H_2O_2$  in DMF (Karipcin, Dede et al. 2010)



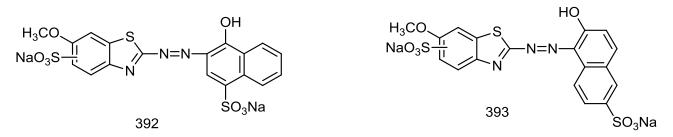
4-(2'-thiazolylazo)halobenzene used as ligand with cis-1,2-bis(diphenylphosphino)ethane)to form complex **390** with copper(II) which exhibit intraligand ( $\pi \rightarrow \pi^*$ ) fluorescence in blue-green region with high quantum yield in DMF solution (Yamgar, Sawant et al. 2010)



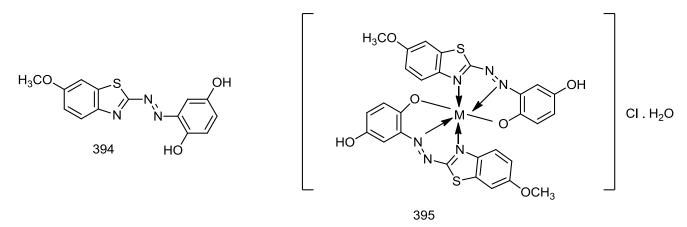
The reaction of thiazolylazo ligands with Zn(II) and Cd(II) salts in presence of 1,2-bis(diphenylphosphino)ethane yields mononuclear complexes **391** which exhibit blue-green emission with high quantum yield (Yamgar, Sawant et al. 2011)



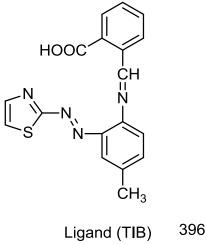
sodium(E)-2-((1-hydroxy-4-sulfonatonaphtalen-2-yl)diazenyl)-6-methoxybenzo[d]thiazole-5 and 7-sulfonate **392** and sodium(E)-2-((2-hydroxy-6-sulfonatonaphtalen-1-yl)diazenyl)-6-methoxybenzo[d]thiazole-5 and 7-sulfonate **393** used as ligand and undergo complexation reaction with Fe(III) ions to form complex Characterized by a thermal resistance up to the temperature of  $190 \, {}^{0}C$  (Chirila, Tulea et al. 2011)



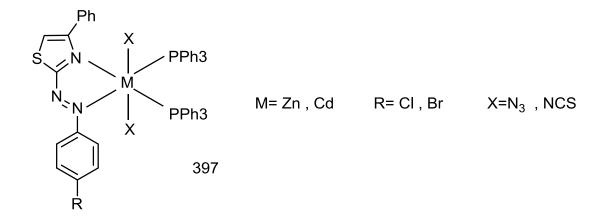
2-[( 6-methoxy-2-benzothiazolyl)azo]-hydroquinone **394** used as ligand to form complexes with Co(II) and Cr(III) ions and used for the spectrophotometric determination of cabalt(II) and chromium(III) (Mussa)



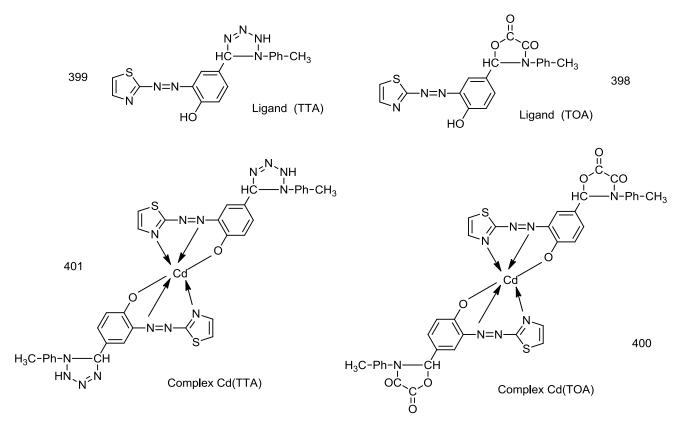
Compound 1-(2 - thiazolazo)-2-(benzoic imine) -5- methyl benzene (TIB) **396** used as ligands with (Ni<sup>+2</sup>) to form complexes (Hassan 2014)



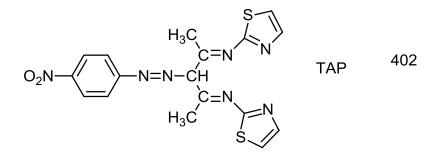
4-(4'-phenyl, 2'-thiazolylazo)substitutedbenzene used as ligand to form complexes 397 which exhibit blue-green



Compounds 2-thiazoleazo [TOA] **398** and 2-thiazoleazo [TTA] **399** used as ligand to form complex **400** and **401** respectively with cadmium (NaghamMahmoodAljamali . 2014)

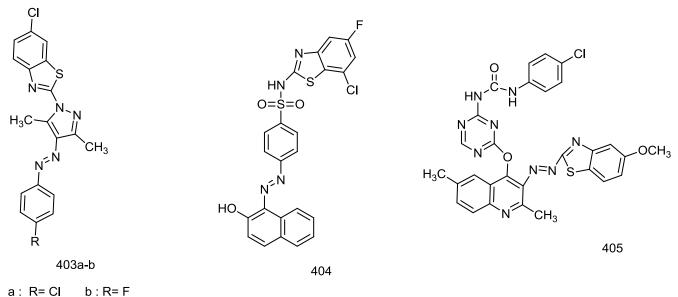


The ligand 3-(4-nito benzene azo)-2,4-bis (2-thiazole imine)-pentane [TAP] **402** was reacted with  $(Mn^{+2},Ni^{+2},Zn^{+2})$  to form metal complexes which show antimicrobial activity against bacteria (*staphyllococcus* .*aureus*) and fungi (Aspergillus *niger*) (Nagham Mahmood Aljamali 2014)

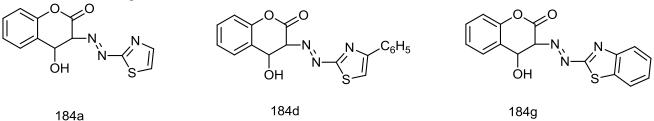


### Aryl azo thiazole used as antimicrobial Activity:-

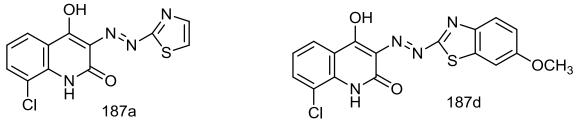
Compound **403a-b** showed significant antibacterial activity, compound (**404**) exhibited good activity against different bacterial and fungal strain and also compound (**405**) increased the antimicrobial activity (Keri, Patil et al. 2015)



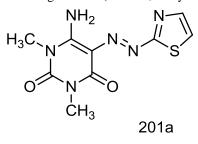
4-hydroxy-3-(thiazol-2-yldiazenyl)-2H-chromen-2-one **184a** and 4-hydroxy-3-[(4-phenylthiazol-2-yl) diazenyl]-2H-chromen-2-one **184d** and 4-hydroxy-3-(benzo[d]thiazol-2-yldiazenyl)-2H-chromen-2-one **184g** inhibited against Staphylococcus aureus, **184a**, **184d** showed good antibacterial activity against Pseudomonas aeruginosa (Sahoo, Kumar Mekap et al. 2015)



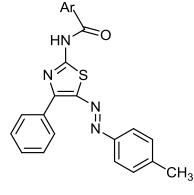
Dye **187a** completely stopped growth by all four bacteria which are E.coli, B. subtilis, M. leuteus and Ps. Aeruginosa. Dye number **187d** stopped all growth by E.coli and M. leuteus and only (Yahyazadeh and Yousefi 2014)



Dye **201a** had some effect against the bacterial growth (Yousefi, Yahyazadeh et al. 2012)



Compounds **318** showed good antimicrobial activity. chloro and nitro derivatives exhibits good activity against *E. coli*, All the derivatives show moderate activity against *S. aureus* and *A. niger*. Dichloro derivatives have good activity against *A. oryzae* (Prajapati and Modi 2011)



Ar =a: Phenyl b: 2-Chlorophenyl c: 4-Chlorophenyl d: 2,4-Dichlorophenyl e: 3-Nitrophenyl f: 4-Nitrophenyl



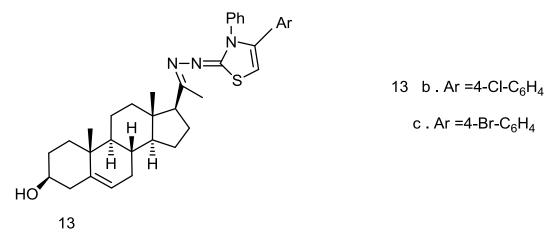
Compounds 2-amino-4-phenyl-5-phenylazothizole **36** and N-(4-phenyl-5-phenylazo-thiazol-2-yl)-substituted arylamide **324** have good anti-bacterial and anti-fungal activity. Therefore, they can be used for the development of new drugs for treatment of bacterial and fungal diseases (Prajapati and Modi 2010)



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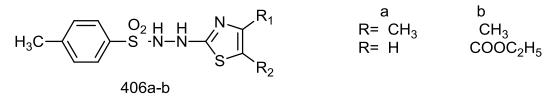
Compounds thiazolyl hydrazonopregnenolone derivatives 13b, 13c, showed high inhibitory effect against tumor cell. (Mohareb, Wardakhan et al. 2012)



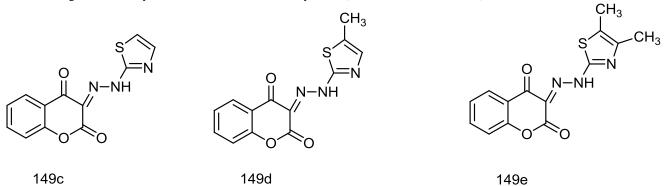
Compound **34d** showed significant antituberculosis activity and Compound **34a** showed notable antituberculosis activity. (Turan-Zitouni, Kaplancikli et al. 2010)



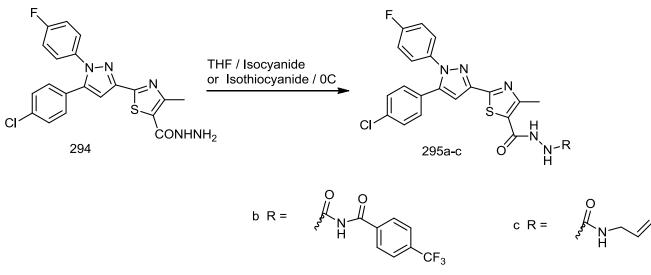
p-toluenesulfonyl-hydrazinothiazoles **406a-b** showed significant anticancer activities on both prostate and hepatocarcinoma cancer cell lines (Zaharia, Ignat et al. 2010)



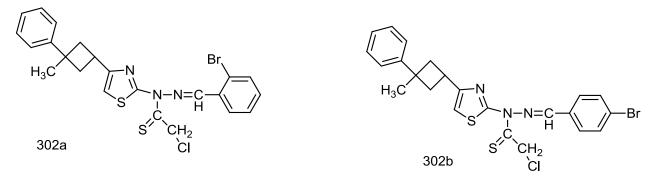
3-[2-(Thiazol-2-yl)hydrazinylidene]chroman-2,4-dione (149c) and 3-[2-(5-Methylthiazol-2-yl)hydrazinylidene] chroman-2,4-dione (149d) and 3-[2-(4,5-Dimethylthiazol-2-yl)hydrazinylidene]chroman-2,4-dione (149e) have a higher efficiency to reduce cancer cell viability (Jashari, Imeri et al. 2014)



Compounds **295b-c** showed good antibacterial activity against S. aureus, E. coli and moderate activity against P. aeruginosa, K. pneumonia. Compounds **295b-c** show pronounced activity against P. marneffei and T. mentagrophytes (Ragavan, Vijayakumar et al. 2010)

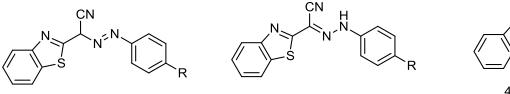


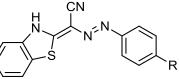
N-(2-bromobenzylidene)-2-chloro-N-(4-(3-methyl-3-phenylcyclobutyl)-thiazol-2-yl) acetohydrazide **302a** showed antibacterial activity against E. coli. N-(4-bromobenzylidene)-2-chloro-N-(4-(3-methyl-3-phenylcyclobutyl)-thiazol-2-yl) acetohydrazide **302b** is very effective against P. aeroginaosa (Cukurovali and Yilmaz 2014)



Aryl azo thiazole used as indicator for analytical chemistry:-

Compounds **407a-g** coexists in the hydrazone and/or azo-enamine-common anion equilibrium or in the solely anionic form. therefore it used as acid–base indicators (Ebead, Selim et al. 2010)





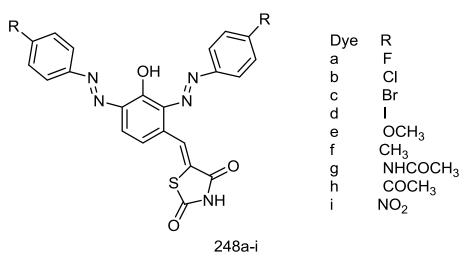
407a**-**g azo

407a-g hydrazone

407a-g azo - enamine

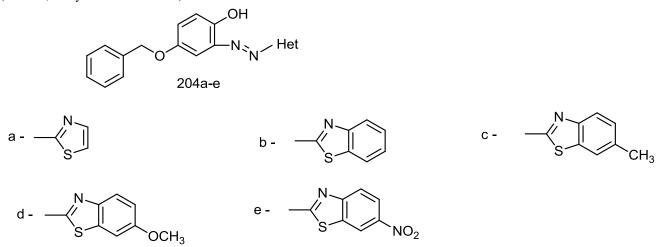
 $\mathsf{R=H}$  ,  $\mathsf{CH}_3$  ,  $\mathsf{OCH}_3$  ,  $\mathsf{COOH}$  ,  $\mathsf{NO}_2$  ,  $\mathsf{CI}$  ,  $\mathsf{OH}$ 

Bis-azo dyes 248a-i used as dyes and can be used as pH indicators. (Mohammadi and Safarnejad 2014)

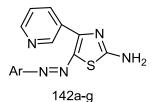


#### Aryl azothiazole with more use:-

Compound **204a-e** is azo dyes used as acid-base endpoint indicator. All the dyes show antibacterial activity. (Yousefi, Yahyazadeh et al. 2013)

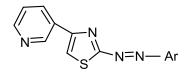


Compounds **142a-g**, **143a-b**, **143e**, **358a-f** used as dyes for polyester fabric. The dyed fabrics exhibit good washing fastness properties, compounds **142a-b**, **142e** and **143b** have significant inhibitory effect on Bacillus subtilis, E.coli,Staphylococcus aureus and Candida albicans. **358c** show Specific antimicrobial activity on Bacillus subtilis. **358e** showed inhibitory effect on E.coli only. (El-Borai, Rizk et al. 2014). 2-amino-5-arylazo-thiazoles (142a-g) 2-arylazo-thiazole (143a-e) 2, 5 diarylazo-thiozoles (358a-f)

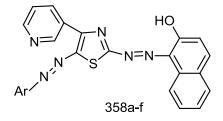


142d-

142a Ar=  $4-C_6H_4CI$ 142b Ar=  $4-C_6H_4OH$ 142c Ar=  $4-C_6H_4OCH_3$  142d Ar=  $4-C_5H_4N$ 142e Ar=  $4-C_6H_4COCH_3$ 142f Ar=  $4-C_6H_4COCH=CH-(4-C_6H_4COCH_3)$ 142g Ar=  $C_{10}H_7$ 

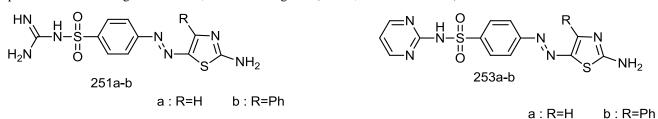


143а-е



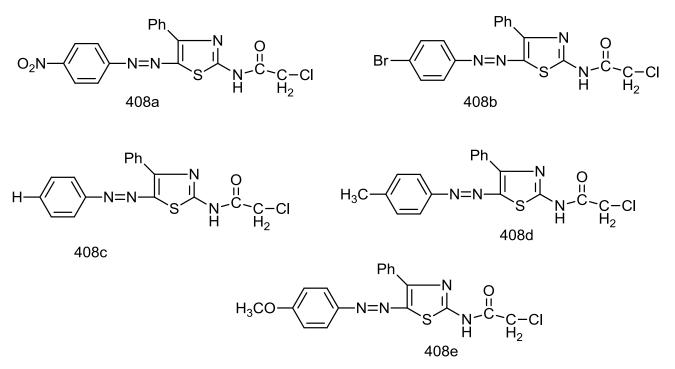
- 143a Ar=  $4-C_{10}H_6OH$ 143b Ar=  $3-CI- 4-C_6H_3OH$ 143c Ar=  $3-NO_2- 4-C_6H_3OH$ 143d Ar=  $3-NH_2 - 4,7SO_3H - 2-C_{10}H_3OH$ 143e Ar=  $4-C_6H_4N(CH_3)_2$
- 358a Ar=  $4-C_6H_4CH_3$ 358b Ar=  $4-C_6H_4OCH_3$ 358c Ar=  $4-C_6H_4CI$ 358d Ar=  $4-C_6H_4COCH_3$ 358e Ar=  $4-C_6H_4COCH_3$ 358e Ar=  $4-C_6H_4COCH=CH-(4-C_6H_4COCH_3)$ 358f Ar=  $C_{10}H_7$

Compounds 2-Amino-5-substituted thiazole **251a**, **251b**, 2-Amino-5-thiazole **253a** and 2-Amino-4-phenyl-5-thiazole **253b** used as dyes and applied as disperse dyes for dyeing polyester fabric, where they exhibited good dye ability and fastness properties, these compounds showed antimicrobial efficiency against selected pathogenic Grampositive and Gram-negative bacteria, as well as fungi (Gaffer, Fouda et al. 2016)

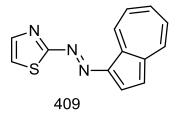


#### Other uses for Aryl azo Thiazole:-

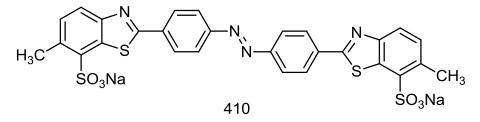
It's found that 5-arylazothiazole derivatives 408a-e act as effective inhibitors for carbon steel dissolution in 0.5M H<sub>2</sub>SO<sub>4</sub> solution. (Abdallah, Al-Tass et al. 2016)



Thiazole in azulene-1-azo-(2-thiazole) **409** enhancing nonlinear optical (NLO) properties of the azulene derivatives. (Essaidi, Niziol et al. 2011)



Compound **410** used as dyes that can be degraded by microbes from petroleum sludge (Huda Alhassani 2014)



# **Conclusion:-**

We have described some recent advances in the synthesis of Aryl Azothiazoles derivatives from thiourea and its derivatives, diazonium salts, benzothiazol and its derivatives and different heterocyclic compounds, also described Chemical reactions of 5-arylazo-thiazole derivatives and 2-arylazo-thiazole derivatives , finally we showed the importance of aryl azothiazoles and its applications as dyes, ligand, indicator in analytical chemistry and inhibitor, in addition to the high value for this compounds in biological activities.

# Acknowledgements:-

The authors are grateful to group members of Prof. Dr. Sayed A. Ahmed for their help.

# **References:-**

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