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Some Nucleophilic Reactions with Isothiocyanatoazobenzene

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The reactivity of Isothiocyanatoazobenzene 2 towards some nucleophiles was investigated. Thus, reaction of isothiocyanate 2 with aromatic amines gave thioureas **3a–d**. The reaction of compound **3a** with arylidenemalononitriles **4a,b** afforded the corresponding 1,3-pyrimidines **7a,b**. Thiosemicarbazide **8** and ethyl thiocarbamate **9** were synthesized by interaction of isothiocyanate 2 with hydrazine hydrate and ethanol, respectively. Cyclocondensation of isothiocyanate 2 with 2-aminophenol and anthranilic acid produced the novel benzoxazole **11** and quinazolinone **13** derivatives, respectively. Finally, treatment of isothiocyanatoazobenzene 2 with compounds **14** and **17** afforded the novel thioureas **15** and **18** derivatives, respectively. The structures of the synthesized compounds were established from their analytical and spectral data.

Keywords Isothiocyanatoazobenzene; pyrimidine derivatives; thiourea

INTRODUCTION

Aromatic azo compounds have attracted great interest in recent years due to their carcinogenic potency¹⁻⁴ and their use in textile⁵ and pharmaceutical industries as dyes, coloring agents,⁶ and analytical reagents.⁷ They can also be used as materials for nonlinear optics and for storage of optical information in laser disks.⁸ In addition, thiourea has a variety of biological activities as antibacterial, antifungal, antitubercular, antithroid, and insecticidal agents.^{9–11} Aryl isothiocyanates¹² are useful and widely used for building blocks in the synthesis of heterocyclic compounds containing nitrogen, sulfur and oxygen.^{13,14}

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Comp. No.	Escherichia coli	Pseudomonas areuginosa	Klebsiella pneumonia
3a	_	_	_
3b	9	10	_
3c	10	10	_
7 A	_	10	_
8	_	10	_
9	14	25	10
11	_	10	9
13	10	20	10
15	_	_	_
18	_	—	—

TABLE I Antibacterial Activity of Some of the Newly SynthesizedCompounds

Diameter of inhibition zone in mm; No inhibition: (---).

As an extension of our interest in the synthesis of heterocyclic compounds exhibiting biological activity,^{15–19} we report herein, the synthesis of new series of thiourea, thiosemicarbazide, benzoxazole, quinazoline, and thiophene derivatives containing aryl azo moiety to evaluate their antimicrobial activity (Table 1).

RESULTS AND DISCUSSION

Isothiocyanatoazobenzene 2 was synthesized by treatment of aminoazobenzene 1 with thiophosgen in chloroform and methylene chloride in the presence of calcium carbonate at room temperature,²⁰ (Scheme 1). The reaction of isothiocyanate 2 with some nucleophiles was studied. Reaction of isothiocyanate 2 with aromatic amines furnished the novel 1, 3-disubstituted thioureas **3a-d** in high yields (Table II) The structures of thioureas 3a-d were established by elemental analysis and spectral data. The infrared spectra of compounds **3a-d** showed the presence of NH, N=N, and C=S functional groups. Cyclization of thiourea derivative **3a** with arylidenemalononitriles **4a**,**b** in refluxing dimethylformamide in the presence of piperidene afforded pyrimidinethione derivatives **7a**,**b**. The molecular structures of pyrimidine derivatives 7 were confirmed on the basis of spectral data. The infrared spectra of the synthesized compounds **7a**,**b** revealed characteristic bands for NH_2 , C=N, C=N and N=N functional groups. Also, the ¹H NMR spectrum of compound 7a in $(DMSO-d_6)$ showed signal characteristic for pyrimidine-H at $\delta = 6.10$ ppm in addition to the presence of methyl, amino and aromatic protons. The formation of pyrimidine derivative 7 is assumed to proceed via the nucleophilic addition of thiourea moiety



 $\begin{array}{l} \text{Sa}_{\text{A}} \sim 2\text{-}\text{Jyrtamy} \\ \text{3b}_{\text{SR}} = 2\text{-Benzothiazolyl} \\ \text{3c}_{\text{SR}} = \text{N}_{\text{c}}(4,5\text{-}\text{Dimethyl-oxazol-2-yl})\text{-benzenesulfonamide} \\ \text{3d}_{\text{SR}} = C_{6}H_{4}\text{SO}_{3}\text{H-p} \\ \text{7a}_{\text{c}}\text{Ar} = C_{6}H_{4}\text{SO}_{3}\text{H-p} \\ \text{7b}_{\text{c}}\text{Ar} = C_{6}H_{4}\text{Cl-p} \\ \text{p}, \ \text{R} = 2\text{-}\text{Pyridinyl} \\ \text{7b}_{\text{c}}\text{Ar} = C_{6}H_{4}\text{Cl-p} \\ \text{p}, \ \text{R} = 2\text{-}\text{Pyridinyl} \\ \end{array}$



to the activated double bond in **4** to form Michael adduct **5** followed by intramolecular cyclization and tautomerization to give **7** (Scheme 1, Table II).

The treatment of isothiocyanate 2 with hydrazine hydrate in ethanol at room temperature yielded the corresponding thiosemicarbazide derivative 8. The molecular structure of compound 8 was identified by analytical and spectral data. Infrared spectrum of compound 8 showed absorption bands for NH, NH₂, and N=N functional groups. Ethyl thiocarbamate derivative 9 was achieved by condensation of isothiocyanate 2 with ethanol as oxygen nucleophiles. (Scheme 2, Table II).

Our investigation was extended to study the reaction of isothiocyanate 2 with 2-aminophenol and anthranilic acid. Cyclocondensation of 2-aminophenol with isothiocyanate 2 in ethanol in the presence of triethylamine at reflux temperature furnished benzoxazole derivative 11. The formation of benzoxazole 11 is assumed to procedure

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TABLE II Characterization Data for Newly Synthesized Compounds

Comp.	M.D.	Solve.	Color.	Mol. formula/	Elen	ental analys	es calcd/found	(%)
no.	(3 ∘)	cry.	yield (%)	(mol. wt.)	С	Н	N	\mathbf{S}
3a	146 - 147	Ethanol	Yellow	$\mathrm{C_{18}H_{15}N_5S}$	64.84	4.53	21.00	9.62
			85	(333.42)	64.90	4.50	21.10	9.60
3b	225 - 226	Ethanol	Yellow	${ m C}_{20}{ m H}_{15}{ m N}_5{ m S}_2$	61.67	3.88	17.98	16.46
			70	(389.50)	61.70	3.90	17.90	16.50
3c	210 - 211	Ethanol	$\operatorname{Reddish}$	${ m C}_{24}{ m H}_{22}{ m N}_{6}{ m O}_{3}{ m S}_{2}$	56.90	4.38	16.59	12.66
			73	(506.61)	56.80	4.40	16.60	12.70
3d	220 - 221	Ethanol	Red	$ m C_{19}H_{16}N_4O_3S_2$	55.33	3.91	13.58	15.55
			65	(412.49)	55.36	3.89	13.60	15.60
7a	105 - 106	Dioxan	Brown	$\mathrm{C}_{29}\mathrm{H}_{23}\mathrm{N}_7\mathrm{S}$	69.44	4.62	19.55	6.39
			68	(501.62)	69.40	4.60	19.60	6.40
7b	115 - 116	Dioxan	Yellow	$ m C_{28}H_{20}CIN_7S$	64.42	3.36	18.78	6.14
			65	(522.04)	64.46	3.40	18.80	6.20
8	198 - 199	Ethanol	Brown	${ m C}_{13}{ m H}_{13}{ m N}_{5}{ m S}$	57.54	4.83	25.81	11.82
			78	(271.35)	57.60	4.90	25.70	11.80
6	138 - 139	Dioxan	Yellow	$C_{15}H_{15}N_{3}OS$	63.13	5.30	14.72	11.24
			76	(285.37)	63.16	5.33	14.75	11.30
11	160 - 161	Ethanol	Brown	${ m C}_{19}{ m H}_{14}{ m N}_4{ m O}$	72.60	4.49	17.82	
			75	(314.35)	72.58	4.52	17.85	
13	>300	Dioxan	Orange	$\mathrm{C}_{20}\mathrm{H}_{14}\mathrm{N}_4\mathrm{OS}$	67.02	3.94	15.63	8.95
			76	(358.42)	67.21	3.90	15.59	8.90
15	188 - 189	Methanol	Yellow	${ m C}_{24}{ m H}_{26}{ m N}_4{ m O}_2{ m S}_2$	61.78	5.62	12.01	13.74
			62	(466.63)	61.80	5.60	12.22	13.69
18	180–181	Methanol	Yellow	$\mathrm{C}_{27}\mathrm{H}_{22}\mathrm{N}_6\mathrm{OS}_2$	63.51	4.34	16.46	12.56
			67	(510.64)	63.60	4.30	16.50	12.60



SCHEME 2

via thiourea intermediate **10** followed by intramolecular cyclization through elimination of hydrogen sulfide,²¹ (Scheme 3). Quinoxaline derivative **13** was obtained by cyclization of anthranilic acid with isothiocyanate **2**, via thiourea intermediate**12** followed by elimination of water. (Scheme 3, Table II).



SCHEME 3

In continuation of this investigation, interaction of 4isothiocyanatoazobenzene **2** with 2-amino-3-ethoxycarbonyl-4, 5, 6, 7-tetrahydrobenzo[b]thiophene²² under reflux in ethanol triethylamine afforded the novel thiourea derivative **15** and tetrahydrobenzo [*b*]thieno[2,3-*d*]pyrimidin-4-one derivative **16** was discarded on the basis of analytical and spectral data. The ¹H NMR spectrum of compound **15** showed signals at δ : 1.23 (t, 3H, CH₃), 4.23 (q, 2H, CH₂), 7.21–7.98 ppm (m, 9H, H-Ar), and 11.29, 12.01 ppm (2s, 2H, 2NH). Condensation of compound **2** with 2-aminothiophene derivative **17** in ethanol and triethylamine yielded the corresponding disubstituted thiourea **18**. The IR spectrum of compound **18** revealed the presence of a strong absorption band characteristic for C=N function at 2203 cm⁻¹, in addition to NH and C=O functional groups (Scheme 4, Table II).



SCHEME 4

ANTIBACTERIAL ACTIVITY

Ten compounds were screened in vitro for their antibacterial against three strains of bacteria: *Escherichia coli*, *Pseudomonas areuginosa* and *Klebsiella pneumonia* by the agar diffusion technique.²³ A 1 mg/mL solution in dimethylformamide was used. The bacteria were maintained on nutrient agar media. Dimethylformamide showed no inhibition zones. The agar media was incubated with different microorganisms culture tested, after 24 h of incubation at 30°C for bacteria, the diameter of inhibition zone (mm) was measured (Table I). Compounds **9** and **13** exhibited antimicrobial activity against all the microorganisms used. Compounds **3b** and **3c** showed weak activities against *Escherichia coli* and *Pseudomonas areuginosa*. The rest of the compounds exhibited from weak to nil activities against all strains of bacteria used.

CONCLUSION

Isothiocyanatoazobenzene (2) was used as starting material for the synthesis of some novel thiourea, aminopyrimidine, thiosemicarbazide, thiocarbamate, benzoxazole, and quinazoline derivatives depend on reaction conditions (temperature and time).

EXPERIMENTAL

All melting points are uncorrected (Stuart Scientific Co., UK). The IR spectra were measured as KBr pellets on Shimadzu IR 200 spectrophotometer. ¹H NMR spectra were recorded in DMSO- d_6 at 200 MHz on a Varian Gemini NMR spectrometer (Varian, UK), using tetramethylsilane as internal reference. Elemental analyses were carried out at the Microanalytical Center of Cairo University. The characteristic data for the synthesized compounds are given in Table II.

N-{4-[Phenyldiazenyl]phenyl}-N-pyridin-3-ylthiourea (3a)

A mixture of **2** (2.39 g, 0.01 mol), 3-aminopyridine (0.94 g, 0.01 mol) in toluene (30 mL) containing few drops of triethylamine was refluxed for 3 h. The precipitate formed after cooling was collected by filtration and recrystallized from ethanol to give **3a** in good yielded (Table II). **3a:** IR (KBr) ν 3219, 3108 (NH), 3045 (CH-arom.), 1594 (N=N) cm⁻¹. ¹H NMR (DMSO-*d*₆) δ 6.67, 6.70, 7.38, 8.10 (s,4H, pyridine), 7.40–7.90 (m, 15H, Ar-H and NH), 10.67 ppm (s, 1H, 1NH).

N-Substituted-N-(4-phenylazophenyl)thiourea (3b–d)—General Procedure

To a suspension of isothiocyanate 2 (2.39 g, 0.01 mol) and aryl amines **3b-d** (1.50, 2.67 and 1.41 g, respectively 0.01 mol) in dioxane (30 mL), triethylamine (0.5 mL) was refluxed until the clear solution was obtained. The solid residue was collected by filtration, washed with cold water and recrystallized from the proper solvent to give compounds **3a-b** (Table II).

N-(1,3-Benzothiazol-2-yl)-N-{4-[phenyldiazenyl]phenyl} thiourea (3b)

IR(KBr) ν 3316, 3159 (NH), 3035 (CH-arom.), 1592 (N=N) cm⁻¹. ¹HNMR (DMSO-*d*₆) δ 7.53–8.10(m, 14H, Ar-H and NH), 10.39 ppm(s, 1H, NH).

N-(4,5-Dimethyl-1,3-oxazol-2-yl)-4-{[({4-[phenyldiazenyl] phenyl}amino)carbono-thioyl]amino}benzenesulfonamide (3c)

IR (KBr) ν 3334, 3280, 3100 (NH), 3040 (CH-arom.), 2940 (CH-aliph.), 1595 (N=N) cm⁻¹. ¹HNMR (DMSO- d_6) δ 1.92 (s, 3H, CH₃), 2.02 (s, 3H, CH₃), 7.29–7.86 (m, 15H, Ar-H and 2NH), 9.45 ppm (b, 1H, NH).

4-{[({4-[phenyldiazenyl] phenyl} amino) carbonothioyl] amino} benzenesulfonic acid (3d)

IR (KBr) ν 3413 (OH), 3307, 3195 (NH), 3037 (CH-arom.), 1592 (N=N) $\rm cm^{-1}.$

6-Amino-3-(4-phenylazo-phenyl)-1-pyridin-2-yl-2-thioxo-4–(4substituted-aryl)-1, 2, 3, 4-tetrahydro-pyrimidine-5carbonitrile (7a, b)—General Procedure

A mixture of thiourea 3a (3.33 g, 0.01 mol), arylidinemalononitril 4a,b (1.68 and 1.89 g, respectively 0.01 mol) and piperidine (0.5 mL) in dimethylformamide (30 mL) was heated under reflux for 6 h. The reaction mixture was then cooled, poured into crushed ice, neutralized with diluted HCl, and the obtained product was collected, washed with cold water and recrystallized from proper solvent to give compound 7 (Table II).

6-Amino-3-(4-phenylazo-phenyl)-1-pyridin-2-yl-2-thioxo-4-ptolyl-1,2,3,4-tetra-hydro-pyrimidine-5-carbonitrile (7a)

IR (KBr) ν 3320, 3246 (NH₂), 3050 (CH-arom.), 2932 (CH-aliph.), 2196 (C=N), 1592 (N=N) cm⁻¹. ¹H NMR (DMSO-*d*₆) δ 2.07 (s, 3H, CH₃), 3.88 (s, 2H, NH₂), 6.10 (s, 1H, 4H-pyrimidine), 6.65, 6.76, 7.45, 8.20 (4H-pyridine), 7.51–7.86 ppm (m, 13H, Ar-H).

6-Amino-4-(4-chloro-phenyl)-3-(4-phenylazo-phenyl)-1pyridin-2-yl-2-thioxo-1, 2, 3, 4-tetrahydro-pyrimidine-5-carbonitrile (7b)

IR (KBr) ν 3335, 3285 (NH₂), 3044 (CH-arom.), 2935 (CH-aliph.), 2210 (C=N), 1595 (N=N) cm⁻¹. ¹H NMR (DMSO-*d*₆) δ 3.91 (s, 2H, NH₂), 5.18 (s, 1H, 4H-pyrimidine), 6.61, 6.70, 7.43, 8.11 (4H-pyridine), 7.48–7.93 ppm (m, 13H, Ar-H).

N-{4-[Phenyldiazenyl]phenyl}hydrazinecarbothioamide (8)

A suspension of 4-isothiocynatoazobenzene **2** (2.39 g, 0.01 mol), hydrazine hydrate (0.5 mL, 0.01 mol) and ethanol (30 mL) was stirred at room temperature for 1 h. The solid that separated out was filtered off and recrystallized from proper solvent to give compound **8** (Table II). IR (KBr) ν 3290, 3248, 3187 (NH, NH₂), 3040 (CH-arom.), 1546 (N=N) cm⁻¹. ¹H NMR (DMSO-*d*₆) δ 3.85 (s, 2H, NH₂), 7.64–7.95 (m, 9H, Ar-H), 9.25 (s, 1H, NH), 11.23 ppm (s, 1H, NH).

N-ethoxy-N-{4-[phenyldiazenyl]phenyl}thiourea (9)

A solution of compound **2** (2.39 g, 0.01 mol) in ethanol (30 mL) was heated under reflux for 3 h and then left to cool; the crystalline product thus formed was filtered, washed with diethyl ether, and recrystallized from dioxan to give compound **9** (Table II). IR (KBr) ν 3227 (NH), 3041(CH-ar.), 2980 (CH-aliph.), 1546 (N=N) cm⁻¹. ¹H NMR (DMSO-*d*₆) δ 1.33 (t, 3H, CH₃), 4.56 (q, 2H, OCH₂), 7.55–7.91 (m, 9H, Ar-H), 11.37 ppm (s, 1H, NH).

N-{4-[Phenyldiazenyl]phenyl}-1,3-benzoxazol-2-amine (11)

A mixture of **2** (2.39 g, 0.01 mol) and 2-aminophenol (1.09 g, 0.01 mol) in dimethyl-formamide containing few drops of triethylamine was refluxed until evolution of H₂S had stopped (lead acetate paper). After cooling, the reaction mixture was poured into cold water and the solid product that obtained was collected by filtration, washed with water and recrystallized from proper solvent to give compound **11** (Table II). IR (KBr) ν 3352 (NH), 3030 (CH-aromatic), 1596 (N=N) cm⁻¹. ¹H NMR (DMSO-*d*₆) δ 7.25–7.95 (m, 13H, Ar-H), 11.26 ppm (s, 1H, NH).

3-{4-[phenyldiazenyl]phenyl}-2-thioxo-2,3-dihydroquinazolin-4(1H)-one (13)

A suspension of 4-isothiocyanatoazobenzene **2** (2.39 g, 0.01 mol) and anthranilic acid (1.37 g, 0.01 mol) in toluene (30 mL) containing triethylamine (0.5 ml). The reaction mixture was heated under reflux for 30 min. The solid product, which produced on heating, was collected and recrystallized from dioxan to give compound **13** (Table II). IR (KBr) ν 3231 (NH), 3030 (CH-arom.), 1658 (C=O), 1596 (N=N) cm⁻¹. ¹H NMR (DMSO-*d*₆) δ 7.33–7.98 (m, 13H, Ar-H), 13.10 ppm (b, 1H, NH).

Preparation of Compounds 15 and 18—General Procedure

A mixture of isothiocyanate **2** (2.39 g, 0.01 mol) and 2-amino-3ethoxycarbonyl-7,5,6,7-tetrahydrobenzo[*b*]thiophene **16** or 2-amino-3cyano-5-methyl-*N*-(4-methyl-phenyl)thiophene-4-carboxamide **17** (2.27 and 2.71 g, respectively 0.01 mol) in ethanol (30 mL) containing triethylamine (0.5 mL) was heated under reflux for 3. The solid obtained was crystallized from proper solvent to give **15** and **18** (Table II).

Ethyl-2-{[(4-[phenyldiazenyl]phenylamino)carbonothioyl] amino}-4,5,6,7 tetrahydro-1-benzothiophene-3-carboxylate (15)

IR (KBr) ν 3240, 3160 (NH), 3042 (CH-arom.), 2933 (CH-aliph.), 1656 (C=O), 1595 (N=N) cm⁻¹. ¹H NMR (DMSO- d_6) δ 1.23 (t, 3H, CH₃), 1.72, 2.49 (8H, m, tetrahydraobenzo), 4.23 (q, 2H, OCH₂), 7.21–7.98 (m, 9H, Ar-H), 11.29 (s. 1H, NH), 12.01 ppm (S, 1H, NH).

3-Cyano-5-methyl-2-{[(4-[methyldiazenyl]phenyl}amino) carbonothioyl]amino-N-(4-methylphenyl)thio-phene-4carboxamide (18)

IR (KBr) ν 3380, 3310, 3203 (NH), 3035 (CH-arom.), 2920 (CH-aliph.), 2203 (C=N), 1660 (C=O), 1540 (N=N) cm⁻¹. ¹H NMR (DMSO-*d*₆) δ 2.08 (s, 3H, CH₃), 2.35 (s, 3H, CH₃), 7.08–7.87 (m, 14H, Ar-H and NH), 9.56, 11.39 ppm (2s, 2H, 2NH).

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