

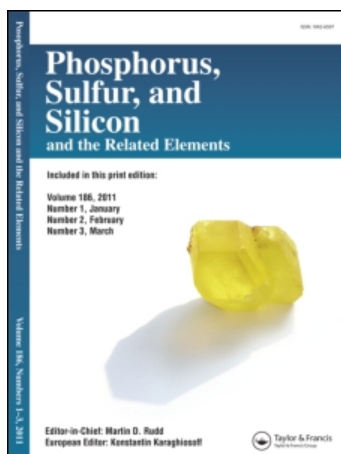
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SYNTHESIS AND CHEMISTRY OF SOME NEW 2-MERCAPTOIMIDAZOLE DERIVATIVES OF POSSIBLE ANTIMICROBIAL ACTIVITY

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4,5-Diaryl-2,3-dihydro-2-mercaptoimidazoles (**2a–e**) were synthesized. They reacted with chloroacetic acid in gl. acetic acid/ Ac_2O in presence of anhyd. sodium acetate afforded 5,6-diaryl-2,3-dihydroimidazo[2,1-b]thiazol-3-ones (**3a–d**). Also these compounds were prepared by the action of chloroacetyl chloride on compounds (**2**) in pyridine. Compounds (**3a–d**) on condensation with aromatic aldehydes yield 2-arylmethylene-5,6-diaryl-2,3-dihydroimidazo[2,1-b]thiazol-3-ones (**4a–q**). The latter compounds were prepared directly by the reaction of (**2**) with chloroacetic acid and the aromatic aldehydes. Compounds (**3a–d**) coupled with aryldiazonium salts in pyridine to give 2-arylhydrazono-5,6-diaryl-2,3-dihydroimidazo[2,1-b]thiazol-3-ones (**5a–r**). Also compounds (**2**) when reacted with 2 or 3-bromopropionic acid afford 2,3-di-hydro-5,6-diaryl-2-methylimidazo[2,1-b]thiazol-3-ones (**6a–d**) and 2,3-di-hydro-6,7-diarylimidazo-[2,1-b]-1,3-thiazin-4-ones (**7a–d**), respectively. Compounds (**3**, **6**, and **7**) have been cleaved by aromatic amines to give the corresponding 2-(4',5'-diaryl-2',3'-dihydroimidazol-2'-yl)thioacetanilide (**8a–f**), 2-(2',3'-dihydro-4',5'-diaryl imidazol-2'-yl)thiopropionamide (**9a–c**), and 3-(2',3'-dihydro-4',5'-diaryl-imidazol-2'-yl)thiopropionamide (**10a–d**) respectively. All the prepared compounds show considerable antimicrobial activity against bacteria, yeast, and fungi.

Keywords: 4,5-Diaryl-2-mercaptoimidazole derivatives

The chemistry of imidazole derivatives has received considerable attention due to their biological activities.^{1–7} Accordingly, we have undertaken the preparation of some new related derivatives for biological evaluation.

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RESULTS AND DISCUSSION

Synthesis of new compounds were achieved by fusion of diaryl benzoin^{8,9} with thiourea at 200°C to give 2,3-dihydro-4,5-diaryl-2-mercaptoimidazoles (**2**). The IR spectra of compounds (**2**) showed absorption bands arounds 3470-3080 cm⁻¹. (cf. Table II). The ¹H NMR spectrum of (**2a**) [DMSO-d₆] led to the following assignments: the 2NH protons as two broad singlets at $\delta = 9.36$ and 9.26 (br., 2H, 2NH) and the aromatic protons as a multiplet (m, 8H, ArH's) (cf. Table II).

Compounds (**2**) were reacted with chloroacetic acid in acetic acid/acetic anhydride in the presence of anhydrous sodium acetate to produce 5,6-diaryl-2,3-dihydroimidazo[2,1-*b*]thiazol-3-ones (**3**); also compounds (**3**) were prepared by the action of chloroacetyl chloride on compounds (**2**) in presence of pyridine. The IR spectra of compounds (**3**) showed the carbonyl absorption bands around 1700 cm⁻¹. The ¹H-NMR spectrum of compound (**3a**) [in DMSO-d₆] showed a methylene group as a singlet, (2H) at 3.25 ppm and the aromatic protons as a multiplet centred in the $\delta = 7.40$ -8.60 (m, 8H, ArH's) (cf. Table II).

The formulation of the cyclised products as (**3**) is favored over the isomeric structures (**3**).¹⁰

Compounds (**3**) were condensed with aromatic aldehydes in acetic acid/acetic anhydride mixture in the presence of anhydrous sodium acetate to produce 2-arylmethylene-5,6-diaryl-2,3-dihydroimidazo[2,1-*b*]thiazol-3-ones (**4a-q**). The latter compounds were prepared directly from (**2**) by the action of chloroacetic acid, aromatic aldehyde and sodium acetate in acetic acid/acetic anhydride mixture. The reaction product was found to be identical in all aspects (m.p., mixed m.p., and IR) with (**4**). The IR spectra of compounds (**4**) showed carbonyl absorption around 1685-1700 cm⁻¹. This shift to lower frequency is due to the conjugation with the exocyclic double bond. The ¹H-NMR spectrum of (**4a**) [DMSO-d₆] revealed the disappearance of the singlet at $\delta = 3.25$ (compared with that of **3a**) belonging to the activated methylene group, besides the appearance of the expected signals at $\delta = 7.0$ -8.60 ppm region (13H) for the aromatic protons and the benzylic proton (cf. Table II).

Compounds (**3**) coupled with aryldiazonium salts in the presence of pyridine to give 2-arylhydrazono-5,6-diaryl-2,3-dihydroimidazo[2,1-*b*]thiazol-3-ones (**5**). The presence of a peak at 2.70 and 3.70 pp in ¹H-NMR spectra, and the absence of NH-groups in compounds (**5n**) and (**5o**) indicate that their coupling products exist in solution in the azo-form¹¹ (cf. Table II).

TABLE I Physical Data of Compounds 2-10

Comp.	Ar/Ar ₁	Yield (%)	Mol. form. (mol. wt.)	m.p. °C Solvent	% Analysis (calcd./found)			
					C	H	N	S
2a	o-C ₅ H ₄ N/ o-C ₅ H ₄ N	95	C ₁₃ H ₁₀ N ₄ S (254)	149	64.41	3.93	22.04	12.59
				AcOH	61.82	3.90	22.00	12.35
2b	p-C ₅ H ₄ N/ p-C ₅ H ₄ N	98	C ₁₃ H ₁₀ N ₄ S (254)	215	61.41	3.93	21.04	12.59
				DMF	62.50	3.97	21.90	12.72
2c	p-C ₆ H ₄ F/ p-C ₆ H ₄ F	98	C ₁₃ H ₁₀ F ₂ N ₂ S (288)	150	62.50	3.47	9.72	11.11
				AcOH	62.30	3.43	9.70	11.00
2d	o-C ₆ H ₄ F/ C ₆ H ₅	96	C ₁₃ H ₁₁ FN ₂ S (270)	200	66.66	4.07	10.37	11.85
				AcOH	66.16	4.00	10.00	12.02
2e	o-C ₄ H ₃ S/ o-C ₄ H ₃ S	97	C ₁₁ H ₈ N ₂ S ₃ (264)	95	50.00	3.03	10.60	36.36
				AcOH	49.83	3.00	10.60	36.24
3a	o-C ₅ H ₄ N/ o-C ₅ H ₄ N	87	C ₁₅ H ₁₀ N ₄ OS (294)	143	61.22	3.40	19.04	10.88
				AcOH	60.95	3.42	19.00	10.41
3b	p-C ₆ H ₄ F/ p-C ₆ H ₄ F	87	C ₁₇ H ₁₀ F ₂ N ₂ OS (328)	108	62.19	3.04	8.53	9.75
				EtOH/AcOH	62.19	3.04	8.52	9.75
3c	o-C ₆ H ₄ F/ C ₆ H ₅	85	C ₁₇ H ₁₁ FN ₂ OS (310)	90	65.80	3.54	9.03	10.32
				EtOH	65.55	3.24	9.03	10.36
3d	o-C ₄ H ₃ S/ o-C ₄ H ₃ S	81	C ₁₃ H ₈ N ₂ OS ₃ (304)	240	51.31	2.63	9.21	31.57
				AcOH	51.20	2.43	9.21	31.57
4a	o-C ₅ H ₄ N/ o-C ₅ H ₄ N, X = 4-Cl	73	C ₂₂ H ₁₃ ClN ₄ OS (416.5)	160	63.38	3.12	13.44	7.68
				AcOH	63.05	3.12	13.43	7.65
4b	o-C ₅ H ₄ N/ o-C ₅ H ₄ N, X = 4-OMe	69	C ₂₃ H ₁₆ N ₄ O ₂ S (412)	280	66.99	3.88	13.59	7.76
				AcOH	67.24	3.90	13.59	7.54
4c	o-C ₅ H ₄ N/ o-C ₅ H ₄ N, X = 2-NO ₂	56	C ₂₂ H ₁₃ N ₅ O ₃ S (427)	192	61.82	3.04	16.39	7.49
				Benzene/pet. ether 40-60°C	61.69	2.98	16.21	7.49
4d	o-C ₅ H ₄ N/ o-C ₅ H ₄ N, X = 4-NO ₂	75	C ₂₂ H ₁₃ N ₅ O ₃ S (427)	172	61.82	3.04	16.39	7.49
				CHCl ₃ /pet. ether 40-60°C	61.69	2.98	16.21	7.49
4e	p-C ₆ H ₄ F/ p-C ₆ H ₄ F, X = 4-Cl	85	C ₂₄ H ₁₃ ClF ₂ N ₂ OS (450.5)	170	63.92	2.88	6.21	7.10
				AcOH	63.70	2.80	6.20	7.10
4f	p-C ₆ H ₄ F/ p-C ₆ H ₄ F, X = 4-OMe	76	C ₂₅ H ₁₆ F ₂ N ₂ O ₂ S (446)	150	76.26	3.58	6.27	7.17
				EtOH	76.00	3.50	6.27	7.15
4g	p-C ₆ H ₄ F/ p-C ₆ H ₄ F, X = 2-NO ₂	71	C ₂₄ H ₁₃ F ₂ N ₃ O ₃ S (461)	110	62.47	2.81	9.11	6.94
				CHCl ₃ /pet. ether 40-60°C	62.47	2.97	9.11	6.82
4h	p-C ₆ H ₄ F/ p-C ₆ H ₄ F, X = 4-NO ₂	87	C ₂₄ H ₁₃ F ₂ N ₃ O ₃ S (461)	235	62.47	2.81	9.11	6.94
				CHCl ₃ /pet. ether 40-60°C	62.47	2.60	9.00	6.72
4i	p-C ₆ H ₄ F/ p-C ₆ H ₄ F, X = 4-Br	80	C ₂₄ H ₁₃ BrF ₂ N ₂ OS (494.9)	158	58.19	2.62	5.65	6.46
				CHCl ₃ /pet. ether 40-60°C	57.87	2.42	5.65	6.73
4j	C ₆ H ₅ / o-C ₆ H ₄ F, X = 4-Cl	87	C ₂₄ H ₁₄ ClFN ₂ OS (432.5)	160	66.58	3.23	6.47	7.39
				AcOH	66.92	3.43	6.47	7.35
4k	C ₆ H ₅ / o-C ₆ H ₄ F, X = 4-OMe	75	C ₂₅ H ₁₇ FN ₂ O ₂ S (428)	140	70.09	3.97	6.54	7.47
				EtOH	70.00	3.90	6.54	7.32

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TABLE I Physical Data of Compounds 2–10 (Continued)

Comp.	Ar/Ar ₁	Yield (%)	Mol. form. (mol. wt.)	m.p. °C Solvent	% Analysis (calcd./found)			
					C	H	N	S
4l	C ₆ H ₅ / o-C ₆ H ₄ F, X = 2-NO ₂	78	C ₂₄ H ₁₄ FN ₃ O ₃ S (443)	120 MeOH	65.01 64.98	3.16 3.03	9.48 9.24	7.22 7.22
4m	C ₆ H ₅ / o-C ₆ H ₄ F, X = 4-NO ₂	81	C ₂₄ H ₁₄ FN ₃ O ₃ S (443)	132 MeOH	65.01 65.01	3.16 2.98	9.48 9.41	7.22 7.00
4n	o-C ₄ H ₃ S/ o-C ₄ H ₃ S, X = 4-Cl	73	C ₂₀ H ₁₁ ClN ₂ OS ₃ (426.5)	190 DMF	56.27 56.22	2.57 2.32	6.56 6.56	22.50 22.00
4o	o-C ₄ H ₃ S/ o-C ₄ H ₃ S, X = 4-OMe	77	C ₂₁ H ₁₄ N ₂ O ₂ S ₃ (422)	245 DMF	59.71 59.21	3.31 2.28	6.63 6.92	22.74 22.93
4p	o-C ₄ H ₃ S/ o-C ₄ H ₃ S, X = 2-NO ₂	79	C ₂₀ H ₁₁ N ₃ O ₃ S ₃ (437)	212 CHCl ₃ /pet. ether 40–60°C	54.91 54.82	2.51 2.32	9.61 9.64	21.96 22.34
4q	o-C ₄ H ₃ S/ o-C ₄ H ₃ S, X = 4-NO ₂	82	C ₂₀ H ₁₁ N ₃ O ₃ S ₃ (437)	182 CHCl ₃ /pet. ether 40–60°C	54.91 54.91	2.51 2.45	9.61 9.48	21.96 22.73
5a	o-C ₅ H ₄ N/ o-C ₅ H ₄ N, X = H	63	C ₂₁ H ₁₄ ClN ₆ O ₃ S (398)	230 AcOH	63.31 63.00	3.51 3.50	21.10 21.00	8.04 8.31
5b	o-C ₅ H ₄ N/ o-C ₅ H ₄ N, X = 4-NO ₂	87	C ₂₁ H ₁₃ N ₇ O ₃ S (443)	118 AcOH	56.88 56.42	2.93 2.81	22.12 22.44	7.22 7.57
5c	o-C ₅ H ₄ N/ o-C ₅ H ₄ N, X = 2-NO ₂	79	C ₂₁ H ₁₃ N ₇ O ₃ S (443)	110 CHCl ₃ /pet. ether 40–60°C	56.88 56.71	2.93 2.92	22.12 22.19	7.22 7.24
5d	o-C ₅ H ₄ N/ o-C ₅ H ₄ N, X = 4-Br	85	C ₂₁ H ₁₃ BrN ₆ OS (476.9)	117 AcOH	52.84 52.95	2.72 2.77	17.61 17.94	6.71 6.79
5e	o-C ₅ H ₄ N/ o-C ₅ H ₄ N, X = 4-Cl	75	C ₂₁ H ₁₃ ClN ₆ OS (432.5)	276 AcOH	58.26 58.62	3.00 3.10	19.42 19.71	7.39 7.75
5f	o-C ₅ H ₄ F/ o-C ₅ H ₄ F, X = H	76	C ₂₃ H ₁₄ F ₂ N ₄ OS (432)	108 CHCl ₃ /pet. ether 40–60°C	63.88 63.95	3.24 3.31	12.96 13.24	7.40 7.76
5g	o-C ₅ H ₄ F/ o-C ₅ H ₄ F, X = 4-NO ₂	89	C ₂₃ H ₁₃ F ₂ N ₅ OS (477)	98 MeOH	57.86 57.79	2.72 2.72	14.67 14.61	6.70 6.92
5h	o-C ₅ H ₄ F/ o-C ₅ H ₄ F, X = 2-NO ₂	81	C ₂₃ H ₁₃ F ₂ N ₅ OS (477)	110 MeOH	57.86 57.52	2.72 2.70	14.67 14.65	6.70 6.96
5i	o-C ₅ H ₄ F/ o-C ₅ H ₄ F, X = 4-Br	93	C ₂₃ H ₁₃ F ₂ BrN ₄ OS (510.9)	128 MeOH	54.02 53.87	2.54 2.78	10.96 10.89	6.26 6.54
5j	o-C ₅ H ₄ F/ o-C ₅ H ₅ , X = H	79	C ₂₃ H ₁₅ FN ₄ OS (414)	112 AcOH	66.66 66.61	3.62 3.62	13.52 13.27	7.72 7.62
5k	o-C ₅ H ₄ F/ C ₅ H ₅ , X = 4-NO ₂	86	C ₂₃ H ₁₄ FN ₅ O ₃ S (459)	190 Benzene/pet. ether 40–60°C	60.13 59.86	3.05 3.00	15.25 15.25	6.97 6.81

(Continued on next page)

TABLE I Physical Data of Compounds 2-10 (Continued)

Comp.	Ar/Ar ₁	Yield (%)	Mol. form. (mol. wt.)	m.p. °C Solvent	% Analysis (calcd./found)			
					C	H	N	S
5l	<i>o</i> -C ₅ H ₄ F/ C ₅ H ₅ , X = 2-NO ₂	83	C ₂₃ H ₁₄ FN ₅ O ₃ S (459)	190 Benzene/pet. ether 40-60°C	60.13 60.34	3.05 3.22	15.25 15.18	6.97 6.99
5m	<i>o</i> -C ₅ H ₄ F/ <i>o</i> -C ₅ H ₅ , X = 4-Br	90	C ₂₃ H ₁₄ BrFN ₄ O ₃ S (492.9)	147 MeOH	55.99 56.17	2.84 2.89	11.36 11.36	6.49 6.75
5n	<i>o</i> -C ₄ H ₃ S/ <i>o</i> -C ₄ H ₃ S, X = H	65	C ₁₉ H ₁₂ N ₄ OS ₃ (408)	156 Benzene/pet. ether 40-60°C	55.88 55.62	2.94 2.92	13.72 13.72	23.52 23.20
5o	<i>o</i> -C ₄ H ₃ S/ <i>o</i> -C ₄ H ₃ S, X = 4-NO ₂	73	C ₁₉ H ₁₁ N ₅ O ₃ S ₃ (453)	163 CHCl ₃ /pet. ether 40-60°C	50.33 50.31	2.42 2.42	15.54 15.43	21.19 21.25
5p	<i>o</i> -C ₄ H ₃ S/ <i>o</i> -C ₄ H ₃ S, X = 2-NO ₂	77	C ₁₉ H ₁₁ N ₅ O ₃ S ₃ (453)	108 CHCl ₃ /pet. ether 40-60°C	50.33 50.48	2.42 2.51	15.54 15.45	21.19 21.23
5q	<i>o</i> -C ₄ H ₃ S/ <i>o</i> -C ₄ H ₃ S, X = 4-Br	81	C ₁₉ H ₁₁ BrN ₄ OS ₃ (486.9)	175 Benzene/pet. ether 40-60°C	46.82 46.75	2.25 2.25	11.50 11.35	19.71 19.50
5r	<i>o</i> -C ₄ H ₃ S/ <i>o</i> -C ₄ H ₃ S, X = 4-Cl	67	C ₁₉ H ₁₁ ClN ₄ OS ₃ (442.5)	128 CHCl ₃ /pet. ether 40-60°C	51.52 51.80	2.48 2.34	12.65 12.63	21.69 21.87
6a	<i>o</i> -C ₅ H ₄ N/ <i>o</i> -C ₅ H ₄ N	65	C ₁₆ H ₁₂ N ₄ OS (308)	132 Pyridine	62.33 62.17	3.89 3.85	18.18 18.16	10.38 10.71
6b	<i>p</i> -C ₆ H ₄ F/ <i>p</i> -C ₆ H ₄ F	91	C ₁₈ H ₁₂ F ₂ N ₂ OS (342)	140 AcOH	63.15 63.11	3.50 3.51	8.18 8.24	9.35 9.30
6c	<i>p</i> -C ₆ H ₄ F/ C ₆ H ₅	89	C ₁₈ H ₁₃ FN ₂ OS (324)	83 EtOH	66.66 66.94	4.01 3.97	8.64 8.51	9.87 10.13
6d	<i>o</i> -C ₄ H ₃ S/ <i>o</i> -C ₄ H ₃ S	76	C ₁₄ H ₁₀ N ₂ OS ₃ (318)	145 CHCl ₃	52.83 52.94	3.14 3.21	8.80 8.78	30.18 30.00
7a	<i>o</i> -C ₅ H ₄ N/ <i>o</i> -C ₅ H ₄ N	57	C ₁₆ H ₁₂ N ₄ OS (308)	118 Pyridine	62.33 62.00	3.89 3.71	18.18 18.20	10.38 10.87
7b	<i>p</i> -C ₆ H ₄ F/ <i>p</i> -C ₆ H ₄ F	82	C ₁₈ H ₁₂ F ₂ N ₂ OS (342)	95 AcOH	63.15 63.00	3.50 3.71	8.18 8.42	9.35 9.31
7c	<i>p</i> -C ₆ H ₄ F/ C ₆ H ₅	74	C ₁₈ H ₁₃ FN ₂ OS (324)	97 EtOH	66.66 66.96	4.01 3.89	8.64 8.51	9.87 9.91
7d	<i>o</i> -C ₄ H ₃ S/ <i>o</i> -C ₄ H ₃ S	69	C ₁₄ H ₁₀ N ₂ OS ₃ (318)	152 CHCl ₃ /pet. ether 40-60°C	52.83 52.71	3.14 3.31	8.80 8.71	30.18 30.43
8a	<i>o</i> -C ₅ H ₄ N/ <i>o</i> -C ₅ H ₄ N	89	C ₂₁ H ₁₇ N ₅ OS (387)	128 EtOH	65.11 64.86	4.39 4.31	18.08 18.00	8.26 8.45
8b	<i>p</i> -C ₆ H ₄ F/ <i>p</i> -C ₆ H ₄ F	98	C ₂₃ H ₁₇ F ₂ N ₃ OS (421)	95 AcOH	65.55 65.50	4.03 3.85	9.97 9.72	7.60 7.51
8c	<i>p</i> -C ₆ H ₄ F/ <i>p</i> -C ₆ H ₄ F, 4-NO ₂	92	C ₂₃ H ₁₆ F ₂ N ₄ O ₃ S (466)	172 MeOH	59.22 59.00	3.43 3.41	12.01 12.37	6.86 6.89
8d	<i>p</i> -C ₆ H ₄ F/ C ₆ H ₅	93	C ₂₃ H ₁₈ FN ₃ OS (403)	105 EtOH	68.48 68.91	4.46 4.53	10.42 10.41	7.94 8.12
8e	<i>p</i> -C ₆ H ₄ F/ C ₆ H ₅ , 4-NO ₂	88	C ₂₃ H ₁₇ FN ₄ O ₃ S (448)	178 AcOH	61.60 61.42	3.79 3.71	12.50 12.30	7.14 7.14

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TABLE I Physical Data of Compounds **2–10** (Continued)

Comp.	Ar/Ar ₁	Yield (%)	Mol. form. (mol. wt.)	m.p. °C Solvent	% Analysis (calcd./found)			
					C	H	N	S
8f	<i>o</i> -C ₄ H ₃ S/ <i>o</i> -C ₄ H ₃ S	91	C ₁₉ H ₁₅ N ₃ OS ₃ (397)	135	57.43	3.77	10.57	24.18
				CHCl ₃ /pet. ether 40–60°C	57.00	3.72	10.23	24.58
9a	<i>o</i> -C ₅ H ₄ N/ <i>o</i> -C ₅ H ₄ N	92	C ₂₂ H ₁₉ N ₅ OS (401)	122	65.83	4.73	17.45	7.98
				CHCl ₃ /pet. ether 40–60°C	65.55	4.71	17.09	7.67
9b	<i>p</i> -C ₆ H ₄ F/ <i>p</i> -C ₆ H ₄ F	98	C ₂₄ H ₁₉ F ₂ N ₃ OS (435)	98	66.20	4.36	9.65	7.35
				AcOH	66.00	4.32	9.63	7.35
9c	<i>o</i> -C ₆ H ₄ F/ C ₆ H ₅	97	C ₂₄ H ₂₀ FN ₃ OS (417)	92	69.06	4.79	10.07	7.67
				Benzene/pet. ether 40–60°C	69.04	4.78	10.00	7.83
10a	<i>o</i> -C ₅ H ₄ N/ <i>o</i> -C ₅ H ₄ N	89	C ₂₂ H ₁₉ N ₅ OS (401)	210	65.83	4.73	17.45	7.98
				CHCl ₃ /pet. ether 40–60°C	65.92	4.71	17.61	7.92
10b	<i>p</i> -C ₆ H ₄ F/ <i>p</i> -C ₆ H ₄ F	97	C ₂₄ H ₁₉ F ₂ N ₃ OS (435)	110	66.20	4.36	9.65	7.35
				AcOH	66.08	4.31	9.25	7.11
10c	<i>o</i> -C ₆ H ₄ F/ C ₆ H ₅	93	C ₂₄ H ₂₀ FN ₃ OS (417)	148	69.06	4.79	10.07	7.67
				Benzene/pet. ether 40–60°C	69.00	4.45	9.73	7.15
10d	<i>o</i> -C ₄ H ₃ S/ <i>o</i> -C ₄ H ₃ S	95	C ₂₀ H ₁₇ N ₃ OS ₃ (411)	135	58.39	4.13	10.21	23.35
				CHCl ₃ /pet. ether 40–60°C	58.80	4.13	10.25	22.87

Compounds (**2**) was reacted with 2- or 3-bromopropanoic acid in refluxing acetic acid/acetic anhydride mixture to produce 5,6-diaryl-2,3-dihydro-2-methylimidazo[2,1-*b*]thiazol-3-ones (**6**) and 6,7-diaryl-2,3-dihydroimidazo[2,1-*b*]-1,3-thiazin-4-ones (**7**) respectively. The IR spectra of compounds (**6**) showed absorption around 1710–1750 cm⁻¹ (C=O) and the ¹H-NMR spectrum of compound (**6a**) [DMSO-*d*₆] showed the following peaks in δ ppm: a doublet at δ 1.8 (3H) for the methyl group, quartet at δ 2.3 (1H) for (CH) of the thiazole ring and a multiplet at δ 7.30–8.70 region (8H) for the aromatic protons (cf. Table II).

The IR spectra of compounds (**7**) showed absorption band at 1700 cm⁻¹ (C=O) and the ¹H-NMR spectrum of compound (**7a**) [DMSO-*d*₆ + CDCl₃] showed the following assignments in δ ppm, a multiplet centered at δ = 2.60–3.40 (4H, thiazin protons) and at δ 7.30–8.70 (m, 8H, Ar's) (cf. Table II).

Compounds (**3**, **6**, and **7**) have been cleaved by aniline to give the corresponding 2-(4',5'-diaryl-2',3'-dihydroimidazol-2'-yl)thioacetanilide (**8**), 2-(4',5'-diaryl-2',3'-dihydroimidazol-2'-yl)thiopropionamide (**9**), and 3-(4',5'-diaryl-2',3'-dihydroimidazol-2'-yl)thiopropionamide derivatives (**10**) respectively. Compounds (**8**, **9**, and **10**) showed correct values in elemental analyses as well as the expect IR peaks. ¹H-NMR [DMSO-*d*₆]

TABLE II IR and ¹H-NMR Spectral Data of Compounds **3–10**

Comp. No.	IR ν [cm ⁻¹]	¹ H-NMR	
		Solvent	[δ ppm]
2a	3160, 3470 (br., NH), 1260 (C=S)	A ^a	9.36–9.56 (br., 2H, 2NH); 7.2–9.0 (m, 8H, ArH's)
2b	3150 (br., NH), 1220 (C=S)	A	3.3 (br., 1H, NH), 5.6 (br., 1H, NH); 7.0–8.6 (m, 8H, ArH's)
2c	3080, 3120 (br., NH), 1260 (C=S)	A	4.2 (s, 1H, NH), 5.9 (s, 1H, NH); 6.65–8.20 (m, 8H, ArH's)
2d	3100 (br., NH), 1220 (C=S)	A	43.10 (br., 1H, NH), 6.8–8.4 (m, 9H, ArH's); 12.4 (br., 1H, NH)
2e	3080 (br., NH), 1220 (C=S)	A	3.3 (br., 2H, 2NH); 6.60–8.00 (m, 6H, ArH)
3a	1710 (C=O), 2925 (CH ₂)	A	3.25 (s, 2H, CH ₂); 7.4–8.6 (m, 8H, ArH's)
3b	1700 (C=O), 2920 (CH ₂)	A	3.75 (s, 2H, CH ₂); 7.0–7.75 (m, 8H, ArH's)
3c	1700 (C=O), 2930 (CH ₂)	A	3.50 (s, 2H, CH ₂); 7.1–8.3 (m, 9H, ArH's)
3d	1700 (C=O), 2920 (CH ₂)	A	3.5 (s, 2H, CH ₂); 6.9–8.3 (m, 6H, thiophene protons)
4a	1700 (C=O)	A	7.0–8.6 (m, 13H, Ar's and benzylic proton)
4b	1675 (C=O), 2910 (CH ₃)	A	3.8 (s, 3H, OCH ₃); 7.06–9.1 (m, 13H, ArH's and benzylic proton)
4c	1690 (C=O)	A	7.4–8.6 (m, 13H, ArH's and benzylic proton)
4d	1685 (C=O)	A	7.8–8.7 (m, 13H, ArH's and benzylic proton)
4e	1700 (C=O)	B ^b	6.8–8.0 (m, 13H, ArH's and benzylic proton)
4f	1700 (C=O), 2920 (CH ₃)	A	3.3 (s, 3H, OCH ₃); 7.1–7.9 (m, 13H, ArH's and benzylic proton)
4g	1740 (C=O)	A	6.9–8.2 (m, 13H, ArH's and benzylic proton)
4h	1740 (C=O)	B	7.0–8.4 (m, 13H, ArH's and benzylic proton)
4i	1680 (C=O)	B	7.1–8.0 (m, 13H, ArH's and benzylic proton)
4j	1730 (C=O)	A	6.9–8.2 (m, 13H, ArH's and benzylic proton)
4k	1700 (C=O), 2940 (CH ₃)	A	3.8 (s, 3H, OCH ₃); 7.00–8.0 (m, 14H, ArH's and benzylic proton)
4l	1740 (C=O)	A	7.2–8.4 (m, 14H, ArH's and benzylic proton)
4m	1730 (C=O)	A	6.09–8.0 (m, 14H, ArH's and benzylic proton)
4n	1690 (C=O)	B	6.9–8.0 (m, 11H, ArH's, thiophene protons and benzylic proton)
4o	1690 (C=O), 2920 (OCH ₃)	A	3.9 (s, 3H, OCH ₃); 7.00–7.9 (m, 11H, ArH's, thiophene protons and benzylic proton)
4p	1685 (C=O)	A	7.1–8.2 (m, 11H, ArH's, thiophene protons and benzylic proton)
4q	1690 (C=O)	B	6.8–7.9 (m, 11H, ArH's, thiophene protons and benzylic proton)
5a	1690 (C=O), 3100 (br., NH)	A	7.0–8.5 (m, 13H, ArH's); 9.4 (s, 1H, NH)

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TABLE II IR and ¹H-NMR Spectral Data of Compounds **3–10** (Continued)

Comp. No.	IR ν [cm ⁻¹]	¹ H-NMR	
		Solvent	[δ ppm]
5b	1680 (C=O), 3250 (br., NH)	A	6.6–9.0 (m, 12H, ArH's); 10.04 (s, 1H, NH)
5c	1680 (C=O), 3150 (br., NH)	A	6.6–9.4 (m, 12H, ArH's); 11.8 (s, 1H, NH)
5d	1680 (C=O), 3280 (br., NH)	A	3.31 (s, 1H, NH); 6.6–8.7 (m, 12H, ArH's)
5e	1685 (C=O), 3230 (br., NH)	A	6.8–8.2 (m, 12H, ArH's); 10.4 (s, 1H, NH)
5f	1680 (br., C=O), 3050 (br., NH)	A	3.39 (br., 1H, NH); 6.6–7.8 (m, 13H, ArH's)
5g	1700 (br., C=O), 3050 (br., NH)	A	3.2 (br., 1H, NH); 6.8–8.7 (m, 12H, ArH's)
5h	1710 (br., C=O), 3060 (br., NH)	A	3.3 (br., 1H, NH); 6.6–8.0 (m, 12H, ArH's)
5i	1680 (br., C=O), 3050 (br., NH)	A	3.2 (br., 1H, NH); 6.9–7.8 (m, 12H, ArH's); 9.4 (s, 1H, NH)
5j	1700 (br., C=O), 3050 (br., NH)	A	2.3 (br., 1H, NH); 6.8–7.8 (m, 14H, ArH's)
5k	1700 (br., C=O), 3100 (br., NH)	B	2.3 (br., 1H, NH); 6.6–7.8 (m, 13H, ArH's)
5l	1700 (br., C=O), 3080 (br., NH)	B	2.3 (br., 1H, NH); 6.5–8.0 (m, 13H, ArH's)
5m	1680 (br., C=O), 3050 (br., NH)	A	3.5 (br., 1H, NH); 6.6–7.8 (m, 13H, ArH's)
5n	1700 (br., C=O)	A	2.7 (s, 1H, CH); 6.2–8.0 (m, 11H, ArH's and thiophene protons)
5o	1700 (br., C=O)	A	3.4 (s, 1H, CH); 6.8–8.5 (m, 10H, ArH's and thiophene protons)
5p	1700 (br., C=O), 3090 (br., NH)	B	3.85 (br., 1H, NH); 6.8–8.5 (m, 10H, ArH's and thiophene protons)
5q	1650 (br., C=O), 3100 (br., NH)	B	6.7–8.0 (m, 10H, ArH's and thio- phene protons); 8.7 (br., 1H, NH)
5r	1685 (C=O), 3150 (br., NH).	A	3.5 (br., 1H, NH); 6.7–8.9 (m, 10H, ArH's and thiophene protons)
6a	1710 (C=O), 2910 (CH ₃)	B	1.8 (d, 3H, CH ₃); 2.3 (q, 1H, CH); 7.3–8.7 (m, 8H, ArH's)
6b	1750 (C=O), 2930 (CH ₃)	A	1.5 (d, 3H, CH ₃); 2.3 (q, 1H, CH); 6.6–8.4 (m, 8H, ArH's)
6c	1710 (C=O), 2920 (CH ₃)	A	1.7 (d, 3H, CH ₃); 2.3 (q, 1H, CH); 6.6–8.2 (m, 9H, ArH's)
6d	1710 (C=O), 2920 (CH ₃)	A	1.7 (d, 3H, CH ₃); 2.4 (q, 1H, CH); 6.8–8.2 (m, 6H, thiophene protones)
7a	1700 (C=O)	A+B	2.6–3.4 (m, 4H, thiazine protons); 7.3–8.7 (m, 8H, ArH's)
7b	1700 (C=O)	B	3.9–4.5 (m, 4H, thiazine protons); 6.9–7.3 (m, 8H, ArH's)

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TABLE II IR and ¹H-NMR Spectral Data of Compounds **3–10** (Continued)

Comp. No.	IR ν [cm ⁻¹]	¹ H-NMR	
		Solvent	[δ ppm]
7c	1700 (C=O)	B	1.8–3.3 (m, 4H, thiazine protons); 6.6–8.1 (m, 9H, ArH's)
7d	1700 (C=O)	B	2.75 (m, 2H, thiazine protons); 6.5–8.0 (m, 6H, ArH's)
8a	1700 (C=O), 3040 (NH), 3400 (NH), 2920 (CH ₂)	A	1.8 (s, 1H, NH); 3.2 (s, 2H, CH ₂); 6.4–8.8 (m, 13H, ArH's); 10.6 (s, 1H, NH)
8b	1680 (C=O), 3090 (br., NH), 2916 (CH ₂)	B	3.6 (s, 1H, NH); 3.8 (s, 2H, CH ₂); 6.7–8.4 (m, 13H, ArH's); 10.7 (s, 1H, NH)
8c	1680 (C=O), 3280 (NH), 2920 (CH ₂)	A	3.4 (s, 1H, NH); 3.9 (s, 2H, CH ₂); 6.9–8.2 (m, 12H, ArH's); 10.5 (s, 1H, NH)
8d	1680 (C=O), 3060 (NH), 3390 (br., NH), 2915 (CH ₂)	B	3.5 (s, 1H, NH); 4.0 (s, 2H, CH ₂); 6.8–8.0 (m, 14H, ArH's); 10.2 (s, 1H, NH)
8e	1680 (C=O), 3100 (NH), 3280 (NH), 2920 (CH ₂)	A	2.5 (s, 1H, NH); 3.35 (s, 2H, CH ₂); 7.2–8.2 (m, 13H, ArH's); 10.5 (s, 1H, NH)
8f	1670 (C=O), 3080 (NH), 3350 (NH), 2915 (CH ₂)	B	1.4 (s, 1H, NH); 1.9 (s, 1H, NH); 4.3 (s, 2H, CH ₂); 5.0–8.0 (m, 11H, ArH's and thiophene protons)
9a	1630 (C=O), 3060 (br., NH), 3400 (br., OH)	A	1.2 (d, 3H, CH ₃); 2.9 (s, 1H, NH); 3.5 (q, 1H, CH); 7.0–8.2 (m, 13H, ArH's); 8.7 (s, 1H, NH)
9b	1740 (C=O), 3050 (br., NH)	B	1.6 (d, 3H, CH ₃); 2.6 (s, 1H, NH); 4.2 (q, 1H, CH); 6.8–7.5 (m, 13H, ArH's); 8.1 (s, 1H, NH)
9c	1740 (C=O), 3060 (br., OH)	B	1.5 (d, 3H, CH ₃); 2.15 (s, 1H, NH); 4.25 (q, 1H, CH); 6.7–7.8 (m, 14H, ArH's); 8.2 (s, 1H, NH)
10a	1690 (C=O), 3060 (br., NH), 3400 (br., OH)	A	1.3 (s, 1H, NH); 2.5–3.6 (m, 4H, 2CH ₂); 6.5–7.8 (m, 13H, ArH's); 8.7 (s, 1H, NH)
10b	1700 (C=O), 3040 (br., NH), 3400 (br., OH)	B	1.25 (s, 1H, NH); 2.1–3.0 (m, 4H, 2CH ₂); 6.7–8.0 (m, 13H, ArH's); 8.4 (s, 1H, NH)
10c	1715 (C=O), 3060 (br., NH), 3400 (br., OH)	B	1.4 (s, 1H, NH); 3.2–3.9 (m, 4H, 2CH ₂); 6.7–7.8 (m, 14H, ArH's); 9.0 (s, 1H, NH)
10d	1680 (C=O), 3040 (br., NH), 3400 (br., OH)	A	1.4 (s, 1H, NH); 3.0–3.7 (m, 4H, 2CH ₂); 6.8–7.8 (m, 11H, ArH's); 8.0 (s, 1H, NH)

^aA = DMSO-d₆.^bB = CDCl₃.

of compound (**8a**) as an example, showed signals at δ 1.80 ppm (s, 1H, NH), δ 3.20 ppm (s, 2H, CH₂), δ 6.40-8.80 ppm (m, 13H, ArH's), and δ 10.60 ppm (s, 1H, NH) (cf. Table II).

The structures of the new compounds were established by chemical analysis, mass spectra, ¹H-NMR and IR spectra. Physical data of compounds (**2-20**) are given in (Table I and II).

Antimicrobial Activity

The antimicrobial activity of the compounds considered was tested on *Bacillus subtilis*, *Staphylococcus aureus*, *Escherichia Coli*, *Proteus mirabilis*, *Candida albicans*, and *Aspergillus niger*. The biological activity was determined according to the cup plate method.¹² The sensitivity of microorganisms to the compound is identified in the following manner:

(+++++) = highly sensitive (inhibition zone > 15 mm);

(++++)= fairly sensitive (inhibition zone > 12 mm);

(++) = slightly sensitive (inhibition zone > 9 mm);

(+) = very slightly sensitive (inhibition zone > 6 mm).

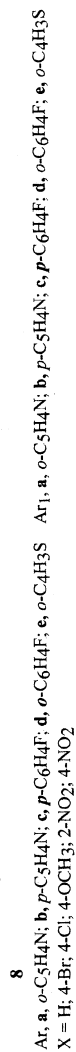
It could be mentioned that, the most determined effect is brought about by compounds **4i**, **4o**, **4f**, **4r**, **5d**, **5i**, **5j**, **5k**, **5o**, **5p**, **5r**, **7d**, **9a**, and **10d** (Table III).

EXPERIMENTAL

All melting points uncorrected. The ¹H-NMR spectra were recorded on a Varian 1H, Gemini 200 Spectrometer (National Research Centre, Egypt) and chemical shifts were expressed as δ -value against TMS as internal standard. The IR spectra (KBr) were recorded on a Perkin-Elmer 1430 Spectrometer (Cairo Univesity). The mass spectra were recorded on Finnigan SSQ 7000 Spectrometer. Microanalytical data performed by the Microanalytical Center at the Faculty of Science, Cairo University.

4,5-Diaryl-2,3-dihydro-2-mercaptoimidazoles **2a-e**

Compounds **2** were prepared from disubstituted benzoin¹³⁻¹⁵ derivatives, by fusion with thiourea at 200°C.¹⁰ Physical data, IR, and ¹H-NMR spectra (Tables I and II).



5,6-Diaryl-2,3-dihydro-2-mercaptoimidazo[2,1-*b*]thiazol-3-ones 3a–d

Method A

A mixture of compounds **2** (0.01 mmol) and chloroacetic acid (0.01 mmol) and 2 g of fused anhyd. Sodium acetate in 15 mL of acetic acid and 7 mL of acetic anhydride was refluxed for 3 h and left to cool. The reaction mixture was poured onto water. The solid obtained was filtered off and crystallized from the proper solvent (Tables I and II).

Method B

Chloroacetyl chloride (0.01 mmol), was added gradually while stirring to a cooled solution of compound **2** (0.01 mmol) in 30 mL pyridine. The reaction mixture was left while cooling for 1 h, then poured into cold water, filtered and crystallised from the proper solvent (Tables I and II).

2-Arylmethylene-5,6-diaryl-2,3-dihydroimidazo[2,1-*b*]thiazol-3-ones 4a–q

Method A

A mixture of compound **3** (0.01 mmol), aromatic aldehyde (0.01 mmol) in a mixture of glacial acetic acid (20 mL) and acetic anhydride (10 mL) in the presence of anhydrous sodium acetate was refluxed for 2 h. After cooling, water was added and the precipitate was filtered off and crystallized from the proper solvent (Tables I and II).

Method B

A mixture of compounds **2** (0.01 mmol), chloroacetic acid (0.01 mmol), aromatic aldehyde and 2 g of anhydrous sodium acetate in a mixture of 20 mL acetic acid/10 mL acetic anhydride was refluxed for 1 h. It was cooled and then poured onto water. The solid obtained was filtered off and crystallized from the proper solvent (Tables I and II).

2-Arylhydrazono-5,6-diaryl-2,3-dihydroimidazo[2,1-*b*]thiazol-3-ones 5a–r

The aromatic amine (0.01 mmol) was dissolved in 3 mL of conc. HCl acid and 2 mL of water, cooled to -10°C and treated with 0.07 g of sodium nitrile in 3 mL of water. The diazotized amine was added gradually while stirring to cold solution of compound **3** (0.01 mmol) in 15 mL of pyridine. The reaction mixture was cooled for 30 min. The product was filtered off and crystallized from the proper solvent (Tables I and II).

TABLE III Antimicrobial Screening for the Selected Compounds Against Bacteria, Yeast and Fungi

Comp.	<i>B. subtilis</i>	<i>S. aureus</i>	<i>E. coli</i>	<i>P. mirabilis</i>	<i>C. albicans</i>	<i>A. niger</i>
Control^a	++++	+++++	+++	+++	++	++
1c	+	+	—	—	+	+
2d	+	+	—	—	+	+
3a	++	++	+	+	++	++
3b	+	+	—	—	+	+
3c	+	+	—	—	+	+
4a	++	++	++	++	++	++
4d	++	++	++	++	++	++
4e	++	++	+	+	++	++
4f	+	+	+	+	++	++
4g	++	++	+	+	+	+
4h	++	++	+	+	++	++
4i	++	++	+	+	+	+
4j	+++	+++	+	+	++	++
4k	++	++	+	+	++	++
4o	+++	+++	+	+	+	+
4p	++	++	+	+	+	+
4q	+	+	+	+	+++	+++
4r	+	+	+	+	+++	+++
5d	++	++	++	++	+++	+++
5f	++	++	+	+	++	++
5g	++	++	+	+	++	++
5h	++	++	+	+	+	+
5i	+++	+++	+	+	+++	+++
5j	++	++	+	+	+++	+++
5k	++	++	++	+++	+++	+++
5l	++	++	+	+	++	++
5n	++	++	+	+	+	+
5o	++	++	++	++	+++	+++
5p	+	+	+	+	+++	+++
5q	+	+	+	+	+++	+++
5r	+++	+++	++	++	+++	+++
6a	++	++	++	++	++	++
6b	+	+	—	—	+	+
6c	++	++	+	+	+	+
7a	++	++	++	++	++	++
7b	+	+	—	—	+	+
7c	+	+	—	—	+	+
7d	++	++	+	+	+++	+++
8b	++	++	++	++	++	++
8c	+	+	—	—	+	+
8d	+	+	—	—	+	+
8e	+	+	—	—	+	+
8f	+	+	+	+	+	+
9a	+++	+++	++	++	+++	+++
9b	+	+	—	—	+	+
10a	+	+	—	—	+	+
10d	++	++	+	+	+++	+++

^aControl Ampicillin.

5,6-Diaryl-2,3-dihydro-2-methylimidazo[2,1-*b*]-thiazol-3-ones 6a–d

Compounds **6** were prepared by the same method used for compounds **3**, but by using 2-bromopropionic acid instead of chloroacetic acid (Tables I and II).

6,7-Diaryl-1,3-thiazino[3,2-*a*]imidazol-4-ones 7a–d

Compounds **7** were prepared by the same method use for compounds **3** but by using 3-bromopropionic acid instead of chloroacetic acid (Tables I and II).

2-(4',5'-diaryl-2',3'-dihydroimidazo-2'-yl thio)acetanilide Derivatives 8a–f

Compounds **3** (0.005 mmol) and (0.006 mmol) of the aromatic amine were refluxed in 15 mL of ethanol for 2 h. The reaction mixture was allowed to cool then poured into water. The solid product formed was collected, washed with little ethanol and crystallized from the proper solvent (Tables I and II).

2-(4',5'-Diaryl-2',3'-dihydroimidazol-2'-yl)-thiopropionamide Derivatives 9a–c

Compounds **9** were prepared by the same method used for compounds **8** (Tables I and II).

3-(4',5'-Diaryl-2',3'-dihydroimidazol-2'-yl)-thiopropionamide 10a–d

Compounds **10** were prepared by the same method used for compounds **8** (Table I and II).

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