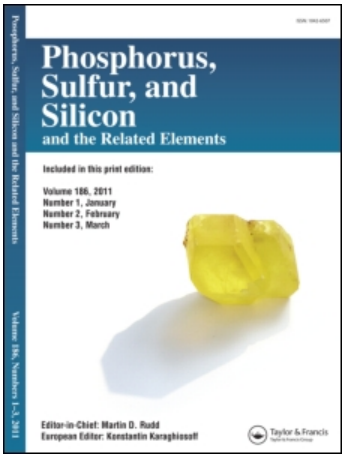

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Compliancy of 1,4-Naphthoquinone for the Synthesis of New Heterocyclic Quinone Compounds Containing Imino Linkage

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Some new β-lactam, thiazolidinone derivatives were synthesized by reaction of new Schiff bases with compounds **4** *and oxazoles, pyrazoles, pyrimidines, and pyrimidinethion were also synthesized by reaction of an arylidino system with hydroxyl amine hydrochloride, hydrazines, urea, and thiourea, which we expected to have biological activities such as bactericidal and fungicidal or other applications of certain interest.*

Keywords Arylidino derivatives; isolated β-lactams; oxazoles; pyrazoles; pyrimidines; pyrimidinethion; Schiff bases; thiazolidinone derivatives

INTRODUCTION

Three membered rings containing nitrogen, oxygen, and sulphur atoms as the heteroatom are called aziridine, oxirane, and thiirane, respectively. A duly examination of the literature for fused three membered heterocycles with other hetero rings indicated that such systems too reave one class of antibiotics.[1] Also β-lactam and related derivatives exhibit antibacterial activities.[2–7] Thiazole derivatives and β-lactam rings were known and used as potent antibiotics,[8] antibiotic activity,[9] and inhibitory activity.[10] Diverse biological activities, such as bactericidal, pesticidal, fungicidal, insecticidal, anticonvulsant, tuberculostatic, anti-inflammatory, and antithydroidal have been found to be associated with thiazolidinone derivatives.[11] Thus, our initial strategy in this

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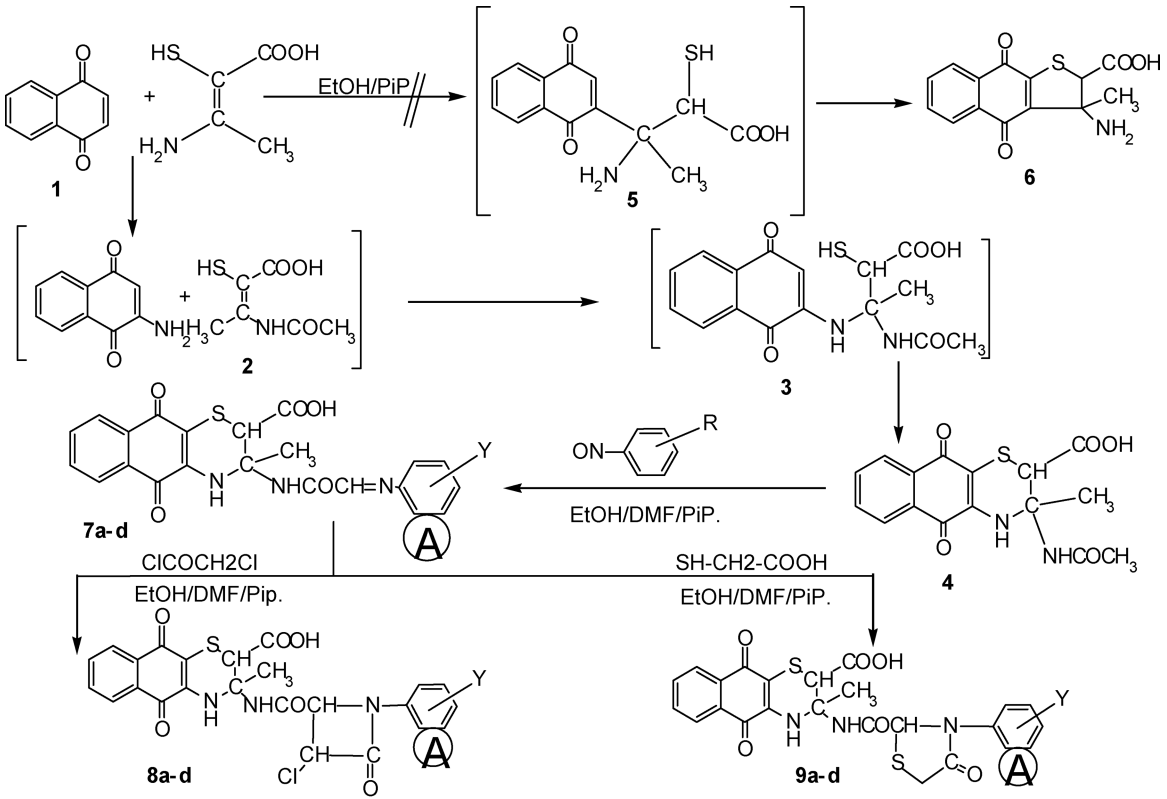
research project was divested to the synthesis of different structural formulas corresponding to the isolated β-lactams, thiazolidinone derivatives, pyrazoles, oxazoles, pyrimidines, and pyrimidinethion.

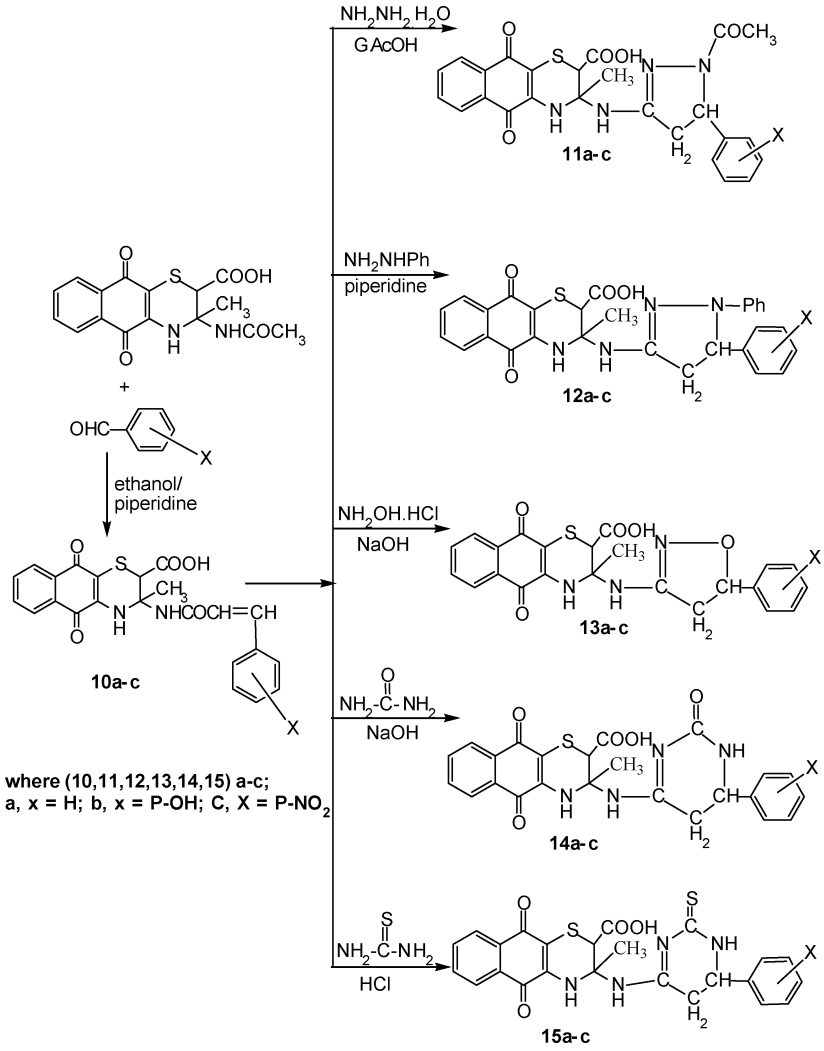
RESULTS AND DISCUSSION

The known importance of these heterocyclic compounds with respect to various types of biological effects promysted us to synthesize new heterocyclic compounds depending upon our previous research in this field.[12–14] Thus, 1, 4-naphthoquinone **1** was reacted with acetamide and mercaptoacetic acid in ethanol at reflux in the presence of piperidine to give 2-amido-2-methyl-2-H-naphtho[2, 3-b][1, 4]thiazine-5, 10-dione, 3-carboxylic acid **4**. The formation of **4** was assumed to proceed via the intermediate **2**, which cyclizes to **4**, (c.f., Scheme 1). The IR spectrum of **4** revealed the presence of absorption bands at 1665, 3421, 3069 cm^{-1} assigned to CO, NH, OH groups, respectively. The ^{1}H-NMR spectrum (DMSO-d6) were showed a singlet at δ 1.02 assigned for methyl group, a singlet at δ 3.43 for a methyl group of the acetyl group, δ 6.01 for (CH-S), multaiplets at δ 7.10–8.10 for aromatic protons, and a broad singlet at δ 9.01 for (NH) protons, respectively, with lack of a peak due to an NH_2 group. The mass spectrum of compound **4** showed m/z at 346 (M^+, 346). The compound **4** contains methyl groups which render it susceptible to react with different nitroso compounds such as β-nitrosoα-naphthol, α-nitrosoβ-naphthol, P-nitrsophenol, and P-nitrso-N,Ndimethyl-aniline in the presence of piperidine as catalyst to give the corresponding Schiff base derivatives **7a–d** (Scheme 1). The reaction of **7a–d** with an equimolar ratio of chloroacetylchloride in a mixture of DMF and ethanol in the presence of piperidine as a catalyst afforded β-lactam derivatives **8a–d** (Scheme 1). Cycloaddition reaction of thioglycolic acid to the previously prepared Schiff base compounds **7a–d** in a mixture of ethanol and DMF in the presence of piperidine as catalyst afforded thiazolidinone derivatives **9a–d** (Scheme 1). The active methyl group in the new compound **4** condensed with different aromatic aldehydes in a mixture of ethanol and DMF under piperidine as a catalyst to yield the corresponding arylidino **10a–c**. The arylidino derivatives **10a–c**, when interacted with hydrazine, hydrate in the presence of acetic acid in ethanol as solvents under piperidine as catalyst, respectively, gave the required N-acetylpyrazolo **11a–d**, and/or N-phenylpyrazolo derivatives **12a–d**, respectively (Scheme 2). The arylidino derivatives **10a–c** when interacted with hydroxylamine hydrochloride in a mixture of ethanol and DMF as solvent under the effect of sodium hydroxide as catalyst, gave the required isooxazolino derivatives **13a–c**.The arylidino



where (7, 8, 9) a-d, a, y, a-OH, A, C_6H_4; b, a-OH, A, C_6H_4, a, y, P-OH, A, H, d, y, P-$N(CH_3)_2$, A, H

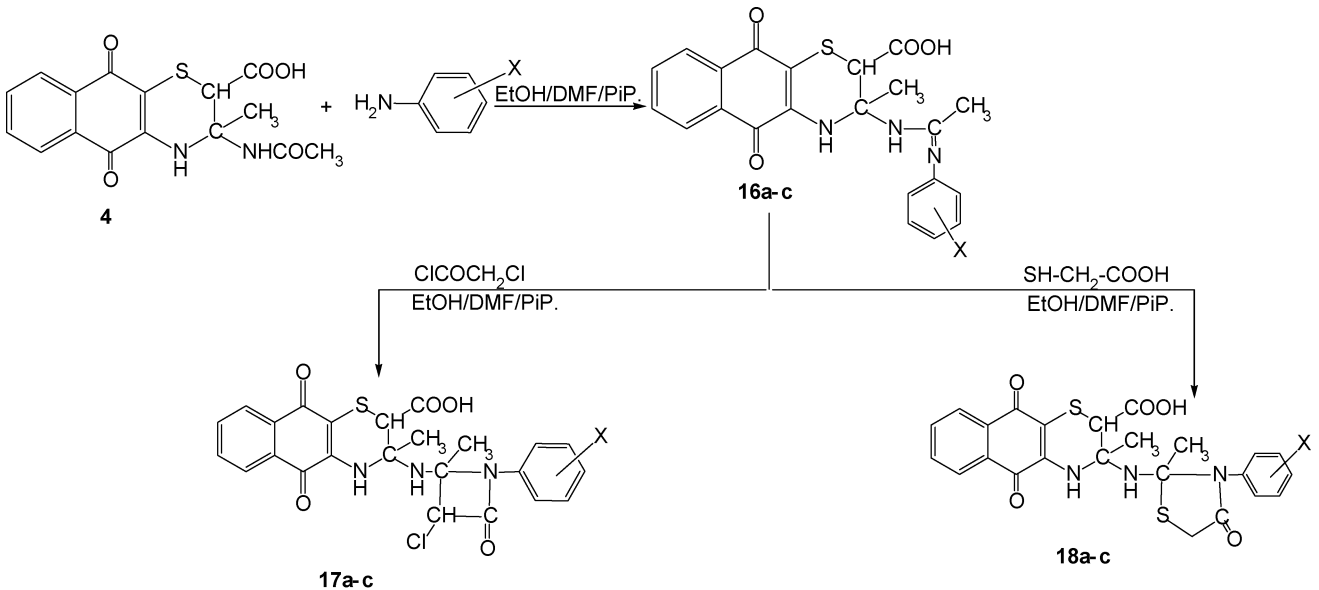
SCHEME 1



SCHEME 2

derivatives **10a–c**, when interacted with urea and/or thiourea in a mixture of ethanol and DMF as solvent under the effect of sodium hydroxide as catalyst, gave the required pyrimidino derivatives **14a–c** or, under the effect of hydrochloric acid as catalyst, gave the required pyrimidinethion derivatives **15a–c**.

The activity of the carbonyl group in compound **4** renders it available to react with different aromatic amine such as aniline, P-chloroaniline, and P-nitroaniline in the presence of a mixture of ethanol and DMF



where 16, 17, 18 a-c; a, x = H; b, x = P-Cl, c; x = P-NO_2

SCHEME 3

as a solvent and piperidine catalyst to give new Schiff base derivatives **16a–c** (Scheme 3). The reaction of **16a–c** with equimolar ratios of chloroacetylchloride in mixture of ethanol and DMF in the presence of piperidine catalyst afforded lactam derivatives **17a–c** (Scheme 3). Cycloaddition reaction of thioglycolic acid to the previously prepared Schiff bases **16a–c** proceeded successfully. Thus thioglycolic acid added to Schiff bases **16a–c** in a mixture of ethanol and DMF in the presence of piperidine as catalyst and afforded thiazolidinone derivatives **18a–c** (Scheme 3).

EXPERIMENTAL

All melting points were uncorrected. IR spectra were recorded on a pye Unicam SP 1100 spectrophotometer using KBr disks. ^{1}HNMR spectra were recorded on a Varian EM390 MHz spectrophotometer using DMSO-d6 as a solvent and TMS as an internal standard. Chemical shifts are expressed as ppm units. Mass spectra were recorded on a HP Ms6988 spectrometer. Analytical data were determined with a CE 440 Elemental Analyzer-Automatic Indicator at Cairo University.

Synthesis of 2-Amido-2-methyl-2-H-naphtho[2, 3-b][1, 4]thiazine-5, 10-dione, 3-carboxylic acid 4

A solution of acetamide (0.59 g, 0.01 mol) and mercaptoaceticacid (0.92 g, 0.01 mol) was treated with 1,4-naphthoquinone (1.58 g, 0.01 mol) in ethanol in the presence of piperidine as catalyst and refluxed for 15 h. The reaction mixture was filtered from unreacted materials. The filtered was poured into ice water acidified by concentrated hydrochloric acid. A reddish brown precipitate separated from the filtrate and was washed several times with water. It was crystallized from ethanol and dimethyl formamide to give compound **8a–d**, mp 290°C (c.f., Table I).

Synthesis of 2-diazaamido-2-methyl-2-H-naphthol[2, 3-b][1, 4]thiazine-5, 10-dione, 3-carboxylic acid 7a–d

Compound **4** (3.46 g, 0.01 mol) and nitroso compounds (1.73 g, 1.73 g, 1.50 g, 0.01 mol) in an equimolar ratio were dissolved in ethanol and dimethylformamide, and 0.5 ml of piperidine as catalyst was added. The mixture was refluxed about 9 h. The reaction mixture was allowed to cool at room temperature, then filtered, washed several times with water, dried, collected, and crystallized from ethanol to give **7a–d** (c.f., Table I).

TABLE I Characterization of Compounds

Compound number	Solvent of crystallization	Melting point	Yield %	Molecular formula	Molecular weight	Mass spectra
4	EtOH/DMF	290	85%	$C_{16}H_{14}O_5N_2S$	346.06	348(M+2)
7a	EtOH/DMF	115	75%	$C_{26}H_{19}O_6N_3S$	501.1	502(M+1)
7b	EtOH/DMF	120	75%	$C_{26}H_{19}O_6N_3S$	501.1	502(M+1)
7c	EtOH/DMF	125	80%	$C_{22}H_{17}O_6N_3S$	451.09	452(M+1)
7d	EtOH/DMF	122	75%	$C_{24}H_{22}O_5N_4S$	478.13	478
8a	EtOH/DMF	260	65%	$C_{28}H_{20}O_7N_3SCl$	577.07	579(M+2)
8b	EtOH/DMF	260	65%	$C_{28}H_{20}O_7N_3SCl$	577.07	579(M+2)
8c	EtOH/DMF	250	67%	$C_{24}H_{17}O_7N_3SCl$	530.10	530
8d	EtOH/DMF	270	70%	$C_{26}H_{23}O_6N_4SCl$	554.10	554
9a	EtOH/DMF	360	70%	$C_{28}H_{21}O_7N_3S_2$	575.08	575
9b	EtOH/DMF	360	73%	$C_{28}H_{21}O_7N_3S_2$	575.08	575
9c	EtOH/DMF	320	75%	$C_{24}H_{19}O_7N_3S_2$	525.06	525
9d	EtOH/DMF	340	72%	$C_{26}H_{24}O_6N_4S_2$	552.11	552
10a	EtOH/DMF	250	80%	$C_{23}H_{18}O_5N_2S$	434.09	432(M-2)
10b	EtOH/DMF	220	82%	$C_{23}H_{18}O_6N_2S$	450.09	449(M-1)
10c	EtOH/DMF	190	78%	$C_{23}H_{17}O_7N_3S$	479.08	479
11a	EtOH/DMF	260	85%	$C_{25}H_{22}O_5N_4S$	490.97	449(M+3)
11b	EtOH/DMF	230	85%	$C_{25}H_{22}O_6N_4S$	506.96	508(M+1)
11c	EtOH/DMF	200	82%	$C_{25}H_{21}O_7N_5S$	535.45	535
12a	EtOH/DMF	230	70%	$C_{29}H_{24}O_4N_4S$	524.15	524
12b	EtOH/DMF	235	68%	$C_{29}H_{24}O_5N_4S$	540.15	540
12c	EtOH/DMF	225	70%	$C_{29}H_{23}O_6N_5S$	569.14	569
13a	EtOH/DMF	200	75%	$C_{23}H_{19}O_5N_3S$	449.11	449
13b	EtOH/DMF	210	73%	$C_{23}H_{18}O_6N_3S$	465.1	465
13c	EtOH/DMF	215	70%	$C_{23}H_{18}O_7N_4S$	494.1	494
14a	EtOH/DMF	220	78%	$C_{24}H_{20}O_5N_4S$	476.11	478(M+2)
14b	EtOH/DMF	225	73%	$C_{24}H_{20}O_6N_4S$	492.11	492
14c	EtOH/DMF	227	70%	$C_{24}H_{19}O_7N_5S$	521.1	521
15a	EtOH/DMF	215	75%	$C_{24}H_{20}O_4N_4S_2$	491.1	491
15b	EtOH/DMF	220	75%	$C_{24}H_{20}O_5N_4S_2$	507.1	507
15c	EtOH/DMF	220	73%	$C_{24}H_{19}O_6N_5S_2$	537.1	537
16a	EtOH/DMF	240	76%	$C_{22}H_{19}O_4N_3S$	421.1	423(M+2)
16b	EtOH/DMF	242	74%	$C_{22}H_{18}O_4N_3SCl$	455.1	455
16c	EtOH/DMF	245	73%	$C_{22}H_{18}O_6N_4S$	466.1	466
17a	EtOH/DMF	210	72%	$C_{24}H_{20}O_5N_3SCl$	497.1	497
17b	EtOH/DMF	215	70%	$C_{24}H_{19}O_5N_3SCl_2$	531.04	531
17c	EtOH/DMF	212	70%	$C_{24}H_{19}O_7N_4SCl$	542.1	542
18a	EtOH/DMF	230	75%	$C_{24}H_{21}O_5N_3S_2$	495.1	495
18b	EtOH/DMF	235	72%	$C_{24}H_{20}O_5N_3S2Cl$	529.05	529
18c	EtOH/DMF	240	70%	$C_{24}H_{19}O_7N_4S_2$	540.1	540

Synthesis of New β-Lactam Derivatives 8a–d

A solution of **7a–d** (5.01 g, 0.01 mol) chloroacetylchloride (1.13 g, 0.01mol) in a mixture of ethanol and dimethylformamide in the

TABLE II Selected HNMR Spectra Data for the New Compounds Listed in Table I

Compound number	^{1}H-NMR d6 (DMSO)
4	δ1.77(s,CH3), δ 3.74 (s,COCH3), δ 4.5 (s,NH), δ 7.01–9 (m,7H, Aromatic protons, CH,NH,OH)
7a	δ1.63(s,CH3), δ 3.54 (s,CH), δ 6.01 (s,NH), δ 7.11–8.07 (m,13H, Aromatic protons, CH,NH,OH), δ 9.673 (s, Carboxylic OH)
7b	δ1.60(s,CH3), δ 3.50 (s,CH), δ 6.02 (s,NH), δ 7.0–8.2 (m,13H, Aromatic protons, CH,NH,OH), δ 9.723 (s, Carboxylic OH)
7c	δ1.4(s,CH3), δ 3.7 (s,CH), δ 5.8 (s,NH), δ 7.2–8.3 (m,11H, Aromatic protons, CH,NH,OH), δ 9.9 (s, Carboxylic OH)
7d	δ1.5(s,CH3), δ 2.34 (s,6H,2CH_3), δ 3.56 (s,CH), δ 5.6 (s,NH), δ 7.1–8.0 (m,10H, Aromatic protons, CH,NH,), δ 9.92 (s, Carboxylic OH)
8a	δ1.64(s,CH_3), δ 2.92 (s,CH), δ 3.41 (s,CH), δ 5.23 (s,NH), δ 7.05–8.7 (m,13H, Aromatic protons, CH,NH,OH), δ 8.85 (s, Carboxylic OH)
8b	δ1.45(s,CH_3), δ 2.83 (s,CH), δ 3.35 (s,CH), δ 5.41 (s,NH), δ 7.1–8.8 (m,13H, Aromatic protons, CH,NH,OH), δ 8.9 (s, Carboxylic OH)
8c	δ1.53(s,CH_3), δ 2.9 (s,CH), δ 3.3 (s,CH), δ 5.52 (s,NH), δ 7–8.3 (m,11H, Aromatic protons, CH,NH,OH), δ 8.98 (s, Carboxylic OH)
8d	δ1.56(s,CH_3), δ 2.3 (s,6H,2CH_3), δ 2.98 (s,CH), δ 3.54 (s,CH), δ 5.64 (s,NH), δ 7–8 (m,10H, Aromatic protons, CH,NH,), δ 9.5 (s, Carboxylic OH)
9a	δ1.62(s,CH_3), δ 2.9 (s,CH), δ 3.42 (s,CH_2), δ 5.24 (s,NH), δ 7.5–8.19 (m,13H, Aromatic protons, CH,NH,OH), δ 8.21 (s, Carboxylic OH)
9b	δ1.62(s,CH_3), δ 2.89 (s,CH), δ 3.42 (s,CH_2), δ 5.23 (s,NH), δ 7.5–8.2 (m,13H, Aromatic protons, CH,NH,OH), δ 8.32 (s, Carboxylic OH)
9c	δ1.63(s,CH_3), δ 2.9 (s,CH), δ 3.43 (s,CH_2), δ 5.23 (s,NH), δ 7.5–8.18 (m,11H, Aromatic protons, CH,NH,OH), δ 8.4 (s, Carboxylic OH)
9d	δ1.66(s,CH_3), δ 2.65 (s,6H,2CH_3), δ 2.95 (s,CH), δ 3.45 (s,CH_2), δ 5.4 (s,NH), δ 7.1–8.3 (m,10H, Aromatic protons, CH,NH,), δ 8.5 (s,Carboxylic OH)
10a	δ1.61(s,CH_3), δ 3.41 (s,NH), δ 5.99 (s,CH), δ 6.49 (s,CH), δ 7.37 (s,SCH), δ 7.45–82 (m,10H, Aromatic protons, NH), δ 10.05 (s, OH)
10b	δ1.6(s,CH_3), δ 3.8 (s,NH), δ 6.31 (s,CH), δ 6.89 (s,CH), δ 7.35 (s,SCH), δ 7.43–8 (m,10H, Aromatic protons, NH,OH), δ 10.24 (s,Carboxylic OH)
10c	δ1.8(s,CH_3), δ 4.2 (s,NH), δ 6.65 (s,CH), δ 6.9 (s,CH), δ 7.3 (s,SCH), δ 7.4–8.01 (m,9H, Aromatic protons, NH), δ 10.47 (s, OH)
11a	δ1.45(s,CH_3), δ 1.65 (s,$COCH_3$), δ 1.7 (s,CH_2), δ 2.4 (s,CH), δ 3.7(s,NH), δ 7.03-8.54 (m,11H,Aromatic protons, CH,NH), δ 10.12 (s,OH)
11b	δ1.53(s,CH_3), δ 1.54 (s,$COCH_3$), δ 1.8 (s,CH_2), δ 2.45 (s,CH), δ 3.73 (s,NH), δ 7.32–8.57 (m,11H, Aromatic protons, CH,NH,OH), δ 10.2 (s,Carboxylic OH)
11c	δ1.6(s,CH_3), δ 1.7 (s,$COCH_3$), δ 1.8 (s,CH_2), δ 2.5 (s,CH), δ 3.8 (s,NH), δ 7.3–8.6 (m,10H, Aromatic protons, CH,NH), δ 10.3 (s, OH)
12a	δ1.47(s,CH_3), δ 1.9 (s,CH_2), δ 2.6 (s,CH), δ 4.01(s,NH), δ 7.1–8.7 (m,11H, Aromatic protons, CH,NH), δ 10.01 (s, OH)
12b	δ1.46(s,CH_3), δ 1.95 (s,CH_2), δ 2.75 (s,CH), δ 4.3 (s,NH), δ 7.3–8.87 (m,11H, Aromatic protons, CH,NH,OH), δ 10.12 (s, Carboxylic OH)
12c	δ1.48(s,CH_3), δ 1.98 (s,CH_2), δ 2.8 (s,CH), δ 4.5 (s,NH), δ 7.4–8.89 (m,10H, Aromatic protons, CH,NH), δ 10.3 (s, OH)

TABLE II Selected HNMR Spectra Data for the New Compounds Listed in Table I *(Continued)*

Compound number	^{1}H-NMR d6 (DMSO)
13a	δ1.23(s,CH_3), δ 1.43(s,CH_2), δ 1.8 (s,CH), δ 3.5 (s,NH), δ 7.2–8.5 (m,11H, Aromatic protons, CH,NH), δ 10.2 (s,OH)
13b	δ1.3(s,CH_3), δ 1.45(s,CH_2), δ 1.9 (s,CH), δ 3.7 (s,NH), δ 7.5–8.7 (m,11H, Aromatic protons,NH,OH), δ 10. 24 (s, Carboxylic OH)
13c	δ1.42(s,CH_3), δ 1.48 (s,CH_2), δ 2.2 (s,CH), δ 3.85 (s,NH), δ 7.7–8.75 (m,10H, Aromatic protons, CH,NH), δ 10.26 (s,OH)
14a	δ1.25(s,CH_3), δ 1.6 (s,CH_2), δ 2.03 (s,CH), δ 3.6 (s,NH), δ 3.85 (s,NH), δ 7.23–8.47 (m,11H, Aromatic protons,CH,NH), δ 10.4 (s,OH)
14b	δ1.27(s,CH_3), δ 1.65 (s,CH_2), δ 2.12 (s,CH), δ 3.67 (s,NH), δ 3.89 (s,NH), δ 7.4–8.7 (m,11H, Aromatic protons, CH,NH,OH), δ 10.45 (s,Carboxylic OH)
14c	δ1.29(s,CH_3), δ 1.7 (s,CH_2), δ 2.3 (s,CH), δ 3.8 (s,NH), δ 3.9 (s,NH), δ 7.6–8.9 (m,10H, Aromatic protons, CH,NH), δ 10.6 (s,OH)
15a	δ1.05(s,CH_3), δ 1.5 (s,CH_2), δ 1.9 (s,CH), δ 3.5 (s,NH), δ 3.8 (s,NH), δ 7.4–8.6 (m,11H, Aromatic protons, CH,NH), δ 10.32 (s,OH)
15b	δ1.08(s,CH_3), δ 1.56 (s,CH_2), δ 1.95 (s,CH), δ 3.84 (s,NH), δ 4.2 (s,NH), δ 7.65–8.85 (m,11H, Aromatic protons, CH,NH,OH), δ 10.45 (s, Carboxylic OH)
15c	δ1.1(s,CH_3), δ 1.63 (s,CH_2), δ 1.98 (s,CH), δ 3.95 (s,NH), δ 4.5 (s,NH), δ 7.68–8.88 (m,10H, Aromatic protons,CH,NH), δ 10.5 (s,OH)
16a	δ1.2(s,CH_3), δ 1.85 (s,CH_3), δ 3.54 (s,NH), δ 7.34–8.23 (m,11H, Aromatic protons, CH,NH), δ 9.57 (s,OH)
16b	δ1.25(s,CH_3), δ 1.9 (s,CH_3), δ 3.98 (s,NH), δ 7.6–8.5 (m,10H, Aromatic protons, CH,NH), δ 9.7 (s,OH)
16c	δ1.4(s,CH_3), δ 2.3 (s,CH_3), δ 4.24 (s,NH), δ 7.7–8.5 (m,10H, Aromatic protons, CH,NH), δ 10.15 (s,OH)
17a	δ1.6(s,CH_3), δ 1.75 (s,CH_3), δ 2.8 (s,CH), δ 3.7 (s,NH), δ 7.43–8.45 (m,11H, Aromatic protons, CH, NH), δ 9.85 (s,OH)
17b	δ1.7(s,CH_3), δ 1.85 (s,CH_3), δ 2.9 (s,CH), δ 3.9 (s,NH), δ 7.5–8.6 (m,10H, Aromatic protons, CH, NH), δ 9.9 (s,OH)
17c	δ1.8(s,CH_3), δ 1.9 (s,CH_3), δ 2.98 (s,CH), δ 3.98 (s,NH), δ 7.6–8.634 (m,10H, Aromatic protons, CH, NH), δ 9.95 (s,OH)
18a	δ1.68(s,CH_3), δ 1.832 (s,CH_3), δ 2.04 (s,CH_2), δ 4.02 (s,NH), δ 7.45–8.23 (m,11H, Aromatic protons, CH,NH), δ 10.2 (s,OH)
18b	δ1.8(s,CH_3), δ 1.867 (s,CH_3), δ 2.34 (s,CH_2), δ 4.21 (s,NH), δ 7.5–8.3 (m,10H, Aromatic protons, CH,NH), δ 10.25 (s,OH)
18c	δ1.85(s,CH_3), δ 1.92 (s,CH_3), δ 2.5 (s,CH_2), δ 4.3 (s,NH), δ 7.6–8.5 (m,10H, Aromatic protons, CH,NH), δ 10.28 (s,OH)

presence of a few drops of piperidine was refluxed for 10–12 h. The filtrate was evaporated under reduced pressure, poured into ice water where the product was separated, filtered, washed, several times with water, and crystallized from ethanol and dimethylformamide (c.f., Table I).

TABLE III Selected IR Spectra, Elemental Analysis for the New Compounds Listed in Table I

Comp. no.	IR υ_{max}Cm $^{-1}$	Calculated % Found % C	H	N	S	Cl
4	1591 (2CO), 1665 (NHCO), 1727	55.65	4.28	8.89	9.85	—
	(COOH), 3069–3450 (NH,OH)	55.45	4.15	8.65	9.50	—
7a	1589 (2CO), 1628 (NHCO), 1665	62.54	3.82	8.39	6.40	—
	(COOH), 3061–3400 (NH,OH)	62.17	3.25	8.11	6.23	—
7b	1590 (2CO), 1630 (NHCO), 1667	62.32	3.82	8.39	6.40	—
	(COOH), 3063–3403 (NH,OH)	62.10	3.34	8.15	6.25	—
7c	1593 (2CO), 1632 (NHCO), 1670	58.58	3.80	9.32	7.12	—
	(COOH), 3063–3405 (NH,OH)	58.18	3.26	9.10	6.85	—
7d	1559 (2CO), 1638 (NHCO), 1673	60.29	4.64	11.72	6.71	—
	(COOH), 3065–3410 (NH,OH)	59.99	4.28	11.22	6.11	—
8a	1590 (2CO), 1631 (NHCO), 1667	58.28	3.49	7.28	5.56	6.14
	(COOH), 2921 (CHCl), 3066–3450 (NH,OH)	57.98	3.15	7.05	5.35	5.74
8b	1591 (2CO), 1632 (NHCO), 1668	58.28	3.49	7.28	5.56	6.14
	(COOH), 2922 (CHCl), 3066–3450 (NH,OH)	58.08	3.15	7.15	5.40	5.74
8c	1592 (2CO), 1632 (NHCO), 1670	54.38	3.23	7.93	6.05	6.69
	(COOH), 2923 (CHCl), 3067–3452 (NH,OH)	54.17	2.85	7.55	5.95	6.50
8d	1594 (2CO), 1634 (NHCO), 1670	56.36	4.18	10.11	5.79	6.40
	(COOH), 2925 (CHCl), 3070–3455 (NH,OH)	55.97	3.99	9.98	5.56	5.99
9a	1590 (2CO),1630 (NHCO), 1667	58.48	3.68	7.31	11.15	—
	(COOH), 3063–3445(NH,OH)	57.99	3.38	6.92	10.75	—
9b	1590 (2CO),1631 (NHCO), 1668	58.48	3.68	7.31	11.15	—
	(COOH), 3063–3445(NH,OH)	57.99	3.38	6.92	10.75	—
9c	1591 (2CO),1632 (NHCO), 1669	54.90	3.65	8.00	12.21	—
	(COOH), 3064–3447(NH,OH)	54.69	3.45	7.74	11.15	
9d	1592 (2CO),1632 (NHCO), 1670	56.56	4.38	10.15	11.61	—
	(COOH), 3065–3450(NH,OH)	56.24	3.99	9.85	11.41	—
10a	1588 (2CO),1662 (NHCO), 1723	63.64	4.18	6.45	7.39	—
	(COOH), 3064–3440 (NH,OH)	63.38	3.98	6.36	7.16	—
10b	1588 (2CO),1661 (NHCO), 1723	61.38	4.03	6.22	7.12	—
	(COOH), 3060–3440 (NH,OH)	61.08	3.94	5.85	6.86	—
10c	1589 (2CO),1663 (NHCO), 1725	59.66	3.58	8.77	6.69	—
	(COOH), 3066–3442 (NH,OH)	59.45	3.26	8.52	6.39	—
11a	1590 (2CO), 1685 (COCH3), 1730	61.16	4.52	11.41	6.53	—
	(COOH), 3060–3430 (NH,OH)	59.96	4.22	11.09	6.25	—
11b	1591 (2CO), 1686 (COCH3), 1731	59.23	4.37	11.05	6.32	—
	(COOH), 3063–3432 (NH,OH)	58.94	4.07	10.85	5.96	—
11c	1592 (2CO), 1688 (COCH3), 1733	56.08	3.95	13.08	5.99	—
	(COOH), 3063–3435 (NH,OH)	55.88	3.73	12.76	5.79	—
12a	1589 (2CO), 1727 (COOH), 3055–	66.45	4.61	10.69	6.12	—
	3460 (NH,OH)	66.15	4.51	10.44	5.85	—
12b	1589 (2CO), 1728 (COOH), 3050–	64.49	4.48	10.37	5.94	—
	3460 (NH,OH)	64.21	4.13	10.03	5.65	—
12c	1590 (2CO), 1729 (COOH), 3056–	61.20	4.08	12.31	5.63	—
	3465 (NH,OH)	60.96	3.78	12.11	5.33	—
13a	1588 (2CO), 1721 (COOH), 3060–	61.15	4.26	9.36	7.14	—
	3465 (NH,OH)	60.83	3.95	9.05	6.84	—

TABLE III Selected IR Spectra, Elemental Analysis for New Compounds Listed in Table I *(Continued)*

Comp. no.	IR υ_{max}Cm $^{-1}$	Calculated % Found %				
		C	H	N	S	Cl
13b	1589 (2CO), 1723 (COOH), 3063–3465 (NH,OH)	59.40 59.26	3.67 3.35	11.34 11.14	6.89 6.66	— —
13c	1589 (2CO), 1725 (COOH), 3060–3460 (NH,OH)	55.91 55.75	3.67 3.36	11.34 10.99	6.49 6.25	— —
14a	1592 (2CO),1667 (NHCO), 1732 (COOH), 3065–3435 (NH,OH)	60.55 60.32	4.23 3.84	11.77 11.52	6.73 6.44	— —
14b	1591 (2CO),1666 (NHCO), 1731 (COOH), 3064–3435 (NH,OH)	58.58 58.24	4.10 3.78	11.39 11.16	6.51 6.25	—
14c	1593 (2CO),1668 (NHCO), 1733 (COOH), 3067–3437 (NH,OH)	55.32 54.98	3.67 3.35	13.44 13.24	6.15 5.75	— —
15a	1589 (2CO),1245 (CS), 1729 (COOH), 3061–3451 (NH,OH)	58.70 58.46	4.10 3.83	11.41 11.24	13.06 12.96	— —
15b	1589 (2CO),1244 (CS), 1730 (COOH), 3062–3453 (NH,OH)	56.85 56.65	3.98 3.58	11.05 10.85	12.64 12.34	— —
15c	1590 (2CO),1242 (CS), 1731 (COOH), 3063–3454 (NH,OH)	53.67 53.47	3.57 3.27	13.04 12.82	11.94 11.76	— —
16a	1589 (2CO),1443(CN), 1667 (COOH), 3063–3405 (NH,OH)	62.75 62.32	4.55 4.36	9.98 9.65	7.61 7.36	— —
16b	1588 (2CO),1445(CN), 1669 (COOH), 3063–3410 (NH,OH)	58.06 57.87	3.99 3.74	9.23 8.99	7.04 6.82	7.79 7.24
16c	1592 (2CO),1446(CN), 1670 (COOH), 3065–3415 (NH,OH)	56.69 56.48	3.89 3.63	12.56 11.89	6.88 6.45	— —
17a	1591 (2CO), 1668 (COOH), 1720 (CO), 2923 (CHCl), 3066–3450 (NH,OH)	57.99 57.76	4.06 3.79	8.45 8.16	6.44 6.25	7.14 6.95
17b	1593 (2CO), 1670 (COOH), 1722 (CO), 2925 (CHCl), 3068–3455 (NH,OH)	54.28 53.97	3.61 3.35	7.91 7.63	6.04 5.85	13.35 13.15
17c	1594 (2CO), 1671 (COOH), 1725 (CO), 2925 (CHCl), 3068–3460 (NH,OH)	53.18 52.88	3.53 3.35	10.34 10.15	5.91 5.71	6.54 6.24
18a	1589 (2CO), 1676 (COOH,CO), 3060–3443 (NH,OH)	58.22 57.88	4.28 3.96	8.49 8.29	12.95 12.66	— —
18b	1592 (2CO), 1678 (COOH,CO), 3065–3445 (NH,OH)	54.49 54.17	3.81 3.61	7.94 7.65	12.12 11.82	6.70 6.36
18c	1594 (2CO), 1679 (COOH,CO), 3066–3446 (NH,OH)	53.37 53.16	3.55 3.25	10.37 10.16	11.87 11.58	— —

Synthesis of New Thiazolidinone Derivatives 9a–d

A solution of **7a–d** (5.01 g, 0.01 mol) and thioglycolic acid (0.92 g, 0.01 mol) in a mixture of ethanol and dimethylformamide in the presence of a few drops of piperidine was refluxed for 8–10 h. The filtrate was evaporated under reduced pressure, poured into ice water where the product was separated, filtered, washed several times with water, and crystallized from the proper solvent to give **9a–d** (c.f., Table I).

Synthesis of 2-Arylidenoamino-2-methyl, 2-H-naphtho[2, 3-b][1, 4]thiazine-5, 10-dione, 3-carboxylic Acid Derivatives 10a–c

A solution of **4** (3.46 g, 0.01 mol) and aromatic aldehydes (1.06 g, 1.22 g, 1.51 g, 0.01 mol) in a mixture of ethanol and dimethylformamide in the presence of two drops of piperidine as catalyst was refluxed for 7–8 h. The mixture was concentrated and then left to cool and pour onto ice water acidified by concentrated hydrochloric acid. The solid product so formed was collected by filtration and crystallized from the proper solvent to give **10a–c** (c.f., Table I).

Synthesis of Acetyl Pyrazolino Derivatives 11a–c

To a solution of arylidino **10a–c** (4.34 g, 4.50 g, 4.79 g, 0.01 mol) in a mixture of ethanol and dimethylformamide as solvent, hydrazine hydrate (0.5 g, 0.01) was added followed by glacial acetic acid (10 ml). The reaction mixture was refluxed for 10–12 h. The reaction mixture was concentrated and filtered. These substances were triturated with water and the precipitates were separated, filtered, washed several times with water, dried, collected, and crystallized from the proper solvent to give **11a–c** (c.f., Table I).

Synthesis of 3-aryl-4-phenylpyrazolino Derivatives 12a–c

To a solution of arylidino **10a–c** (4.34 g, 4.50 g, 4.79 g, 0.01 mol) in a mixture of ethanol and dimethylformamide as solvent, added phenyl hydrazine (1.08 g, 0.01 mol) in the presence of 0.5 ml of piperidine as catalyst. The reaction mixture was refluxed for 9–11 h. The reaction mixture was concentrated, cooled, poured onto water acidified by concentrated hydrochloric acid. The solid substances were collected by filtration, washed with water for several times, and crystallized from the proper solvent to give **12a–c** (c.f., Table I).

Synthesis of 3-arylisoxazolo Derivatives 13a–c

A solution of arylidino **10a–c** (4.34 g, 4.50 g, 4.79 g, 0.01 mol) was refluxed with hydroxylamine hydrochloride (0.69 g, 0.01 mol) in the presence of sodium hydroxide as catalyst and a mixture of ethanol and dimethylformamide as solvent for 6–8 h. The reaction mixture was filtered hot, the filtrate was concentrated and poured onto ice water, where the products were separated, filtered, washed several times with water, and crystallized from the proper solvent to give **13a–c** (c.f., Table I).

Synthesis of 3-arylpyrimidino Derivatives 14a–c

A solution of arylidino **10a–c** (4.34 g, 4.50 g, 4.79 g, 0.01 mol) was refluxed with urea (0.6 g, 0.01 mol) in the presence of sodium hydroxide as catalyst and a mixture of ethanol and dimethylformamide as solvent for 8–10 h. The reaction mixture was filtered from unreacted materials, and the filtrate was concentrated and poured on to ice water, and the products were separated, filtered, washed several times with water, and crystallized from the proper solvent to give **14a–c** (c.f., Table I).

Synthesis of 3-arylthiopyrimidino Derivatives 15a–c

A solution of arylidino **10a–c** (4.34 g, 4.50 g, 4.79 g, 0.01 mol) was refluxed with thiourea (0.67 g, 0.01 mol) in the presence of hydrochloric acid as catalyst and a mixture of ethanol and dimethylformamide as solvent for 8–10 h. The reaction mixture was filtered hot, cooled, the filtrate was concentrated, poured onto ice/water, and the products were separated, filtered, washed several times with water, and crystallized from the proper solvent to give **15a–c** (c.f., Table I).

Synthesis of New Schiff Base Derivatives 16a–c

Compound **4** (3.46 g, 0.01 mol) and aromatic amine (0.93 g, 1.27 g, 1.38 g, 0.01 mol) in an equimolar ratio were dissolved in ethanol and dimethylformamide and 0.5 ml of piperidine as catalyst was added. The mixture was refluxed about 10 h. The reaction mixture was allowed to cool at room temperature, then filtered, washed several times with water, dried, collected, and crystallized from the proper solvent to give **16a–c** (c.f., Table I).

Synthesis of New β-Lactam Derivatives 17a–c

A solution of (4.21 g, 4.55 g, 4.66 g, 0.01 mol) of chloroacetylchloride (1.13 g, 0.01 mol) in a mixture of ethanol and dimethylformamide in the presence of a few drops of piperidine was refluxed for 12–14 h. The filtrate was evaporated under reduced pressure, poured into ice/water where the product was separated, filtered, washed several times with water and crystallized from the proper solvent to give **17a–c** (c.f., Table I).

Synthesis of New Thiazolidinone Derivatives 18a–c

A solution of **16a–c** (4.21 g, 4.55 g, 4.66 g, 0.01 mol) and thioglycolic acid (0.92 g, 0.01 mol) in a mixture of ethanol and dimethylformamide

in the presence of a few drops of piperidine was refluxed for 11–13 h. The filtrate was evaporated under reduced pressure, poured into ice/water where the product was separated, filtered, washed several times with water, and crystallized from the proper solvent to give **18a–c** (c.f. Table I).

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