REACTIONS OF ACENAPHTHENEQUINONE DERIVATIVES WITH SOME AROMATIC AND ALIPHATIC AMINES

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Reaction of acenaphthenequinone and aceanthrenequinone (**1a,b**) with diaminomaleonitrile at reflux temperature gave acenatho[1,2-b]pyrazine-8,9-dicarbonitrile and aceanthryleno[1,2-b]pyrazine-10-11-dicarbonitrile (**2a,b**), respectively. The reaction of **2a,b** with hydrazine hydrate afforded the corresponding cyclic products, 8,11-diaminoacenatho[1,2-b]pyrazino[2,3-d]pyridazine and 10,13-diaminoaceanthryleno[1,2b]pyrazino[2,3-d]pyridazine (**3a,b**). The reaction of **1a,b** with *p*-bromoaniline in presence of $ZnCl_2$ afforded complexes bis(*p*-bromophenylimino)acenaphthene and -aceanthrene (**7a,b**). We have also described the synthesis of spiro[2H-aceanthrene-2,2'-thiazolidine]-1,4'-dione derivatives (**8a,b**). Reaction of **1b** with 1-amino-3-(N,N-dimethylamino)propane, benzylhydrazine and *p*-bromophenylhydrazine has been investigated for studying the utility of products as pharmacological agents. Chemical and spectroscopic evidences for the structures of the new compounds are presented.

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INTRODUCTION

Recent studies have shown that acenaphthenequinone and its derivatives exhibit various biological activities,[1-6] such as bactericidal, antihypoxic, fungicidal, and are useful as phospholipase A2 inhibitors.[7] Acenaphthenequinones hydrogensulfite had a narcotic effect on mice and it inhibited the growth of transplanted tumours.[3] In addition, the condensation product of acenaphthenequinone with 2,3-diaminopyrazine has been used to provoke ataxia by lowering central nervous system activity.[6] In the literature, there is an abundance of reports dealing with the chemistry of acenaphthenequinone, but very little is known about the reaction of benzoacenaphthenequinone (aceanthrene-quinone) and its derivatives. Moreover, aceanthrenequinone derivatives have been extensively utilized as intermediate for the synthesis of fused aceanthrenes of potential biological activity.[8,9] In view of these findings and our interest in the synthetic potential of fused nitrogen heterocyclic compounds,[10,11] we have studied the synthesis of some differently fused acenaphthene and aceanthrene derivatives for studying their utility as pharmacological agents.

EXPERIMENTAL

General

Melting points were measured on a Kofler hot stage microscope (Reichert, Vienna) and are uncorrected. ^1H and ^{13}C NMR spectra were recorded on a Bruker DPX 200 spectrometer at 200 MHZ (^1H) and 50 MHZ (^{13}C). Chemical shifts are given in δ units relative to internal TMS at 295 K. IR spectra were obtained on a Biorard FT-IR-45 instrument. All experiments were carried out with exclusion of moisture. For all newly synthesized compounds satisfactory elemental analyses were obtained.

Reaction of 1,2-diketones 1a,b with diaminomaleonitrile (General Method)

A mixture of **1a** or **1b** (1 mmol) and 1 mmol of diaminomaleonitrile in 50 mL acetic acid was heated under reflux for 3 h. The solvent was reduced under reduced pressure and the solid product obtained was filtered off and recrystallized from suitable solvent to give the corresponding condensed products **2a** or **2b** respectively.

Acenaphtho[1,2-b]pyrazine-8,9-dicarbonitrile, 2a.

Prepared from 0.25 g acenaphthenequinone (1 mmol); crystallization from DMF / H_2O gave red crystals; Yield (88 %); m.p.: 238 °C; IR (KBr): 3065, 2238, 1614, 1488, 1421 cm^{-1} ; ^{1}HNMR ($CDCl_3$): δ = 7.93-8.01 (t,$2H_{ar}$), 8.31-8.35 (d, $2H_{ar}$), 8.52-8.56 (d, $2H_{ar}$) ppm.

Aceanthryleno[1,2-b]pyrazine-10,11-dicarbonitrile,2b.

Prepared from 0.26 g aceanthrenequinone **1** (1 mmol); crystallization from benzene gave brown crystals; Yield (85 %); m.p.:322 °C; IR (KBr) :3065, 2234, 1625, 1577, 1521, 1431 cm^{-1} ; ^{1}HNMR ($CDCl_3$) : δ = 7.77-7.79 (t, $1H_{ar}$), 7.96-7.98 (m, $2H_{ar}$), 8.28-8.32 (d, $1H_{ar}$), 8.48-8.52(d, $1H_{ar}$), 8.66-8.69 (d, $1H_{ar}$), 8.97 (d, $1H_{ar}$), 9.34-9.38 (d, $1H_{ar}$) ppm.

Reaction of 1,2-dicarbonitriles 2a,b with hydrazine hydrate (General Method)

A mixture of **2a** or **2b** (1 mmol) and 1.5 mmol of hydrazine hydrate in 50 mL toluene was heated under reflux for 3h. The solvent was evaporated under reduced pressure

and the solid product obtained was filtered off and recrystallized from suitable solvent to give the corresponding condensed products **3a** or **3b** respectively.

8,11-Diaminoacenatho[1,2-b]pyrazino[2,3-d]pyridazine, 3a.

Prepared from acenatho[1,2-b]pyrazine-8,9-dicarbonitrile (1 mmol); crystallization from ethanol gave dark red crystals; Yield (72 %); m.p.: 289 °C; U.V.(DMSO): λ 316, 448 nm. IR (KBr): 3439, 3269, 3115, 1666, 1608, 1466, 1456 cm^{-1} ; ^{1}HNMR (DMSO): δ = 6.21 (br, $2NH_2$), 7.99-8.05 (t, $2H_{ar}$), 8.39-8.42 (d, $2H_{ar}$), 8.49-8.52 (d, $2H_{ar}$) ppm.

10,13-Diaminoaceanthryleno[1,2-b]pyrazino[2,3-d]pyrazidine, 3b.

Prepared from aceanthryleno[1,2-b]pyrazine-10,11-dicarbonitrile (1 mmol); crystallization from acetic acid gave dark brown crystals; Yield (75 %); m.p.: 349 °C; IR (KBr): 3442, 3367, 3118, 1661, 1624, 1577, 1541, 1489 cm^{-1}; ^{1}HNMR ($CDCl_3$): δ = 6.11 (br, $2NH_2$), 7.56-7.70 (m, $3H_{ar}$), 7.72-7.92 (d, $1H_{ar}$), 8.05-8.14 (m, $2H_{ar}$), 8.52 (s, $1H_{ar}$), 9.07-9.10 (d, $1H_{ar}$) ppm.

8,11-Diacetamidoacenatho[1,2-b]pyrazino[2,3-d]pyridazine, 4.

A mixture of 8,11-diaminoacenatho[1,2-b]pyrazino[2,3-d]pyrazine (1 mmol) and 20 ml acetic anhydride was refluxed for 3 h. After cooling, the precipitate was filtered to give **4**. Crystallization from acetic acid gave dark brown crystals: Yield (75 %); m.p.: 349 °C; IR (KBr): 3367, 3017, 1672, 1610, 1542, 1487 cm^{-1}; ^{1}HNMR (DMSO): δ = 2.37 (s, $2CH_3$), 8.05 (t, $2H_{ar}$), 8.45 (d, $2H_{ar}$), 8.60 (d, $2H_{ar}$), 10.50 (br, 2NH) ppm.

Bis(*p*-bromophenylimino)acenaphthene and aceanthrene, 5a,b.

(i) A mixture of **1a** or **1b** (5 mmol), 0.86 g anhydrous ZnCl2 (6 mmol) and 2.25 g of p-bromoaniline (12 mmol) in 30 mL acetic acid was heated under reflux for 1h. The suspension was cooled to 20 °C and the solid filtered off. The product was washed with acetic acid and diethyl ether and air dried, to give the complexes **5a** or **5b** respectively, as an orange solids (95 %). Compound **5a**, 1HNMR ($CDCl_3$): δ = 7.47-7.73 (m, $12H_{ar}$), 8.18 (d, $2H_{ar}$) ppm. Compound **5b**, 1HNMR ($CDCl_3$): δ = 6.96 (m, $4H_{ar}$), 7.37-7.85 (m, $8H_{ar}$), 8.01-8.33 (m, $4H_{ar}$), 8.74 (s, $1H_{ar}$), 9.23 (d, $1H_{ar}$) ppm.

(ii) Compound **5a** or **5b** (6.4 mmol) was added to a solution of 25 g K_2CO_3 in 25 mL water and the mixture was heated at reflux with vigorous stirring. After 2 h the mixture was cooled to 20 °C, the solid product filtered off and washed with water (5 x 30 mL). The product was extracted with boiling ethanol (200 mL), until the ethanol extracts were almost colorless. The combined ethanol extracts were evaporated to 120 mL and set aside at -20 °C. After one day the product was filtered and dried in vacuo, to give compounds **6a,b**.

Bis(*p*-bromophenylimino)acenaphthene, 6a.

Yield (62 %); m.p.: 311 °C; IR (KBr): 3092, 1658, 1637, 1579, 1485, 1459, cm^{-1}. ^{1}HNMR ($CDCl_3$): δ = 6.95 (m, $3H_{ar}$), 7.55-7.59 (m, $4H_{ar}$), 7.84 (m, $2H_{ar}$), 8.03-8.28 (m, $5H_{ar}$) ppm.

Bis(*p*-bromophenylimino)aceanthrene, 6b.

Yield (62 %); m.p.: 298 °C; IR (KBr): 3090, 1665, 1637, 1565, 1487, 1461 cm^{-1}. ^{1}HNMR ($CDCl_3$): δ = 6.92 (d, $2H_{ar}$), 7.01 (d, $2H_{ar}$), 7.56–8.31 (m, $10H_{ar}$), 8.75 (d, $1H_{ar}$), 9.12(d, $1H_{ar}$) ppm.

3'-Arylspiro[2H-aceanthrene-2,2'-thiazolidine]-1,4'-diones, 7a,b.

A mixture of 0.10 g **1b** (0.4 mmol) and 0.4 mmol of the aromatic amine namely, aniline, *p*-bromoaniline or *p*-chloroaniline was dissolved in 50 mL of benzene. The reaction mixture was heated under reflux for 5 h in presence of 1.0 mL acetic acid. The solvent was evaporated under reduced pressure and 0.05 g mercaptoacetic acid (0.5 mmol) was added to the residue dissolved in 50 mL of benzene. The reaction mixture was refluxed until no more water was collected in a Dean-Stark separator. The solvent was evaporated in vacuo and the yellowish solid obtained was filtered off and recrystallized from suitable solvent to give the corresponding condensed products **7a** and **7b** respectively.

3'-Phenylspiro[2H-aceanthrene-2,2'-thiazolidine]-1,4'-diones (**7a**) was crystallized from toluene to give yellowish crystals; Yield (62 %); m.p.: 298 °C; IR (KBr): 3090, 2933, 1699-1680, 1627, 1579, 1485, 1459 cm^{-1} . ^{1}HNMR ($CDCl_3$): δ = 3.85 (d, $1H_{ar}$), 4.38 (d, 1H), 7.15 (d, $2H_{ar}$), 7.33-7.74 (m, $7H_{ar}$), 7.94 (d, $1H_{ar}$), 8.15 (d, $1H_{ar}$), 8.65 (s, $1H_{ar}$), 9.08 (d, $1H_{ar}$) ppm.

3'-(p-Bromophenyl)spiro[2H-aceanthrene-2,2'-thiazolidine]-1,4'-diones (**7b**) was crystallized from toluene to give yellowish crystals; Yield (45 %); m.p.: >300 °C; IR (KBr): 3050, 2980, 1690-1680, 1618, 1580, 1485, cm^{-1} . ^{1}HNMR ($CDCl_3$): δ = 3.90 (d, 1H), 4.45 (d, 1H), 6.90 (d, $2H_{ar}$), 7.17 (d, $2H_{ar}$), 7.62-7.68 (m, $3H_{ar}$), 7.78 (t, $1H_{ar}$), 7.96 (d, $1H_{ar}$), 8.15 (d, $1H_{ar}$), 8.70 (s, $1H_{ar}$), 9.07 (d, $1H_{ar}$) ppm.

2-(3-Dimethyamino-propylimino)-2H-aceanthrylen-1-one, 9.

A mixture of 0.10 g **1b** (0.4 mmol) and 0.5 mmol of 1-amino-3-(N,N-dimethylamino)propane in 50 mL benzene was heated under reflux for 3 h until no more water was collected in a Dean-Stark separator. The solvent was evaporator under reduced pressure and the yellow solid obtained was filtered off and purified from methanol/$CHCl_3$ to give the corresponding condensed products **9**. Yield (35 %); m.p.: >250 °C; IR (KBr): 3046, 2932, 1710, 1665, 1625, 1585, 1532, 1489 cm^{-1} . ^{1}HNMR (CD_3COCD_3): δ = 1.83 (q, CH_2), 2.19 (s, $2CH_3$), 2.35 (t, CH_2), 4.41 (t, CH_2), 7.55-7.74 (m, $3H_{ar}$), 8.01(d, $1H_{ar}$), 8.28 (d, $1H_{ar}$), 8.52 (d, $1H_{ar}$), 8.77 (s, $1H_{ar}$), 9.78 (d, $1H_{ar}$) ppm. ^{13}C NMR (CD_3COCD_3): 35.50 (CH_2), 48.16 (CH_2), 54.36($2CH_3$), 66.80 (CH_2), 124.36,

133.46, 134.90, 135.44, 135.69, 136.40, 137.38, 137.93, 139.14, 140.37, 141.74, 142.36, 144.55, 145.83(aryl), 172.39 (C=N), 174.12 (C=O) ppm.

Reaction of aceanthrenequinone 1 with aroyl hydrazine derivatives

A mixture of 0.10 g aceanthrenequinone (0.4 mmol) and 0.4 mmol aroyl hydrazine derivatives namely, benzoyl hydrazine, p-methylbenzoyl hydrazine, p-bromobenzoyl hydrazine or m-chlorobenzoyl hydrazine was refluxed for 3 h in 30 mL of dry methanol and 1 mL acetic acid. After cooling, the precipitate was filtered to give **10a-d**.

Compound **10a** was crystallized from benzene to give orange crystals; Yield 2.28 g (65 %); m.p.: >250 °C; IR (KBr): 3239, 3046, 1710, 1695, 1625, 1612, 1585, 1522, 1489 cm^{-1}. Elemental analysis for $C_{23}H_{14}N_2O_2$ (350.38), Calcd. C, 78.84; H, 4.02; N, 7.99; found C, 78.70; H, 4.20; N, 7.80.

Compound **10b** was crystallized from methanol to give orange crystals; Yield 2.18 g (60 %); m.p.: >265 °C; IR (KBr): 3242, 3046, 2943, 1710, 1693, 1628, 1610, 1585, 1522, 1482 cm^{-1}. Elemental analysis for $C_{24}H_{16}N_2O_2$ (364.40), Calcd. C, 79.11; H, 4.42; N, 7.69: found C, 79.00; H, 4.50; N, 7.80 .

Compound **11c** was crystallized from toluene to give orange crystals; Yield 3.86 g (90 %); m.p.: 240 °C; IR (KBr): 3239, 3046, 1710, 1695, 1625, 1612, 1585, 1522, 1489 cm^{-1}. ^{1}HNMR ($CDCl_3$): δ = 7.65–7.71(m, 5Har + NH), 7.80 (m, $2H_{ar}$), 8.00 (d, $2H_{ar}$), 8.10 (d, $1H_{ar}$), 8.20 (d, $1H_{ar}$), 8.80 (s, $1H_{ar}$), 9.12(d, $1H_{ar}$) ppm. The mass spectrum showed molecular ion peak at m/e 430 [M^+ + 1], 245 [M^+ - C_6H_4Br], base peak at 217 [M^+ -COC_6H_4Br] [100 %], 189 [M^+ -$N_2COC_6H_4Br$], 157, 139, 104 and 75. Elemental analysis for $C_{23}H_{13}BrN_2O_2$ (429.27), Calcd. C, 64.25; H, 3.05; N, 6.53, found C, 64.40; H, 3.00; N, 6.40.

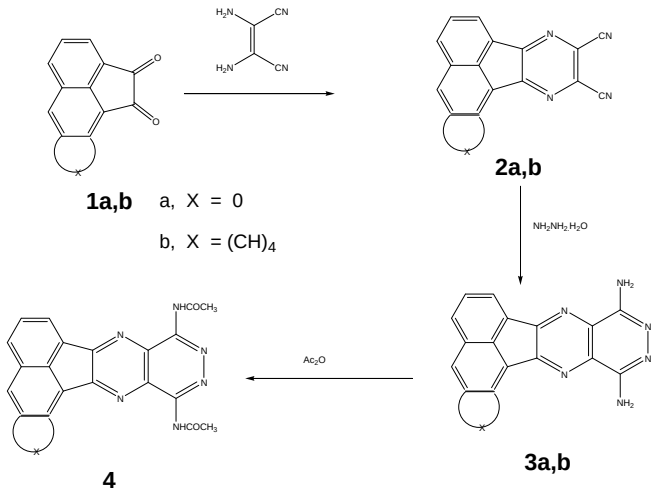
Compound **11d** on crystallization from benzene gave red yellow crystals; Yield 2.89 g (75 %); m.p.: 238 °C; IR (KBr): 3239, 3046, 1710, 1695, 1625, 1612, 1585, 1522, 1489 cm^{-1}. ^{1}HNMR ($CDCl_3$): δ = 7.52–8.11(m, $9H_{ar}$ + NH), 8.20 (d, $1H_{ar}$), 8.80 (s, $1H_{ar}$), 9.12 (d, $1H_{ar}$) ppm. The mass spectrum showed molecular ion peak at m/e 384 [M^+], 247 [M^+-C_6H_4-Cl], base peak at 217 [M^+-COC6H4Cl] [100 %], 189 [M^+-N_2COC6H4-Cl], 139, 101 and 75. Elemental analysis for $C_{23}H_{13}ClN_2O_2$ (384.82), Calcd. C, 71.79; H, 3.40; N, 7.28; found C, 71.90; H, 3.30; N, 7.50.

Reaction of 10b with malononitrile

A mixture of hydrazone **10b** (1mmol) and malononitrile (1 mmol) in 30 mL acetic acid was heated under reflux for 1 h. The solid formed during heating was filtered off and recrystallized from aqueous DMF to give the condensed product **11;** Yield (80 %); m.p.: 340 °C; IR (KBr): 3360, 3240, 3055, 2266, 2193, 1689, 1665, 1589, 1520 cm^{-1}. ^{1}HNMR ($CDCl_3$): δ = 7.63-7.72(m, $4H_{ar}$), 7.78-7.83 (t, $1H_{ar}$), 7.94-7.96 (d, $2H_{ar}$), 8.05-8.07 (d, 2H), 8.10-8.18 (d, 1Har), 8.77(s, $1H_{ar}$) , 9.04-9.06 (d, 2Har), 14.50(s, 1H, NH) ppm.^{13}CNMR ($CDCl_3$): δ = 108, 119.50, 123.76, 124.22, 127.02, 127.46, 127.81, 128.02, 128.50, 129.25, 129.47, 129.89, 130.45, 131.19, 132.27, 133.19, 134.90, 142.56, 160.20, 190.10 (aryl and C=O) ppm.

RESULTS AND DISCUSSION

The condensation of the 1,2-diketones with aliphatic diamine was carried out in the manner described by Chiodini[12] to give fused pyrazine derivatives in good yield. Based on these results, it was found that treatment of acenaphthenequinone and aceanthrenequinone (**1a,b)** with diaminomalenonitrile at reflux temperature gave acenaphtho[1,2-b]-pyrazine-8,9-dicarbonitrile and aceanthryleno[1,2-b]pyrazine-10,11-dicarbonitrile (**2a,b),** respectively. The reaction of **2a,b** with hydrazine hydrate afforded the corresponding cyclic product namely, 8,11-diamino-acenatho[1,2-b]pyrazino[2,3-d]pyridazine and 10, 13-diamino-aceanthryleno[1,2-b]pyrazino[2,3-d]pyridazine (**3a,b**). Treatment of **3a** with acetic anhydride under reflux temperature readily afforded 8,11-diacetamido-acenaphtho[1,2-b]pyrazino[2,3-d]pyridazine (**4)** (Scheme 1).



Scheme 1

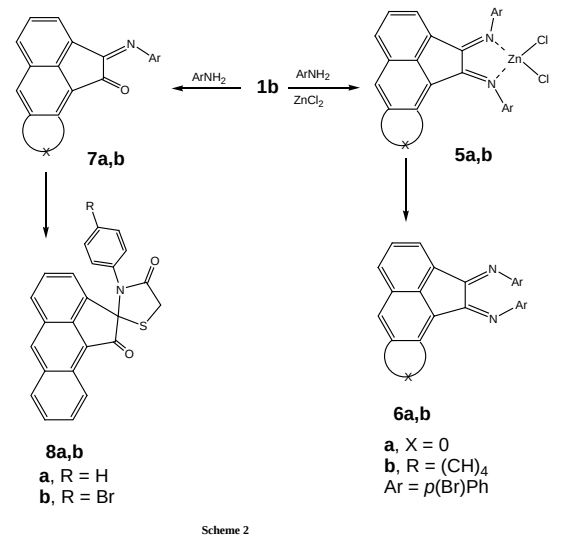
We have been interested in the complexation properties of bis(arylimino)-acenaphthene and aceanthrene (**6a,b**) due to the presence of two exocyclicimine functionalities, which are not being part of a heteroaromatic ring system, is expected to lead to better O-donating and better π-accepting properties as compared with bipyridyl. Also, the rigid acenaphthene and aceanthere backbone prevents rotation around the imine carbon-carbon bond and as a consequence both imine-N atoms remain in a fixed *cis* orientation favouring chelating coordination to a metal center.

Compounds **6a,b** could not be obtained directly by the reaction of 1,2-diketones **1a,b** with *p*-bromoaniline under different conditions. In all cases reaction occurred to give monoimino compounds with mixtures of several compounds were formed, which were not further investigated. Compounds **5a,b** were synthesized form **1a,b** and *p*-bromoaniline in boiling acetic acid and zinc chloride. Compounds **6a,b** were synthesized from 5**a,b** by replacing the zinc chloride with aqueous potassium carbonate.[13]

Table 1. Antimicrobial activity of some synthesized compounds

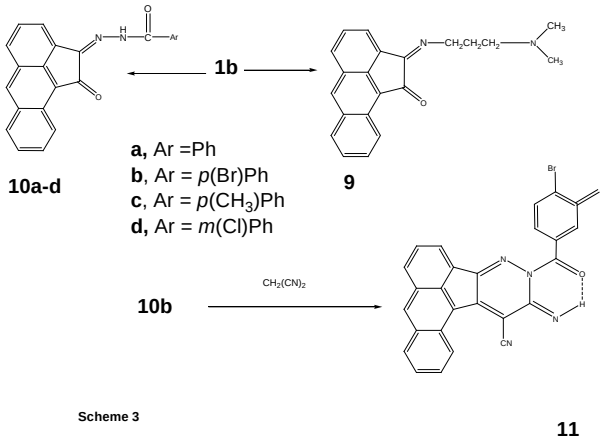
Compd. No	Zone of inhibition				
	Sarcina Lutea	***B. Megaterium***	***B. Cerius***	***B. Subtilis***	***Pseudomonas Aeruginosa***
6a	10	6	-	-	5
6b	10	5	-	-	5
8a	13	10	10	10	15
8b	16	12	10	10	15
9	15	8	-	-	13

A recent observation by Diurno and co-workers[14,15] that spirothiazolidinone derivatives have antimicrobial and antifungal activities prompted us to synthesize 3'-aryl-spiro[2H-aceanthrene-2,2'-thiazolidine]-1,4'-diones (**8a,b)** via the Schiff-bases of aceanthrenequinone with arylamines, followed by cyclization with mercaptoacetic acid in refluxing benzene with removal of water from the reaction mixture. The chemical structure of compound **8b** was confirmed by ^{1}H-COSY spectroscopy (Scheme 2). The structure of all newly synthesized compounds was confirmed by their elemental and spectroscopic data.



Scheme 2

Treatment of **1b** with 1-amino-3-(*N*,*N*-dimethylamino)propane under reflux afforded *2*-(imino-N,N-dimethylpropylamine)aceanthrene **(9)**.



Scheme 3

Furthermore, condensation reaction of aceanthrene-quinone **1b** with aroyl hydrazine derivatives namely, benzoyl hydrazine, *p*-toluoyl hydrazine, *p*-bromobenzyoyl hydrazine and m-chlorobenzoyl hydrazine afforded the corresponding hydrazone derivatives **10a-d** respectively. Finally, our study was extended to prepare the aceanthryleno[1,2-c]-pyridazine derivative **11** by reaction of hydrazone **10b** with malononitrile to give the corresponding product (Scheme 3).

Screening for antimicrobial activities

The antimicrobial activity of some of the prepared compounds was determined by cup-plate technique (BPC, 1963) using Cork borer for making wells in agar plates. The sample of the compounds **7-10** were dissolved in DMF (20 % conc.). 0.1 cm^3 of each sample was used for some Gram-positive (*Sarina lutea*, *Bacillus Megaterium*, *Bacillus cerius* and *Bacillus subtilis*) and Gram-negative (*Pseudomonas Aeruginosa*) bacteria under aseptic conditions. The medium for cultivation of the test organisms was nutrient agar (APHA, 1985). Bacteria were incubated at 30 °C for 24 h and the diameters of the inhibition zones were measured in mm. Compounds **7-10** showed antimicrobial activity against both Gram-Positive and Gram-negative bacteria as shown in Table 1**.**

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