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Synthesis And Antimicrobial Activity Of N-{4-[3-(4-Chlorophenyl)-Ethylene Oxide-Carbonyl]-Phenyl}-2-(1,1,3-Trioxo-1,3-Dihydro-Benzo[D]Isothiazol-2-Yl)-Acetamide

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ABSTRACT

Epoxide (2) was used to synthesize a number of new condensed and noncondensed heterocyclic systems. Thus, reaction of epoxide (2) with amines, hydrazines, active methylene compounds and Friedel-Craft's have been studied. On the other hand, epoxide (2) reacted with Grignard reagents and afforded (7a,b). However, epoxide (2) condensed with thiourea and glycine gave oxazolinethione and 2-morpholinone derivatives (8) and (9), respectively. Also, epoxide (2) reacted with acetic acid and gave (10). Action of different nitriles with epoxide (2) gave the hydroxy amide derivatives (11a-c), which underwent cyclization and afforeded 2-oxazoline derivatives (12a-c).

KEYWORDS

Epoxide, isothiazole, acetamide, furanone, morphilinone, oxazolinethione

INTRODUCTION

The reported pharmaceutical properties[1-4] analgesic agents, enzyme inhibitors and anxiolytic agent of 1, 1, 3-trioxo-1,3-dihydro-benzo [d] isothiazole and its derivatives promoted our interest for the synthesis of title and its derivatives and a variety of stablized carbanions have been widely used for the intera molecular ring opining of 1,2-epoxides. Most commonly these carbanions are stabilized by adjacent electron-withdrawing groups (EWGs) such as cyano, sulfonyl or sulfur-containing groups[5]. When, the EWGs is a carbonyl, some ambiguity may result from the presence of two nucleophilic sites (C and O) which may intra moleculary displace the oxirane ring leading to the corresponding C- or O- alkylation products, respectively.

Our program is study the effect of bulky heteryl moiety at the β-position on the behaviour of the oxirane ring towards different nitrogen and carbon nucleophiles and its behaviour towards the expected biological activity of some synthesized compounds. Treatment of an alcoholic solution of chalcone (**1**) with hydrogen peroxide in alkaline medium yielded N-{4-[3-(4-chlorophenyl)-ethylene oxide-carbonyl]-phenyl}-2-(1,1,3-trioxo-1,3-dihydro-benzo[d]isothiazol-2-yl)-acetamide (**2**). The IR spectrum of **2** showed absorption bands at 1750-1730 cm^{-1}(ν CO, cyclic imide), at 1680 cm^{-1}(ν CO, ketone), at 1660 cm^{-1}(ν CO, amide), at 1320 and 1120 cm^{-1} (ν SO_2), at 1070 cm^{-1}(ν C-O-C) and 3150 cm^{-1}(ν NH). The mass spectrum of **2**

showed peaks at m/z (relative intensity %) 496.5 (0.20)(M^+), 372(0.50), 371(0.11), 374(0.17), 315(2.15), 196(100.00), 183(2.60), 168(6.70), 141(2.95), 140(8.30), 99(41.10), 91(30.40), 77(36.50), 75(13.50), 65(21.20) and 57(44.30).

It has been reported [6] that, the oxirane ring of α, β-epoxy- β -aroylacrylic acid was opened by nucleophilic reagents. In the present investigation, the epoxide **(2)** was allowed to react with p-bromoaniline and benzylamine in boiling n-butanol and yielded N-{4-[3-(4-chlorophenyl)-α-arylamino- β -hydroxy-carbonyl-ethylene oxide]-phenyl}-2-(1,1,3-trioxo-1,3-dihydro-benzo [d] isothiazol-2-yl)-acetamide **(3a,b)**.The structure of **3a**,**b** was proved by: (i) IR spectrum, which showed absorption, bands at 3450-3350 cm^{-1} (ν OH) and disappear (ν C-O-C). (ii) ^1H NMR spectrum of **3a** showed signals at δ (ppm). 3.1(d, 1H,α-CH), 3.3 (d, 1H,β-CH), 4.5 (s, 2H, NCH_2CO), 7.8 (m, 16H, Ar-H), 10.2 (d, 2H, NH) and 11.2(s, 1H, OH). Recently[7], it has been shown that, the oxirane ring of α, β -epoxy ketone was opened by action of hydrazines. Thus, the epoxide **(2)** was reacted with hydrazine hydrate and phenyl hydrazine and afforded N-{4-[5-(4-chlorophenyl)-4-hydroxy-4,5-dihydro(1H or 1-phenyl)pyrazoline-3-yl]-phe-nyl}-2-(1,1,3-trioxo-1,3-dihydro benzo[d]isothiazol-2-yl)acetamide **(4a,b)**.The structure of **4a** and **4b** was supported by: (i) IR spectrum, which revealed absorption bands at 1740-1730 cm^{-1} (ν CO of cyclic imide), 1320 and 1130 cm^{-1} (ν SO_2), 1670 cm^{-1} (ν CO of amide), 1630 cm^{-1} (ν C=N) and 3200 cm^{-1} (ν NH). (ii) ^1H NMR spectrum of **4b** showed signals at δ (ppm). 2.6 –2.8 (d, 2H, 2CH cyclic), 4.6 (s, 2H, NCH_2CO), 5.9 (s, 1H, OH), 7.2-7.9 (m, 17H, Ar-H), 10.3(s, 1H, CONH). (iii) Mass spectrum of **4a** showed peaks at m/z (relative intensity %) 510.5 (0.03)(M^+), 358(0.21), 341(2.86), 340 (4.70), 316(11.50), 197(3.20), 196(25.40), 183(100.00), 141(7.60), 133(31.70), 105 (36.40), 91 (27.80), 77 (43.03), 65(13.90) and 57(21.11).

On the other hand, the reactivity of the oxirane ring towards active methylene compounds has been studied. When the epoxide **(2)** was reacted with ethyl acetoacetate and ethyl bromoacetate with the fission of the oxirane ring, followed by ring closure furnished N-{4-[-4-(4-chlorophenyl)-3-(aceto or bromo)-2-oxo-2,3,4,5-tetrahydro-furan-carbon-yl-5-yl]-phenyl}-2-(1,1,3-trioxo-1,3-dihydro-benzo[d]isothia-zol-2-yl)-acetamide **(5a,b).** The structure of **(5a)** and **(5b)** was supported by: (i) IR spectrum which showed absorption bands at 1765-1755 cm^{-1} (attributable to ν CO of furanones). (ii) Mass spectrum of 5a showed peaks at m/z (relative intensity %) 580 (0.01)(M^+), 400(0.02), 372(0.24), 371(0.84), 343(0.13), 315(0.50), 183(0.19), 168(6.20), 141(8.90), 117(0.44), 105 (100.00), 77(41.93) and 65(16.20).

The reaction of epoxides with aromatic hydrocarbons in the presence of anhydrous aluminum chloride under Friedel-Craft's conditions has been reported [8]. Thus,toluene was arylated by using the epoxide **(2)** and yielded N-{4-[3-(4-chlorophenyl)-α-(4-methylphenyl)-β-(4-methylphenyl)-acryloyl]-phenyl}-2-(1,1,3-trioxo-1,3-dihydroben-zo[d]isothiazol-2-yl)-acetamide **(6)**.The structure of **6** was proved by: (i) IR spectrum which showed absorption bands at 1605 cm^{-1} (ν C=C). (ii) ^1H NMR spectrum of **6** showed signals at δ (ppm). 2.1(s, 6H, $2CH_3$), 4.4(s, 2H, NCH_2O), 7.2-7.8(m, 20 H, Ar-H) and 9.8(s, 1H, CONH).

Previously[9], it was reported that, epoxides underwent ring opening by action of Grignard reagents. So, the epoxide **(2)** was reacted with ethyl magnesium bromide and phenylmagnesium bromide and afforded N-{4-[3-(4-chlorophenyl)-α-(4-alkyl or aryl)-β-hydroxy-proponoyl]-phenyl}-2-(1,1,3-trioxo-1,3-dihydro-benzo[d]isothiazol-2-yl)-acetamide **(7a,b).** The structure of **7a** and **7b** was proved by: (i) IR spectrum, which showed absorption bands at 3450 cm^{-1} (ν OH). (ii) ^1H NMR spectrum of **7a** showed signals at δ (ppm). 1.3 (t, 3H, CH_2CH_3), 3.1 (d, 1H, α-CH), 3.3 (d, 1H, β-CH), 4.1 (q, 2H,CH_2CH_3), 4.5 (s,2H,NCH_2CO), 7.2-7.9 (m,12H,Ar-H) and 9.2 (s,1H, CONH) and 11.1 (s, 1H,OH).

Reactions of the epoxides with thiourea and glycine were reported[10-12]. So, conden-sation of epoxide **(2)** with thiourea and glycine in the presence of dimethyl formamide (DMF) as a

solvent and anhydrous aluminum chloride as a catalyst furnished N-{4-[4-(4-chlorophenyl)-2-thioxo-2,3,4,5-tetrahydro-oxazolin-4-yl]-phenyl}-2-(1,1,3-trioxo-1 ,3-dihydrobenzo [d] isothiazol-2-yl)-acetamide **(8)**.and N-{4-[5-(4-chlorophenyl)- 2-oxo-2,3,4,5,6-pentahydro-morpholin-carbonyl-6-yl]-phenyl}-2-(1,1,3-trioxo-1,3-dihydro-benzo[d]isothiazol-2-yl)-acetamide **(9)**. The structure of **8** and **9** was established by: (i) IR spectrum of **8** showed absorption bands at 1260 cm^{-1} (ν C=S) and IR spectrum of **9** showed absorption bands at 1710 cm^{-1} (ν CO of α-lactone). (ii) Mass spectrum of **9** showed peaks at m/z (relative intensity %) 353.5 (3.80)(M^+), 414(6.20), 400(12.40), 372(7.80), 371(15.30), 343(2.70), 315(8.90), 196(41.60), 168(39.90), 140(47.50), 119(43.70), 104(32.60), 91(44.30), 76(100.00) and 65(28.50).

The reaction of the epoxide **(2)** with acetic acid at room temperature afforded N-{4-[3-(4-chlorophenyl)-α-acetoxy-β-hydroxy-proponoyl] phenyl}-2-(1,1,3-trioxo-1,3-dihydro-benzo[d]isothiazol-2-yl)-acetamide **(10).** The structure of **(10)** was supported by: (i) IR spectrum of **(10)** showed absorption bands at 3450-3350 cm^{-1} (ν OH). (ii) ^{1}H NMR spectrum of **(10)** showed signals at δ (ppm). 2.1(s, 3H, $COCH_3$), 3.15(d, 1H,α-CH), 3.35(d, 1H,β-CH), 7.8(m, 12H,Ar-H) and 9.1(s, 1H,CONH) and 11.2(s, 1H,OH).

The interest in the biological and industrial potential[13] of 2-oxazolines has been resulted in various synthetic procedures for the introduction of five-membered nitrogen and oxygen containing heterocycles i.e. 2-oxazoline into hydrocarbon chain. Different studies of the reactions of various short chain epoxides with nitriles in the presence of a catalyst leading to the formation of 2-oxazolines have been described[14]. These observations prompted us to carry out the conversion of α, β-epoxy ketone **(2)** into 2-oxazolines via the reaction of epoxide **(2)** with benzonitrile, acetonitrile and acrylonitrile in the presence of boron triflouride-etherate as catalyst and yielded the corresponding α,-hydroxy-β -amido derivatives **(11a-c)**, which underwent cyclization by subjected to fusion in an oil bath at 210-220°C and furnished N-{4-[4-(4-chloro-phenyl)-4,5-dihydro-oxazolincarbonyl-5-yl]phenyl}-2-(1,1,3-trioxo-1,3-dihydro-benzo [d]isothiazol-2-yl)-acetamide **(12a-c)**. The structure of **11a-c** and **12a-c** was supported by: (i) IR spectrum of **11a-c** showed absorption bands at 3400-3150 cm^{-1} (νNH) and (νOH), IR spectrum of **12a-c** showed absorption bands at 1630 cm^{-1} (ν C=N), 1210 cm^{-1} (ν C-O-C) and disappear (ν OH). (ii) ^{1}H NMR spectrum of **12c** showed signals at δ(ppm). 2.3(d, 2H, 2CH cyclic), 4.6(s, 2H, NCHCO), 6.2 (t, 1H,CH=CH), 6.9 (d, 1H, CH_2=CH-) 7.9 (m, 12H,Ar-H) and 9.9 (s, 1H,CONH). (iii) Mass spectrum of **12b** showed peaks at m/z (relative intensity %) 537.5(2.80)(M^+), 372(5.70), 371(11.50), 343(3.70), 315(15.50), 196 (24.00), 183(100.00), 168(12.20), 141(9.60), 140(18.50), 120(13.30), 119(49.10), 105(27.30), 91(30.20), 77(38.50), 65(22.60) and 57(47.40).

ANTIMICROBIAL ACTIVITY

The antimicrobial activities of all synthesized compounds were determined by using the hole plate and filter paper disc method. The tested compounds were dissolved in 10% acetone. The concentrations chosen were (125, 250 ìg/ml). The results are summarized in TABLE (1).

TABLE 1: Activity (A) and minimum inhibitory concentration (MIC)

Compd. No.	Asperigillus flauus		E-coli		Staphylococcus aureus		Bacillus circulans	
	A	MIC	A	MIC	A	MIC	A	MIC
3a	+	250	-	-	+	250	++	125
4b	++	125	+	250	+	250	+	250
6b	++	250	-	-	-	-	++	250
7	+	250	+	250	-	-	+	125
8	+	250	-	-	+	250	-	-
9	++	125	+	250	++	125	+	250
11a	+	250	-	-	+	125	+	125
11b	++	125	++	250	+++	125	++	125
11c	++	125	+	125	++	250	+	250
12c	++	250	++	250	++	250	++	125

The width of the zone of inhibition indicates the potency of antimicrobial activity, (-) no antimicrobial activity, (+) week activity with diameter equal to (0.5-0.7cm), (++) moderate activity with the diameter zone equal to (1.0-

1.2cm), (+++) marked activity with the diameter zone equal to (1.6-1.8cm).

Origin of cultures: Botany Department, Faculty of Science, Benha University, Egypt.

EXPERIMENTAL

All melting points are uncorrected. The IR spectra in KBr were recorded on a Shimadzu 470 Spectrometer. The ^1H NMR were measured on Varian EM-390-90 MHz a spectrometer using TMS as internal reference and the chemical shifts are expressed as ä (ppm). The mass spectra were recorded on HP Model: MS 5988 at 70 eV. The physical data are listed in TABLE (2).

N-{4-[3-(4-Chlorophenyl)-ethylene oxide-carbonyl]-phenyl}- 2-(1, 1, 3-trioxo-1, 3-dihydrobenzo [d] isothiazol-2-yl)-acetamide (2)

A solution of chalcone **(1)** (0.01mole) in acetone (40 ml) was mixed with 8% aqueous sodium hydroxide (12 ml) followed by addition of hydrogen peroxide (30% 5 ml). The solution was shaken and heated for 2hr, then allowed to stand overnight at room temperature, water then added and the solution acidified with dil HCl. The mixture was extracted with ether and the solid separated was crysalized from the proper solvent to give **(2)**.

N-{4-[3-(4-Chlorophenyl)-á-arylamino-â-hydroxy-carbonyl-ethylene oxide]-phe-nyl }-2-(1,1,3-trioxo-1,3-dihydrobenzo [d] isothiazol-2-yl)-acetamide (3a,b)

Epoxide **(2)** (0.01mole) in n-butanol was treated with p-bromoaniline and benzylamine (0.01mole). The solution was heated under reflux for 3hr. The solid products after concentration and cooling were crystallized from the proper solvent to give **(3a,b)**.

N-{4-[5-(4-Chlorophenyl)-4-hydroxy-4,5-dihydro-1H or 1-phenyl-pyrazolin-3-yl]-phenyl}-2-(1,1,3-trioxo-1,3-dihydro benzo [d] isothiazol-2-yl)-acetamide (4a, b).

A solution of epoxide **(2)** (0.01mole) was refluxed in n-butanol with hydrazine hydrate and phenylhydrazine (0.05mole) for 8hr, then the reaction mixture was poured into ice-water. The products were separated and crystallized from the proper solvent to give **(4a,b)**.

N-{4-[4-(4-Chlorophenyl)-3-(aceto or bromo)-2-oxo-2,3,4,5-tetrahydro-furan-car-bonyl-5-yl]-phenyl}-2-(1,1,3-trioxo-1,3-dihydro-benzo[d]isothiazol-2-yl)-acetami-de (5a,b)

The epoxide **(2)** was added to a cold stirred of sodium hydroxide (0.02 mole), acetic acid (30ml) with ethyl acetoacetate and ethyl bromoacetate (0.015 mole). The reaction mixture was stirring for 15 min, then warmed to 50^0C for 6hr. The complex was decomposed with a mixture of water and dil HCl, then extracted with benzene and washed with aqueos $NaHCO_3$ solution (10%) and water. The products obtained was crystallized from the proper solvent to give **(5a,b)**.

N-{4-[3-(4-Chlorophenyl)-α,-(4-methylphenyl)-β-(4-methylphenyl)-acr-yloyl]-phenyl}-2-(1,1,3-trioxo-1,3-dihydro-benzo [d] isothiazol-2-yl)-acetamide (6).

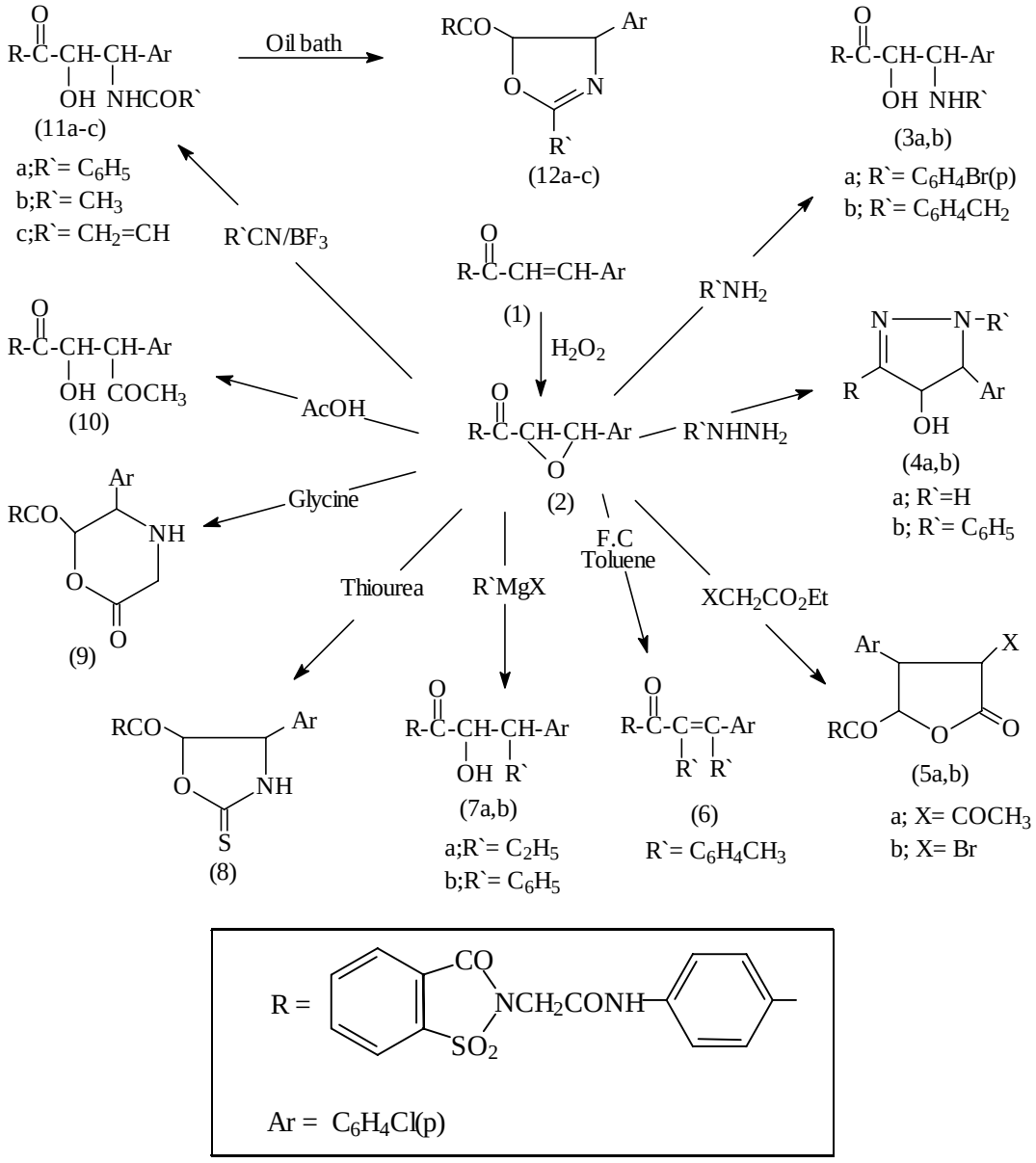
A solution of epoxide **(2)** in dry benzene (20 ml), toluene (0.03 mole) and anhydrous $AlCl_3$ (0.01mole) was added then the reaction mixture was reflux for 4hr. the solide that separated on cooling was crystallized from the proper solvent to give **(6)**.

N-{4-[3-(4-Chlorophenyl)- α-(4-alkyl or aryl)-β-hydroxyproponoyl]phenyl}-2-(1,1 ,3-trioxo-1,3-dihydro-benzo [d] isothiazol-2-yl)-acetamide (7a,b).

To a suspension of epoxide **(2)** (0.01 mole) in dry ether (50 ml) was added an ethereal solution of ethyl magnesium bromide and phenyl magnesium bromide (0.03 mole), the reaction mixture was refluxed on a steam-bath for 4hr, then decomposed with a saturated solution of ammonium chloride, extrated with ether.The solid obtained was crystallized from the proper solvent to give **(7a,b)**.

N-{4-[4-(4-Chlorophenyl)-2-thioxo-2,3,4,5-tetrahydro-oxazolin-4-yl]-phenyl}-2-(1, 1,3-trioxo-1,3-dihydro-benzo[d]isothiazol-2-yl)-acetamide (8) and N-{4-[5-(4-chlo-rophenyl)-2-oxo-2,3,4,5,6-pentahydro-morpholin-carbonyl-6-yl]-phenyl}-2-(1,1 ,3 - trioxo-1,3-dihydro-benzo [d] isothiazol-2-yl)-acetamide (9).

Equimolar amounts of epoxide **(2)**, thio-



SCHEME

urea and glycine (0.01 mole) were heated under reflux in DMF (20 ml) in the presence of catalytic amount of $AlCl_3$ (0.05 gm) for 3hr. The reaction mixture was poured into water and extracted with ether. The solid obtained was crystallized from the proper solvent and give **(8)** and **(9)**.

N-{4-[3-(4-Chlorophenyl)- α-acetoxy-β-hydroxy-proponoyl]phenyl}-2-(1,1,3-trioxo-1,3-dihydro-benzo [d] isothiazol-2-yl)-acetamide (10)

A solution of epoxide **(2)** and acetic acid (30 ml) was refluxed for 4hr.The reaction mixture was cooling and poured into water.The separated solid was crystallized from the proper solvent to give **(10)**.

α-Hydroxy-β-amido derivatives (11a-c)

An equimolar amounts of epoxide **(2)** (0.03 mole) and BF_3-etherate (0.03 mole) as catalyst stirred at room temperature for 5hr. The reaction mixture was poured into aqueous $NaHCO_3$ (5%) and extracted with ether. The solid obtained was crystallized from the proper solvent to give **(11a-c)**.

N-{4-[4-(4-Chlorophenyl)-4,5-dihydro-oxazolin-carbonyl-5-yl]-phenyl}-2-(1,1,3-trioxo-1,3-dihydro-benzo[d] isothiazol-2-yl)-acetamide (12a-c)

TABLE 2:Physical data of various compounds prepared

Compd. No.	Mol. Formula (M.Wt)	Solvent Yield %	M.P. (Colour)	Analysis data Calcd. / Found %		
				C	H	N
2	$C_{24}H_{17}ClN_2O_6S$ (496.5)	B 65	105 (y)	58.0 58.0	3.4 3.4	5.6 5.6
3a	$C_{30}H_{23}ClN_3O_6BrS$ (668.5)	B 66	167 (b)	53.9 53.9	3.4 3.4	6.3 6.3
3b	$C_{31}H_{25}ClN_3O_6S$ (602.5)	X 64	139 (y)	61.7 61.6	4.1 4.1	7.0 7.0
4a	$C_{24}H_{19}ClN_4O_5S$ (510.5)	E 58	145 (y)	56.4 56.3	3.7 3.6	11.0 11.0
4b	$C_{30}H_{23}ClN_4O_5S$ (595.5)	E 60	187 (y)	60.5 60.5	3.9 3.9	9.4 9.3
5a	$C_{28}H_{21}ClN_2O_8S$ (580.5)	B 61	191 (y)	57.9 57.9	3.6 3.7	4.8 4.8
5b	$C_{26}H_{18}ClN_2O_7BrS$ (617.5)	B 63	203 (b)	50.2 50.1	2.9 2.8	4.5 4.5
6	$C_{38}H_{29}ClN_2O_5S$ (660.5)	E 60	121 (y)	69.0 69.0	4.4 4.4	4.2 4.2
7a	$C_{26}H_{23}ClN_2O_6S$ (526.5)	E 67	198 (y)	59.3 59.3	4.4 4.3	5.3 5.2
7b	$C_{30}H_{23}ClN_2O_6S$ (574.5)	E 74	217 (y)	62.7 62.7	4.0 4.1	4.9 4.9
8	$C_{25}H_{18}ClN_3O_6S_2$ (555.5)	X 75	161 (b)	54.0 54.1	3.2 3.1	7.6 7.5
9	$C_{26}H_{20}ClN_3O_7S$ (553.5)	B 63	183 (y)	56.4 56.3	3.6 3.5	7.6 7.6
10	$C_{26}H_{21}ClN_2O_7S$ (540.5)	E 78	211 (y)	57.7 57.6	3.9 3.8	5.2 5.2
11a	$C_{31}H_{24}ClN_3O_7S$ (617.5)	B 67	173 (y)	60.2 60.2	3.9 3.8	6.8 6.7
11b	$C_{26}H_{22}ClN_3O_7S$ (555.5)	B 65	219 (y)	56.2 56.2	4.0 4.1	7.6 7.5
11c	$C_{27}H_{22}ClN_3O_7S$ (567.5)	B 59	179 (y)	57.1 57.2	3.9 3.8	7.4 7.5
12a	$C_{31}H_{22}ClN_3O_6S$ (599.5)	B 61	134 (y)	66.7 66.7	3.7 3.6	7.0 7.0
12b	$C_{26}H_{20}ClN_3O_6S$ (537.5)	B 60	159 (y)	58.0 58.1	3.7 3.7	7.8 7.7
12c	$C_{27}H_{20}ClN_3O_6S$ (649.5)	B 59	151 (b)	59.0 50.1	3.6 3.5	7.6 7.5

Benzene = B; Xylene = X; Ethanol = E;
brown = b; yellow = y

The hydroxy amides **(11a-c)** were heated at 210-220°C for 6-8hr, in an oil bath.The cooled pyrolysate was dissolved in ether, filtered and dried over anhydrous sodium sulphat.The solvent was evaporated and gave the solid product which crysallized from the proper solvent to give **(12a-c)**.

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