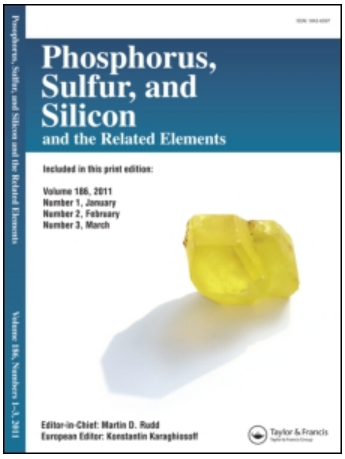

Phosphorus, Sulfur, and Silicon and the Related Elements

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SYNTHESIS OF N-ARYL SUBSTITUTED 4*H*-1,4-BENZOTHIAZINE 1,1-DIOXIDE 2-CARBOXYLIC ACID-ESTERS

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SYNTHESIS OF N-ARYL SUBSTITUTED 4*H*-1,4-BENZOTHIAZINE 1,1-DIOXIDE 2-CARBOXYLIC ACID-ESTERS

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A group of N-aryl substituted 1,4-benzothiazine-1,1-dioxide-2 carboxylic acid esters is reported. This is the first example of N-aryl-derivatives of the 4*H*-1,4-benzothiazine nucleus; the key step is the cyclization of N-aryl-phenylsulfonyl-acrylates **4–8** using potassium carbonate in acetonitrile/18-crown-6-ether to the corresponding title compounds in moderate yields.

Keywords: 4*H*-1,4-Benzothiazine-1,1-dioxide; N-aryl-phenylsulfonylacrylates; cyclization reaction

Recently, there has been great interest in sulfur-containing heterocyclic compounds, especially those having a sulfone functionality, because of their importance in medicinal chemistry[1–3]. During our research, involved in the synthesis of novel biologically active sulfur heterocycles[4], we explored a synthetic approach to obtain N-aryl substituted 4*H*-1,4-benzothiazine-1,1 -dioxide-2-carboxylic acid-esters.

4*H*-Benzothiazines possess a variety of pharmacological and biological activities similar to that of structurally related phenothiazines[5]. A few examples of N-alkylated 4*H*-1,4-benzothiazine-1,1-dioxide-2-carboxylic acids have appeared in the literature[6,7]. The usual procedure for their synthesis involves the N-alkylation of 4*H*-1,4-benzothiazine-2-carboxylated compounds[6,7]. Alternatively, some N-alkyl substituted 1,4-benzo-

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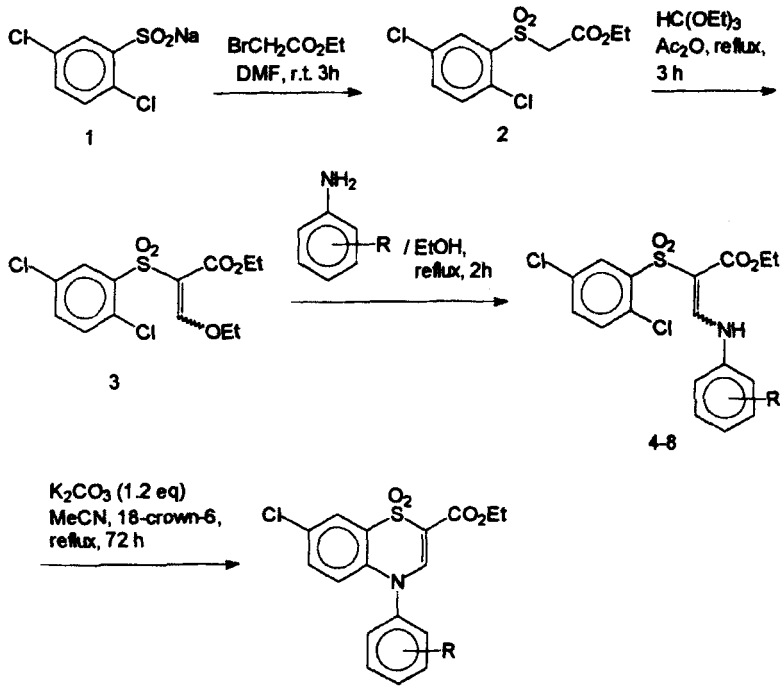
thiazine-2-carboxylic acid 1-oxides with antibacterial properties were synthesized by intramolecular cyclization of their corresponding N-alkyl-phenyl-sulfinyl acrylates, using sodium hydride in refluxing toluene for 2 hours in low yields[8]. Until now, no reports of N-arylated 4*H*-1,4-benzothiazine-1,1-dioxides 2-carboxylic acid-esters have appeared, thus it was interesting for us to explore their synthesis.

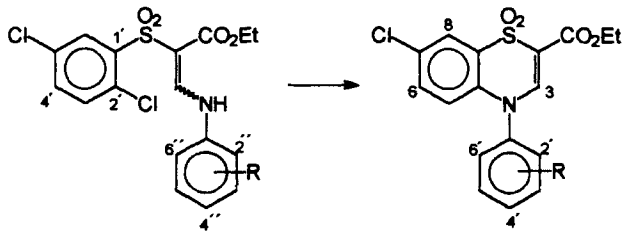


SCHEME

Our synthetic approach, which was achieved in a 4-step procedure is outlined in the SCHEME. First, treatment of sodium 2,5-dichlorobenzene-sulfinate **1** with ethylbromoacetate in DMF at room temperature afforded ethyl-2,5-dichlorophenyl-sulfonylacetate **2.** Then, the enol ether **3** was obtained by the reaction of **2** with triethyl orthoformate in acetic anhydride, which, upon evaporation of the solvent to dryness, was allowed to react as a crude with an appropiately substituted aniline in ethanol at

reflux for 2 hours using a catalytic amount of concentrated sulphuric acid, yielding phenylsulfonyl-acrylates **4–8** as sole products (E or Z stereochemistry was not determined). Finally, we used different procedures to cyclisize these intermediates. The results are summarized in the TABLE Favourable conditions were obtained using 1.2 mol-eq. of potassium carbonate in acetonitrile and a small amount of 18-crown-6 ether under reflux for 72 hr.

TABLE Conditions for the cyclization of the compounds **4–8** to **9–13**



4-8 **9-13**

Compd. No.	R	Base, mol-eq.	Solvent	Time(h), temp.	Yield (%)
9	4′-Br	NaH, 1.2	Toluene	24, reflux	8
9	4′-Br	NaH, 1.2	Toluene	72, reflux	15
9	4′-Br	NaH, 1.2	DMF	24, reflux	17
9	4′-Br	NaH, 1.2	DMF	72, reflux	20
9	4′-Br	K_2CO_3, 1.2	Dioxane	24, reflux	14
9	4′-Br	K_2CO_3, 1.2	Dioxane	72, reflux	27
9	4′-Br	K_2CO_3, 1.2	MeCN, 18-crown-6	72, reflux	50
10	4′-Cl	K_2CO_3, 1.2	MeCN, 18-crown-6	72, reflux	48
11	3′-Cl	K_2CO_3, 1.2	MeCN, 18-crown-6	72, reflux	57
12	4′-OMe	K_2CO_3, 1.2	MeCN, 18-crown-6	72, reflux	61
13	4′-F	K_2CO_3, 1.2	MeCN, 18-crown-6	72, reflux	41

This cyclization is more difficult to achieve than that in the synthesis of the corresponding N-aryl-quinolones which are the carbonyl isosteres of our target compounds[9,10]. Classical procedures for the N-aryl-quinolones, including sodium hydride in refluxing toluene[9], sodium hydride/DMF[10], as well as the use of potassium carbonate in dioxane were employed for cyclization of phenyl-sulfonyl-acrylate **4,** giving poor yields of the desired N-arylated 1,4-benzothiazine-sulfone **9,** some uncyclisized **4,** and decomposition products (not isolated) in all cases. But on using potassium carbonate (1.2 eq) in 18-crown-6/acetonitrile, an enhanced yield of **9** was obtained and so this method was employed for the cyclization of the remaining sulfonyl-acrylates **5–8.** Studies of this cyclization reaction are now in progress in our laboratories with some other polyfunctionalizated N-aryl-sulfonyl-acrylates.

EXPERIMENTAL

Melting points were determined with a Fischer-Johns micro hot-stage apparatus and are uncorrected. The IR spectra were recorded as KBr pellets using a NICOLET Magna-FT/IR 550 spectrometer. Proton NMR spectra (NMR) were recorded on a JEOL GSX (270 MHz) spectrometer; δ values in ppm relative to tetramethylsilane are given. When reported, mass spectra were recorded on a Hewlett-Packard HP5971A Mass Selective Detector conected to a Gas Chromatograph HP5970 Series II with EI(70 eV). Elemental analysis were performed by Laboratorio de Servicios, Facultad de Ciencias, Escuela de Quimica, Universidad Central de Venezuela (Caracas, Venezuela); results fill in the range $\pm$ 0.4% of the theoretical values. Silica gel plates Merck F_{254} (Merck, Darmstadt, Germany) were used for TLC controls. Column Chromatography was performed with Kieselgel 60 (70–230 mesh) silica gel (Merck) and hexanes-ethyl acetate (8:2) as an eluant. Reagents were obtained from Aldrich (USA) and used without further purification. Solvents were distilled prior to use. Sodium 2,5-dichlorobenzenesulfinate **1** was synthesized according to literature procedure[11].

Ethyl 2,5-dichlorophenyl-sulfonylacetate 2

Sodium 2,5-dichlorobenzenesulfinate **1** (3.95 g, 17.03 mmol) was dissolved in DMF (40 mL), then ethyl-bromoacetate (2.85 g, 17.03 mmol)

was slowly added and the reaction mixture was stirred at room tempertature for 3 h. When reaction time was completed, the mixture was poured into ice-crushed water; the white precipitate formed was filtered, washed twice with water and dried under vacuo, giving a white powder. Yield: 3.78 g (75%); mp 75–76°C. IR (KBr, cm^{-1}): ν=1733 (C=O, ester); 1370,1295 (SO_2); 1167,1105 (SO_2). MS (EI): m/z = 297 (M^+). ^1H NMR ($CDCl_3$/TMS): δ = 1.14 (t, 3H, CH_3), 4.10 (c, 2H, O-CH_2-), 4.42 (s, 1H, methylene CH_2), 7.49 (d, 1H, ar.3′-H, J= 9.2 Hz), 7.56 (dd, 1H, ar.4′-H, J= 9.2 Hz, J=2.2 Hz), 8.09 (d, 1H, ar.6′-H, J= 2.2 Hz).

Anal. Calcd. for $C_{10}H_{10}Cl_2O_4S$: C, 40.42; H, 3.37.
Found: C, 40.33 ; H, 3,48.

Ethyl 2-(2,4-dichloro-phenylsulfonyl)-3-(substituted-anilino)-acrylates 4–8

General procedure

A mixture of **2** (2.0 g, 6.73 mmol), acetic anhydride (1.5 g, 10.10 mmol) and triethylortoformate (1.65 g, 16.15 mmol) was stirred under reflux using a Dean-Stark trap for 3 h. Then, solvent was removed under vacuo and the oil obtained used as a crude for the next step. The above mentioned oil was dissolved in ethanol (50 mL), substituted-aniline (6.73 mmol) was slowly added and to the mixture I drop of concentrated sulphuric acid. Reaction was stirred under reflux for 2 h., then allowed to cool at room temperature and the precipitated solid thus obtained filtered, washed with ethanol, and dried under vacuo to give **4–8**.

Ethyl 2-(2,4-dichloro-phenylsulfonyl)-3-(4-bromoanilino)-acrylate 4

Yield: 2.32 g (72%); mp: 225–226 °C (recrystallized from EtOH). IR (KBr, cm^{-1}): ν = 3248 (NH); 1674 (C=O, ester); 1623 (C=C); 1340,1320 (SO_2); 1155,1136 (SO_2). ^1H NMR ($CDCl_3$/TMS): δ = 1.12 (t, 3H, CH_3), 4.09 (c, 2H, CH_2), 7.06 (d, 2H, 2″-H, 6″-H, J=8.7 Hz), 7.36 (d, 1H, 3′-H, J=8.4 Hz), 7.45 (dd, 1H, 4′-H, J= 8.4 Hz, J= 2.5 Hz), 7.52 (d, 2H, 3″-H, 5″-H, J= 8.7 Hz), 8.26 (d, 1H, 6′-H, J=2.5 Hz), 8.57 (d, 1H, vinyl CH, J= 13.8 Hz), 10.71 (d, 1H, NH, J= 13.8 Hz).

Anal. Calcd. for $C_{17}H_{14}Cl_2BrNO_4S$: C, 40.97; H, 2.81; N, 2.81.
Found: C, 40.84; H, 2.80; N, 2.80

Ethyl 2-(2,4-dichloro-phenylsulfonyl)-3-(4-chloroanilino)-acrylate 5

Yield: 2.05 g (70 %); mp: 227–228 °C (recrystallized from EtOH). IR (KBr, cm^{-1}): ν= 3254 (NH); 1672 (C=O, ester); 1622 (C=C); 1323,1301 (SO_2);1154,1130(SO_2). ^1H NMR ($CDCl_3$/TMS): δ = 1.12 (t, 3H, CH_3), 4.09 (c, 2H, CH_2), 7.1(d, 2H, 2″-H, 6″-H, J=8.6 Hz), 7.38 (d, 2H, 3″-H, 5″-H, J= 8.6 Hz), 7.36 (d, 1H, 3′-H, J= 8.4 Hz), 7.45 (dd, 1H, 4′-H, J= 8.4 Hz, J= 2.4 Hz), 8.27 (d, 1H, 6′-H, J= 2.4 Hz), 8.56 (d, 1H, vinyl CH_2, J= 13.8 Hz), 10.73 (d, 1H, NH, J= 13.8 Hz).

Anal. Calcd. for $C_{17}H_{14}Cl_3NO_4S$: C, 46.99; H, 3.22; N, 3.22.

Found: C, 47.12; H, 3.21; N, 3.23.

Ethyl 2-(2,4-dichloro-phenylsulfonyl)-3-(3 -chloroanilino)-acrylate 6

Yield: 2.19 g (75 %); mp: 220–221°C (recrystallized from EtOH). IR (KBr, cm^{-1}): ν = 3265 (NH); 1682 (C=O, ester); 1623 (C=C); 1324,1307 (SO_2); 1160,1139 (SO_2), ^1H NMR ($CDCl_3$/TMS): δ = 1.12 (t, 3H, CH_3), 4.09 (c, 2H, CH_2), 7.06 (d, 2H, 2″-H, 6″-H, J= 8.6 Hz), 7.14–7.39 (m, 3H, arom. 3′-H, 3″-H, 5″-H, J= 8.6 Hz), 7.36 (d, 1H, 3′-H, J= 8.4 Hz), 7.44 (dd, 1H, 4′-H, J= 8.4 Hz, J= 2.2 Hz), 8.27 (d, 1H, 6′-H, J= 2.2 Hz), 8.58 (d, 1H, vinyl CH_2, J= 13.8 Hz), 10.73 (d, 1H, NH, J= 13.8 Hz).

Anal. Calcd. for $C_{17}H_{14}Cl_3NO_4S$: C, 46.99; H, 3.22; N, 3.22

Found: C, 47.06; H, 3.22; N, 3.21

Ethyl 2-(2,4-dichloro-phenylsulfonyl)-3-(4-methoxyanilino)-acrylate 7

Yield: 2.18 g (78%); mp:196–197 °C (recrystallized from EtOH). IR (KBr, cm^{-1}): ν = 3273 (NH); 1686 (C=O, ester); 1625 (C=C); 1330,1310 (SO_2); 1164,1151 (SO_2). ^1H NMR ($CDCl_3$/TMS): δ = 1.12 (t, 3H, CH_3), 3.81 (s, 3H, OCH_3), 4.11 (c, 2H, CH_2), 6.92 (d, 2H, 2″-H, 6″-H, J= 8.9 Hz), 7.12 (d, 2H, 3″-H, 5″-H, J= 8.9 Hz), 7.36 (d, 1H, 3′-H, J= 8.4 Hz), 7.44 (dd, 1H, 4′-H, J= 8.4 Hz, J= 2.5 Hz), 8.27 (d, 1H, 6′-H, J= 2.4 Hz), 8.52 (d, 1H, vinyl CH_2, J= 14.1 Hz), 10.69 (d, 1H, NH, J= 14.1 Hz).

*Anal.*Calcd. for $C_{18}H_{17}Cl_2NO_5S$: C, 42.11; H, 3.31; N, 2.73

Found: C, 42.18; H, 3.30; N, 2.74

Ethyl 2-(2,4-dichloro-phenylsulfonyl)-3-(4-fluoroanilino)-acrylate 8

Yield: 1.83 g (65 %); mp: 228–229 °C(recrystallized from EtOH). IR (KBr, cm^{-1}): ν = 3273 (NH); 1684 (C=O, ester); 1627 (C=C); 1329,1307 (SO_2); 1156,1138 (SO_2). ^1H NMR ($CDCl_3$/TMS): δ = 1.12 (t, 3H, CH_3), 4.09 (c, 2H, CH_2), 7.04–7.15 (m, 4H, arom. 2″-H, 3″-H, 5″-H, 6″-H), 7.36 (d, 1H, 3′-H, J= 8.6 Hz), 7.44 (dd, 1H, 4′-H, J= 8.6 Hz, J= 2.2 Hz), 8.27 (d, 1H, 6′-H, J= 2.2 Hz), 8.53 (d, 1H, vinyl CH_2, J= 14. 1 Hz), 10.72 (d, 1H, NH, J= 14. 1 Hz).

Anal. Calcd. for $C_{17}H_{14}Cl_2FNO_4S$: C, 46.47; H, 3.20; N, 3.20

Found: C, 46.62; H, 3.19; N, 3.20

Ethyl 4-(substituted-aryl)-7-chloro-4*H*-1,4-benzothiazine-2-carboxylate 1,1-dioxides 9–13

General procedure

A mixture of ethyl 2-(2,5-dichloro-phenylsulfonyl)-3-(substituted-anilino)-acrylate **4–8** (2. 1 mmol), potassium carbonate (3.15 mmol, 1.2 eq.) and 18-crown-6 (0.12 mmol) in acetonitrile (20 mL) was refluxed for 72 hours. Then, solvent was removed under vacuo and the solid residue thus obtained washed with 25% aqueous EtOH. The residue was purified by column chromatography (silica gel) eluting with hexanes-EtOAc (80:20) to afford desidered **9–13** as white solids.

Ethyl 4-(4-bromophenyl)-7-chloro-4*H*-1,4-benzothiazine-2-carboxylate 1,1-dioxide 9

Yield: 0.46 g (50%); mp: 215–216 °C.; IR (KBr, cm^{-1}): ν = 1700 (C=O, ester); 1620 (C=C); 1280,1270 (SO_2); 1140,1130 (SO_2). ^1H NMR ($CDCl_3$/TMS): δ = 1.39 (t, 3H, CH_3); 4.41 (c, 2H, CH_2); 6.61 (d, 1H, 5-H, J= 9.2 Hz); 7.25 (d, 2H, 2′-H, 6′-H, J= 8.4 Hz); 7.36 (dd, 1H, 6-H, J= 9.2 Hz, J= 2.2 Hz); 7.75 (d, 2H, 3′-H, 5′-H, J=8.4 Hz); 7.88 (s, 1H, vinyl 3-H); 8.12 (d, 1H, 8′-H, J=2.2 Hz).

Anal. Calcd. for $C_{17}H_{13}ClBrNO_4S$: C, 42.41; H, 2.70; N, 2.91

Found: C, 42.35; H, 2.69 ; N, 2.90.

Ethyl 4-(4-chlorophenyl)-7-chloro-4*H*-1,4-benzothiazine-2-carboxylate 1,1-dioxide 10

Yield: 0.44 g (48%); mp: 212–213 °C; IR (KBr, cm^{-1}): ν = 1690 (C=O, ester); 1625 (C=C); 1295,1280 (SO_2); 1150,1140 (SO_2). ^1H NMR ($CDCl_3$/TMS): δ = 1.38 (t, 3H, CH_3); 4.39 (c, 2H, CH_2); 6.60 (d, 1H, 5-H, J= 9.2 Hz); 7.29–7.36 (m, 3H, 2′-H, 6′-H, 6-H); 7.59 (d, 2H, 3′-H, 5′-H, J=8.4 Hz); 7.88 (s, 1H, vinyl 3-H); 8.1 1 (d, 1H, 8′-H, J= 2.2 Hz).

Anal. Calcd. for $C_{17}H_{13}Cl_2NO_4S$: C, 46.73; H, 2.98; N, 3.20
Found: C, 46.87; H, 2.99; N, 3.21.

Ethyl 4-(3-chlorophenyl)-7-chloro-4*H*-1,4-benzothiazine-2-carboxylate 1,1-dioxide 11

Yield: 0.52 g (57 %); mp: 193–194 °C.; IR (KBr, cm^{-1}): ν = 1690 (C=O, ester); 1620 (C=C); 1290,1270 (SO_2); 1150,1145 (SO_2). ^1H NMR ($CDCl_3$/TMS): δ = 1.40 (t, 3H, CH_3); 4.39 (c, 2H, CH_2); 6.62 (d, 1H, 5-H, J=9.2 Hz); 7.28–7.58 (m, 5H, arom., 2′-H, 4′-H, 5′-H, 6′-H, 6-H); 7.89 (s, 1H, vinyl 3-H) ; 8.14 (d, 1H, 8′-H, J= 2.2 Hz).

Anal. Calcd. for $C_{17}H_{13}Cl_2NO_4S$: C, 46.73; H, 2.98; N, 3.20
Found: C, 46.89; H, 2.98 ; N, 3.21

Ethyl 4-(4-methoxyphenyl)-7-chloro-4*H*-1,4-benzothiazine-2-carboxylate 1,1-dioxide 12

Yield: 0.52 g (61 %); mp: 192–193 °C.; IR (KBr, cm^{-1}): ν = 1690 (C=O, ester); 1623 (C=C); 1300,1287 (SO_2); 1154,1146 (SO_2). ^1H NMR ($CDCl_3$/TMS): δ = 1.38 (t, 3H, CH_3); 3.88 (s, 3H, OCH_3); 4.38 (c, 2H, CH_2); 6.63 (d, 1H, 5-H, J=9.2 Hz); 7.07 (d, 2H, 2′-H, 6′-H, J=8.9 Hz); 7.22 (d, 2H, 3′-H, 5′-H, J=8.9 Hz); 7.27 (dd, 1H, 6-H, J= 9.2 Hz; J= 2.2 Hz); 7.90 (s, 1H, vinyl 3-H); 8.12 (d, 1H, 8′-H, J= 2.2 Hz).

*Anal.*Calcd. for $C_{18}H_{16}ClNO_5S$: C, 54.89; H, 4.10; N, 3.56
Found: C, 54.68 ; H, 4.09; N, 3.56

Ethyl 4-(4-fluorophenyl)-7-chloro-4*H*-1,4-benzothiazine-2-carboxylate 1,1-dioxide 13

Yield: 0.38 g (41 %); mp: 216–217 °C.; IR (KBr, cm^{-1}): ν = 1695 (C=O, ester); 1620 (C=C); 1295,1283 (SO_2); 1149,1138 (SO_2). ^1H NMR

($CDCl_3$/TMS): δ = 1.39 (t, 3H, CH_3); 4.42(c, 2H, CH_2); 6.58(d, 1H, 5-H, J=8.9 Hz); 7.29–7.39 (m, 5H, arom., 2′-H, 3′-H, 5′-H, 6′-H, 6-H); 7.88 (s, 1H, vinyl 3-H); 8.13 (d, 1H, 8′-H, J= 2.5 Hz).

Anal. Calcd. for $C_{17}H_{13}ClFNO_4S$: C, 53.48; H, 3.43; N, 3.67
Found: C, 53.60; H, 3.44; N, 3.66

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