THE SYNTHESIS AND REACTIONS OF TRISUBSTITUTED N-(X-2-BENZOTHIAZOLYL)FORMAMIDINES

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The modified Vilsmayer–Haack reagent ANC(O)H—$ClSO_2C_6H_5$ (A = $(CH_3)_2$, $(C_2H_5)_2$, $(CH_2)_5$, $CH_2CH_2OCH_2CH_2$) reacts with 2-amino-X-benzothiazole in pyridine and ether to give three types of products in high yields depending on the character of substituents A and X. Isolated were: N,N-dialkyl-N′-(X-2-benzothiazolyl)formamidines, N,N′-bis(X-2-benzothiazolyl)formamidines, and N,N,-dialkyl-N′,N″-bis(X-2-benzothiazolyl)triaminomethanes. Investigated were the conditions and reaction products of N,N-dialkyl-N′,N″-bis(X-2-benzothiazolyl)triaminomethanes with phenylethanoyl, phenoxyethanoyl, 4-chlorophenoxyethanoyl, and N-phthalimidoethanoyl chlorides either in ether or in pyridine. Mutual transformations of the individual products are described.

Formamidines are well frequented in the literature, since they use to be utilized as reactive intermediates[1,2]. Some of them are, moreover, highly biologically active compounds, *e.g.* pharmaceuticals[3,4], pesticides[5–7], and bactericides[8,9]. Formamidines also undergo delocalization to form a system with two centres of nucleophility. Formamidine derivatives with benzothiazole ring attached in position 2 have the delocalization extended to the heterocycle and therefore, also the heteroring nitrogen is able to react[10]. As known, also many compounds related to benzothiazole reveal biological activity[8,11]. So far, not too much information is available on the reactivity, synthesis and relationship between structure and biological activity. Two papers appearing in this field deal with the synthesis of formamidine derivatives of 2-aminobenzothiazole[8,11]. The latter describes the synthesis of N,N′-bis(6-methyl)- or (6-methoxy)-2-benzothiazolylformamidines from the corresponding aminobenzothiazole and triethyl formate, the former concerns the preparation of N,N-dimethyl-N′-(6-X-2-benzothiazolyl)formamidines with the goal to investigate their bactericide effects.

This paper was aimed to find out reaction conditions leading to formation of trisubstituted formamidines related to 2-benzothiazole in high yields and purity and to verify the suitability of $R_2NC(O)H$—benzenesulfonyl chloride reagent for this purpose.

Benzenesulfonyl chloride (*II*) is rarely used for the synthesis of formamidines, since results are ambiguous and concurrent benzenesulfonation takes frequently place. Treatment of this reagent with aniline and its nitro derivatives showed in line with[12] that the concurrent benzenesulfonation preferentially occured with more basic amines, whilst the less basic ones afforded formamidines. The ANC(O)H—benzene-

sulfonyl chloride belongs to mild Vilsmayer–Haack reagents and therefore, it is suitable for investigating changes in the reaction direction regarding the conditions; it is also well suited to react with compounds sensitive towards $POCl_3$, PCl_5, and $SOCl_2$. 2-Aminobenzothiazole and especially its derivatives substituted in positions 4 or 6 by an electron accepting group belong to weak bases which can tautomerize into their imino forms[13]. Alkylation was reported to take place at nitrogen in position 3 and acylation at the amino group in position 2 (ref.[14]). As found, formation of formamidines employing this reaction occurred in position 2 of the primary amino group.

Compound *I* was reacted with dialkylformamide (*III*) in pyridine (*IV*) or in ether. The reagent has to be prepared in pyridine under cooling prior to addition of 2-aminobenzothiazole. No formamidines could be obtained in the absence of pyridine or benzenesulfonyl chloride. Advantage of this procedure is the isolation of formamidines as free bases. It is evident that the 2-benzothiazolyl skeleton stabilizes the formamidines obtained due to its electron-accepting character; pyridine proved advantageous since reaction can proceed in a homogeneous phase. The compounds prepared are sufficiently stable, especially in solid state and therefore, they are convenient synthons.

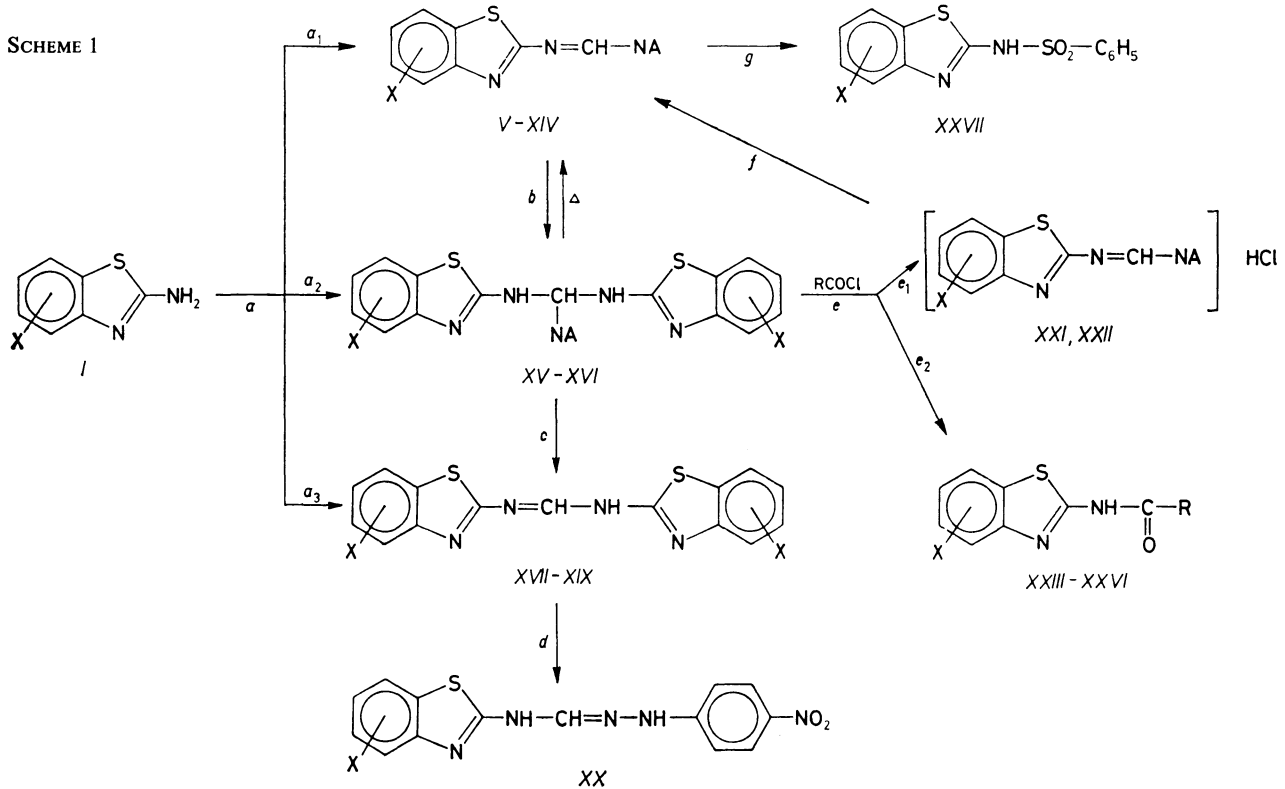
Employed were 2-aminobenzothiazole and its 4-chloro, 6-chloro, 5-bromo and 6-bromo derivatives, dimethyl, diethyl, pentamethylene, and 3-oxapentamethyleneformamides. The reaction was carried out with all chemicals under the same conditions: three types of products were isolated depending on the position of the substituent at the starting benzothiazole. Yields of products were very high (80–100%, reactions $a_1 - a_2$).

N,N-Dialkyl-N'-(X-2-benzothiazolyl)formamidines (*V–XIII*) were the products when reacting compound *I* with the above-mentioned formamides with the exception of diethylformamide. The latter reacted with 4-chloro derivative of *I* to yield N,N'--diethyl-N',N''-bis(4-chloro-2-benzothiazolyl)triaminomethane (*XV*). N,N-Diethyl--N'-(4-chloro-2-benzothiazolyl)-formamidine (*XIV*) was not succeeded to obtain by a direct treatment of the chloro derivative *I* with diethylformamide neither at 0°C; all attempts resulted in formation of *XV*.

The unsubstituted and 6-chloro derivatives of *I* afforded with diethylformamide products of the a_3 reaction, *i.e.* N,N'-bis(X-2-benzothiazolyl)formamidines *XVII* and *XVIII*; it could be anticipated that they are the final compounds of reactions a_1, *b*, *c* (*cf.* Scheme 1). This conclusion is backed by results of two reactions *b* and *c*. Formamidine *V* reacts with 2-amino-4-chlorobenzothiazole in pyridine (reaction *b*) to give *XVI*, and compound *XIX* originated from *XV* and *XVI* due to decomposition in acetic acid (reaction *c*).

Decomposition of formamidines in the excess of benzenesulfonyl chloride and formation of benzenesulfonyl derivatives from the starting primary amines *via* N,N-dialkyl-N'-(X-2-benzothiazolyl)formamidines according to reaction *g* let us suggest this scheme of arylsulfonation to be effective with more basic amines.

SCHEME 1



V, *XVI*, *XXII*, X = 4-Cl; A = $(CH_3)_2$
XIV, *XV*, *XXI*, X = 4-Cl; A = $(C_2H_5)_2$
VII, X = 4-Cl; A = $O(CH_2CH_2)_2$
VIII, X = 6-Cl; A = $(CH_2)_5$
IX, X = 6-Cl; A = $O(CH_2CH_2)_2$
X, X = 5-[illegible] A = $O(CH_2CH_2)_2$
VI, X = 4-Cl; A = $(CH_2)_5$

XI, X = 6-Br; A = $O(CH_2CH_2)_2$
XII, X = H; A = $O(CH_2CH_2)_2$
XIII, X = H; A = $(CH_2)_5$
XVII, X = H
XVIII, X = 6-Cl
XIX, X = 4-Cl

XX, X = H
XXIII, X = 4-Cl; R = $CH_2C_6H_5$
XXIV, X = 4-Cl; R = $CH_2OC_6H_5$
XXV, X = 4-Cl; R = $CH_2OC_6H_4Cl$-(4)
XXVI, X = 4-Cl; R = $CH_2N(CO)_2C_6H_5$

In formula *XXVI* for R = $CH_2N(CO)_2C_6H_5$ read R = $CH_2N(CO)_2C_6H_4$.

Compounds *XV* and *XVI* were found to be little stable in solutions and on crystallization; anyhow, compound *XVI* is more stable. In polar solvents as *e.g.* in ethanol and dimethyl sulfoxide they are relatively stable and could crystallize from the former solvent. Recrystallization or a longer heating resulted in formation of formamidines *XIV*, *V* and the starting *I*. Decomposition proceeds much faster in little polar solvents as *e.g.* benzene or chloroform, from which only formamidines *V*, *XIV* and compound *I* could be obtained. Compound *XV* undergoes decomposition at standing in chloroform at room temperature and therefore, its ^{1}H NMR spectrum was recorded in dimethyl sulfoxide. Heating in acid medium (acetic acid) caused splitting of dialkylamine (reaction *c*).

Substances *XV* and *XVI* were acylated with phenylacetic, phenoxyacetic, 4-chlorophenoxyacetic, and phthalimidoacetic chlorides either in ether or in pyridine. In ether, 4-chloro-2-aminobenzothiazole was eliminated from *XV* and *XVI* and acylated; the second moiety passed into trisubstituted formamidine hydrogen chlorides (*XXI*, *XXII*), reaction e_1. In pyridine, compounds *XV* and *XVI* afforded with a two-molar amount of the acylating agent 4-chloro-2-acylamidobenzenethiazoles *XXIII*–*XXVI* in quantitative yield. Direct acylation of 4-chloro-2-aminobenzothiazole with acyl chlorides proceeded under more drastic conditions in c. 60% yield[15]. Free bases *V* and *XIV* were obtained from hydrogen chlorides *XXI* and *XXII* by treatment with sodium hydrogen carbonate.

N,N'-Bis(2-benzothiazolyl)formamidine is a good component for addition-elimination reactions; with 4-nitrophenylhydrazine it furnished the stable red $N_{(3)}$-(2-benzothiazolyl)-$N_{(1)}$-(4-nitrophenyl)formamidrazone (*XX*) in almost quantitative yield.

All reactions excepting e_1 (Scheme 1) led to one product only, this being checked by analysis of the reaction residues. The ^{1}H NMR spectra showed a well distinguishable signal of the methine group between nitrogen atoms at $\delta = 8{\cdot}40-8{\cdot}80$ ppm. The IR spectrum of formamidines revealed the stretching C=N vibration of medium intensity at 1 605–1 612 cm^{-1}, the ν(NH) appeared at 3 274 and 3 100 cm^{-1}.

EXPERIMENTAL

The ^{1}H NMR spectra of saturated dimethyl sulfoxide solutions were measured with a Tesla BS 487 apparatus operating at 80 MHz; the internal reference was hexadeuteriodimethyl sulfoxide. The IR spectra of Nujol mull were taken with a Perkin–Elmer, model 567 spectrophotometer in the 400–4 000 cm^{-1} range.

N,N-Dialkyl-N'-(X-2-benzothiazolyl)formamidine *V*–*XIII*

Benzenesulfonyl chloride (19 mmol) was added to a cooled and stirred solution of dialkylformamide (19 mmol) in pyridine at 0°C. The mixture was then stirred at 30°C and for 20 min and cooled to 0°C; the respective derivative of *I* (16 mmol) was added during 10 min and the stirred mixture was kept at 55–60°C for 1 h. The cooled mixture was poured into cold water (40 ml) and neutra-

TABLE I

Characteristic data of compounds *V*—*XIII* and *XXI*—*XXVI*

Compound	Formula M_r	M.p. °C	Yield %	Calculated/found					^{1}H NMR spectral data δ, ppm
				% C	% H	% N	% S	% X	
V	$C_{10}H_{10}ClN_3S$ 239·7	110—111	97·5	50·10 50·23	4·20 4·17	17·51 17·53	13·37 13·62	14·78 15·02	3·00 (3 H, s), 3·12 (3 H, s), 6·97—7·37 (2 H, m), 7·65 (1 H, q), J = 3 Hz, 8·40 (1 H, s)
VI	$C_{13}H_{14}ClN_3S$ 279·8	112—114	95·6	55·80 55·75	5·04 5·01	15·01 15·14	11·45 11·50	12·67 12·88	1·42 (6 H, s), 3·32—3·71 (4 H, m), 6·87—7·75 (3 H, m), 8·40 (1 H, s)
VII	$C_{12}H_{12}ClN_3OS$ 281·8	111—112	96	51·15 51·20	4·29 4·24	14·91 14·86	11·37 11·30	12·58 12·89	3·62 (8 H, d), 6·97—7·72 (3 H, m), 8·47 (1 H, s)
VIII	$C_{13}H_{14}ClN_3S$ 279·8	111—113	90	55·80 55·87	5·04 5·03	15·01 15·00	11·45 11·65	12·67 12·88	1·37—1·70 (6 H, s), 3·32—3·77 (4 H, m), 7·13—7·75 (3 H, m), 8·39 (1 H, s)
IX	$C_{12}H_{12}ClN_3OS$ 281·8	146—147	92	51·15 51·32	4·29 4·25	14·91 14·88	11·37 11·60	12·58 12·23	3·50—3·75 (8 H, m), 7·15—7·80 (3 H, m), 8·50 (1 H, s)
X	$C_{12}H_{12}BrN_3OS$ 326·16	136—138	93	44·18 44·42	3·70 3·56	12.88 13·21	9·82 10·13	24·49 23·96	3·62 (8 H, s), 7·40—8·20 (3 H, m), 8·50 (1 H, s)
XI	$C_{12}H_{12}BrN_3OS$ 326·16	148—150	94	44·18 44·27	3·70 3·57	12·88 13·14	9·82 10·21	24·49 23·84	3·64 (8 H, s), 7·37—8·17 (3 H, m), 8·50 (1 H, s)

XII	$C_{12}H_{13}N_3OS$ 247·2	131–132	97	58·29 58·44	5·26 5·28	16·99 17·03	12·96 13·23	— —	3·60 (8 H, d), J = 2 Hz, 7·00–7·73 (4 H, m), 8·47 (1 H, s)
XIII	$C_{13}H_{15}N_3S$ 245·3	76–79	84	63·64 63·63	6·16 6·31	17·12 17·12	13·06 13·21	— —	1·27–1·70 (6 H, m), 3·30–3·73 (4 H, m), 6·87–7·75 (4 H, m), 8·39 (1 H, s)
XXI	$C_{12}H_{15}Cl_2N_3S$ 304·2	163–165 (decomp.)	45	47·37 47·08	4·93 4·65	13·82 13·59	10·52 10·80	23·60 24·01	1·10–1·33 (6 H, m), 3·52–3·80 (4 H, m), 5·57 (NH, broad), 7·10–7·48 (2 H, m), 7·75–7·90 (1 H, d), J = = 8 Hz, 8·17–8·77 (1 H, d)
XXII	$C_{10}H_{11}Cl_2N_3S$ 276·0	165–168 (decomp.)	48	43·46 43·78	3·98 3·69	15·22 15·48	11·62 11·90	25·69 24·95	3·35 (3 H, s), 3·42 (3 H, s), 6·72 (NH, broad), 7·50–7·05 (2 H, m), 7·62–7·92 (1 H, m), 8·20, 8·87 (1 H, d)
XXIII	$C_{15}H_{11}ClN_2OS$ 302·7	183–185	90[a]	59·52 59·40	3·63 3·59	9·25 9·16	10·59 10·28	11·71 11·82	3·77 (2 H, s), 7·07–7·87 (9 H, m)
XXIV	$C_{15}H_{11}ClN_2O_2S$ 318·5	147–148 (ethanol)	92[a]	56·53 56·48	3·45 3·51	8·78 8·93	10·06 10·14	11·12 11·34	4·87 (2 H, s), 6·72–7·90 (9 H, m)
XXV	$C_{15}H_{10}Cl_2N_2O_2S$ 352·9	176·178	90[a]	51·02 50·91	2·83 2·79	7·92 7·58	9·07 8·91	20·07 19·54	4·84 (2 H, s), 6·75–7·87 (8 H, m)
XXVI	$C_{17}H_{10}ClN_3O_3S$[b] 371·8	331–332	95[a]	54·91 54·89	2·71 2·51	11·30 11·28	8·62 8·59	9·53 9·31	

[a] *via* path e_2 (*cf.* Scheme 1); [b] insoluble.

lized with saturated $NaHCO_3$ solution. The pasty product gradually solidified on standing in the $NaHCO_3$ solution. The precipitate was filtered off and crystallized from ethanol. Yields varied within 95—98%.

N,N-Diethyl-N′,N″-bis(4-chloro-2-benzothiazolyl)triaminomethane (*XV*)

Two procedures were applied for preparation of the title product, the first being that given for compounds *V*—*XIII* with N,N-diethylformamide as the reagent. Yield 98%. The second procedure differed from the first one in the temperature only, which was kept at 0°C. Yield 83%, m.p. 135—137°C. The product was crystallized from ethanol, since in benzene a partial decomposition occured leading to compounds *I* and *XIV*. For $C_{19}H_{19}Cl_2N_5S_2$ (452·4) calculated: 50·44% C, 4·23% H, 15·47% N, 15·67% Cl, 14·17% S; found: 50·12% C, 4·25% H, 15·29% N, 15·54% Cl, 14·17% S. ^{1}H NMR spectrum: (δ, ppm): 1·00—1·20, (6 H, m), 3·25—3·57 (4 H, m), 6·77—7·87 (8 H, m), 8·37 (1 H, s); (C^2HCl_3): 1·05—1·37 (6 H, m), 3·27—3·75 (4 H, m), 6·67—7·65 (6 H, m), 8·45 (1 H, s). IR spectrum (cm^{-1}): 3 274 ν(NH), 3 100 ν(NH), 2 950 ν(CH), 1 608 ν(C=N).

N,N-Dimethyl-N′,N″-bis(4-chloro-2-benzothiazolyl)triaminomethane (*XVI*)

Compound *I* (2·6 mmol) was added to the solution of *V* (2·6 mmol) in pyridine (20 ml) and the mixture was stirred at 30°C for 20 min and at 50°C for additional 2 h. The cooled solution was poured into water (30 ml) and stirred at 10°C for 2 h, the product was filtered off and crystallized from ethanol. Yield 98—100%, m.p. 146—147°C. For $C_{17}H_{15}Cl_2N_5S_2$ (424·4) calculated: 48·22% C, 3·33% H, 16·54% N, 16·74% Cl, 15·14% S; found: 48·22% C, 3·28% H, 16·28% N, 16.41% Cl, 15·10% S. ^{1}H NMR spectrum (δ, ppm): 3·02 (1 H, s), 3·15 (3 H, s), 6·75—8·20 (8 H, m), 8·40 (1 H, s); ($CDCl_3$): identical with that of *V*. IR spectrum (cm^{-1}): 3 220 ν(NH), 3 090 ν(NH), 1 622 (ν(C=N).

N,N′-Bis(2-benzothiazolyl)formamidine (*XVII*)

According to procedure given for compounds *V*—*XIII* and employing diethylformamide (1·63 g, 16·1 mmol), benzenesulfonyl chloride (2·84 g, 16·1 mmol), 2-aminobenzothiazole (2 g, 13·4 mmol), and pyridine (8 ml), the product was obtained in 79% yield; m.p. 261—262°C (decomp., ethanol). For $C_{15}H_{10}N_4S_2$ (310·4) calculated: 58·04% C, 3·24% H, 18·04% N, 20·65% S; found: 57·94% C, 3·17% H, 18·16% N, 20·75% S. ^{1}H NMR spectrum (δ, ppm): 7·12—7·93 (9 H, m), 8·80 (1 H, d). IR spectrum (cm^{-1}): 3 210, 3 060 ν(NH), 1 605 ν(C=N).

N,N′-Bis(6-chloro-2-benzothiazolyl)formamidine (*XVIII*)

Applying procedure for synthesizing *XVII*, the title product was obtained from 6-chloro-2-aminobenzothiazole (2·5 g, 13·4 mmol) in 78% yield, m.p. 325—227°C (decomp., tetrahydrofuran). For $C_{15}H_8Cl_2N_4S_2$ (379·3) calculated: 47·50% C, 2·12% H, 14·77% N, 16·90% S; found: 47·58% C, 1·90% H, 14·83% N, 17·28% S. IR spectrum (cm^{-1}): 3 220, 3 100 ν(NH), 2 980 ν(CH), 1 604 ν(C=N).

N,N′-Bis(4-chloro-2-benzothiazolyl)formamidine (*XIX*)

Compound *XV* or *XVI* (4 mmol) was dissolved in acetic acid (20 ml) at an ambient temperature, and heated to 90°C for 30 min while stirred. Product *XIX* gradually separated as a light-yellow precipitate in 90% yield, m.p. 323—325°C (decomp.). For $C_{15}H_8Cl_2N_4S_2$ (379·3) calculated: 47·50% C, 2·12% H, 14·77% N, 16·90% S; found: 47·54% C, 1·82% H, 14·67% N, 17·29% S.

N,N-Diethyl-N′-(4-chloro-3-benzothiazolyl)formamidine (*XIV*)

Compound *XXI* (4 mmol) in 2% $NaHCO_3$ (30 ml) was stirred at 25°C for 10 min. Product *VXI* separating during stirring was filtered off and crystallized from ethanol. M.p. 38—42°C, yield 100%; yield *per* compound *I via* reactions a_2 and e_1 was 45%. For $C_{12}H_{14}ClN_3S$ (267·7) calculated: 53·79% C, 5·04% H, 15·69% N, 11·95% S; found: 53·84% C, 5·04% H, 15·86% N, 12·09% S. ^{1}H NMR spectrum (δ, ppm): 0·97—1·27 (6 H, m), 3·23—3·63 (4 H, m), 6·87—7·35 (2 H, m), 7·55—7·65 (1 H, m), 8·37 (1 H, s).

$N_{(3)}$-(2-Benzothiazolyl)-$N_{(1)}$-(4-nitrophenyl)formamidrazone (*XX*)

4-Nitrophenylhydrazine (3 mmol) was added to *XVII* (3 mmol) in ethanol (15 ml) and refluxed for 30 min. The red, gradually separating compound was filtered off and crystallized from tetrahydrofuran; m.p. 248 C (decomp.). For $C_{14}H_{11}N_5SO_2$ (313·2) calculated: 53·67% C, 3·51% H, 22·36% N, 10·22% S; found: 53·81% C, 3·53% H, 22·15% N, 10·00% S.

N-(4-Chloro-2-benzothiazolyl)benzenesulfoammide (*XXVII*)

A mixture of *II* (10 mmol), *I* (10 mmol), *III* (20 mmol), and *IV* (15 ml) was stirred at 90°C for 2h, cooled and poured into cold water. The separated product was filtered off and crystallized from ethanol; m.p. 219—221°C. For $C_{13}H_9ClN_2O_2S_2$ (324·8) calculated: 48·07% C, 2·74% H, 8·62% N, 10·92% Cl, 19·74% S; found: 47·96% C, 2·56% H, 8·67% N, 10·80% Cl, 19·56% S.

Acylated N,N-Dialkyl-N′,N″-bis(chloro-2-benzothiazolyl)triaminomethanes

a) Applying procedure e_1 (compounds *XXI*—*XXIV*): an ethereal solution of phenylacetic, or phenoxyacetic chloride (9·0 mmol, 20 ml) was dropwise added to *XV* or *XVI* (4·4 mmol) dissolved in ether (20 ml). The temperature was raised and the stirred solution was refluxed for 2 h, cooled and left to stand overnight in a refrigerator. The separated substance was filtered off and dissolved in ether (60 ml) under reflux. The insoluble hydrogen chloride *XXI* or *XXII* was filtered off, whilst the acyl derivatives *XXIII* or *XXIV*, separating after removal of the solvent were crystallized from ethanol (Table I).

b) According to procedure e_2 (compounds *XXIII*—*XXVI*): an ethereal solution of either phenylacetic or phenoxyacetic or 4-chlorophenoxyacetic, or phthalimidoacetic chloride (9 mmol) was added at 0°C to *XV* or *XVI* (4·4 mmol) dissolved in ether (20 ml), pyridine (2 ml). The mixture was then refluxedfor 3 h and the solvent was removed. Cold water (50 ml) and $NaHCO_3$ (10 mmol) were added to the residue, the content was homogenized, the separated precipitate was filtered off and crystallized from ethanol (Table I).

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