BEHAVIOR OF 1,5-DIHYDRO-6-OXO-1, 2,4-TRIAZINO [3,4-B] BENZOISOTHIAZOLE SULPHUR DIOXIDE TOWARDS CARBON ELECTROPHILES, CHLORINE NUCLEOPHILES AND SOME STUDIES WITH THE PRODUCTS

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Abstract: The reaction of triazino benzo isothiazole sulphur dioxide with carbon electrophiles namely, α-cyanocinnamonitrile, α-ethoxycarbonyl cinnamonitrile, α-cyanocinnamamide, formaldehyde piperidine, and ethyl chloroacetate has been investigated. Also it reacted with chlorine nucleophile and gave the chloro derivative. Its behavior towards nitrogen nucleophiles namely, acylhydrazine, aminoacids, aminophenols, hydrazine hydrate, ammonium acetate and sodium azide has been discussed.
Key words: Isothiazole; triazinoamine; triazino (4, 3-b) saccharine

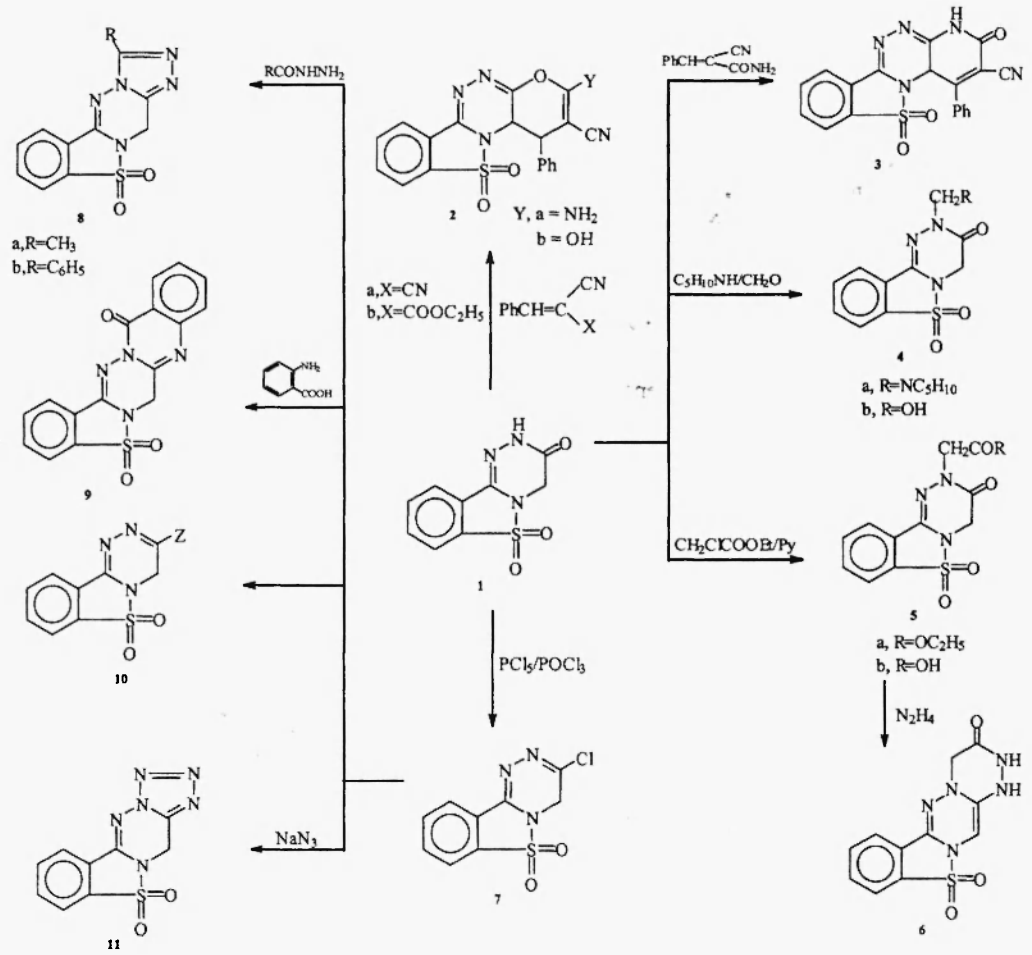
Introduction

The title compound contains saccharine counter part and has multi-function groups. This prompted us to use it as a key starting material for synthesis of a diverse of some heterocyclic compounds with the expected pharmaceutical action. [1-4]

Thus when compound 1,5-dihydro-6-oxo-1,2,4-triazino [3,4-b] benzoisothiazole sulphur dioxide (**1**) was allowed to react with α-cyanocinnamonitrile and/or α-carboethoxycinnamonitrile in n-butanol in the presence of triethylamine as a catalyst [5,6] afforded 6-amino-5-cyano-4-phenyl-3,4-dihydropyrano[2,3-e]1,5-dihydro-6-oxo-1,2,4-triazino [3,4-b] benzoisothiazole sulphur dioxide (**2a**) and /or 6-hydroxy-5-cyano-4-phenyl-3,4-dihydropyrano[2,3-e]1,5-dihydro-6-oxo-1,2,4-triazino [3,4-b] benzo isothiazole sulphur dioxide (**2b**) . IR spectra of compounds (**2**) exhibits strong absorption bands at 2250,1610 attributable to vC N and vC=N respectively, a band at 3100 vNH for (**2a**) and a band at 3400 cm^{-1} vOH for 2b. Such IR data agreed well with the proposed structure. The ^{1}H-NMR spectrum of compound (**2a**) when run in DMSO shows the following signals at δ2.1(m, 2H,methine protons); δ5.6 (broad singlet,2H, NH_2; due to quadrapole of nitrogen); δ7.2-8.1(m, 9H, aromatic protons). Similarly, the compound (**1**) has been reacted with α-nitilecinnamamide [7-9] under practically the same experimental conditions and afforded 5-cyano-6-oxo-4-phenyl-1,3-dihydropyridino[2,3-e]1,5-dihydro-6-oxo-1,2, 4-triazino [3,4-b] benzoisothiazole sulphur dioxide (**3**). Its IR spectrum exhibits absorption bands at 3320, 3100 (nonbonded and bonded vNH), 2250 (vC N), 1670 (vC=O) and 1620 cm^{-1}(vC=N). The ^{1}H-NMR spectrum showed the following signals δppm 2.7 (s, 1H, methine proton, 5.7(broad singlet NH proton due to quadrapole of nitrogen which disappear completely when spectrum runs in D_2O); and 7.4-8.1(m, 9H, aromatic protons). Such spectroscopic data is consistent with the proposed structure.

When compound (**1**) was allowed to react with piperidine and formaldehyde in boiling ethanol [10] yielded the Mannich base "1–piperidino methyl-5H-6-oxo-1,2,4-triazino [3,4-b] benzo isothiazole sulphur dioxide" (**4a**) through an isolated intermediate 1-hydroxymethyl-5H-6-oxo-1,2,4-triazino [3,4-b] benzoisothiazole sulphur dioxide (**4b**), which when submitted to react with piperidine under the same condition afforded (**4a**) which identified via melting point and mixed melting point determination. The IR spectrum of (**4a**) revealed strong absorption bands at 1670, 1620cm^{-1} attributable to vC=O and vC=N respectively.

On the other hand, when compound (**1**) was submitted to react with ethylchloroacetate in the presence of pyridine as a catalyst [11] gave 1-ethoxycarbonylmethyl-5H-6-oxo-1,2,4-



10a,Z = $NHC_6H_4COOH(m)$
b,Z = $NHC_6H_4COOH(p)$
c,Z = $NHCH_2COOH$
d,Z = $NHC_6H_4OH(o)$
e,Z = $NHC_6H_4OH(m)$
f,Z = $NHC_6H_4OH(p)$
g,Z = $NH\text{-}NH_2$
h,Z = $NHN=CHC_6H_5$
i,Z = $NHN=C_6H_4Cl(p)$
j,Z = $NHN=CHC_6H_4OCH_3(p)$
k,Z = NH_2

triazino [3,4-b] benzoisothiazole sulphur dioxide (**5a**). IR spectrum of compound (**5a**) exhibits strong absorption bands at 1742, 1670, 1620cm^{-1} attributable to ν_{max} of two carbonyl groups and νC=N respectively. ^{1}H-NMR spectrum of (**5a**) showed the following signals δppm 1.1(t, 3H $CH_2\underline{CH_3}$); 2.7(s, 2H, CH_2-CO-); 4.1(q, 2H, $\underline{CH_2}CH_3$); 4.5(s, 2H, N-CH_2-COOEt); and 7.2-7.9 (m, 4H, ArH).

Also structure of compound (**5a**) was proved chemically via: (i) The alkaline hydrolysis which afforded the corresponding acid 1-carboxymethyl-5H-6-oxo-1,2,4-triazino [3,4-b] benzoisothiazole sulphur dioxide (**5b**), its IR spectrum exhibits strong absorption bands at 1715, 1670, 1615 due to ν_{max} of two carbonyl and νC=N, (ii) Hydrazinolysis of (**5a**) gave the nitrogen bridgehead compound 1,2,5-trihydro-6-oxo-1,2,4-triazino[3,4-f](1,2,4-triazino[3,4-b]benzoisothiazole sulphur dioxide) (**6**) via hydrazide formation, followed by ring closure [12]. IR spectrum of compound (**6**) revealed strong absorption bands at 3380, 3150 (bonded and nonbonded NH), 1660, 1620 cm^{-1}due to νC=O and νC=N respectively. ^{1}H-NMR spectrum in DMSO showed signals at δ3.3(s, 2H, methylene protons), 5.5(s, 2H, broad 2NH, due to quadrapole of nitrogen), 6.1(s, 1H,olefinic proton), and δ7.1-7.8(m, 4H, aromatic protons).

Treatment of compound (**1**) with a mixture of PCl_5/$POCl_3$ on boiling water bath [13] afforded 5-hydro-6-chloro [1,2,4-] triazino [4,3-b] benzoisothiazole sulphur dioxide (**7**). Structure of compound (**7**) was inferred from its behavior towards nitrogen nucleophiles.

Thus when compound (**7**) was allowed to react with acylhydr-azines[14 15] namely, acetyl hydrazine, and benzoyl hydrazine in boiling butanol yielded heteroannulated compounds 5-methyl-1,2,4-triazolo [3,4-f]-5H-1,2,4-triazino [3,4-b] benzoisothiazole sulphur dioxide (**8a**) and 5-phenyl-1,2,4-triazolo [3,4-f]-5H-1,2,4-triazino [3,4-b] benzoisothiazole sulphur dioxide(**8b**) respectively. IR spectra of compounds (**8a,b**) exhibit strong absorption in the region 1615-1630 due to νC=N. ^{1}H-NMR spectrum of (**8b**) when runs in DMSO exhibits signals at δppm 2.7(s, 2H, methylene protons) and 7.1-8.2 (m, 9H,aromatic protons).

Anthranilic acid has been reacted also with chloro derivative (**7**) by fusion into an oil bath at 170 °C [16,17] and gave 4-oxoquinazolino[2,3-f]-5H-1,2,4-triazino [3,4-b] benzoisothiazole sulphur dioxide (**9**) whose structure was inferred from its IR spectrum which reveals strong absorption bands at 1675 and 1615 cm^{-1} due to νC=O and νC=N respectively.

On the other hand, m-aminobezoic and p-aminobenzoic have been reacted with the chloro derivative (**7**) under the same condition and gave the corresponding carboxyphenyl triazinoamine derivatives 6-(3-carboxy aniline) 5-hydro-1,2,4- triazino [4,3-b] benzoisothiazole sulphur dioxide (**10a**) and 6-(4-carboxy aniline) 5-hydro-1,2,4-triazino [4,3-b] benzoisothiazole sulphur dioxide (**10b**). The IR spectra of compounds (**10a**) and (**10b**) showed strong absorption bands in the region 3400-3350 (basin peak); 3200-3100; 1685-1680; 1620-1615 cm^{-1} attributable to νOH, νNH, νC=O and νC=N respectively. Simillary, glycine reacted with compound (**7**) and yielded 6-(carboxy methyl aniline) 5-hydro-1,2,4-triazino [4,3-b] benzoisothiazole sulphur dioxide (**10c**). Its IR spectrum exhibits absorption bands at 1715 and 1625 due to νC=O and νC=N.

Recently, it was reported that N-alkyl/or arylamino derivatives of analogous triazine have antidepressant activity (on the CNS, central nervous system). In this article the outhor thought to synthesise N-alkyl or aryl amino triazinobenzoisothiazole sulphur dioxide in purpose of their expected pharmaceutical activity. Thus when the chloro derivative (**7**) was allowed to react with aminophenols namely, o-, m- and p- aminophenols in an oil bath at 170 °C afforded 6-substituted-5-hydro-1,2,4-triazino [4,3-b] benzoisothiazole sulphur dioxide (**10d-f**). The IR spectra of compounds (**10d-f**) reveal strong absorption bands at 3400-3420; 3200-3260; 1620-1625cm^{-1}attributableto νOH, νNH and νC=N respectively.

When the chloro derivative (7) was submitted to react with hydrazine hydrate by fusion, it yielded 6-hydrazino-5-hydro-1,2,4-triazino[4,3-b] benzoisooxazole sulphur dioxide (**10g**), its structure was supported from: (i) Its IR spectrum showed absorption bands at 3300, 3210, 3150 and 1620 cm^{-1} due to ν_{max} of NH and C=N respectively. (ii) When the hydrazino derivative was allowed to react with aromatic aldehydes namely, bezaldehyde, p-chlorobezaldehyde and p-methoxy-benzaldehyde afforded the 6-substituted-5-hydro-1,2,4-triazino [4,3-b] benzoisothiazole sulphur dioxide (**10h-j**) respectively. Similarly, ammonolysis of chloro derivative **7** via its treatment with ammonium acetate at 170 °C in an oil bath gave 6-amino-5-hydro-1, 2,4-triazino [4,3-b] benzisothiazole sulphur dioxide (**10k**). Its IR spectrum reveals strong absorption bands at 3350, 3200 and 1620 cm^{-1} attributable to vNH (nonbonded and bonded) and vC=N respectively.

Finally when the chloro derivative (7) reacted with sodium azide in boiling acetic acid [18 19] it yielded 1,2,3,4-tetrazolo[4,5-f]- 5-hydro-1, 2,4-triazino [4,3-b] benzoisothiazole sulphur dioxide (**11**). IR spectrum of compound (**11**) reveals absorption bands at 1620 and cm^{-1} due to vC=N and tetrazole moiety.

EXPERIMENTAL

All melting points reported are uncorrected. The IR absorption spectra were determined with a "Pye Unicam Ltd." Spectro-photometer using KBr pellet wafer technique. ^{1}HNMR data were recorded on a Varian instrument division EM-360, 60 MHz NMR spectrophotometer, (DMSO) TMS an internal reference or/and the chemical shifts are reported in PPM (δ) relative to internal TMS-characterization and physical data are listed in table 1.

Reaction of Saccharinotriazino (1) with α-cyanocinnamonitrile, α-ethoxy carboxycinnamonitrile and cinnamonitrile amide.
Formation of compounds (2a-b) and (3)

A solution of saccharinotriazinone (1) (0.01mole) in n-butanol (30ml) containing trimethylamine (0.5ml) was treated with α-cyanocinn- amonitrile, α-ethoxy carboxy cinnamonitrile and/or cinnamonitrile amide, the whole was heated under reflux for 4-6 hrs. The solid products obtained after cooling was crystallized from the proper solvents to give (**2a-b**) and (**3**).

Action of piperidine on saccharinotriazinone (1) in presence of form-aldehyde.
Formation of Mannich base N-methyl piperidinotriazinone (4a-b)

A mixture of (1) (0.01mole), formaldehyde (0.01mole) and piperi-dine (0.01mole) in boiling ethanol (30ml) was refluxed for 3 hrs, after cooling the separated product was crystallized from the proper solvent to give Mannich base (**4a-b**).

Action of ethylchloroacetate on saccharinotriazinone (1).

Formation of compound (5a)

A mixture of saccharinotriazinone (1) (0.01mole) and ethylchloro acetate (0.01mole) in least amount of pyridine was heated on a steam bath for 3 hrs. The reaction mixture poured into water-dil.HCl. The separated solid was crystallized from ethanol to give (**5a**).

Alkaline hydrolysis of ester (5a). Formation of compound (5b)

The ester (**5a**) was refluxed with 10% aqueous sodium hydroxide solution (10ml per gm of ester) for 3 hrs. The alkaline solution was acidified with HCl and extracted with ether. The separated solid after evaporation the ether was crystallized from ethanol to give the corresponding acid (**5b**).

Table 1. Microanalysis of Prepared Compounds

Comp	Formula of M. Wt	m.p.°C Colour	Solvent Yield %	Analysis % Required / Found				
				C	H	N	S	Cl
1	$C_9H_7N_3O_3S$ 251	145 yellowish brown	EtOH 35	43.0 42.9	2.79 2.76	16.7 16.4		
2a	$C_{19}H_{13}N_5O_3S$ 391	85 golden yellow	Benz. 70	58.3 58.9	3.3 3.7	17.9 18.3	8.18 8.33	
2b	$C_{19}H_{12}N_4O_4S$ 392	205 yellow	Toluene 73	58.1 58.5	3.1 3.3	14.2 14.9	8.2 8.4	
3	$C_{19}H_{11}N_5O_3S$ 389	109 yellowish white	Benz. 68	58.6 58.9	2.8 3.1	17.9 18.3	8.2 8.6	
4a	$C_{15}H_{18}N_4O_3S$ 334	70 yellowish brown	Toluene 45	53.8 53.7	5.39 5.1	16.7 16.4	9.5 9.1	
4b	$C_{10}H_9N_3O_4S$ 267	92 yellow	Toluene 60	44.9 44.6	3.4 3.1	15.7 15.4	11.9 11.5	
5a	$C_{13}H_{13}N_3O_5S$ 333	210 pale yellow	EtOH 70	46.8 46.5	3.9 4.2	12.6 12.2	9.6 9.4	
5b	$C_{11}H_9N_3O_5S$ 305	195 pale yellow	EtOH 65	43.2 43.6	2.9 2.6	13.7 13.3	10.5 10.2	
6	$C_{11}H_9N_5O_3S$ 291	95 yellow	EtOH 70	45.3 45.7	3.1 3.4	24.0 24.3	10.1 10.4	
7	$C_9H_6N_3O_2SCl$ 255.5	105 brown	Benz. 75	42.3 41.9	2.3 2.1	16.4 16.2	12.5 12.3	13.9 13.6
8a	$C_{11}H_9N_5O_2S$ 275	190 yellow	Toluene 80	48.0 48.3	3.2 3.5	25.4 25.7	11.6 11.9	

Table 1. (Cont.).

8b	$C_{16}H_{11}N_5O_2S$ 337	170 golden yellow	Benz. 70	56.9 57.2	3.2 3.5	20.7 20.9	9.4 9.7	
9	$C_{16}H_{10}N_4O_3S$ 338	238 buff	Toluene 73	56.8 56.4	2.96 2.91	16.5 16.2	9.4 9.1	
10a	$C_{16}H_{12}N_4O_4S$ 356	225 pink	Benz. 68	53.9 53.6	3.37 3.11	15.7 15.4	8.9 8.6	
10b	$C_{16}H_{12}N_4O_4S$ 356	250 deep yellow	Toluene 60	53.9 53.7	3.37 3.11	15.7 15.5	8.8 8.5	
10c	$C_{11}H_{10}N_4O_4S$ 244	320 pale yellow	ACOH 73	44.9 44.6	3.4 3.11	15.7 15.4	13.1 12.8	
10d	$C_{15}H_{12}N_4O_3S$ 328	115 yellow	Ether 45	54.8 54.5	3.66 3.41	17.1 16.8	9.7 9.4	
10e	$C_{15}H_{12}N_4O_3S$ 328	110 brown	EtOH 60	54.8 54.6	3.65 3.42	17.1 16.7	9.7 9.4	
10f	$C_{15}H_{12}N_4O_3S$ 328	108 pale yellow	EtOH 70	54.8 54.6	3.65 3.42	17.1 16.7	9.7 9.4	
10g	$C_9H_9N_5O_2S$ 251	75 buff	Butanol 75	43.1 42.7	3.8 3.5	27.9 27.6	12.7 12.5	
10h	$C_{16}H_{13}N_5O_2S$ 339	120 yellow	EtOH 40	56.6 56.3	3.8 3.6	20.6 20.4	9.4 9.1	
10i	$C_{16}H_{12}N_5O_2SCl$ 373.5	200 yellow	EtOH 63	51.4 51.2	3.2 2.9	18.7 18.4	8.5 8.2	9.5 9.1
10j	$C_{17}H_{15}N_5O_3S$ 369	130 pale yellow	ACOH 55	55.3 55.0	4.0 3.8	18.9 18.6	6.8 6.6	
10k	$C_9H_8N_4O_2S$ 236	247 Brown	Toluene 75	45.7 45.5	3.4 3.1	23.7 23.4	13.9 13.6	
11	$C_9H_6N_6O_2S$ 262	130 pale yellow	EtOH 62	41.2 40.8	2.3 2.0	32.1 31.8	12.2 11.9	

Condensation reaction of N-hydroxy carboxy methyl saccharino tria-zinone (5a) with hydazine hydrate. Formation of compound (6)

A mixture of N-hydroxy carboxy methyl saccharinone (**5a**) (0.01mole) submitted to react with hydrazine hydrate (0.015mole) by heating under refluxing in ACOH for 3 hrs. After filtration, concentration and cooling the separated solid was crystallized from ethanol to give (**6**)

Action of $POCl_3/PCl_5$ on the saccharinotriazinone (1). Formation of chloro saccharinotriazinone (7)

A mixture of (1) (0.01mole), phosphorous oxytrichloride (0.02mole) and phosphorous pentachloride (1gm) was refluxed on a steam bath for 3 hrs. Then poured slowly into ice-cold water. The solid that separated was washed several times with water, dried and crystallized from benzene to give (7).

Action of acylhydrazine on chloro saccharinotriazinone (7).
Formation of compounds (**8a**,b)

A mixture of (7) (0.01mole) and acyl hydrazine namely, acetyl hydrazine and benzoyl hydrazine in n-butanol (50ml) was heated under reflux for 5 hrs. The separated solid after concentration and cooling was filtered off and crystallized from proper solvent to give (**8a,b**)

Condensation reaction of aromatic amino acids with chloro sacchar-inotriazine (7).
Formation of compounds (9,10a,b)

A mixture of chloro saccharinotiazine (7) (0.01mole) and amino acids namely,anthranilic acid, m-aminobenzoic acid and p-aminobenzoic acid (0.01mole) in ethanol (30ml) was heated under reflux into oil bath at 170°C for 3 hrs. After cooling water was added and the solid obtained filtered off and was crystallized from proper solvent to give (**9,10a** and **b**) respectively.

Reaction of chloro saccharinotriazine (7) with glycine.
Formation of compound (10c)

A mixture of (7) (0.01mole) and glycine (0.015mole) was heated under reflux for 3 hrs into ethanol (30ml). The solid that separated after concentration and cooling filtered off and crystallized from suitable solvent to give compound (**10c**).

Action of aminophenols on chloro saccharinotriazine (7).
Formation of compounds (10d-f)

A solution of (7) (0.01mole) and o-, p- and m-aminophenols (0.015mole) into benzene (50ml) was heated under reflux for 5 hrs. Then the separated solid after concentration and cooling was crystallized from suitable solvent to give (**10d-f**).

Reaction of hydrazine hydrate with chloro saccharinotriazine (7).
Formation of compound (10g)

A mixture of chloro saccharinotriazine (7) (0.01mole) and hydrazine hydrate (0.015mole) into n-butanol was heated under reflux for 4 hrs. Then the mixture was poured in water. The precipitated solid was filtered off, dried and crystallized from acetic acid to give (**10g**)

Reaction of hydrazine derivative (10g) with aromatic aldehyde.
Formation of compounds (10h-j)

A mixture of hydrazine (**10g**) (0.01mole) and aromatic aldehydes, namely, benzaldehyde, p-chlorobenzaldehyde and p-methoxy benzald-ehyde (0.015mole) was heated under reflux for 4 hrs into n-butanol. The solid that separated after concentration and cooling was crystallized from proper solvent to give compounds (**10h-j**)

Treatment of chloro derivative (7) with ammonium acetate.

Formation of compound (10k)

A mixture of chloro saccharainotriazine (7) (0.01mole) and ammonium acetate (0.012mole) was fused at 170 °C on oil sand bath for 2 hrs, with continuously stirring. Poured into water, then the precipitated solid was filtered off and crystallized from toluene to give (**10k**)

Reaction of sodium azide with chloro saccharinotriazine (7).

Formation of compound (11)

A mixture of chloro derivative (7) (0.01mole) and sodium azide (0.015mole) was heated under reflux for 3 hrs, into acetic acid.The solid that separated after concentration and cooling was filtered off and crystallized from suitable solvent to give compound (11).

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