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A Novel Synthesis of Heterocycles from Thiocarbohydrazides

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Summary. The reaction of thiocarbohydrazides **1a** and thiocarbazones **1b** with tetracyanoethylene (*TCNE*) afforded the thiazol, thiadiazole, thiazine, and thiadiazepine derivatives **4**–**7**. 2-Dicyanomethyleneindane-1,3-dione (*CNIND*) reacted with **1a,b** to give aminoindenopyrazolopyridazinone (**12**) and phenyl-1,2,3,4-tetraazacyclopentafluorene (**13**). The indazole and oxathiadiazole derivatives **17** and **19** were formed during the reaction of **1b** with 2,3-dichloro-5,6-dicyano-1,4-benzoquinone (*DDO*). 6,7-Dichloro-5-phenylpyrazolophthalazinol (**21**) was obtained from the reaction of **1b** with 2,3,5,6-tetrachloro-1,4-benzoquinone (*CHL-p*). The oxidative cyclization of thiodicarbazones **2a**–**d** with the above acceptors afforded the thiadiazole and thiadiazine derivatives **8** and **10**.

Keywords. Thiocarbohydrazides; π-Acceptors; Heterocyclic compounds.

Eine neue Synthese von Heterocyclen aus Thiocarbohydraziden

Zusammenfassung. Die Reaktion der Thiocarbohydrazide **1a** und der Thiocarbazone **1b** mit Tetracyanoethylen (*TCNE*) ergab die Thiazol-, Thiadiazol-, Thiazin- und Thiadiazepinderivate **4**–**7**. 2-Dicyanomethylenindan-1,3-dion (*CNIND*) liefert mit **1a,b** Aminoindenopyrazolopyridazinon (**12**) und Phenyl-1,2,3,4-tetraazacyclopentafluoren (**13**). Die Indazol- und Oxathiadiazolderivate **17** und **19** wurden durch Reaktion von **1b** mit 1,3-Dichlor-5,6-dicyano-1,4-benzochinon (*DDQ*) gebildet. 6,7-Dichlor-5-phenylpyrazolophthalazinol (**21**) wurde aus **1b** und 2,3,5,6-Tetrachlor-1,4-benzochinon (*CHL-p*) erhalten. Die oxidative Cyclisierung der Thiodicarbazone **2a**–**d** mit den obengenannten Akzeptoren ergab die Thiadiazol- und Thiadiazinderivate **8** und **10**.

Introduction

In recent years there has been increasing interest in the synthesis of heterocyclic compounds by cyclization of appropriate linear compounds. Organosulfur compounds play an important role in modern organic synthesis, not only because they constitute a particularly useful class of synthons [1], but also because they are of great biological interest [2]. Semi- and thiosemicarbazones of aldehydes are suitable substrates for the preparation of five or six membered heterocyclic rings containing three heteroatoms [3–5].

Recently, we have described the synthesis of thiazine, thiadiazine, thiadiazole, indazole, and pyridazine as well as pyridazino[4,5-*d*]pyridazine derivatives by the interaction of *TCNE* and benzo- as well as naphthoquinones with thiosemicarbazide, dithiocarbazate, and its azomethine derivatives [6–9].

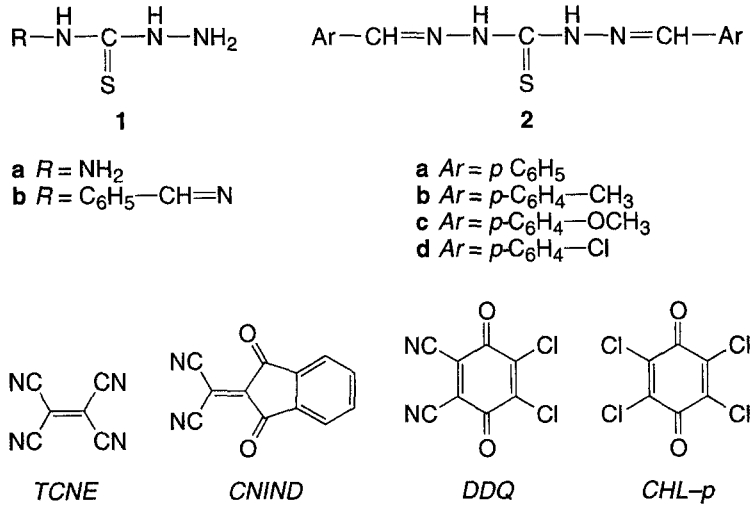


Fig. 1

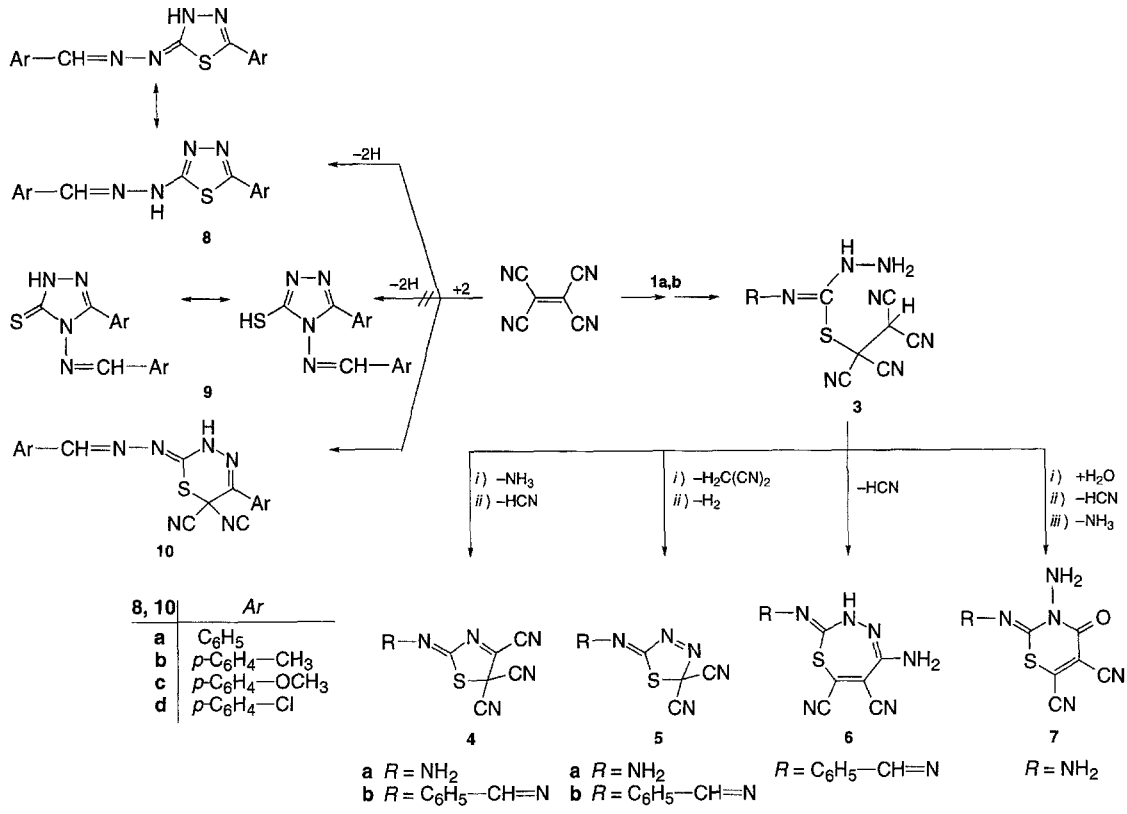
In the light of the above observations, we have selected the biologically active thiocarbohydrazide [10] and thiodicarbazone derivatives **1** and **2** (Fig. 1) as starting compounds for the synthesis of a variety of new heterocyclic ring systems with the expectation that they will be of biological interest.

Results and Discussion

When compounds **1a,b** were allowed to react with *TCNE* followed by chromatographic separation of the reaction mixture, several zones were observed from which products **4–7** could be isolated (Scheme 1). Initially, thiocarbazides **1a,b** and *TCNE* gave the intermediate tetracyanoethane derivatives **3**. Elimination of one molecule of HCN and one of ammonia from **3** afforded 2-hydrazino- and benzalhydrazono-4,5,5-tricyanothiazole (**4a,b**). 2-Hydrazino- and benzalhydrazono- 5,5-dicyanothiadiazole (**5a,b**) were formed after elimination of a molecule of malononitrile from **3** followed by dehydrogenation. Cyclization of the intermediate **3** afforded 3*H*-2-benzalhydrazono-5-amino-6,7-dicyano-1,3,4-thiadiazepine (**6**) or 2-hydrazino-3-N-amino-5,6-dicyano-1,3-thiazin-4-one (**7**) *via* elimination of HCN as well as abstraction of H_2O and HCN and NH_3, respectively.

Assignment of the structures of compounds **4–7** was based on their spectroscopic data and combustion analysis (see Tables 1 and 2).

For example, the ^{1}H NMR spectrum of **6** confirmed the presence of a thiadizepine-NH at $\delta = 10.05$ ppm and an amino group at 4.50 ppm in addition to the aromatic protons as well as an azomethine-CH. The ^{1}H NMR spectrum of **7** revealed the presence of two amino groups, one at $\delta = 5.10$ ppm and the other at 6.14 ppm. The IR spectra of **6** and **7** showed the presence of conjugated cyano groups at 2235, 2207, and 2242 cm^{-1}, in addition to a carbonyl group at 1708 cm^{-1} in compound **7**. The mass spectra of **6** and **7** gave the correct molecular weight which was also supported by combustion analysis.



Scheme 1

CNIND reacted with **1a,b** to give 1-aminoindeno[1,2-*c*]pyrazolo[4,3-*e*]pyridazin-10(1*H*)-one (**12**) and 5-phenyl-2*H*-1,2,3,4-tetraazacyclopenta[*jk*]fluorene (**13**, Scheme 2).

The ^{1}H NMR spectra of **12** and **13** revealed the presence of a broad peak at $\delta = 13.40$–13.50 ppm due to the pyrazole-NH, and another one in **12** at $\delta = 6.30$ ppm due to the NH_2 group, in addition to the aromatic protons. The IR spectra clearly indicate the presence of a carbonyl group in **12** and the absence of a cyano one. However, in compound **13** both carbonyl and cyano groups were absent. The elemental analyses of **12** and **13** showed that no sulfur is present. This was confirmed by the mass spectrum which exhibited a correct molecular ion peak.

The formation of the indazole and oxathiadiazole derivatives **17** and **19** (Scheme 3) requires the intermediate formation of a thiocarbohydrazide radical (**15**). Aerial oxidation of the latter, accompanied by loss of H_2O and dehydrogenation, produced the oxathiadiazole derivatives **19**. On the other hand, **15** could be dimerized, and extraction of sulfur takes place easily; reaction with *DDO* under elimination of HCl and H_2O gives the indazole derivatives **17**.

Similarly, *CHL-p* interacted with **1b** followed by an interamolecular condensation reaction to yield the final product 6,7-dichloro-5-phenylpyrazolo-[3,4,5-*de*]phthalazin-8-ol (**21**, Scheme 4).

Table 1. ^{1}H NMR, IR, and mass spectroscopic data of compounds **4–8, 10, 12, 13, 17, 19,** and **21**

	^{1}H NMR (δ, *TMS*)[1]	IR (KBr, cm^{-1})	MS (m/z%)
4a	4.70 (br, 2H, NH_2)	3339, 3193 (NH_2), 2200, 2195 (CN), 1641 (C = N)	190 (M^+, 22), 174 (7), 148 (6), 122 (3), 96 (4), 44 (100)
4b	7.54–7.78 (m, 5H, Ar–H), 8.14 (s, 1H, azomethine-CH)	2205, 2200 (CN), 1622 (ArC = C, C = N)	279 (M^+, 79), 252 (5), 200 (28), 177 (62), 160 (78), 118 (65), 103 (93), 90 (100)
5a	4.87 (br, 2H, NH_2)	3320, 3200 (NH_2), 2220 (CN), 1634 (C = N)	166 (M^+, 54), 148 (100), 134 (11), 71 (44), 43 (76)
5b	7.46–7.71 (m, 5H, Ar–H), 8.12 (s, 1H, azomethine-CH)	2227, 2213 (CN), 1652, 1622 (Ar–C = C, C = N)	254 (M^+, 18), 228 (22), 202 (31), 103 (96), 76 (62), 44 (100)
6	4.50 (br, 2H, NH_2), 7.35–7.65 (m, 5H, Ar–H), 8.10 (s, 1H, azomethine-CH), 10.05 (br, 1H, NH)	3414, 3312, 3195 (NH_2, NH), 2235, 2207 (CN), 1635, 1596 (Ar–C = C, C = N	295 (M^+, 100), 279 (6), 161 (73), 103 (31), 77 (39)
7	5.10 (br, 2H, NH_2), 6.14 (br, 2H, NH_2)	3411, 3171 (NH_2), 2242 (CN), 1708 (CO), 1636 (C = N)	209 (M^+, 69), 194 (100), 168 (54), 154 (9), 136 (11), 56 (79), 42 (65)
8a[2]	7.40–7.87 (m, 10H, Ar–H), 8.13 (s, 1H, azomethine-CH), 12.60 (br, 1H, thiadiazole-NH)	3410 (NH), 3061 (Ar–CH), 1611, 1589 (Ar–C = C)	280 (M^+, 41), 177 (50), 121 (28), 118 (46), 104 (40), 103 (100), 90 (56), 76 (50)
8b	2.30 (s, 6H, $2CH_3$), 7.36–7.84 (m, 8H, Ar–H), 8.16 (s, 1H, azomethine-CH), 12.54 (br, 1H, thiadiazole-NH)	3380 (NH), 3025 (Ar–CH), 2919 (Ali-CH), 1611 (Ar–C = C)	308 (M^+, 100), 217 (18), 191 (72), 177 (14), 132 (77), 118 (43), 104 (99), 91 (44), 65 (17)
8c	3.87 (s, 6H, $2OCH_3$), 7.33–7.76 (m, 8H, Ar–H), 8.12 (s, 1H, azomethine-CH), 12.50 (br, 1H, thiadiazole-NH)	3420 (NH), 3003 (Ar–CH), 2934–2836 (Ali-CH), 1609 (Ar–C = C	
8d	7.44–7.69 (m, 8H, Ar–H), 8.18 (s, 1H, azomethine-CH), 12.64 (br, 1H, thiadiazloe-NH)	3385 (NH), 1616 (Ar–C = C)	352 (12), 351 (13), 349 (M^+, 61), 348 (85), 347 (11), 237 (17), 211 (65), 197 (21), 152 (77), 140 (18), 139 (33), 138 (33), 134 (56), 124 (100), 102 (25), 89 (64)

(*Continued*)

Table 1. (*Continued*)

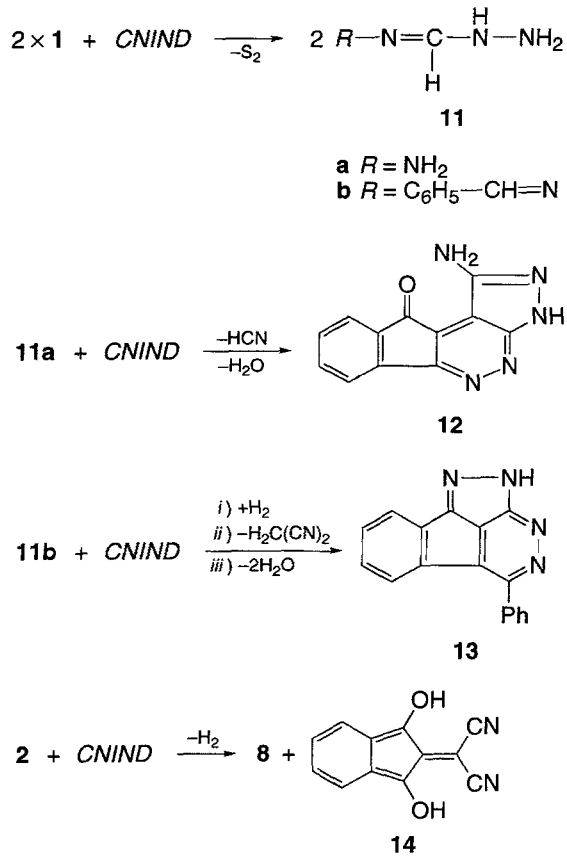
10a	7.35–7.95 (m, 10H, Ar–H), 8.43 (s, 1H, azomethine-CH), 10.54 (s, 1H, thiadiazine-NH)	3447 (NH), 2210 (CN), 1621 (Ar–C = C)	344 (M^+, 8), 243 (40), 140 (33), 103 (100), 76 (43), 43 (38)
10b	2.28 (s, 6H, 2 CH_3), 7.28–7.79 (m, 8H, Ar–H), 8.35 (s, 1H, azomethine-CH), 10.30 (s, 1H, thiadiazine-NH)	3450, 3313 (NH), 2217 (CN), 1649, 1609 (Ar–C = C, C = N)	372 (M^+, 22), 254 (51), 118 (100)
10c	3.83 (s, 6H, 2OCH_3), 7.19–7.79 (m, 8H, Ar–H), 8.34 (s, 1H, azomethine-CH), 10.35 (br, 1H, thiadiazine-NH)	3445–3316 (NH), 2915 (Ali–CH), 2210 (CN), 1648, 1616 (Ar–C = C, C = N)	404 (M^+, 38), 270 (61), 134 (100)
10d	7.40–7.80 (m, 8H, Ar–H), 8.40 (s, 1H, azomethine-CH), 10.38 (br, 1H, thiadiazine-NH)	3400, 3200 (NH), 3056 (Ar–CH), 2213 (CN), 1616 (Ar–C = C)	
12	6.30 (br, 2H, NH_2), 7.35–7.48 (m, 4H, Ar–H), 13.50 (br, 1H, pyrazole-NH)	3460–3265 (NH_2, NH), 1690 (CO), 1635, 1587 (Ar–C = C, C = N)	237 (M^+, 3), 221 (10), 193 (20), 160 (31), 147 (38), 128 (17), 104 (38), 76 (45), 64 (100)
13	7.40–7.60 (m, 4H, Ar–H), 13.40 (br, 1H, pyrazole-NH)	3446, 3251 (NH), 1634, 1588 (Ar–C = C, C = N)	271 (M^+, 67), 270 (M^+, 3), 255 (24), 227 (6), 185 (36), 158 (17), 129 (31), 102 (36), 64 (100), 44 (98)
17	7.35–7.66 (m, 5H, Ar–H), 8.12 (s, 1H, azomethine-CH), 9.39 (br, 1H, OH)	3487 (OH), 2212 (CN)	346 (12), 344 (M^+, 26), 342 (14), 318 (18), 230 (21), 162 (25), 103 (39), 73 (39), 44 (100)
19	7.38–7.78 (m, 5H, Ar–H), 8.17 (s, 1H, azomethine-CH)	3117 (Ar–CH), 1620 (Ar–C = C)	207 (M^+, 70), 178 (24), 162 (35), 131 (100), 120 (45), 104 (67), 77 (93)
21	7.28–7.71 (m, 5H, Ar–H), 9.51 (br, 1H, OH)	3449 (OH), 1611 (Ar–C = C)	319 (14), 317 (M^+, 27), 315 (16), 300 (6), 264 (18), 177 (28), 160 (19), 103 (100), 76 (46)

[1]All compounds were measured in *DMSO*-d_6 except **4a, 5a** (*DMF*-d_7), **6** (Acetone-d_6), and **10a–d** ($CDCl_3$); [2] ^{13}C NMR (*DMSO*-d_6) of **8a**: 126.43, 126.57, 126.68, 128.87, 129.27, 129.70, 130.11, 130.12, 130.55, 134.09, 169.71 ppm

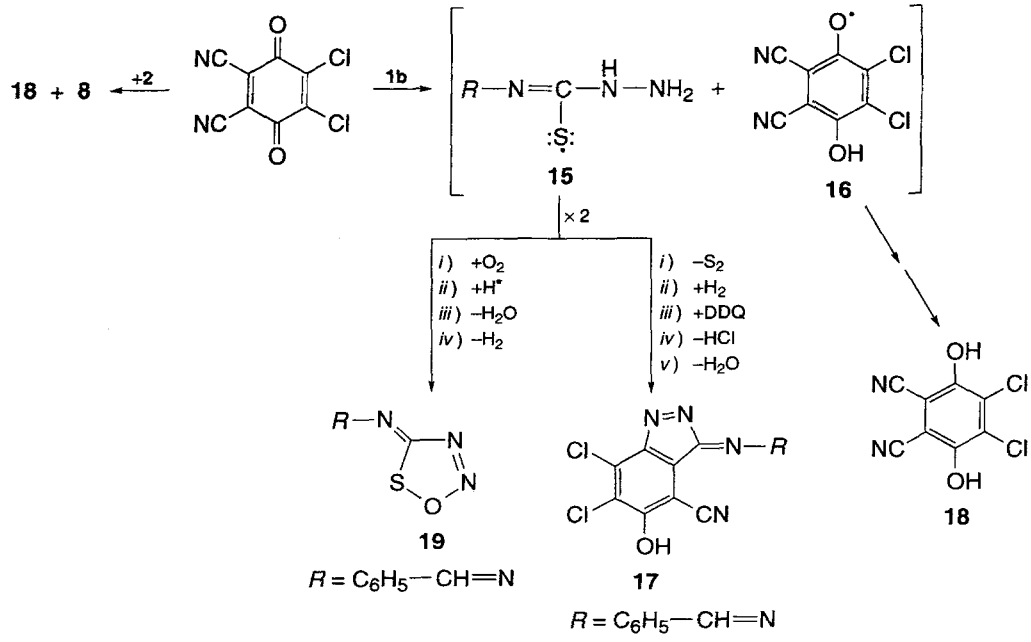
Table 2. Physical data of compounds **4–8, 10, 12, 13, 17, 19,** and **21**

	Yield (%)	m.p. (°C)	Colour	Solvent	Mol. Formula (M. Wt.)
4a	44	328–330	Yellowish-brown	*DMF*/Ethanol	$C_6H_2N_6S$ (190.188)
4b	35	301–303	Orange	Acetonitrile	$C_{13}H_6N_6S$ (278.297)
5a	26	339–341	Pale yellow	Acetonitrile	$C_4H_2N_6S$ (166.166)
5b	28	323–325	Yellowish brown	Ethanol	$C_{11}H_6N_6S$ (254.275)
6	29	341–343	Reddish brown	Ethanol	$C_{13}H_9N_7S$ (295.327)
7	24	246–248	Yellow	Ethanol	$C_6H_4N_6OS$ (208.203)
8a	71	169–171	Pale yellow	Acetonitrile	$C_{15}H_{12}N_4S$ (280.353)
8b	69	238–240	Pale yellow	Ethyl acetate	$C_{17}H_{16}N_4S$ (308.406)
8c	75	163–165	Pale yellow	Acetonitrile	$C_{17}H_{16}N_4O_2S$ (340.405)
8d	67	218–220	Pale yellow	Acetonitrile	$C_{15}H_{10}Cl_2N_4S$ (349.243)
10a	19	217–219	Yellow	Ethanol	$C_{18}H_{12}N_6S$ (344.399)
10b	21	192–194	Yellow	Ethanol	$C_{20}H_{16}N_6S$ (372.447)
10c	16	178–180	Yellow	Ethanol	$C_{20}H_{16}N_6O_2S$ (404–451)
10d	19	205–207	Yellow	Ethanol	$C_{18}H_{10}C1_2N_6S$ (413.289)
12	71	322–324	Reddish-brown	*DMF*/Ethanol	$C_{12}H_7N_5O$ (237.220)
13	69	310–312	Red	Acetonitrile	$C_{17}H_{10}N_4$ (270.293)
17	47	309–311	Yellow	Methanol	$C_{15}H_7C1_2N_5O$ (344.159)
19	27	211–213	Pale yellow	Ethanol	$C_8H_6N_4OS$ (206.228)
21	58	202–204	Brown	Acetonitrile	$C_{14}H_6C1_2N_2O$ (317.134)

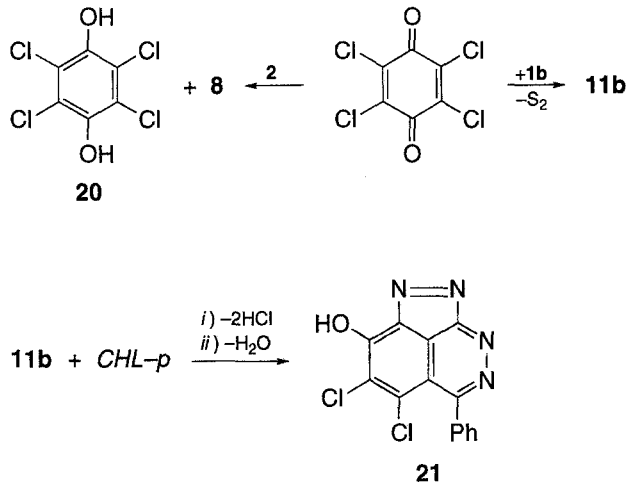
Recently, the oxidative cyclization of some semicarbazones of aldehydes induced by metallic salts and the regiochemistry of this reaction have been discussed [11, 12]. In order to gain further information on the course of the oxidative cyclization reactions, we have studied the behaviour of thiodicarbazones **2a–d** with the previous acceptors. In continuation of our research in this area [6, 7] we expected that **2** could



Scheme 2



Scheme 3



Scheme 4

lead to the 1,2,4-triazole derivatives **9** upon reaction with *TCNE*. However, two possible tautomeric structures were considered (*c.f.* structures **8** and **9** in Scheme 1).

In the ^{1}H NMR spectra, the ring NHs are clearly represented between $\delta = 12.50$ and 12.64 ppm. On the other hand, the aromatic SH and exocylic NH appear at $\delta = 2.00$–4.00 and 10.31–10.52 ppm, respectively [13, 14]. Thus, one can conclude that the triazole derivatives **9** exist in their thione form rather than in the thiol one in *DMSO* solution. It is difficult to differentiate between the two structures on the basis of IR or ^{1}H NMR spectra, because both structures have the same functional groups and the same types of hydrogen atoms. The structure of **8** has unambiguosly proven by its ^{13}C NMR spectrum which clearly revealed no signals between 180 and 190 ppm due to C=S [13], but instead a peak at 169.70 ppm for the Ph–C(S)=N carbon atom of the thiazdiazole ring system [13]. The remaining chemical shift values in the ^{13}C NMR spectrum are given in Table 1.

Scheme 1 shows also the successful isolation of the minor products, thiadiazine derivatives **10**, from the reaction of **2** with *TCNE*.

Both the NH and CN groups in **10a** appeared in the IR spectrum at 3447 and 2210 cm^{-1}, respectively. The ^{1}H NMR spectrum ($CDCl_3$) exhibited a singlet at $\delta = 8.43$ ppm corresponding to azomethine-CH, and a broad peak at 10.54 ppm due to thiadiazine-NH, besides that of the aromatic protons at 7.35–7.95 ppm. The mass spectrum showed a molecular ion peak at $m/z = 344$, confirming the structural features.

Experimental

All melting points are uncorrected; IR spectra: Shimadzu 470 spectrophotometer (KBr); ^{1}H NMR and ^{13}C NMR spectra: Bruker AC 200 (200 MHz) and AM 400 (400 MHz) instruments with *TMS* as internal standard (δ in ppm); MS: Finnigan MAT 8430 at 70 eV; elemental analyses: Microanalytical Center at Cairo University; preparative thin layer chromatography (PTLC) was carried out on 48 cm wide and 20 cm high glass plates covered with a 1 mm thick layer of silica gel (Merck PF 254, fluorescent indicator) with toluene/ethyl acetate as the solvent system. *TCNE*, *DDQ*, *CHL-p*, and *CNIND* were purified and prepared as described before [16]. Thiocarbohydrazide was purchased from Aldrich. **1b** and **2** were prepared according to published procedures [14]. All compounds gave satisfactory elemental analyses (C, H, N, Cl).

Reaction of TCNE with **1a**

Into a stirred solution of *TCNE* (2 mmol) in 10 ml *DMF*, thiocarbohydrazide (**1a**, 1 mmol) in 15 ml *DMF* was added at room temperature. The mixture was stirred for 3 h and then allowed to stand for 48 h during which time a crystalline product separated. The resulting solid material was filtered, washed with ethanol, dried, and recrystallized from a suitable solvent to give the thiazole derivative **4a**. The filtrate was evaporated, and the residue was chromatographed (PTLC) using toluene/ethyl acetate (3:1) to give two zones (first: thiazinone **7**; second: thiadiazole **5**). The products **5** and **7** were extracted with acetone and recrystallized from an appropriate solvent.

Reaction of TCNE with **1b**

To a solution of 256 mg *TCNE* (2 mmol) in dry ethyl acetate (15 ml), thiocarbazone **1b** (1 mmol) in 20 ml dry ethyl acetate was added with stirring at room temperature. The stirring was continued for 2 h, and the mixture was then set aside overnight. The solvent was evaporated, and the residue was chromatographed (PTLC) using toluene/ethyl acetate (5:1) as eluent to give numerous zones, three of which were extracted. The first one contained the thiadiazepine derivative **6**, the second the substituted thiadiazole **5b**, and the migrating zone slowest contained the major product (**4b**). The three zones were extracted with acetone and recrystallized from suitable solvents.

Reaction of CNIND with **1a**

To a solution of 416 mg (2 mmol) *CNIND* in 15 ml *DMF*, 106 mg (1 mmol) of **1a** in 15 ml *DMF* were added. The mixture was stirred at room temperature for 72 h during which time the reddish-brown crystals of aminoindenopyrazolopyridazinone (**12**) separated. The resulting crude mass was collected, washed with cold ethanol, and recrystallized from a suitable solvent.

Reaction of CNIND with **1b**

A solution of **1b** (1 mmol) in dry ethyl acetate (20 ml) was added dropwise to a stirred solution of *CNIND* (2 mmol) in 20 ml dry ethyl acetate at room temperature. Stirring was continued for 30 min. The mixture was then set aside for 3 h during which time red crystals of phenyltetraazacyclopentafluorene (**13**) separated. The product obtained was recrystallized to give the pure compound.

Reaction of DDQ with **1b**

To a stirred solution of 340.50 mg (1.5 mmol) *DDQ* in 20 ml dry ethyl acetate, thiocarbazone (**1b**, 1 mmol) in 20 ml dry ethyl acetate was added dropwise at room temperature. The reaction mixture was stirred for 72 h and then filtered off. The precipitate was washed with cold ethyl acetate and recrystallized from a suitable solvent to give indazole derivative **17**. The filtrate was concentrated, and the residue was then chromatographed (PTLC) using toluene/ethyl acetate (3:1) as eluent to give two zones (first zone: oxathiadiazole derivative **19**; second zone: dihydroquinone **18** (89 mg, 39%). The zones were extracted with acetone and recrystallized from an appropriate solvent.

Reaction of CHL-p with **1b**

Thiocarbazone **1b** (1 mmol) in 20 ml dry ethyl acetate was added to 492 mg (2 mmol) *CHL-p* in 30 ml ethyl acetate at room temperature. The reaction mixture was stirred for 3 h. After standing for 48 h brown crystals were collected and washed with cold ethyl acetate, giving the phenylpyrazolophthalazinol derivative **21**.

Reaction of **2** *with TCNE, DDQ, CNIND, and CHL-p*

A solution of **2a–d** (1 mmol) in 15 ml dry ethyl acetate was added to a solution of each of the above acceptors (2 mmol) in 20 ml dry ethyl acetate. The reaction mixture was then set aside for 2 h during which time the crystals of thiadiazole deivatives **8a–d** separated. The filtrate of the reaction mixture of **2a–d** and *TCNE* was concentrated, and the residue was then chromatographed (PTLC) using toluene/ethyl acetate (10:1) as eluent to give only one zone containing the thiadiazine derivatives **10a–d** as minor products. In the case of *CNIND* and 2, the filtrate contained 1,3-dihydroxy-2*H*- (inden-2-ylidene)malononitrile (**14**, [17]), whereas in the case of *DDQ* and *CHL-p* with **2** the filtrate contained the dihydroquinones **18** and **20**.

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