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Synthesis of 3,4,4'- and 2',3,4-Triaminodiphenyl Ethers

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Abstract — Procedures were developed for preparing 3,4,4'- and 2',3,4-triaminodiphenyl ethers by the reaction of 4-acetylaminophenol with, respectively, 4-nitro- or 2-nitrochlorobenzene, followed by nitration of the resulting 4-acetylamino-4'- or -2'-nitrodiphenyl ether, hydrolysis of the product, 4-acetylamino-3,4'- or -2',3-dinitrodiphenyl ether, and reduction of 4-amino-3,4'- or -2'-3-dinitrodiphenyl ether.

3,4,4'-Triaminodiphenyl ether **I** and 2',3,4-triaminodiphenyl ether **II** are used as intermediates in production of antihelminth preparations [1, 2]. These compounds are prepared [3] by reaction of alkali metal 2- or 4-aminophenolate with 2-nitro-5-chloroaniline, followed by reduction of the resulting 3,4'- or 2',3-diamino-4-nitrodiphenyl ether over Raney nickel. The major disadvantage of this procedure, in our opinion, is the use of difficultly available 2-nitro-5-chloroaniline.

Procedures for preparing 4-amino-4'-nitrodiphenyl ether **III**, 4-amino-2'-nitrodiphenyl ether **IV**, and 4-acetylamino-4'-nitrodiphenyl ether **V** have been reported. Ethers **III** and **IV** are prepared by the reaction of 4-nitrochlorobenzene **VI** or 2-nitrochlorobenzene **VII** with sodium (or potassium) 4-aminophenolate in DMSO on heating [4, 5]. Also, ether **III** is prepared by heating nitro compound **VI** and sodium 4-aminophenolate in DMSO in the presence of KOH [6], in chlorobenzene [7], or in DMSO [8] in the presence of a phase-transfer catalyst.

In another procedure, ethers **III** and **IV** are prepared in two steps by the reaction of sodium *p*-nitrophenolate with 4-nitrochlorobenzene **VI** or 2-nitrochlorobenzene **VII**, followed by reduction of the resulting 4,4'-dinitrodiphenyl ether **VIII** or 2',4-dinitrodiphenyl ether **IX**. 4,4'-Dinitrodiphenyl ether **VIII** is also prepared by the reaction of potassium nitrophenolate with nitrochlorobenzene **VI** in the presence of activated copper powder [9]. Dinitrodiphenyl ether **VIII** is then reduced with sodium disulfide in acetone [9], with hydrogen sulfide in ammonia [10], with phenylhydrazine in *p*-dichlorobenzene [11], or with iron and concentrated HCl in methanol [12]. In another procedure, 4,4'-dinitrodiphenyl ether **VIII** and 2',4-dinitrodiphenyl ether **IX** are prepared by the reaction of nitrochlorobenzene **VI** with, respectively, sodium 4-nitrophenolate and 2-nitrophenolate in DMSO in the presence of sodium nitrite [5].

Procedures for preparing 4-acetylamino-4'-nitrodiphenyl ether **V** and 4-acetylamino-2'-nitrodiphenyl ether **X** from 4-amino-4'-nitrodiphenyl ether **III** and 4-amino-2'-nitrodiphenyl ether **IV** have not been reported; only the synthesis directly from 4-acetylamino-4'-nitrodiphenyl ether **V** was published. Ether **V** is prepared by condensation of 4-acetylaminophenol **XI** with 4-nitrobromobenzene [13] or 4-nitrofluorobenzene [14] in methanol in the presence of NaOH and copper powder, and also by the reaction of acetylaminophenol **XI** with nitrochlorobenzene **VI** in the presence of alkali or, which is preferable, of potassium carbonate in neutral solvents such as DMF, *o*-dichlorobenzene, xylene, or diglyme [15].

Thus, published data suggest that the most convenient route to 4-acetylamino-4'-nitrodiphenyl ether **V** is single-step synthesis from acetylaminophenol **XI** and nitrochlorobenzene **VI** [15]. Procedures for preparing 4-amino-4'-nitrodiphenyl ether **III** and 4-amino-2'-nitrodiphenyl ether **IV** from sodium 4-aminophenolate and 4-nitrochlorobenzene **VI** or 2-nitrochlorobenzene **VII** [5] also deserve attention.

It also seems feasible to prepare triaminodiphenyl ethers **I** and **II** from 4-amino- or 4-acetylaminophenol and 4-nitrochlorobenzene **VI** or 2-nitrochlorobenzene **VII** by Scheme 1.

We prepared diphenyl ethers **V** and **X** according to [15] from acetylaminophenol **XI** by its reaction with **VI** or **VII** in DMF in the presence of anhydrous potassium carbonate; the yields were 96 and 93%, respectively. To develop a process for commercial synthesis of **V** and **X**, we optimized the reaction parameters. The

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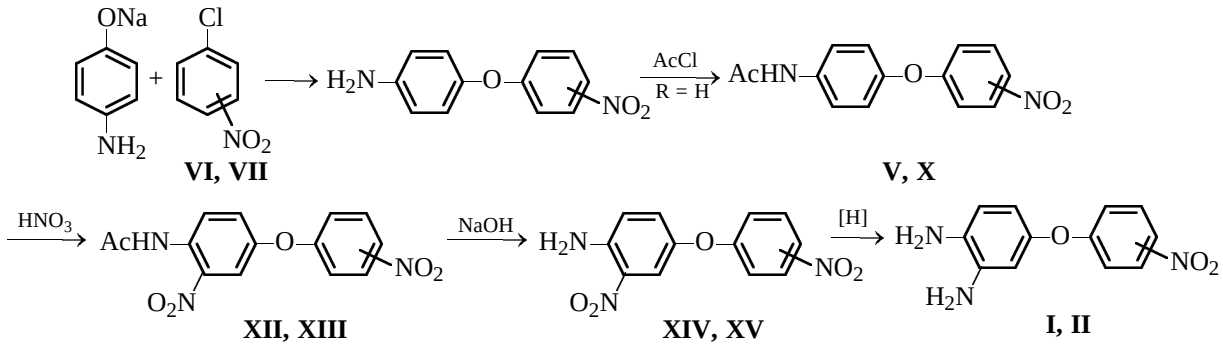
Scheme 1.



p-NO_2 (**V, VI, XII, XIV**), *o*-NO_2 (**X, VII, XIII, XV**); *p*-NH_2 (**I**), *o*-NH_2 (**II**).

Table 1. Synthesis of 4-acetylamino-2'-nitrodiphenyl ether **X**

Molar ratio **VII** : **XI**	Molar ratio **VII** : K_2CO_3	Weight ratio **VII** : DMF	*T*, °C	Time, h	Yield of X, %[a]
1.0 : 1.0	1.0 : 1.20	1 : 5.34	145	8.6	87 (90)
1.0 : 1.0	1.0 : 1.20	1 : 5.34	145	8.2	87 (90)
1.0 : 1.0	1.0 : 1.20	1 : 5.34	146	7.0	86 (90)
1.0 : 1.0	1.0 : 1.20	1 : 5.34	149	13.5	96 (94)
1.0 : 1.03	1.0 : 0.90	1 : 4.44	145	7.0	94 (93)
	1.0 : 0.1 NaOH				
1.0 : 1.07	1.0 : 1.0	1 : 4.91	145	10	93 (96)
	1.0 : 0.4 NaOH				
1.0 : 1.06	1.0 : 1.02	1 : 4.91	145	8.4	94 (93)
	1.0 : 0.28 NaOH				
1.0 : 1.09	1.0 : 0.33	1 : 4.91	110	16	76 (88)
	1.0 : 0.33 NaOH				
1.0 : 1.09	1.0 : 0.45	1 : 4.91	120	17	79 (88)
	1.0 : 0.28 NaOH				
1.0 : 1.10	1.0 : 0.45	1 : 3.69	110	14	75 (91)
	1.0 : 0.28 NaOH				
1.0 : 1.10	1.0 : 0.45	1 : 3.69	120	12.5	84 (92)
	1.0 : 0.28 NaOH				
1.0 : 1.10	1.0 : 1.02	1 : 4.91	145	8	93 (93)
	1.0 : 0.28 NaOH				
1.0 : 1.10	1.0 : 1.10	1 : 5.34	145	10	93 (95)
1.0 : 1.10	1.0 : 1.20	1 : 5.34[b]	81	13	50 (75)
1.0 : 1.10	1.0 : 1.20	1 : 5.34	145	9.70	95 (94)
1.0 : 1.10	1.0 : 1.20	1 : 5.34	145	8.20	95 (94)
1.0 : 1.10	1.0 : 1.20	1 : 5.34	145	6.75	81 (92)
1.0 : 1.10	1.0 : 1.20	1 : 5.34	150	10	94 (94)
1.0 : 1.10	1.0 : 1.20	1 : 5.34	145	3.75	43 (96)
1.0 : 1.10	1.0 : 1.20	1 : 5.34	145	6.50	81 (92)

[a] Yield of the crude product; the main substance content (wt %) is given in parentheses. [b] Acetonitrile was taken instead of DMF.

results concerning synthesis of **X** are listed in Table 1. The optimal reaction parameters are as follows: 140–150°C, 8–10 h, molar ratios **XI** : **VII** (1.03–1.10) : 1.00, **VII** : K_2CO_3 1.0 : (1.1–1.2), **VII** : NaOH 1.00 : (0.28–0.31); weight ratio **VII** : DMF 1.0 : (4.4–5.4).

Ether **V** was prepared similarly, and also by independent synthesis: acylation with acetic anhydride of 4-amino-4'-nitrodiphenyl ether **III** (prepared according to [4] in 84% yield) in aprotic organic solvents under the conditions suggested previously for acylation of

Table 2. Nitration of 4-acetylamino-2'-nitrodiphenyl ether **X** (40–45°C)

Molar ratio **X** : HNO_3	Molar ratio CH_3CO_2H : HNO_3	Molar ratio HNO_3 : $(CH_3CO)_2O$	HNO_3 concentration, wt %	Time, h	Yield of crude **XIII**, %	Main substance content in crude **XIII**, wt %
1.0 : 3.12	1 : 3.11	1.0 : 2.35	57	5.0	91.5	97.0
1.0 : 3.12	1 : 3.11	1.0 : 2.35	57	5.5	91.7	96.0
1.0 : 3.10	1 : 2.09	1.0 : 2.04	57	7.0	83.9	91.0
1.0 : 3.00	1 : 4.94	1.0 : 2.65	57	8.0	76.3	99.0
1.0 : 3.01	1 : 2.77	1.0 : 2.65	57	9.0	75.3	99.0
1.0 : 2.94	1 : 4.73	1.0 : 0.34	90	3[a]	89.9	99.0
1.0 : 2.87	1 : 5.13	1.0 : 0.37	90	1.0	93.0	97.0
1.0 : 2.75	1 : 5.11	1.0 : 0.37	90	1.5	94.4	97.7
1.0 : 2.74	1 : 5.11	1.0 : 0.36	90	1.0	79.3	98.1
1.0 : 2.74	1 : 5.11	1.0 : 0.36	90	1.5	93.1	98.6
1.0 : 2.74	1 : 5.11	1.0 : 0.36	90	2.0	94.9	99.0
1.0 : 2.77	1 : 5.12	1.0 : 0.36	90	1.75	93.3	98.6
1.0 : 2.70	1 : 5.13	1.0 : 0.37	90	1.5	92.0	98.0
1.0 : 2.70	1 : 5.13	1.0 : 0.37	90	1.5	92.3	98.3
1.0 : 2.68	1 : 2.12	1.0 : 2.10	57	7.0	85.4	99.3
1.0 : 2.65	1 : 2.12	1.0 : 1.77	57	4.5	95.7	96.1
1.0 : 2.65	1 : 2.12	1.0 : 1.77	57	5.5	95.1	97.0
1.0 : 2.65	1 : 2.13	1.0 : 1.77	57	6.5	95.3	98.2
1.0 : 2.64	1 : 2.12	1.0 : 2.09	57	7.0	85.6	99.5
1.0 : 2.64	1 : 2.12	1.0 : 1.76	57	6.5	85.3	98.2

[a] At 32°C.

4-amino-4'-nitrodiphenyl sulfide [16]. The reaction occurs readily with a qantitative yield.

4-Acetylamino-3,4'-dinitrodiphenyl ether **XII** and 4-acetylamino-2',3-dinitrodiphenyl ether **XIII** were prepared by nitration of ethers **V** and **X**, respectively. When optimizing the nitration conditions, we found that, under the conditions used previously for nitration of 4-acetylamino-4'-nitrodiphenyl sulfide [16] with dilute nitric acid (ρ 1.35 g cm^{-3}), the reaction is slow, and heating causes removal of the acetyl protecting group.

When the nitration was performed under more rigorous conditions, i.e., with more concentrated HNO_3 (ρ 1.5 g cm^{-3}) and at higher temperature (50–60°C), the reaction was noticeably faster and gave virtually pure dinitrodiphenyl ethers **XII** and **XIII** in 94 and 97% yields, respectively. When the nitration is performed at a lower temperature (35–40°C), the mixture contains a large amount of *N*-nitro compounds [16], which rearrange into the *C*-nitro compounds under these conditions very slowly.

Experiments showed that nitration of **X** can be performed most readily in a mixture of acetic acid and acetic anhydride. The results of nitration of ether **X** in this medium are given in Table 2.

Thus, ether **X**, compared to the related sulfide [16], undergoes nitration more readily, with the higher yield and better quality of the reaction product. The process parameters can be varied in a fairly wide range without noticeable deterioration of the results. The following parameters can be recommended as optimal for nitration of **X**: 40–45°C, 4.5–6.5 h, HNO_3 concentration ≥90%; molar ratios **X** : HNO_3 1.0 : (2.7–2.9), HNO_3 : acetic acid 1.0 : (2.1–5.1), HNO_3 : acetic anhydride 1.0 : (0.3–1.7).

4-Acetylamino-3,4'-dinitrodiphenyl ether **XII** was prepared in a quantitative yield by nitration of **V** with HNO_3 (ρ 1.50 g cm^{-3}) in chloroform (molar ratio **V** : HNO_3 : chloroform 1 : 3 : 17) in the presence of acetic anhydride at 50–60°C.

Hydrolysis of **XII** with 30% aqueous KOH at 100°C [molar ratio **XII** : KOH 1 : (6–7)] gave 4-amino-3,4'-dinitrodiphenyl ether **XIV** in 96% yield. In the course of the reaction, first a solution is formed and then a solid gradually precipitates. Formation of the homogeneous solution can probably be explained by initial formation of the potassium salt with deprotonation of the acetanilide moiety. The precipitating hydrolysis product is insoluble in water and aqueous alkali.

Table 3. Synthesis of 4-amino-2,3'-dinitrodiphenyl ether **XV** (70–75°C)

Molar ratio **XIII** : NaOH	Weight ratio **XIII** : $(CH_3)_2CHOH$	Time, h	Yield of crude **XV**, %	Main substance content in crude **XV**, wt %
1.0 : 1.96	1 : 4.10	1.5	83.4	99.1
1.0 : 1.81	1 : 4.10	4.0	92.7	97.4
1.0 : 1.81	1 : 4.10	3.0	93.0	98.1
1.0 : 1.81	1 : 4.10	2.0	93.0	97.5
1.0 : 1.81	1 : 4.10	2.0	92.6	97.4
1.0 : 1.81	1 : 4.10	1.6	92.5	97.0
1.0 : 1.81	1 : 4.10	1.5	93.1	98.4
1.0 : 1.81	1 : 4.10	1.5	92.6	97.2
1.0 : 1.81	1 : 4.10	1.5	92.4	97.4
1.0 : 1.81	1 : 4.10	1.4	88.7	98.5
1.0 : 1.81	1 : 4.10	1.0	87.7	97.0
1.0 : 1.81	1 : 4.10	1.0	89.8	97.2
1.0 : 1.81	1 : 4.10	2.2[a]	81.2	85.8
1.0 : 1.81	1 : 4.10	3.0[b]	87.2	97.5
1.0 : 1.81	1 : 4.10	1.4[c]	51.6	92.5
1.0 : 1.81	1 : 4.10	2.2[d]	86.7	98.7
1.0 : 1.81	1 : 4.10	1.2[e]	88.0	98.6
1.0 : 1.66	1 : 4.10	3.0	79.3	98.7
1.0 : 1.66	1 : 4.10	1.0	71.3	95.3
1.0 : 1.50	1 : 4.10	4.0	79.0	97.3
1.0 : 1.50	1 : 4.10	2.0	69.0	97.3
1.0 : 1.35	1 : 4.10	2.0	81.2	92.5

[a] At 36°C. [b] At 45°C. [c] At 44°C. [d] At 55°C. [e] At 65°C.

With the aim to replace KOH by cheaper NaOH and reduce the consumption of alkali, we performed experiments on deprotection of **XIII** in 2-propanol. We found that the most promising procedure for deacetylation of **XIII** is treatment of its solution in 2-propanol with aqueous NaOH. 4-Amino-2',3-dinitrodiphenyl ether **XV** is thus obtained in 96% yield. At slower (within 50 min) addition of the NaOH solution, by-products are formed in smaller amounts.

The results concerning synthesis of **XV** are given in Table 3. The optimal reaction conditions are as follows: 70–75°C, 1.5–2.5 h, molar ratio **XIII** : alkali 1.0 : 1.8, weight ratio **XIII** : 2-propanol 1.0 : 4.1.

The last step of the synthesis of **I** and **II** is reduction of **XIV** and **XV**. The reduction was performed in a refluxing mixture of dioxane with ethylene glycol or in refluxing 1-butanol with hydrazine hydrate in the presence of $FeCl_3$ and activated carbon under the conditions similar to those of reduction of 4-amino-3,4'-dinitrodiphenyl sulfide [17]; ethers **I** and **II** were obtained in 87 and 84% yields, respectively.

EXPERIMENTAL

The crude reaction products were analyzed with an Altex liquid chromatograph (model 100 pump; model 153 UV detector; model 210 20-μl loop dosing unit; 30-, 50-, and 100-μl SNR Hamilton syringes; stainless steel column 0.25 m long, 4.6 mm i.d.).

Analysis of mixtures of **VI**, **VII**, and **XI**, of nitrodiphenyl ethers **V** and **X**, of dinitrodiphenyl ethers **XII**–**XV**, and of triaminodiphenyl ethers **I** and **II** is based on HPLC separation on a column packed with Rsil reversed phase or Ultrasphere ODS in the eluent system 65% methanol + 35% water with addition of 25 ml of a 10% solution of 18-crown-6 in methanol per liter of the eluent. The chromatograms were calculated by internal normalization of the peak areas.

4-Acetylamino-2'-nitrodiphenyl ether X and 4-acetylamino-4'-nitrodiphenyl ether V. To 1.2 l of DMF, we added with vigorous stirring 2.24 mol of 4-acetylaminophenol **V**, 2.04 mol of nitrobenzene **VI** or **VII**, and 2.45 mol of anhydrous potassium carbonate. The mixture was heated to 145–150°C and vigorously stirred at this temperature for 8–10 h. The

reaction completion was judged from the disappearance of **VI** or **VII**. When residual amounts of **VI** or **VII** remained, the mixture was alkalized with a small amount of 44% aqueous NaOH and stirred for 1 h. After complete conversion of **VI** or **VII**, the mixture was placed in a crystallizer with water and cooled to room temperature; the precipitated ether **V** or **X** was filtered off, washed with 0.5 N alkali and water, and dried. Ether **V**: yield 93%, R_f 0.36 (benzene–hexane, 3 : 7), mp 153–154°C. Found, %: C 61.78; H 4.45; N 10.22. $C_{14}H_{12}N_2O_4$. Calculated, %: C 61.76; H 4.44; N 10.29. Ether **X**: yield 96%, R_f 0.30 (benzene–hexane, 10 : 2), mp 128–129°C. Found, %: C 61.81; H 4.42; N 10.26. $C_{14}H_{12}N_2O_4$. Calculated, %: C 61.76; H 4.44; N 10.29.

4-Acetylamino-3,4'-dinitrodiphenyl ether XII. To a suspension of 0.69 mol of **V** in 950 ml of chloroform, we added 66 ml of acetic anhydride and then, dropwise, 90 ml of HNO_3 (ρ 1.5 g cm^{-3}) at 45–50°C, after which the mixture was stirred at this temperature for 6 h. Then 500 ml of water was added, and the precipitate was filtered off. Yield 97%, R_f 0.7 (benzene–ethanol, 10 : 2), mp 121–123°C. Found, %: C 52.98; H 3.46; N 13.30. $C_{14}H_{11}N_3O_6$. Calculated, %: C 53.00; H 3.49; N 13.24.

4-Acetylamino-2',3-dinitrodiphenyl ether XIII. To 350 ml of acetic acid, we added with stirring 0.6 mol of **X**. After that, we added 66 ml of acetic anhydride and then, dropwise with vigorous stirring at 10–15°C over a period of 40–50 min, 77 ml of HNO_3 (ρ 1.5 g cm^{-3}). The resulting mixture was stirred for 20–25 min at 20–25°C, heated to 40–45°C, and vigorously stirred at this temperature for an additional 4–6 h. The reaction progress was monitored by TLC. When no ether **X** was longer detected, the mixture was placed in a crystallizer with cold water, and the precipitate of **XIII** was filtered off, thoroughly washed on the filter with water, and dried. Yield 92%, R_f 0.56 (benzene–ethanol, 10 : 2), mp 98–99°C. Found, %: C 53.05; H 3.52; N 13.28. $C_{14}H_{11}N_3O_6$. Calculated, %: C 53.00; H 3.49; N 13.24.

4-Amino-3,4'-dinitrodiphenyl ether XIV and 4-amino-2',3-dinitrodiphenyl ether XV. To 600 ml of 2-propanol, we added 0.39 mol of **XII** or **XIII**. The mixture was vigorously stirred at 25–30°C, and a solution of 0.40 mol of NaOH in 66 ml of water was gradually added. The resulting mixture was heated to 70–75°C, vigorously stirred at this temperature for 1.5–3 h, and placed in a crystallizer with cold water. The precipitate was filtered off, washed on the filter with water, and dried. Ether **XIV**: yield 96%, R_f 0.53 (benzene–ethanol, 10 : 2), mp 179–181°C. Found, %: C 52.42; H 3.32; N 15.47. $C_{12}H_9N_3O_5$. Calculated, %: C 52.36; H 3.29; N 15.27. Ether **XV**: yield 95%, R_f 0.78 (benzene–ethanol, 10 : 2), mp 162–163°C. Found, %: C 52.30; H 3.21; N 15.20. $C_{12}H_9N_3O_5$. Calculated, %: C 52.36; H 3.29; N 15.27.

3,4,4'-Triaminodiphenyl ether I and 2',3,4-triaminodiphenyl ether II. Nitro ethers **XIV** and **XV** were reduced to amino ethers **I** and **II** according to [17]. Ether **I**: yield 87%, R_f 0.14 (benzene–dioxane, 10 : 1), mp 162–163°C. Found, %: C 66.85; H 6.24; N 19.44. $C_{12}H_{13}N_3O$. Calculated, %: C 66.95; H 6.09; N 19.52. Ether **II**: yield 84%, R_f 0.23 (benzene–ethanol, 10 : 2), mp 147–148°C. Found, %: C 66.81; H 5.91; N 19.53. $C_{12}H_{13}N_3O$. Calculated, %: C 66.95; H 6.09; N 19.52.

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