Oxidation of Arylacetylhydrazones of Carbonyl Compounds with Lead Tetraacetate

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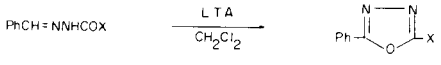
Oxidation of the title compounds **8**, **9** with lead tetraacetate at room temperature gives a variety of products depending on the substituents on the carbonyl carbon atom. Thus, on oxidation of the aldehyde derivatives **8** 1,3,4-oxadiazolo derivatives **10** are obtained in good yields. However in some cases formation of *N*-acetyl-*N*-arylacetyl-*N'*-benzoylhydrazines **11** is also observed, whereas oxidation of the ketone hydrazones **9** gives in good yields the 2H,5H-1,3,4-oxadiazoles **15** or substituted monoacetoxy- **17** and diacetoxyalkanes **18**. The reaction mechanisms are also discussed.

J. Heterocyclic Chem., **19**, 705 (1982).

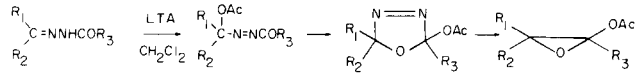
It is known that benzaldehyde benzoylhydrazones **1** and semicarbazones **2** undergo (1,2,3) oxidative cyclization when reacted with lead tetraacetate at room temperature to give 1,3,4-oxadiazoles **3**.



1, X = Ph
2, X = NH_2

3

On the contrary, oxidation (3,4,5) of ketone benzoylhydrazones **4** with lead tetraacetate leads to the highly reactive azoacetates **5** which at ~-20° cyclize to 2*H*,5*H*-1,3,4-oxadiazoles **6**. The oxadiazoles **6** are unstable and decompose between room temperature and 50° to the epoxides **7**.



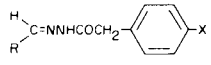
4 **5** **6** **7**

2*H*,5*H*-1,3,4-Oxadiazoles are also formed by the oxidation with lead tetraacetate of ketosemicarbazones (5,6,7) and of other analogous compounds (2,8).

It has also been shown previously (9) that lead tetraacetate oxidation of bisaroylhydrazones of α-dicarbonyl compounds leads to the formation of 1,2,3-triazolylisoimides, whereas insertion (10) of a methylene group in the aroyl unit leading to bisarylacetylhydrazones gives by the oxidation with lead tetraacetate 1-(*N*-arylacetylamino)-1,2,3-triazoles.

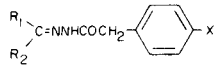
In the course of further work we have undertaken the preparation and oxidation with lead tetraacetate of some acetylhydrazones of several aldehydes **8** and ketones **9** (Table 1) in order to study if the insertion of a methylene group changes the reaction sequence.

The oxidation of aldehyde arylacetylhydrazones **8** follows an analogous pathway to that observed (1,2) in the oxidation of benzaldehyde benzoylhydrazones **1** leading to the formation of the corresponding 1,3,4-oxadiazoles **10** (Tables II and III). However, in the case of the oxidation of



8

a, R = CH_3, X = H
b, R = CH_3, X = OMe
c, R = CH_3, X = NO_2
d, R = Ph, X = H
e, R = Ph, X = OMe
f, R = Ph, X = NO_2

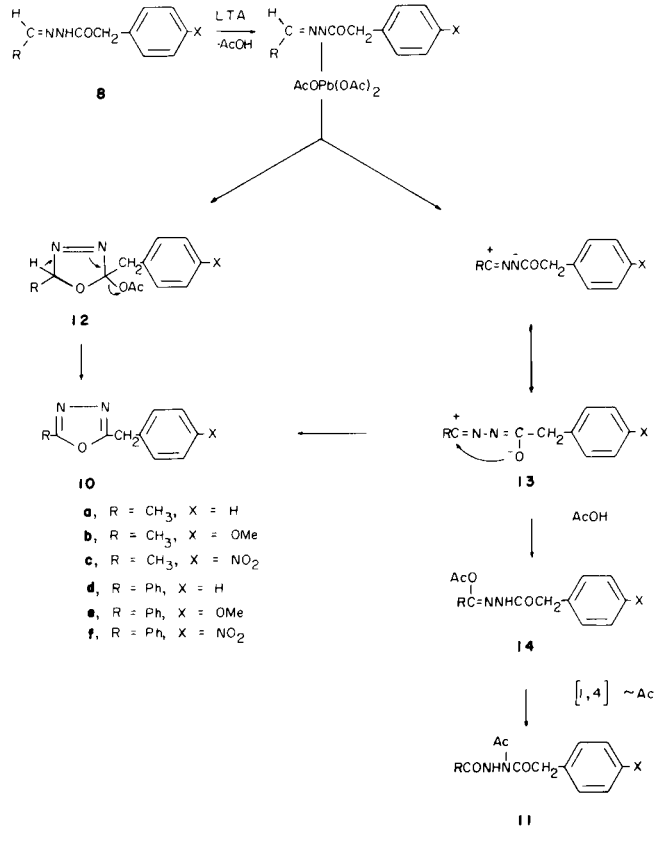


9

a, $R_1 = R_2 = CH_3$, X = H
b, $R_1 = R_2 = CH_3$, X = OMe
c, $R_1 = R_2 = CH_3$, X = NO_2
d, $R_1 = CH_3$, R_2 = Ph, X = H
e, $R_1 = CH_3$, R_2 = Ph, X = OMe
f, $R_1 = CH_3$, R_2 = Ph, X = NO_2
g, $R_1 = R_2$ = Ph, X = OMe
h, $R_1 = R_2$ = Ph, X = NO_2

the arylacetylhydrazones **8d**, **8e** and **8f** a second product was isolated. To this product the structure of *N*-acetyl-*N*-

Scheme I



8

12

10

a, R = CH_3, X = H
b, R = CH_3, X = OMe
c, R = CH_3, X = NO_2
d, R = Ph, X = H
e, R = Ph, X = OMe
f, R = Ph, X = NO_2

13

14

[1,4] ~Ac

11

Table I

Analytical Data for the Acylhydrazones **8** and **9**

Compound	Mp °C	Yield	IR (Nujol) cm⁻¹	NMR (Deuteriochloroform)				MS m/e	Formula	Molecular Weight	Analysis % Calcd.			Found		
			ν NH ν C=O δ CH_3	δ CH_2	δ OCH_3	δ CH	δ aromatic protons	(R.I. %)			C	H	N	C	H	N
8a	148-149 (lit (14) 145-146)	87	3200 1660 1.85 [3] d (a)	3.85 [2] s		ν_A 7.42 ν_B 7.55 [1] AB system J_{AB} 6 Hz	7.32 [5] s	M⁺ 176 (19) 161 (3) 135 (24) 118 (33) 91 (100)	$C_{10}H_{12}N_2O$	176.21						
8b	153-154	62	3200 1655 1.86 [3] d (a)	3.72 [2] s	3.72 [3] s	ν_A 7.37 ν_B 7.52 [1] AB system J_{AB} 6 Hz	ν_A 6.87 ν_B 7.19 [4] AB system J_{AB} 9 Hz	M⁺ 206 (65) 191 (1) 164 (44) 148 (18) 121 (100)	$C_{11}H_{14}N_2O_2$	206.24	64.06	6.84	13.58	64.35	6.85	13.61
8c	185-186	88	3200 1670 1.88 [3] d (a)	3.65 [2] 4.02 two s		ν_A 7.39 ν_B 7.55 [1] AB system J_{AB} 5 Hz	ν_A 7.66 ν_B 8.29 [4] AB system J_{AB} 8 Hz	M⁺-1 220 (18) 205 (11) 179 (100) 162 (16) 136 (85)	$C_{10}H_{11}N_3O_3$	221.21	54.29	5.01	19.00	54.37	5.10	19.08
8d	139-140 and 149-150 (lit (15) 148)	80	3160 1660	4.07 [2] s		included in the aromatics	6.95-7.85 [11] m	M⁺ 238 (20) 135 (86) 120 (70) 103 (79) 91 (100)	$C_{15}H_{14}N_2O$	238.28						
8e	168-170	59	3180 1660	4.03 [2] s	3.73 [3] s	included in the aromatics	ν_A 6.89 ν_B 7.73 [4] AB system J_{AB} 8 Hz 7.23-7.85 [6] m	M⁺ 268 (100) 164 (100) 148 (91) 121 (100) 103 (43)	$C_{16}H_{16}N_2O_2$	268.30	71.62	6.01	10.44	71.35	5.86	10.18
8f	208-209 (lit (17) 204-205)	82	3240 1670	4.08 [2] s		included in the aromatics	7.18-8.42 [10] m	M⁺-1 282 (21) 179 (100) 136 (49) 103 (43)	$C_{15}H_{13}N_3O_3$	283.28						
9a	107-109 (lit (16) 108-110)	77	3230 1650 1.57 [6] 1.80 three s 2.02	3.67 [2] 3.97 two s			7.30 [5] s	M⁺ 190 (100) 175 (91) 150 (60) 134 (20) 118 (96) 91 (100)	$C_{11}H_{14}N_2O$	190.24						
9b	128-130	41	3170 1635 1.60 [6] 1.80 three s 2.01	3.61 [2] 3.90 two s	3.78 [3] s		ν_A 6.86 ν_B 7.26 [4] AB system J_{AB} 9 Hz	M⁺ 220 (100) 205 (12) 164 (88) 148 (53) 121 (100)	$C_{12}H_{16}N_2O_2$	220.26	65.43	7.32	12.72	65.24	7.08	12.57
9c	186-188	91	3220 1640 1.85 [6] 2.02 two s	4.08 [2] s			ν_A 7.51 ν_B 8.16 [4] AB system J_{AB} 8Hz	M⁺ 235 (100) 220 (89) 136 (40)	$C_{11}H_{13}N_3O_3$	235.24	56.16	5.57	17.86	56.36	5.70	18.00
9d	159-161 (lit (17) 158-160.5)	68	3180 1650 2.27 [3] s	4.20 [2] s			7.30-8.00 [10] m	M⁺ 252 (100) 237 (11) 134 (88)	$C_{16}H_{16}N_2O$	252.30						

								133 (71) 118 (23) 91 (54)									
9e	173-174	62	3180 1650	2.20 [3] s	4.05 [2] s	3.77 [3] s	6.80-8.00 [9] m	M+ 282 (100) 267 (2) 164 (57) 148 (43) 133 (91) 121 (94)	$C_{17}H_{18}N_2O_2$	282.33	72.32	6.43	9.92	72.40	6.40	9.84
9f	170-172	85	3180 1665	2.25 [3] s	4.22 [2] s		7.10-8.30 [9] m	M+ 297 (83) 282 (13) 136 (15) 133 (100)	$C_{16}H_{15}N_3O_3$	297.30	64.63	5.09	14.14	64.46	5.00	14.20
9g	138-139	21	3320 1690		3.53 [2] 4.12 two s	3.76 [3] s	6.60-7.80 [14] m	M+ 344 (72) 195 (100) 180 (12) 164 (20) 149 (20) 121 (67)	$C_{22}H_{20}N_2O_2$	344.40	76.72	5.85	8.13	76.52	5.92	8.17
9h	186-187	40	3290 1680		4.27 [2] s		7.05-7.70 [14] m	M+ 359 (84) 195 (100) 180 (23) 165 (36)	$C_{21}H_{17}N_3O_3$	359.37	70.18	4.77	11.69	70.32	4.73	11.75

(a) The nmr solvent is dimethylsulphoxide.

Table II

Products obtained by the oxidation of Acylhydrazones **8** and **9**

Hydrazone	Oxadiazole	Yield %	Acetylarylacetylbenzoyl-hydrazine	Yield %	2H,5H-Oxadiazole	Yield %
8a	**10a**	23	—	—	—	—
8b	**10b**	60	—	—	—	—
8c	**10c**	80	—	—	—	—
8d	**10d**	74	**11a**	13	—	—
8e	**10e**	70	**11b**	12	—	—
8f	**10f**	57	**11c**	12	—	—
9a	—	—	—	—	**15a**	31
9b	—	—	—	—	**15b**	40
9c	—	—	—	—	**15c**	61

Table III

Analytical Data for 1,3,4-Oxadiazoles **10**

Compound	Mp °C	NMR (Deuteriochloroform) δ CH_3	δ OCH_3	δ CH_2	δ aromatic protons	MS m/e (R.I. %)	Formula	Molecular Weight	Analysis % Calcd. C	H	N	Found C	H	N
10a	liquid purified by tlc (lit (a) bp 163-164)	2.22 [3] s		3.99 [2] s	7.17 [5] s	M⁺ 174 (100) 145 (32) 132 (25) 119 (17) 91 (70) 83 (90)	$C_{10}H_{10}N_2O$	174.20						
10b	liquid purified by tlc	2.83 [3] s	3.74 [3] s	3.97 [2] s	ν_A 6.70 ν_B 7.06 $[4]^{AB}_{system}$ J_{AB} 9 Hz	M⁺ 204 (100) 189 (25) 175 (2) 162 (6) 148 (18) 121 (69) 83 (15)	$C_{11}H_{12}N_2O_2$	204.22	64.69	5.92	13.72	65.03	5.68	13.33
10c	79-81	2.52 [3] s		4.29 [2] s	ν_A 7.52 ν_B 8.26 $[4]^{AB}_{system}$ J_{AB} 9 Hz	M⁺ 219 (100) 204 (4) 190 (4) 164 (2) 136 (10) 83 (36)	$C_{10}H_9N_3O_3$	219.20	54.79	4.14	19.17	54.28	4.19	18.98
10d	103-104 (lit (a) 101-102.5)			4.35 [2] s	7.33-8.35 [10] m	M⁺ 236 (100) 207 (14) 145 (24) 119 (5) 105 (26) 91 (33)	$C_{15}H_{12}N_2O$	236.26						
10e	89-90		3.73 [3] s	4.16 [2] s	ν_A 6.78 ν_B 7.28 $[4]^{AB}_{system}$ J_{AB} 9 Hz 7.25-8.08 [5] m	M⁺ 266 (100) 251 (7) 237 (2) 149 (45) 145 (17) 121 (62) 105 (12)	$C_{16}H_{14}N_2O_2$	266.29	72.16	5.30	10.52	71.99	5.10	10.38
10f	132-133			4.40 [2] s	ν_A 7.60 ν_B 8.26 $[4]^{AB}_{system}$ J_{AB} 9 Hz 7.50-8.20 [5] m	M⁺ 281 (100) 252 (4) 145 (52) 136 (14) 105 (85)	$C_{15}H_{11}N_3O_3$	281.26	64.05	3.94	14.94	63.85	4.04	14.78

(a) R. Huisgen, J. Sauer, H. J. Sturm and J. H. Markgraf, *Chem. Ber.*, **93**, 2106 (1960).

Table IV

Analytical Data for the Acetylarylacetylbenzoylhydrazines **11**

Compound	Mp °C	IR (Nujol) cm⁻¹ ν NH	IR (Nujol) cm⁻¹ νC=O	NMR (Deuteriochloroform) δ COCH₃	NMR (Deuteriochloroform) δ OCH₃	NMR (Deuteriochloroform) δ CH₃	NMR (Deuteriochloroform) δ aromatic protons	MS m/e (R.I. %)	Formula	Molecular Weight	Analysis % Calcd. C	Calcd. H	Calcd. N	Found C	Found H	Found N
11a	104-105	3230	1660 1720	2.30 [3] s		3.98 [2] s	7.10-7.90 [10] m	M⁺ 296 (4) 254 (13) 236 (10) 118 (98) 105 (100) 91 (66)	$C_{17}H_{16}N_2O_3$	296.31	68.90	5.44	9.45	68.59	5.52	9.39
11b	84-87	3240	1660 1725	2.42 [3] s	3.75 [3] s	4.00 [2] s	ν_A 7.13 ν_B 6.85 [4] $_{system}^{AB}$ J_{AB} 9 Hz 7.25-8.00 [5] m	M⁺ 326 (<0.5) 284 (<0.5) 266 (<0.5) 148 (99) 121 (98) 105 (100)	$C_{18}H_{18}N_2O_4$	326.34	66.24	5.56	8.58	65.87	5.84	8.88
11c	143-145	3210	1660 1720	2.43 [3] s		4.24 [2] s	ν_A 7.38 ν_B 8.12 [4] $_{system}^{AB}$ J_{AB} 9 Hz 7.50-8.10 [5] m	M⁺ 341 (5) 299 (100) 281 (100) 145 (64) 136 (100) 105 (100)	$C_{17}H_{15}N_3O_5$	341.31	59.82	4.43	12.31	59.49	4.43	12.30

Table V

Analytical Data for the 2*H*,5*H*-1,3,4-Oxadiazoles **15**

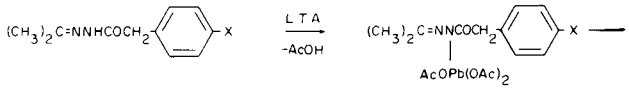
Compound	Mp °C	IR (Nujol) cm⁻¹ ν CO	NMR (Deuteriochloroform) δ CH₃	NMR (Deuteriochloroform) δ COCH₃	NMR (Deuteriochloroform) δ OCH₃	NMR (Deuteriochloroform) δ CH₂	NMR (Deuteriochloroform) δ aromatic protons	MS m/e (R.I. %)	Formula	Molecular Weight	Analysis % Calcd. C	Calcd. H	Calcd. N	Found C	Found H	Found N
15a	43-45	1755	0.95 [3] s 1.41 [3] s	1.98 [3]		3.67 [2] s	7.17 [5] s	M⁺ 248 (<0.5) 220 (5) 189 (2) 177 (19) 119 (18) 100 (65) 91 (100)	$C_{13}H_{16}N_2O_3$	248.27	62.89	6.50	11.28	63.00	6.52	11.33
15b	64-66	1750	1.13 [3] s 1.53 [3] s	2.08 [3] s	3.83 [3] s	3.72 [2] s	ν_A 6.82 ν_B 7.18 [4] $_{system}^{AB}$ J_{AB} 9 Hz	M⁺ 278 (0.5) 250 (44) 219 (5) 192 (75) 150 (97) 121 (100) 100 (22)	$C_{14}H_{18}N_2O_4$	278.30	60.42	6.52	10.07	60.65	6.66	10.14
15c	84-86	1750	1.20 [3] s 1.53 [3] s	2.03 [3] s		3.83 [2] s	ν_A 7.48 ν_B 8.16 [4] $_{system}^{AB}$ J_{AB} 9 Hz	M⁺ (-) 265 (4) 234 (14) 208 (60) 164 (60) 136 (77) 100 (100)	$C_{13}H_{15}N_3O_5$	293.27	53.24	5.16	14.33	53.63	5.25	14.42

arylacetyl-*N'*-benzoylhydrazine (**11**) (Tables II and IV) was attributed by analogy with the products obtained by the oxidation with lead tetraacetate of some monosubstituted aldehyde hydrazones (3), of benzil monophenylhydrazones (3) and of some monobenzoylhydrazones of α-dicarbonyl compounds (11) and on the basis of its spectral data. The formation of the products **10** and **11** can be explained by reaction Scheme I.

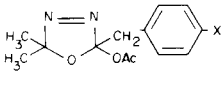
The first step of the reaction leads to the formation of a bond between the nitrogen of the NH group and lead. Cleavage of the N-Pb bond results to the cyclic compound **12** which looses acetic acid and is converted to the 1,3,4-oxadiazole **10**. On the other hand the acetylaryl-acetylbenzoylhydrazines **11** are considered (3) to arise from addition of acetic acid to the nitrilamides **13** followed by an [1,4]-acyl migration in the compounds **14** with the formation of **11**. However, 1,3,4-oxadiazoles **10** can also be formed *via* the nitrilamides **13**.

Oxidation of arylacetylhydrazones of ketones **9** with lead tetraacetate leads to a variety of products the formation of which greatly depends on the type of the ketone. In particular, the acetone hydrazones **9a, 9b** and **9c** give upon oxidation good yields of the 2*H*,5*H*-1,3,4-oxadiazoles **15** (Tables II and V) according to reaction Scheme II. This structure **15** was attributed to the oxidation products

Scheme II



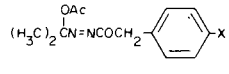
9a–9c



15

a, X = H
b, X = OMe
c, X = NO_2

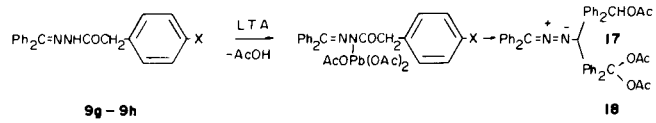
instead of the azoacetate structure **16** on the basis of their spectral data. In particular, only one signal was observed for the carbonyl carbon in the ^{13}C nmr spectrum and two signals in the ^{1}H nmr spectrum for the $(CH_3)_2$-C group.



16

On the other hand acetophenone hydrazones **9d, 9e** and **9f** decompose on oxidation to the starting ketone. Benzophenone hydrazones **9g** and **9h** give on oxidation with lead tetraacetate *O*-acetylbenzydrol **17** according to scheme III.

Scheme III



9g – 9h **18**

However, by the oxidation of the hydrazone **9h** the biacetylated product **18** was also isolated. A similar behaviour was observed (3) by the oxidation with lead tetraacetate of benzophenone hydrazones where the reaction products were substituted monoacetoxy- or diacetoxy-alkanes and also recently (12) by the oxidation of benzil monosemicarbazones where *O*-acetylbenzoin was the main reaction product. In addition oxidation (13) of diphenyl-diazomethan with lead tetraacetate leads to the formation of diphenylmethanediol diacetate (**18**).

From the experimental results it is suggested that for the formation of 1,3,4-oxadiazoles **10** the aldehydic hydrogen is necessary. Absence of the aldehydic hydrogen leads to the formation of 2*H*,5*H*-1,3,4-oxadiazoles **15** as in the case of the acetone hydrazones **9a-9c**. However, when the acetone moiety [$(CH_3)_2$-C=] is replaced with the acetophenone [CH_3(Ph)-C=] or the benzophenone [$(Ph)_2$-C=] the acetylation of the ketonic carbon atom does not take place due to steric hindrance according to scheme III. The diazocompound ($Ph_2C=\overset{+}{N}=\bar{N}$) is stable enough (14) so that acetoxylation can take place to give the *O*-acetylbenzydrol **17** which on further acetoxylation gives **18**. However, the diazocompound [CH_3-(Ph)-$C=\overset{+}{N}=\bar{N}$] being less stable looses nitrogen to give finally acetophenone.

The spectral data for the hydrazones **8** and **9** and the oxidation products **10, 11** and **15** are given in Tables I, III, IV and V. In addition ^{13}C nmr spectrum in deuteriochloroform solution of 2,2-dimethyl-4-acetoxy-4-benzyl-2*H*,5*H*-1,3,4-oxadiazole (**15a**) showed three signals at 21.72, 23.37 and 24.25 ppm for the methyl carbon atoms, one signal at 41.19 ppm for the methylene carbon atom, four signals at 127.26, 128.09, 130.68 and 133.16 ppm for the aromatic carbon atoms and one signal at 167.82 ppm for the carbonyl carbon atom. The two quaternary carbons are probably masked by the methyl carbons.

EXPERIMENTAL

All melting points are uncorrected and they were obtained with a Kofler hot stage apparatus. The ir spectra were obtained with a Perkin-Elmer Model 257, whereas nmr spectra reported in δ units with a Varian Associates A-60A spectrometer with TMS as internal reference. The mass spectra were obtained with a Hitachi-Perkin-Elmer Model RMU-6L

spectrometer with ionization energy 70eV. Analyses were performed with a Perkin-Elmer Model 240 CHN Analyser.

Preparation of arylacetylhydrazones of aldehydes **8** and ketones **9**. The hydrazones (Table I) were prepared according to well known procedures (14-17), namely by reacting the aldehyde or the ketone with the corresponding acid hydrazide in ethanol. Only the benzophenone hydrazones were prepared in a sealed tube at 110°.

Oxidation of Arylacetylhydrazones with Lead Tetraacetate.

A general procedure (9) is described. To a suspension of 0.02 moles of hydrazone in 40 ml of methylene chloride, a solution of 0.02 moles of lead tetraacetate in 40 ml methylene chloride was added and the mixture was stirred at room temperature for 4 hours. The methylene chloride solution was treated with water, filtered and the organic layer was washed with sodium carbonate solution, water, and then dried. The oil which was left behind was subjected to column chromatography on silica gel (petroleum ether-ethyl acetate 5:1) and the products were isolated (Tables II, III, IV, V). By the oxidation of acetophenone hydrazones **9d**, **9e**, **9f** only the starting ketone was isolated in 40%. The benzophenone hydrazone **9g** gave *O*-acetylbenzydrol (**17**) (18) in 54% yield, mp 39-41° (lit (18) mp 42°) and the hydrazone **9h** gave *O*-acetylbenzydrol (**17**) in 40% yield and diphenylmethylene diacetate (**18**) (13) in 30% yield, mp 121-122° (lit (13) mp 121°).

REFERENCES AND NOTES

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