Preparation of *trans-o*-(Bromomethyl)ethyl Cinnamate (7). Wohl–Ziegler bromination[25] of **6** yielded **7**. A mixture of **6** (10.0 g, 53 mmol), *N*-bromosuccinimide (9.7 g, 55 mol) (Aldrich), dibenzoyl peroxide (0.8 g), and carbon tetrachloride (200 ml) was heated under reflux until formation of succinimide was complete (3 h). The mixture was cooled and the succinimide filtered and washed with carbon tetrachloride. A preliminary vacuum distillation (short path) afforded two fractions: bp 100–130 °C (0.4 mm) (2.5 g) and bp 130–154 °C (0.4 mm) (10.24 g). The higher boiling fraction was crystallized from methanol (75 ml) at −65 °C to give a 65% yield (9.3 g) of **7**: mp 33.0–33.5 °C; IR (CCl_4) 2995, 1720, 1640 cm^{-1}; NMR (CCl_4) δ 1.31 (t, 3 H, CH_3), 4.22 (q, 2 H, CH_2), 4.5 (s, 2 H, CH_2), 6.30 (d, 1 H, C=CH) (J = 16 Hz), 7.28 (m, 4 H, aromatic), 7.90 (d, 1 H, C=CH) (J = 16 Hz).

Preparation of *o*-Formylbenzyl Bromide (8). A solution of **7** (5.38 g, 20 mmol), methanol (80 ml), and methylene chloride (20 ml) was ozonolyzed at −63 °C. The blue color from the presence of excess ozone indicated that ozonolysis was complete after 0.25 h. After reduction with dimethyl sulfide (10 ml) was complete (0.5 h) (in a preparative reaction, this methanol solution ordinarily was used without further purification), the solution was diluted with benzene (25 ml). The organic phase was washed with water (25 ml) and concentrated to give 1.02 g of clear, yellow liquid. Crude **8** was distilled (bulb to bulb) to give a 10% yield (0.4 g) of brown liquid: bp 70–75 °C (0.4 mm); IR (neat) 2840, 2770, 1700 cm^{-1}; NMR (CCl_4) δ 4.9 (s, 2 H, CH_2), 7.55 (m, 4 H aromatic), 10.1 (s, 1 H, CHO).

Preparation of 2,2-Diformyldibenzyl Diselenide Dimethyl Acetal (10). A solution of **7** (4.0 g, 14.8 mmol) and methanol (100 ml) was ozonolyzed at −60 °C. The blue color from the presence of excess ozone indicates that ozonolysis was complete after 20 min. Reduction with dimethyl sulfide (10 ml) was complete in 0.5 h.

To the stirred reaction mixture, trimethyl orthoformate (28 ml) and *p*-toluenesulfonic acid (0.8 g) were added at room temperature. After stirring for 48 h, the crude product was neutralized with solid potassium carbonate and methanol (80 ml) was distilled off at atmospheric pressure.

The crude acetal in methanol (21 ml) was added dropwise to a freshly prepared solution of bismethoxymagnesium diselenide[26] (50 mmol) at room temperature. After the resulting dark solution was stirred at room temperature for 18 h, water (1 l.) was added to the reaction mixture and the crude product was extracted with diethyl ether (4 × 125 ml). The combined, yellow ether extracts were washed with saturated sodium bicarbonate solution (75 ml) and saturated sodium chloride solution (75 ml), and dried ($MgSO_4$). Concentration of the filtered ether extracts afforded a 26% yield (0.94 g) of crude **10**, which was applied to a hexane-packed column (17 × 9.3 cm) of basic II alumina (Woelm) and eluted with hexane/benzene (30:70). A 10.8% yield (0.39 g) of **10** was obtained as an oil: IR (neat) 2840, 1200, 1110, 1080, 1060 cm^{-1}; NMR ($CDCl_3$) δ 3.22 (s, 12 H, OCH_3), 4.11 (s, 4 H, CH_2), 5.54 (s, 2 H, CH), 7.34 (m, 8 H, aromatic); mass spectrum *m/e* (rel intensity) 490 (39, M^+), 245 (22), 165 (100), 134 (39), 119 (26), 105 (79).

Preparation of 2,2′-Diformyldibenzyl Diselenide (4). A solution of **10** (40 mg) and deuterium oxide (2 drops) in deuterioacetone (360 mg) was added to an NMR tube. Hydrolysis was effected with trifluorodeuterioacetic acid (1 drop) after approximately 2 h at room temperature (a brown, uncharacterized sediment which settled out of the tube at this time was removed from the reaction mixture). The resultant solution was used directly for determining the NMR spectrum. This solution was then concentrated, diluted with anhydrous diethyl ether (2 ml), and dryed over $MgSO_4$. The filtered ether solution crystallized in the freezer and yielded yellow-tan crystals, mp 50–64 °C. Two recrystallizations from anhydrous diethyl ether afforded yellow-tan crystals of **4**: mp 72.5–73.5 °C; IR (neat) 2750, 1695 cm^{-1}; NMR ($CLCl_3$) δ 4.30 (s, 4 H, CH_2), 7.50 (m, 8 H, aromatic), 10.1 (s, 2 H, CHO); mass spectrum *m/e* (rel intensity) 398 (6, M^+), 199 (0.3), 119 (100), 91 (42), 65 (6) (mp of Magdasieva and Vdovin product,[3] 71–72 °C).

2-Selenaphthalide was synthesized from phthalide (Aldrich Chemical Co.) by the method of Gunther.[4b] Samples recrystallized from petroleum ether were used for IR and NMR spectral determinations.

2-Thiaphthalide was synthesized from phthalide (Aldrich Chemical Co.) by the method of Prey.[10] Samples purified by benzene elution from alumina columns gave mp 55.5–56.5 °C (reported 55–60 and 60 °C).[27]

Registry No.—**1**, 35951-68-9; **2**, 58534-05-7; **4**, 60633-89-8; **5**, 939-57-1; **6**, 24393-48-4; **7**, 60633-90-1; **8**, 60633-91-2; **10**, 60633-92-3.

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Homogeneous Catalytic Cyclization and Oxidation of Diols

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The 1,4-diketone unit is an important synthon for a variety of synthetic quests. Such diones are difficult to prepare directly via oxidation of 1,4-diols because other products are actually favored.[2] Hence, several indirect approaches have been developed.[3] These routes are stoichiometric and often involve several steps. We sought an alternative procedure, one that would be catalytic; palladium chloride oxidation of diols was chosen because Pd(II) is a mild oxidizing agent that can be made catalytic in an oxygen environment by the addition of copper chloride. Also, unlike many other oxidizing agents, palladium is not known to effect cleavage of the carbon skeleton in 1,2-diols, so it was considered that Pd(II) might be useful for preparing a variety of diones of differing structural relationships.

The oxidation of simple alcohols with palladium salts has been reported by Lloyd.[4] He found that primary and secon-

Table I. Reactions of Diols with Palladium Catalysts

Registry no.	Entry	Diol	Catalyst	Temp, °C	Time, h	Conversion, %	Ether (%)[a]	Other (%)
513-85-9	1	2,3-Butanediol	*b*	150	24	12		2,3-Butanedione (8) (431-03-8)[f]
1460-57-7	2	*trans*-1,2-Cyclohexanediol	*c*	200	96	Trace		
107-88-0	3	1,3-Butanediol	*c*	150	30	15		2-Butenal (5) (4170-30-3)[f]
110-63-4	4	1,4-Butanediol	*b*	125	24	42	Tetrahydrofuran (40) (109-99-9)[f]	Trace oxidation
	5		*c*	150	6	97	Tetrahydrofuran (95)	
2935-44-6	6	2,5-Hexanediol	*b*	100	24	52	2,5-Dimethyltetrahydrofuran (35) (2144-41-4 (cis))[f]	2,5-Hexanedione (15) (110-13-4)[f]
	7		*c*	150	6	97	2,5-Dimethyltetrahydrofuran (82–92) (2390-94-5 (trans))[f]	
	8		*d*	150	6	40	2,5-Dimethyltetrahydrofuran (39)	
	9		*e*	150	6	85	2,5-Dimethyltetrahydrofuran (80)	
110-03-2	10	2,5-Dimethyl-2,5-hexanediol	*b*	150	6	90	2,2,5,5-Tetramethyldihydrofuran (70) (15045-43-9)[f]	Unidentified
931-71-5 (cis) 6995-79-5 (trans)	11	1,4-Cyclohexanediol (cis and trans)	*c*	150	24	15		See text
111-29-5	12	1,5-Pentanediol	*b*	150	6	100	Tetrahydropyran (75–80) (142-68-7)[f]	Unidentified polymer
	13		*c*	150	6	68	Tetrahydropyran (58)	Unidentified polymer
629-11-8	14	1,6-Hexanediol	*b*	150	24	63	Oxacycloheptane (10) (592-90-5)[f]	1,7-Dioxacyclotetradecane (48) (1080-24-6)[f]
	15		*c*	150	24	56	Oxacycloheptane (8)	1,7-Dioxacyclotetradecane (35)

[a] Yields are based on millimoles of starting diol even when the conversion was <100%. [b] 0.047 M in $\mathrm{PdCl_2}$ and 0.063 M in $\mathrm{CuCl_2}$. [c] 0.047 M in $\mathrm{PdCl_2}$, 0.063 M in $\mathrm{CuCl_2}$, and 0.09 M in $\mathrm{Cu(NO_3)_2}$. [d] 0.047 M in $\mathrm{PdCl_2}$ and 0.09 M in $\mathrm{Cu(NO_3)_2}$. [e] 0.047 M in $\mathrm{PdCl_2}$ and 0.13 M in NaCl. [f] Registry no.

dary alcohols are oxidized to the corresponding acetals and ketones using mild conditions, typically 70–120 °C. Tertiary alcohols were found to be unreactive. Oxidation of ethylene glycol produced low yields of the cyclic acetal of its glycolic aldehyde, 2-methylol-1,3-dioxolane. Oxidation of 1,4-butanediol eventuated in low yields of γ-butyrolactone and 2-(ω-hydroxybutoxy)tetrahydrofuran.

Because we wished to avoid lactone formation, our study began with a secondary diol, 2,5-hexanediol. Upon heating the diol to 90 °C with a $\mathrm{PdCl_2–CuCl_2–Cu(NO_3)_2–O_2}$ catalyst system, oxidation was observed to be very slow; only by raising the temperature to 130 °C, whereupon a brown gas was evolved, was a fairly rapid reaction rate observed. After the reaction was completed, instead of finding oxidation to be the primary process, it was discovered that cyclization of the diol had occurred to form 2,5-dimethyltetrahydrofuran (a mixture of cis and trans isomers) in very high yield. In view of the mildness of this conversion and its specificity our attention shifted to the ether-forming process. To determine the generality of this observation, a variety of diols were subjected to similar "oxidative" conditions. The products reported in Table I were obtained by distillation and identified by IR, ^{1}H NMR, and mass spectroscopy.

From the data in Table I it is evident that 1,2- and 1,3-diols react very slowly. *trans*-1,2-Cyclohexanediol was virtually inert to oxidation, even at 200 °C. 2,3-Butanediol was oxidized to its dione in low conversion; other unidentified products were also formed, though in lesser amounts. A great variety of unidentified products were formed from 1,3-butanediol. Carbonyl formation was apparent; 1,2-dihydration was also evidenced as olefins were detected. Other products appeared to be derived from diol–olefin and diol–carbonyl condensations.[5] With the exception of 1,4-cyclohexanediol, 1,4-diols were found to undergo facile reaction and form ethers in high yields. Examples in Table I demonstrate that primary, secondary, and tertiary alcohols all form ethereal products. Cyclization of 1,5-pentanediol occurs readily to generate tetrahydropyran in excellent yield. 1,6-Hexanediol reacts more slowly than 1,4- or 1,5-diols and produces the seven-membered cyclic ether in low yield; actually the 14-membered cyclic diether is the major product. Linear ethers, lactones, or acetals were not detected, though they may have been too nonvolatile to isolate via distillation. Pot residues showed absorptions from 1730 to 1700 cm^{-1}, indicating oxidation. Yields of these residue products are low.

Upon treatment with the palladium "oxidative" catalyst, 1,4-cyclohexanediol (a mixture of cis and trans isomers) conceivably could have produced 1,4-cyclohexanedione and/or 7-oxabicyclo[2.2.1]heptane; instead, after 24 h and a 15% conversion, the only products isolated were high boiling (160–220 °C, 1 mm) and appeared (IR and NMR) to be monoones, acetals, and hemiacetals. Formation of the bicyclic ether apparently was precluded by ring strain.

Concerning these results, several observations are noteworthy. (1) The trend in the ease of cyclization is consistent with that noted for several other reactions in which the ease of ring formation follows the order $4 < 5 \sim 6 > 7$.[6] (2) For 1,4-, 1,5-, and 1,6-diols oxidation is a very minor process at 150 °C.

At lower temperatures (entries 4 and 6) oxidation of a 1,4-diol becomes significant, and modest yields of dione are obtained. However, considering the extended reaction time required to effect a minor conversion, this approach is not considered to be synthetically useful; the only reasonably attractive feature is that this route is catalytic. (3) The reaction is sensitive to the identity of the anion: the rate of cyclization is slower in the presence of nitrate and chloride than chloride alone. Lloyd observed a faster rate of oxidation in the presence of both nitrate and chloride.[4] This difference indicates that the palladium-diol complex undergoing dehydration (cyclization) has more chloride ligands than does the favored palladium-alcohol complex eventuating in carbonyl formation. (4) If copper is omitted (entry 9) the palladium precipitates out after a few minutes. Cyclization continues albeit at a slower rate; heterogeneous catalytic cyclization is well documented for palladium-aluminum alloys, though these are generally employed at significantly higher temperatures.[7] (5) 1,2-Dehydration was observed only with 1,3-butanediol and was slow. Pincol rearrangement products were not observed for any 1,2-diol. (6) The role of Pd(II) in the cyclization is considered to be simply that of a Lewis acid.

Experimental Section

Reactions under Oxygen. Oxidations were carried out in a standard, low-pressure catalytic apparatus (Parr Instrument Co., Model 3911). In typical run a 500-ml glass reactor vessel was charged with 30.0 g of diol along with the desired amounts of catalyst, and the system was sealed, purged three times with oxygen, then pressurized to 60 psig oxygen pressure and rapidly brought to the desired temperature by means of a heating mantle. The temperature was established by standardizing the mantle and variac without pressurizing the system. After the desired time had passed, the reactor was cooled and the contents were collected for analysis.

Product separations and yields were determined by distillation and/or gas chromatography. Product identities were determined by IR, NMR, and mass spectral data.

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A Novel Reaction of Some Fluorospiro[isobenzofuran-1(3*H*),4'-piperidin]-3-ols in 97% Formic Acid

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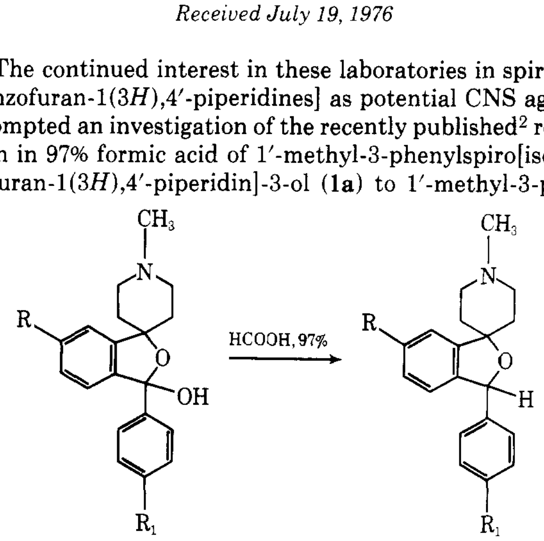
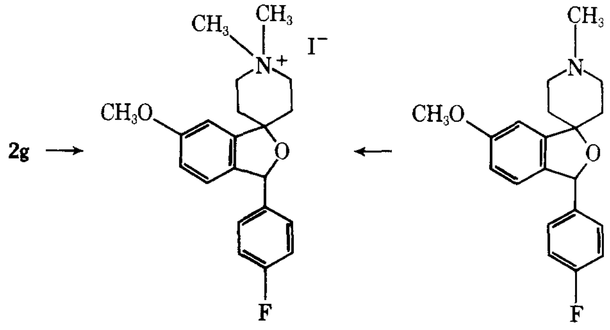
The continued interest in these laboratories in spiro[isobenzofuran-1(3*H*),4'-piperidines] as potential CNS agents[1] prompted an investigation of the recently published[2] reduction in 97% formic acid of 1'-methyl-3-phenylspiro[isobenzofuran-1(3*H*),4'-piperidin]-3-ol (**1a**) to 1'-methyl-3-phenylspiro[isobenzofuran-1(3*H*),4'-piperidine] (**2a**). We have duplicated the published[2] yield of **2a** using identical procedures. Furthermore, we have found that the 6-chloro- and 3-(4-chlorophenyl) analogues of **1a** are reduced cleanly under these conditions to the corresponding analogues of **2a** in recrystallized yields of 85 and 82%, respectively. TLC and NMR showed no evidence of anomalous by-products in either case. However, the reduction of the 3-(4-fluorophenyl) analogue (**1b**)[2] or the 6-fluoro analogue (**1c**) led not only to the corresponding fluorospiro[isobenzofuran-1(3*H*),4'-piperidines] (**2b** and **2d**, in 71 and 36% yields, respectively) but also led to the hydroxy analogues (**2c** and **2e**, in 14 and 41% yields, respectively). Furthermore, both **2f** and **2g** were isolated (in 42 and 23% yields, respectively) from the reduction of the difluoro analogue (**1d**).



1a, R = R^1 = H
b, R = H; R^1 = F
c, R = F; R^1 = H
d, R = R^1 = F

2a, R = R^1 = H
b, R = H; R^1 = F
c, R = H; R^1 = OH
d, R = F; R^1 = H
e, R = OH; R^1 = H
f, R = R^1 = F
g, R = OH; R^1 = F

The structures of **2b–f** were proved by comparison of melting points and infrared and ^{1}H NMR spectra with samples obtained in alternate unambiguous syntheses.[1] The orientation of the hydroxy and fluoro substituents of **2g** was demonstrated by conversion to the methoxy methiodide (**3**). This was shown to be identical with the methiodide obtained from **4** that had been prepared unambiguously.[1]



3 **4**

In the case of the reduction of **1c** to **2d** and **2e**, **2d** could not