Reductions of Benzene Derivatives Whose Benzylic Positions Bear Oxygen Atoms under Mild Conditions

by **Abdullah Menzek***, **Melek Gökmen Karakaya**, and **Afşin Ahmet Kaya**

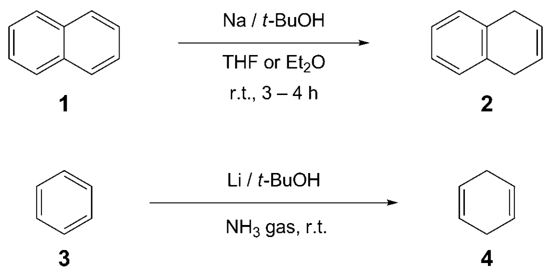
Department of Chemistry, Faculty of Science, Atatürk University, TR-25240 Erzurum
(phone: +90 4422314423; fax: +90 4422360948; e-mail: amenzek@atauni.edu.tr)

Reductions of compounds whose benzylic positions bear O-atoms, such as benzyl alcohol, dibenzyl ether, styrene oxide, benzaldehyde, acetophenone, and benzophenone, to the corresponding non-conjugated dienes were performed by using *t*-BuOH, Li, and gaseous NH_3 in THF at room temperature. In these reductions, it was observed that the functional groups at benzylic positions were reduced first.

Introduction. – As is well known, the reduction of aromatic compounds is very important in organic chemistry and therefore these reactions have been investigated by many chemists [1]. The *Birch* reactions are very commonly used for the reduction of aromatic compounds. However, they generally require harsh conditions such as high temperatures or liquid NH_3 [1][2].

The formation of *Birch* reduction products of naphthalene (**1**) depends on the reaction conditions [1a]. High temperatures (60–145°) have been used in the reduction of naphthalene to 1,4-dihydronaphthalene (**2**) since 1942 [3]. We performed the reduction of naphthalene and naphthalene derivatives with Na and *t*-BuOH to give the corresponding 1,4-dihydronaphthalene and naphthalene derivatives in high yields at room temperature as the sole product (*Scheme 1*) [4].

Scheme 1



Benzene (**3**) and especially benzene rings with electron-donating groups are reduced by alkali metal in liquid NH_3 to the corresponding products [1][2][5]. The temperature is usually below −33° in these reactions. We also performed the reduction of benzene and its derivatives with lithium and *t*-BuOH to the corresponding non-conjugated dienes **4** as the sole product in high yield at room temperature (*Scheme 1*) [6].

Reduction reactions performed at room temperature have certain advantages [4][6]. Reduction reactions of compounds whose benzylic positions bear heteroatoms such as O- and N-atoms may be important under mild conditions. In these compounds, two centres, the benzylic C-atom and the benzene ring, will be reduced, and their reactivities will be different. Therefore, reductions of compounds whose benzylic positions bear O-atoms, such as benzyl alcohol (BnOH), benzaldehyde, benzophenone, styrene oxide, and Bn_2O, were investigated at room temperature with gaseous NH_3 and Li.

Results and Discussion. – Solutions of benzyl alcohol (**5**), dibenzyl ether (**6**), and styrene oxide (**7**) in THF and *t*-BuOH were reacted with metallic Li and in an NH_3 atmosphere at room temperature (*Scheme 2*, *Table*). In these reduction reactions, NH_3 is found in atmospheres and solutions of the reactions as gaseous and dissolved, respectively. 1-Methylcyclohexa-1,4-diene (**8**) was obtained from the reduction of BnOH in 54% yield and is a reduction product of toluene [6]. The ^{1}H-NMR spectrum of the mixture formed in the reduction of Bn_2O (**6**) showed that it contains toluene (**9**) and 1-methylcyclohexa-1,4-diene (**8**) [6][7] in an 8 : 92 ratio (*Table*). In the same way, it was observed that 2-phenylethanol (**10**) and 2-(cyclohexa-1,4-diene-1-yl)ethanol (**11**) were formed from styrene oxide (**7**). Compound **11** [8] is a reduction product of alcohol **10**. To test the formation of **11** from alcohol **10**, we studied the reduction of **10** under the same conditions. It was observed that the reduction products **11** [8] and **12** [9] in a 31 : 7 ratio were present in the reaction mixture (*Scheme 2*). When the reduction of

Scheme 2

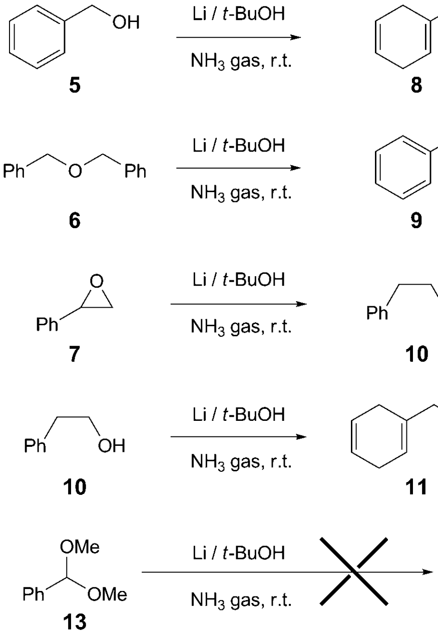


Table. *Reduction Reactions of Benzene Derivatives Whose Benzylic Positions Bear O-Atoms*

	Li (equiv.)	*t*-BuOH (equiv.)	Time [h]	Ratio of the compounds in the mixture (yield [%])[a]
5	7	8	20	**5/9/8**[b]) 0 : 0 : 100
6	11	10	12	**6/9/8** 0 : 8 : 92
7	2.9	2.8	24	**7/10/11** 23 : 71 : 6
10	8	7	10	**10/11/12** 24 : 62 : 14
13				No reaction
20	4.5	4	22	**20/5/9/8** 26 : 31 : 19 : 24
21	7	7	7	**21/23/24/25** 27 : 8 : 56 : 9
25	3.5	3	24	**25/26/27/28/29** 7 : 1 : 2 : 0 : 0
25	8	7	24	**25/26/27/28/29** 0 : 0 : 62 : 14 : 24
25	17	15	24	**25/26/27/28/29** 0 : 0 : 0 : 59 : 41

[a]) Equivalents of benzene derivatives were taken as one, and yields of the products in the mixtures [%] were determined from their ^{1}H-NMR spectra. [b]) Obtained by work-up procedure *A* as described in the *Exper. Part.*

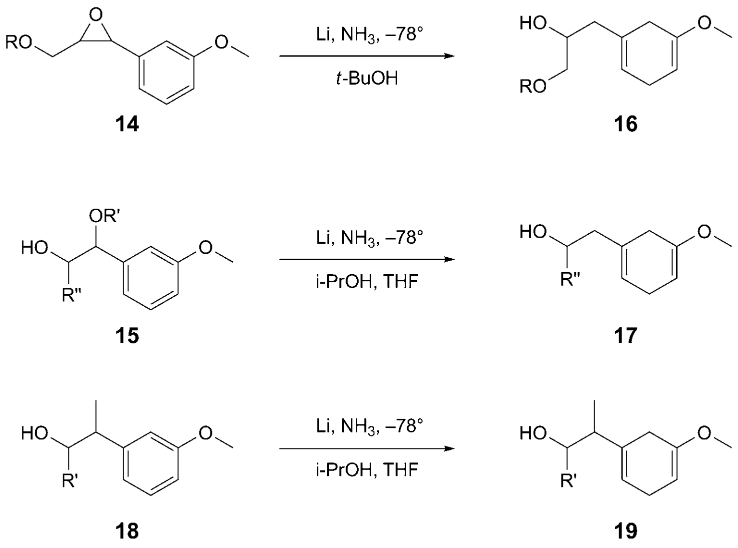
(dimethoxymethyl)benzene (**13**) was attempted at room temperature with gaseous NH_3 atmosphere with different equivalents of *t*-BuOH and Li, in all cases only starting material was found. In the reductions of **5**–**7**, functional groups such as hydroxide, ether, and epoxide in benzylic positions were removed. However, ether and OH groups in β-position (with respect to Ph) of **7** and **10** were not affected by the reduction.

Similar reactions are known (*Scheme 3*) [10]. Compounds **14** and **15** bear O-atoms in benzylic positions as in the form of an epoxide and an ether group, respectively, and they were reduced to the corresponding products **16** and **17**, respectively. Only the aromatic ring was reduced in the case of **18** [10a]. In **15** and **18**, the OH groups in β-position of the aromatic rings were not removed by reduction. These results support our observations.

Solutions of benzaldehyde (**20**), acetophenone (**21**), and benzophenone (**22**) in THF and *t*-BuOH were reacted with metallic Li at room temperature in an NH_3 atmosphere (*Scheme 4*, *Table*). From these reduction reactions, benzylic alcohols, alkyl benzenes, and 1-alkylcyclohexa-1,4-dienes were obtained as the reduction products. Therefore, the products of benzaldehyde (**20**) were **5**, **8** [6][7], and **9**, while the products of acetophenone (**21**) were **23**, **24**, and **25** [11]. The reduction of benzophenone (**22**) gave 1,1′-methanediyldibenzene (**26**) and the cyclohexadiene derivatives **27**, **28**, and **29**.

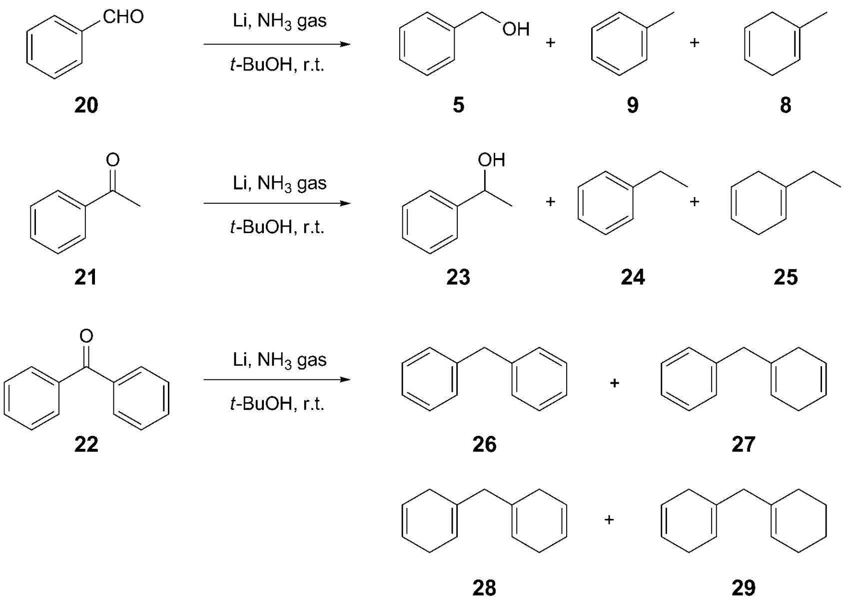
According to reduction reactions of benzene derivatives with C=O groups in the benzylic position, *e.g.*, **20**, **21**, and **22**, the sequence of reduction products formed should be benzylic alcohols, alkyl benzenes, and 1-alkylcyclohexa-1,4-dienes. However, the ratio of the products should depend on equivalents of reagents. Therefore, we studied the reduction reactions of benzophenone (**22**) with different equivalents of *t*-BuOH and Li (*Table*). 1,1′-Methanediyldibenzene (**26**) and (cyclohexa-1,4-dien-1-ylmethyl)benzene (**27**) were present in the reaction mixture of benzophenone (1 equiv.) with *t*-BuOH (3 equiv.), and Li (3.5 equiv.), while products **28** and **29** were absent. The products **28** and **29** were only present under reaction conditions with *t*-BuOH (15 equiv.) and Li (17 equiv.). The reduction products **26**–**29** should be formed in the

Scheme 3



R = SiR'_3, R' = $CH(Me)_2$, R'' = CH_2CH_2–cyclohexyl

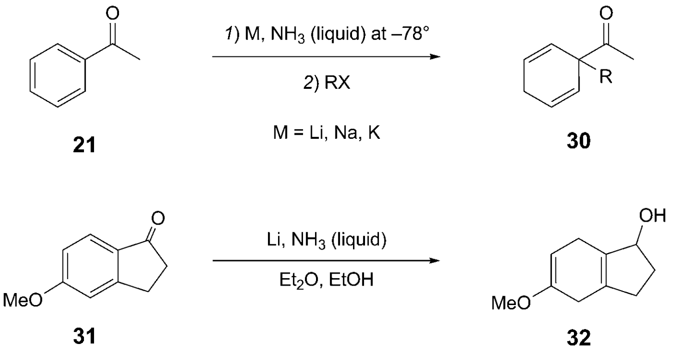
Scheme 4



reduction of benzophenone *via* diphenylmethanol, which was not observed in these reactions. It has been reported that diphenylmethanol was formed in 3% yield in the conversion of benzophenone with Na into 1,1′-methanediyldibenzene [12].

We have not observed any diene or alkene with C=O or OH functional groups formed in the reaction mixtures of benzyl alcohol (**5**), dibenzyl ether (**6**), styrene oxide (**7**), benzaldehyde (**20**), acetophenone (**21**), and benzophenone (**22**) with *t*-BuOH, metallic Li and in an NH_3 atmosphere at room temperature. It has been reported that acetophenone (**21**) was converted into dienone **30** [13] by reductive alkylation and that the indanone derivative **31** was reduced to dienol **32** [1e] (*Scheme 5*). The reason why dienes or alkenes with CO or OH functional groups were not observed in our reactions may be that intermediates such as anions or radical anions in benzene rings are not more stable than those in benzylic position.

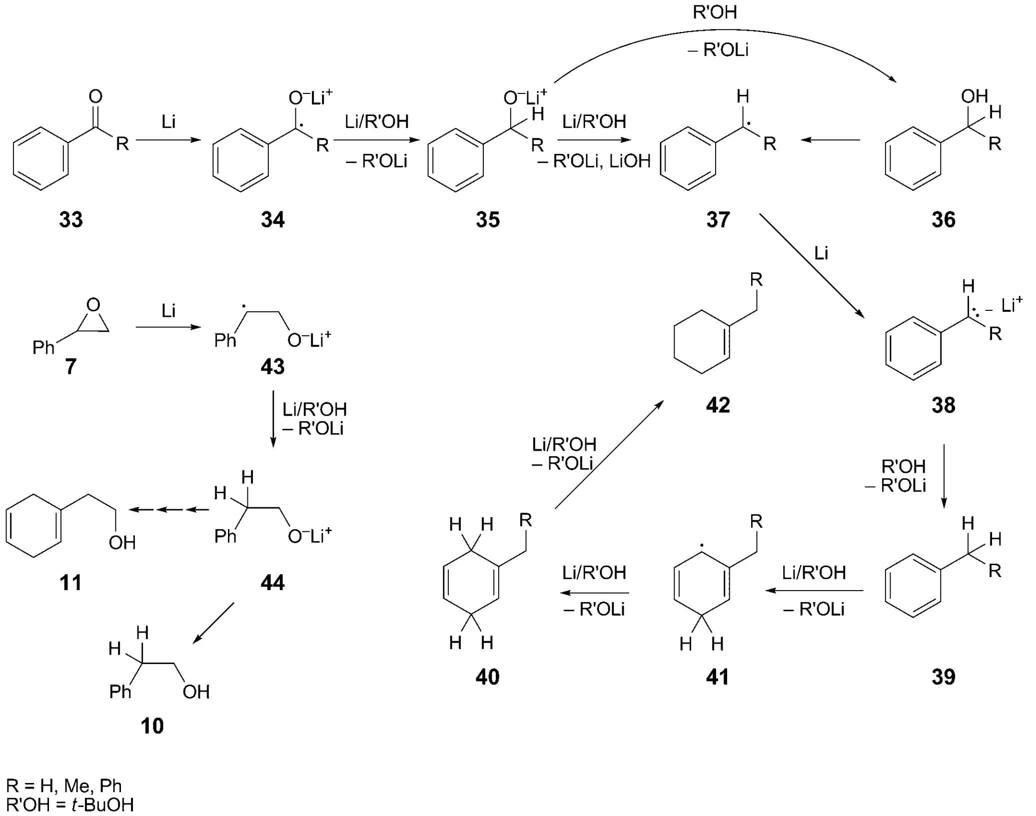
Scheme 5



To rationalize the formation of the reduction products, the following reaction mechanism was proposed (*Scheme 6*). Li attacks the C=O group in aldehyde or ketone **33** to give the radical lithium alkoxide **34**. Intermediate **34** reacts with Li/R′OH to convert into intermediate **35** *via* the dianion. Intermediate **35** can react with both a proton (from R′OH), to give benzyl alcohol derivative **36**, and Li/R′OH, to give radical **37**. However, a benzyl alcohol derivative **36** can also be converted into intermediate **37** under the reaction conditions. Intermediate **37** can react with Li and R′OH (as proton source), respectively, to give, *via* lithium phenylmethanides **38**, alkyl benzene **39**. As earlier described [1][6], alkyl benzenes **39** were reduced with Li/R′OH to the corresponding cyclohexa-1,4-dienes **40** *via* intermediate **41**. Alkene **42** is produced from cyclohexa-1,4-dienes **40** by reduction of one C=C bond.

In the reduction of epoxide **7**, Li attacks the benzylic C-atom of the epoxide in **7** to give radical lithium alkoxide **43** which is a stabilized intermediate [14]. The latter also reacts with Li to give a dianion, and then this dianion is converted with alcohol (R′OH) into intermediate **44**. Intermediate **44** can react with R′OH (as proton source) to give 2-phenylethanol (**10**), which is converted into alcohols **11** and **12** [7] by reduction of the aromatic ring. While OH groups at benzylic C-atoms were reduced, the OH groups at other positions were not reduced. However, radicals, radical anions, or dianions such as **34**, **37**, and **43** may resonate with their aromatic rings.

Scheme 6



As reported in the literature [13], it was expected that 1,4-diene derivatives with C=O, OR, and OH groups positioned at the C-atom neighboring the ring would be formed in these reactions. They were not observed, however.

Conclusions. – We have performed reductions of compounds whose benzylic positions bear O-atoms, such as BnOH, Bn_2O, styrene oxide, benzaldehyde, acetophenone, and benzophenone, with gaseous NH_3 atmosphere and Li to yield the corresponding nonconjugated dienes at room temperature. As reported in [13], it was expected that 1,4-diene derivatives with C=O, OR, and OH groups neighboring the ring would be formed in these reactions, but they were not observed. The reason for this may be that intermediates such as anions or radical anions in benzene rings are not more stable than those in benzylic position. Therefore, it was observed that functional groups at benzylic positions were reduced first, and only then the benzene rings were reduced. Ratios of reduction products depend on equivalents of reactants. A reduction product(s) may be obtained as the major or sole product if the equivalents of reactants were adjusted.

However, 2-phenylethanol (**10**) and a cyclohexadiene derivative with a OH group, **11**, were obtained in the reduction of styrene oxide (**7**) as a mixture. In order to test the formation of **11** [8] from alcohol **10**, the reduction of alcohol **10** was studied under the same conditions, and alcohol **11** was obtained from this reaction. It was seen that the OH group at non-benzylic C-atoms was not reduced, while those at benzylic position were reduced.

Experimental Part

All chemicals and solvents are commercially available and were used after distillation or treatment with drying agents except for NH_3 gas. There is no need to purify the commercial NH_3 gas by distilling. Free oil on lithium wire was removed with hexane, and lithium was freshly cut into pieces in hexane before use. All reactants (BnOH, Bn_2O, styrene oxide, benzaldehyde, acetophenone, benzophenone, (dimethoxymethyl)benzene and benzophenone) and some products (**5**, **9**, **10**, **22**, **23**, and **26**) are commercial products. The other products (**8** [6][7], **11** [8], **12** [9], and **24** [11]) are known. Column chromatography (CC) was performed on silica gel (SiO_2; 60 mesh, *Merck*). Preparative thick-layer chromatography (PLC): 1 mm of silica gel *60 PF* (*Merck*) on glass plates. ^1H- and ^{13}C-NMR Spectra: 200 (50)-MHz *Varian* spectrometer; δ in ppm, with Me_4Si as the internal standard. Elemental analyses were performed on *a Leco CHNS-932* apparatus.

General Procedure. In a two-necked, round-bottomed flask fitted with stirring bar were placed dry THF, *t*-BuOH, and compounds whose benzylic positions bear O-atoms. The flask was attached to a balloon filled with NH_3 gas whose pressure was *ca.* 1 atm, and the resulting soln. was stirred and cooled in an ice–water bath. After the mixture was stirred in NH_3 atmosphere for 15–20 min, freshly cut small pieces of wire lithium were added within 3–5 min. The temp. of the bath was allowed to rise gradually to r.t. Then, the mixture was stirred for the appropriate time and then cooled in an ice–water bath again. Then, the NH_3-gas tube was closed, the balloon was removed, and cold H_2O or HCl (1%) was added carefully drop by drop to the flask until all the Li was consumed. The mixture was poured into a mixture of H_2O and ice. The workup was carried out in two different ways: *A*) The org. layer was separated and washed several times with ice–water to remove *t*-BuOH and THF. Then, it was dried (Na_2SO_4) to give the reduction product(s). *B*) The mixture was extracted with a solvent ($CHCl_3$, CH_2Cl_2, or others), and the extracts were washed several times with ice–water to remove *t*-BuOH and THF. The soln. was dried (Na_2SO_4) and the solvent removed to yield the reduction product(s). Ratios and yields of the products in the mixtures were determined by ^1H-NMR spectroscopy.

Reduction of BnOH (*= Phenylmethanol*; **5**). According to the *General Procedure*, in a 250 ml-flask, with *t*-BuOH (27.45 g, 370 mmol, 8 equiv.), dry THF (150 ml), and benzyl alcohol (5.0 g, 46 mmol, 1 equiv.), and Li (3.24 g, 460 mmol, 10 equiv.); reaction time 20 h. Workup procedure *A* (2 × 50 ml of H_2O for wash); *1-methylcyclohexa-1,4-diene* (**8**) [6][7] (2.5 g, 54%).

Reduction of 2-Phenyloxirane (**7**). According to the *General Procedure*, in a 250 ml-flask, with *t*-BuOH (17.27 g, 233 mmol), dry THF (50 ml), and 2-phenyloxirane (10 g, 83.3 mmol), and Li (1.74 g, 240 mmol); reaction time 24 h. Workup procedure *B*; product mixture (7.74 g) of *2-phenyloxirane* (**7**), *2-phenylethanol* (**10**), and a reduction product of 2-phenylethanol, *2-(cyclohexa-1,4-dien-1-yl)ethanol* (**11**) [8] with a ratio of 23:71:6.

Reduction of 2-Phenylethanol (**10**). According to the *General Procedure*, in a 250 ml-flask, with *t*-BuOH (12.74 g, 172 mmol, 7 equiv.), dry THF (50 ml), and 2-phenylethanol (3.3 g, 24.6 mmol, 1 equiv.), and Li (1.38 g, 196 mmol, 8 equiv.); reaction time 10 h. Workup procedure *B*; product mixture (2.94 g) of 2-phenylethanol (**10**), 2-(cyclohexa-1,4-dien-1-yl)ethanol (**11**) [8] (60%), and *2-(cyclohex-1-en-1-yl)ethanol* (**12**) [9] (13%) with a ratio of 24:62:14.

Reduction of Bn_2O (**6**). According to the *General Procedure*, in a 100 ml-flask, with *t*-BuOH (7.5 g, 100.9 mmol, 10 equiv.), dry THF (40 ml), and dibenzyl ether (2.0 g, 10.1 mmol, 1 equiv.), and Li (777 mg, 111 mmol, 11 equiv.); reaction time 24 h. Workup procedure *B*; product mixture (643.44 mg) of toluene (**9**) (3.0%) and 1-methylcyclohexa-1,4-diene (**8**) [6][7] (33.0%), in a ratio of 8:92.

Reduction of Benzaldehyde (**20**). According to the *General Procedure*, in a 250 ml-flask, with *t*-BuOH (44.47 g, 0.6 mol), dry THF (110 ml), and benzaldehyde (15.9 g, 0.15 mol), and Li (4.725 g, 0.675 mol); reaction time 22 h. Workup procedure *B*; product mixture (5.20 g) of benzaldehyde (**20**) (9.1%), phenylmethanol (**5**) (10.9%), toluene (**9**) (6.7%), and 1-methylcyclohexa-1,4-diene (**8**) [6][7] (8.4%), in a ratio of 26 : 31 : 19 : 24.

Reduction of Acetophenone (=*1-Phenylethanone*; **21**). According to the *General Procedure*, in a 250 ml-flask, with *t*-BuOH (62.16 g, 0.84 mol, 7 equiv.), dry THF (50 ml), acetophenone (24 g, 0.12 mol, 1 equiv.), and Li (5.88 g, 0,84 mol, 7 equiv.); reaction time 7 h. Workup procedure *B*; product mixture (7.64 g) of acetophenone (**21**) (9.3%), *ethylbenzene* (**24**) (19.2%), *1-phenylethanol* (**23**) (2.8%), and *1-ethylcyclohexa-1,4-diene* (**25**) [10] (3.1%), in a ratio of 27 : 56 : 8 : 9.

Reduction of Benzophenone (= *Diphenylmethanone*; **25**). Benzophenone was reduced under three different conditions (see below). Workup procedure *A* was used for these reactions. Crude mixtures were submitted to PLC or CC (SiO_2) with hexane. The reduced products *diphenylmethane* (**26**), *(cyclohexa-1,4-dien-1-ylmethyl)benzene* (**27**), *1,1′-methanediylbis(cyclohexa-1,4-diene)* (**28**) and *1-(cyclohexen-1-ylmethyl)cyclohexa-1,4-diene* (**29**) were obtained. However, ratios of the products in the mixtures were determined on the basis of their ^{1}H-NMR spectra.

1) In a 250-ml flask, with *t*-BuOH (2.444 g, 32.967 mmol, 3 equiv.), dry THF (80 ml), and benzophenone (2.0 g, 10.989 mmol, 1 equiv.), and Li (267.2 mg, 38.462 mmol, 3.5 equiv.); reaction time 24 h. The residue (2.145 g) was obtained, and the ratio of the products **25**, **26**, and **27** was 7 : 1 : 2.

2) In a 250-ml flask, with *t*-BuOH (5.702 g, 76.923 mmol, 7 equiv.), dry THF (80 ml), and benzophenone (2.0 g, 10.989 mmol, 1 equiv.), and Li (615.4 mg, 87.912 mmol, 8 equiv.); reaction time 24 h. The residue (1.550 g) was obtained, and ratios of the products **27**, **28**, and **29** was 62 : 14 : 24.

3) In a 250-ml flask, with *t*-BuOH (12.217 g, 164.8 mmol, 15 equiv.), dry THF (80 ml), and benzophenone (2.0 g, 10.989 mmol, 1 equiv.), and Li (1.308 g, 186.8 mmol, 17 equiv.); reaction time 24 h. The residue (1.636 g) was obtained, and ratio of the products **28** and **29** was 59 : 41.

(Cyclohexa-1,4-dien-1-ylmethyl)benzene (**27**). Colorless liquid. IR ($CHCl_3$): 3051, 2897, 2871, 2820, 2382, 1661, 1610, 1507, 1455, 1429, 1095, 1043, 966. ^{1}H-NMR (200 MHz, $CDCl_3$): 7.30 – 7.19 (*m*, 5 arom. H); 5.70 (br. *s*, 2 olefinic H); 5.60 (br. *s*, 1 olefinic H); 3.30 (br. *s*, 2 aliphatic H); 2.73 – 2.50 (*m*, 4 aliphatic H). ^{13}C-NMR (50 MHz, $CDCl_3$): 141.7 (C); 136.5 (C); 131.0 (CH); 130.6 (CH); 130.3 (CH); 128.0 (CH); 126.3 (CH); 126.1 (CH); 122.3 (CH); 46.3; 30.8; 29.9. Anal. calc. for $C_{13}H_{14}$: C 91.71, H 8.29; found: C 91.44, H 8.27.

1,1′-Methanediylbis(cyclohexa-1,4-diene) (**28**). Colorless liquid. ^{1}H-NMR (200 MHz, $CDCl_3$): 5.70 (br. s, 4 olefinic H); 5.48 (br. s, 2 olefinic H); 2.72 – 2.54 (m, 10 aliphatic H). ^{13}C-NMR (50 MHz, $CDCl_3$): 134.8 (CH); 126.5 (CH); 126.1 (CH); 122.2 (CH); 48.2; 30.6; 28.9. Anal. calc. for $C_{13}H_{16}$: C 90.64, H 9.36; found: C 90.41, H 9.38.

1-(Cyclohexen-1-ylmethyl)cyclohexa-1,4-diene (**29**). Colorless liquid. ^{1}H-NMR (200 MHz, $CDCl_3$): 5.71 (br. *s*, 2 olef. H); 5.45 (br. *s*, 2 olef. H); 2.72 – 2.51 (*m*, 6 aliph. H); 2.00 – 1.87 (*m*, 4 aliph. H); 1.67 – 1.52 (*m*, 4 aliph. H). ^{13}C-NMR (50 MHz, $CDCl_3$): 137.5 (C); 135.4 (C); 126.6 (CH); 126.1 (CH); 124.9 (CH); 121.8 (CH); 48.8; 30.6; 29.8; 28.9; 27.4; 25.0; 24.5. Anal. calc. for $C_{13}H_{18}$: C 89.59, H 10.41; found: C 89.94, H 10.37.

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