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Synthesis of substituted bis(3,3-dialkyl-3,4-dihydro-1-isoquinolyl)methanes

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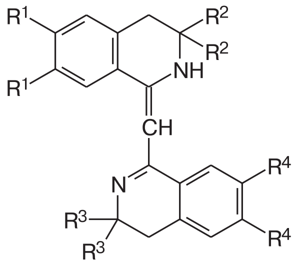
Symmetrical and non-symmetrical substituted bis(3,4-dihydro-1-isoquinolyl)methanes were synthesized by fusion of substituted 1-methylthio-3,4-dihydroisoquinolines with 1-methyl-3,4-dihydroisoquinolines and by the Ritter reaction of 1,1-dialkyl-2-arylethanols with 1-cyanomethylidene-1,2,3,4-tetrahydroisoquinoline or malononitrile.

Key words: isoquinolines, nitriles, malononitrile, the Ritter reaction.

Porphyrins, phthalocyanines, and some alkaloids contain two and more heterocycles linked to each other through the methine or methylene bridge. As a rule, such substances manifest pronounced complex-forming properties. It has previously[1] been shown that bis(3,3-dimethyl-3,4-dihydro-1-isoquinolyl)methane (**1a**) forms a complex with cobalt chloride. Compound **1a** was prepared by heating of 3,3-dimethyl-1-methylthio-3,4-dihydroisoquinoline with malonic acid[2] or in the reaction of 3,3-dimethyl-1-methylthio-3,4-dihydroisoquinoline with methylmagnesium iodide or with 1-methyl-3,4-dihydroisoquinoline enamine anion.[3] The present study is aimed at searching for different routes of synthesis of bis(3,3-dimethyl-3,4-dihydro-1-isoquinolyl)methane (**1a**) and developing of preparative methods for the synthesis of other symmetrically and non-symmetrically substituted bis(3,3-dialkyl-3,4-dihydro-1-isoquinolyl)methanes **1b—h**.



1a—h

$R^1 = R^4 = H, R^2 = R^3 = Me$ (**a**);
$R^1 = R^4 = H, R^2 = Me, (R^3)_2 = (CH_2)_4$ (**b**);
$R^1 = R^4 = H, R^2 = Me, (R^3)_2 = (CH_2)_5$ (**c**);
$R^1 = OMe, R^2 = R^3 = Me, R^4 = H$ (**d**);
$R^1 = R^4 = OMe, R^2 = R^3 = Me$ (**e**);
$R^1 = OMe, R^2 = Me, (R^3)_2 = (CH_2)_4, R^4 = H$ (**f**);
$R^1 = OMe, R^2 = Me, (R^3)_2 = (CH_2)_5, R^4 = H$ (**g**);
$R^1 = R^4 = H, (R^2)_2 = (R^3)_2 = (CH_2)_4$ (**h**)

Results and Discussion

The Ritter reaction of malononitrile (**2**) with the corresponding tertiary or secondary alcohols (Scheme 1, method ***A***) seems to be a convenient method for the synthesis of symmetrical bis(3,3-dialkyl-3,4-dihydro-1-isoquinolyl)methanes **1a,e,h**. For example, the reaction of nitrile **2** with alcohols **3a,b**, **4** in the presence of H_2SO_4 affords compounds **1a,e,h** in 78, 20, and 56% yields, respectively. Synthesis of **1a,e** is impeded by the formation of intermediate nitriles **5a,b**, which were identified in the reaction mixture by TLC, and other by-products. The reaction can easily be monitored by TLC because nitriles **5a,b** are colored blue upon treatment with chloranil[4] and bis(3,3-dialkyl-3,4-dihydro-1-isoquinolyl)methanes **1a,e,h** become intensely green.

The Ritter reaction of nitriles **5a,b** with the corresponding alcohols **3a—c**, **4** affords pure compounds **1a—g** in good yields, producing both symmetrical (**1a,e**) and non-symmetrical (**1b—d,f,g**) bis(3,3-dialkyl-3,4-dihydro-1-isoquinolyl)methanes (see Scheme 1, method ***B***). Compounds **1d,e** are obtained in low yield because their crystallization is difficult.

Compound **1a** was also synthesized in 80% yield by fusion (160—180 °C) of 3,3-dimethyl-1-methylthio-3,4-dihydroisoquinoline (**6a**) with 1,3,3-trimethyl-3,4-dihydroisoquinoline (**7a**) (see Scheme 1, method ***C***). This procedure can be recommended for the synthesis of compounds **1b,c,h**, which are poorly soluble in EtOH and, hence, can easily be separated from resinous by-products of the thermal reaction by recrystallization, and for the synthesis of bis(dihydroisoquinolyl)methanes **1d—f**, which are difficult to crystallize. In the latter case, the target products require chromatographic purification, which lowers the preparative value of the method.

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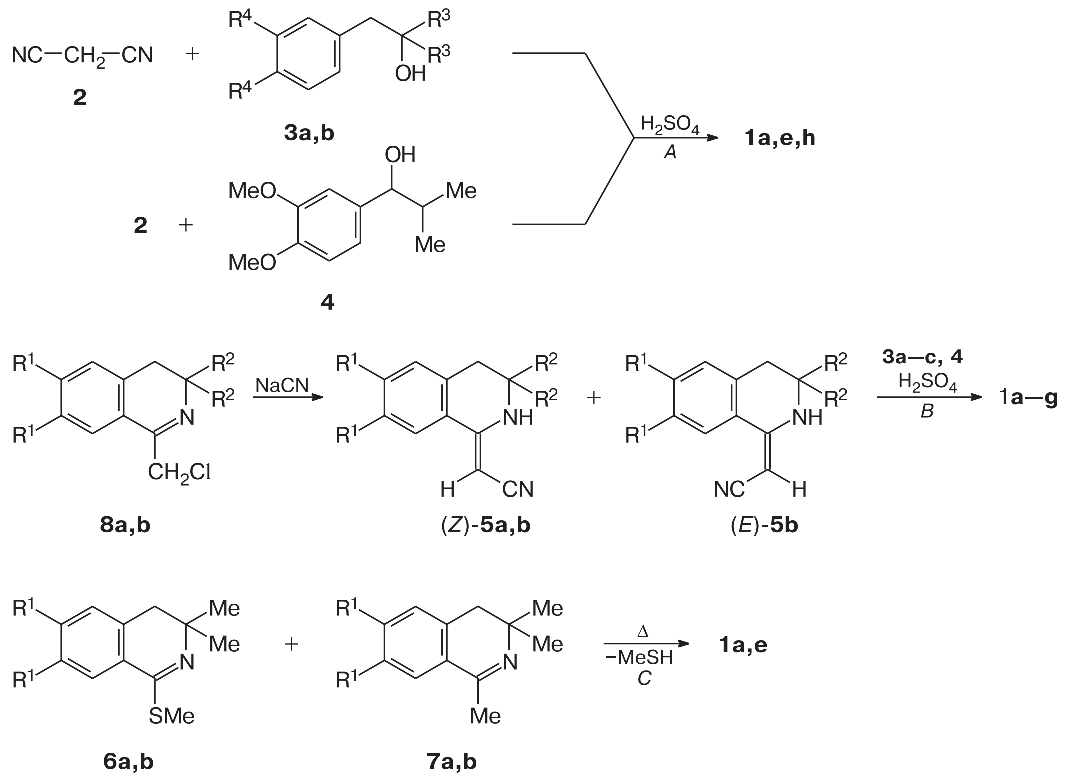
Scheme 1



3: R^3 = Me, R^4 = H (**a**); $(R^3)_2$ = $(CH_2)_4$, R^4 = H (**b**); $(R^3)_2$ = $(CH_2)_5$, R^4 = H (**c**)
5, 8: R^1 = H, R^2 = Me (**a**); R^1 = OMe, R^2 = Me (**b**); **6, 7:** R^1 = H (**a**); R^1 = OMe (**b**)

Table 1. Yields, melting points, and elemental analysis data for compounds **1b—h** and **5a,b**

Compound	Starting reactants	Yield (%) (method)	M.p./°C	Found/Calculated (%) C	H	N	Molecular formula
1b	**5a** + **3b**	88 (*B*)	133—134	84.19 84.23	7.87 7.92	7.99 7.86	$C_{25}H_{28}N_2$
1c	**5a** + **3c**	52 (*B*)	125—126	84.21 84.28	8.13 8.16	7.40 7.56	$C_{26}H_{30}N_2$
1d	**5b** + **3a**	14 (*B*)	120—122	76.91 76.89	7.81 7.74	7.25 7.17	$C_{25}H_{30}N_2O_2$
1e	**2** + **4** **5b** + **4** **6b** + **7b**	20 (*A*) 28 (*B*) 20 (*C*)	112—114	72.09 71.97	7.48 7.61	6.20 6.22	$C_{27}H_{34}N_2O_4$
1f	**5b** + **3b**	51 (*B*)	94—97	77.80 77.85	7.66 7.74	6.85 6.72	$C_{27}H_{32}N_2O_2$
1g	**5b** + **3c**	69 (*B*)	141—143	78.14 78.10	8.12 7.96	6.48 6.51	$C_{28}H_{34}N_2O_2$
1h	**2** + **3b**	56 (*A*)	159—161	84.90 84.77	7.94 7.90	7.37 7.32	$C_{27}H_{30}N_2$
5a	**8a**	70	110—111	78.81 78.75	7.20 7.12	14.17 14.13	$C_{13}H_{14}N_2$
5b	**8b**	68	125—126	69.77 69.74	7.13 7.02	10.85 10.84	$C_{15}H_{18}N_2O_2$

Nitriles **5a,b** were synthesized[5] from 1-chloromethyl-3,3-dimethyl-3,4-dihydroisoquinolines **8a,b** and sodium cyanide.

The yields and melting points of synthesized compounds **1b—h**, **5a,b** are presented in Table 1, and the parameters of their IR, mass, and ^{1}H NMR spectra are given in Table 2. The IR spectra of nitriles **5a,b** contain weak absorption bands of the C≡N groups at 2340 cm^{-1}. Absorption bands of the C=N groups in compounds **1a—h** lie in the interval of 1605—1620 cm^{-1}.

According to the ^{1}H NMR spectroscopic data, compound **5b** is formed as an inseparable mixture of the *Z*- and *E*-isomers in the 6 : 1 ratio (signals for both forms are presented in Table 2). The signals were assigned[6—8] using the data on enamines of the 3,4-dihydroisoquinoline series. The anisotropic influence of the C≡N group is the

Table 2. Spectral characteristics of synthesized compounds **1b—h** and **5a,b**

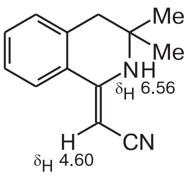
Compound	IR, ν/cm^{-1}	^{1}H NMR (DMSO-d$_6$, δ)						MS, m/z ($I_{\rm rel}$ (%))
		C(4)H$_2$ (s, 2 H)	R^2, R^3	NH (s, 1 H)	CH (s, 1 H)	H arom.	OMe (s, 3 H)	
1b	1615, 1585, 1570, 1250, 1305, 1245, 1170, 1065, 1040	2.75, 2.85	1.25 (s, 6 H, Me$_2$); 1.60—1.90 (m, 8 H, (CH$_2$)$_4$)	12.12	5.85	7.20 (m, 2 H); 7.31 (m, 4 H); 7.78 (m, 2 H, H(8), H(8´))	—	356 [M]$^+$ (42), 341 [M – Me]$^+$ (100)
1c	1610, 1580, 1570, 1305, 1245, 1175, 1135, 1065, 1045, 970	2.75 [a]	1.25 (s, 6 H, Me$_2$); 1.32—1.74 (m, 10 H, (CH$_2$)$_5$)	11.90	5.89	7.18 (m, 2 H); 7.30 (m, 4 H); 7.80 (m, 2 H, H(8), H(8´))	—	370 [M]$^+$ (58), 355 [M – Me]$^+$ (100), 327 (49)
1d	1605, 1585, 1520, 1305, 1270, 1240, 1210, 1160, 1140, 1090, 1060, 1010	2.65, 2.74	1.22 (s, 12 H, Me$_4$)	12.20	5.72	6.75 (s, 1 H, H(5)); 7.15 (m, 1 H); 7.30 (m, 3 H); 7.84 (m, 1 H, H(8´))	3.80, 3.87	—
1e [b]	1620, 1590, 1575, 1515, 1270, 1210, 1150, 1130 w, 1090, 1045, 1000, 860, 825	2.53 [a]	1.17 (s, 12 H, Me$_4$)	11.03	5.47	6.42 (s, 2 H, H(5), H(5´)); 7.02 (s, 2 H, H(8), H(8´))	3.75, 3.79	—
1f	1610 sh, 1595, 1570 w, 1515, 1270, 1235, 1140, 1085, 1035	2.68, 2.80	1.20 (s, 6 H, Me$_2$); 1.57—1.90 (m, 8 H, (CH$_2$)$_4$)	12.20	5.75	6.76 (s, 1 H, H(5´)); 7.15—7.37 (m, 4 H); 7.81 (s, 1 H, H(8´))	3.78, 3.85	—
1g	1610, 1580, 1560, 1510, 1270, 1235 w, 1140, 1090, 1065, 1040, 1010	2.65, 2.75	1.22 (s, 6 H, Me$_2$); 1.30—1.75 (m, 10 H, (CH$_2$)$_5$)	11.95	5.80	6.77 (s, 1 H, H(5)); 7.15—7.35 (m, 4 H, H(5´), H(6´), H(7´), H(8)); 7.82 (m, 1 H, H(8´))	3.80, 3.87	—
1h	1615, 1575, 1565, 1520, 1300, 1240, 1040, 1110, 955	2.83 [a]	1.55—1.83 (m, 16 H, 2 (CH$_2$)$_4$)	12.08	5.90	7.22 (m, 2 H); 7.32 (m, 4 H); 7.85 (m, 2 H, H(8), H(8´))	3.80, 3.87	382 [M]$^+$ (100), 353 [M – Et]$^+$ (22), 339 [M – Pr]$^+$ (7), 325 [M – Bu]$^+$ (11)
(*Z*)-5a	3310 (NH), 2340 (CN), 1610, 1570, 1300 w, 1240 br, 1145, 1320, 965	2.62	1.40 (s, 6 H, Me$_2$)	6.56	4.60	7.27 (m, 2 H); 7.42 (m, 1 H); 7.72 (m, 1 H, H(8))	—	—
(*Z*)-5b	3320 (NH), 2340 (CN), 1595, 1570, 1520, 1270, 1235,	2.73	1.25 (s, 6 H, Me$_2$)	6.14	4.47	6.72 (s, 1 H, H(5)); 7.95 (s, 1 H, H(8))	3.80 [c]	—
(*E*)-5b	1220, 1160, 1090, 875	2.70	1.25 (s, 6 H, Me$_2$)	7.29	4.05	6.76 (s, 1 H, H(5)); 7.12 (s, 1 H, H(8))	3.82 [c]	—

[a] 4 H.

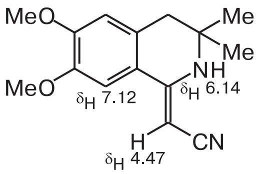
[b] The spectrum was recorded in CDCl$_3$.

[c] s, 6 H, (OMe)$_2$.

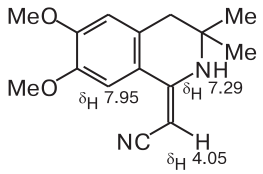
characteristic feature of the ^{1}H NMR spectrum of nitrile **5b**. This group shields the proton of the NH group in the *Z*-isomer (δ 6.14 instead of 7.29 for the *E*-isomer) and deshields the H(8) proton in the *E*-isomer (δ 7.95 instead of 7.12 for the *Z*-isomer). The chemical shifts of compound **5a** indicate that it exists as *Z*-isomer.



(*Z*)-**5a**



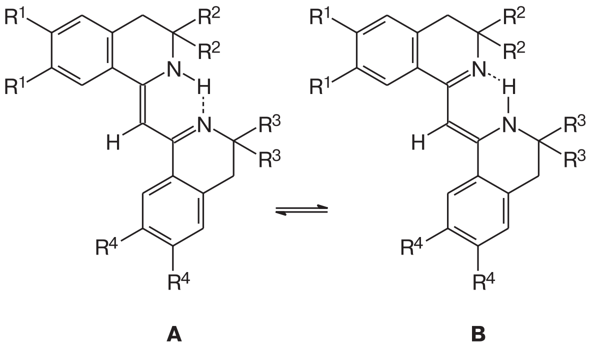
(*Z*)-**5b**



(*E*)-**5b**

The ^{1}H NMR spectra of **1a—h** (see Table 2) show that these compounds are tautomers with the exocyclic double bond in which one of the rings has the imine form, whereas another ring exists as enamine (distinct singlets of the NH (δ 11.03—12.20) and CH= groups (δ 5.72—5.90)). Meanwhile, the IR spectra recorded in both Nujol and CCl_4 solutions exhibit no absorption of the NH groups in the 2400—3600 cm^{-1} region. Probably, the bands of ν(NH) stretching vibrations coincide with the bands of stretching vibrations of the CH bonds (2840—3000 cm^{-1}) or are of very low intensities, indicating that the NH group forms a strong intramolecular hydrogen bond of the chelate type[9] (Scheme 2). Such an intramolecular hydrogen bond suggests the transfer of a proton from one isoquinoline ring to another accompanied by the tautomeric rearrangement of the molecule. However, these data alone for non-symmetrical diisoquinolylmethanes **1b—d,f,g** do not allow unambiguous assignments of the forms **A** or **B** to particular compounds.

Scheme 2



The mass spectra of compounds **1b,c,h** contain peaks of molecular ions corresponding to the calculated values (see Table 2). Compound **1d** gives no molecular ion peak.

Thus, our studies suggest the optimum methods for the synthesis of bis(3,4-dihydro-1-isoquinolyl)methanes **1**. These involve the reaction of malononitrile with alcohols **3a,b** (method ***A***) for symmetrical compounds **1a,h** and the Ritter reaction of alcohols **3a—c**, **4** with nitriles **5a,b** (method ***B***) for other compounds of the type **1**.

Experimental

IR spectra were recorded on a UR-20 instrument for suspensions in Nujol and solutions in CCl_4. Mass spectra were obtained on a Finnigan MAT (EI, 70 eV) instrument. ^{1}H NMR spectra were recorded on a Bruker WM-250 spectrometer at 25 °C using HMDS as the internal standard. The course of the reactions and purity of the products were monitored by TLC on Silufol plates ($CHCl_3$—acetone, 9 : 1). The substances were detected with a 3% solution of chloranil in toluene. Commercially available alcohol **3a** (Lancaster) was used as received.

1-Cyanomethylidene-6,7-dimethoxy-3,3-dimethyl-1,2,3,4-tetrahydroisoquinoline (5b). A mixture of chloride **8b** [10] (5.1 g, 20 mmol) and NaCN (1.1 g, 22 mmol) in MeOH (70 mL) was refluxed for 3 h and cooled. Water (50 mL) was added to the solution, and crystals that precipitated were separated and recrystallized from MeOH. Compound **5b** was obtained (3.5 g, 68%) as a mixture of *Z*- and *E*-isomers in a ratio of 6 : 1.

Nitrile 5a was obtained as *Z*-isomer similarly from chloride **8a**.[11]

Synthesis of compounds 1a—h

Method *A*. A solution of alcohol **3a** (15.0 g, 0.1 mol) in toluene (30 mL) was added dropwise to a solution of malononitrile **2** (3.3 g, 50 mmol) in 98% H_2SO_4 (30 mL) with intense stirring. The mixture was stirred for 1 h and poured in 300 mL of water. The organic layer was separated, and the aqueous layer was washed with toluene (2×20 mL) and neutralized with $(NH_4)_2CO_3$ to pH ≈ 8. The precipitate that formed was filtered off, washed with water, and recrystallized from EtOH. **1-(3,3-Dimethyl-3,4-dihydro-1-isoquinolyl)methylidene-3,3-dimethyl-1,2,3,4-tetrahydroisoquinoline (1a)** was obtained in 78% yield (12.1 g) with m.p. 175—177 °C (*cf.* Ref. 3: m.p. 175—177 °C).

Condensation of nitrile **2** (3.3 g, 50 mmol) with alcohol **4** [12] (20.8 g, 0.1 mol) in 30 mL of 98% H_2SO_4 was performed analogously. (Alcohol **4** was prepared from veratraldehyde and Pr^{i}MgBr in diethyl ether according to a described procedure[13] and used without additional purification after the ether was distilled off.) The reaction product was washed with water and dried. The yellow precipitate (14.9 g) was recrystallized twice from decane (with decantation from resin) to give 4.95 g (20%) of **1-(6,7-dimethoxy-3,3-dimethyl-3,4-dihydro-1-isoquinolyl)methylidene-6,7-dimethoxy-3,3-dimethyl-1,2,3,4-tetrahydroisoquinoline (1e).**

1-{Spiro[cyclopentane-1,3´-(3,4-dihydro-1-isoquinolyl)]}methylidenespiro[cyclopentane-1,3´-(1,2,3,4-tetrahydroisoquinoline)] (1h) (10.7 g, from EtOH) was obtained similarly from nitrile **2** (3.3 g, 50 mmol) in 98% H_2SO_4 (30 mL) and 16.6 g (0.1 mol) of 1-benzylcyclopentanol (**3b**)[14] (m.p. 47—50 °C, used without additional purification after the Grignard synthesis and distillation of diethyl ether).

Method *B*. A solution of nitrile **5a** (3.0 g, 15 mmol) and 2-benzylpropan-2-ol (**3a**) (2.3g, 15 mmol) in toluene (40 mL) was added dropwise to 10 mL of 98% H_2SO_4 with intense stirring and cooling with water. After 30 min, the reaction mixture was poured into water (80 mL), the organic layer was separated, and the aqueous layer was extracted with toluene (2×15 mL). The aqueous layer was neutralized with $(NH_4)_2CO_3$ to pH ≈ 8, and the yellow precipitate that formed was filtered off and washed with water. Recrystallization from EtOH gave compound **1a** (4.95 g, 65%) with m.p. 175—177 °C (*cf.* Ref. 3: m.p. 175—177 °C). Its ^{1}H NMR spectrum in $CDCl_3$ coincides with that of the authentic sample described previously.[3]

3,3-Dimethyl-1-{spiro[cyclopentane-1,3´-(3,4-dihydro-1-isoquinolyl)]}methylidene-1,2,3,4-tetrahydroisoquinoline (1b) (0.82 g) was obtained similarly from nitrile **5a** (0.52 g, 2.62 mmol) and alcohol **3b** (0.46 g, 2.62 mmol) in 98% H_2SO_4 (15 mL) after crystallization from 80 mL of EtOH.

3,3-Dimethyl-1-{spiro[cyclohexane-1,3´-(3,4-dihydro-1-isoquinolyl)]}methylidene-1,2,3,4-tetrahydroisoquinoline (0.87 g) was obtained similarly from nitrile **5a** (0.88 g, 4.5 mmol) and 0.85 g (4.5 mmol) of 1-benzylcyclohexanol (**3c**)[15] (m.p. 45—48 °C from hexane, used without additional purification after the Grignard synthesis and distillation of diethyl ether) in 98% H_2SO_4 (20 mL).

1-(3,3-Dimethyl-3,4-dihydro-1-isoquinolyl)methylidene-6,7-dimethoxy-3,3-dimethyl-1,2,3,4-tetrahydroisoquinoline (1d) (0.27 g) was obtained similarly from nitrile **5b** (1.29 g, 5 mmol) and alcohol **3a** (0.75 g, 5 mmol) after crystallization from EtOH in the cold (−10 °C).

Compound **1e** was synthesized similarly from nitrile **5b** (a mixture of isomers) and alcohol **4**.

6,7-Dimethoxy-3,3-dimethyl-1-{spiro[cyclopentane-1,3´-(3,4-dihydro-1-isoquinolyl)]}methylidene-1,2,3,4-tetrahydroisoquinoline (1f) and **6,7-dimethoxy-3,3-dimethyl-1-{spiro[cyclohexane-1,3´-(3,4-dihydro-1-isoquinolyl)]}methylidene-1,2,3,4-tetrahydroisoquinoline (1g)** were obtained similarly from nitrile **5b** (a mixture of isomers) and alcohols **3b,c**.

Method *C*. Sulfide **6a** [16] (2.08 g, 10 mmol) and 1,3,3-trimethyl-3,4-dihydroisoquinoline **7a** [17] (1.73 g, 10 mmol) were heated on a metallic bath for 3 h at 160—180 °C. The warm melt was poured into 30 mL of EtOH, and the precipitate that formed was filtered off and recrystallized. Compound **1a** was obtained in 60% yield (1.98 g) with m.p. 175—177 °C (*cf.* Ref. 3: m.p. 175—177 °C).

Sulfide **6b** [18] and 6,7-dimethoxy-1,3,3-trimethyl-3,4-dihydroisoquinoline **7b** [19] (10 mmol) were condensed similarly. The melt formed in the reaction was heated to boiling with decane (40 mL) and decanted from resin. Double repetition of this procedure gave 0.9 g of compound **1e**.

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