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The first regio- and diastereoselective synthesis of homochiral perhydroimidazoisoxazoles via the 1,3-dipolar cycloaddition of imidazoline 3-oxides with (1*S*)-(−)-β-pinene

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Abstract—The 1,3-dipolar cycloaddition of imidazoline 3-oxides **1** with (1*S*)-(−)-β-pinene proceeds regio- and diastereoselectively to give homochiral perhydroimidazoisoxazole derivatives **3** in high yields in the cases of imidazoline 3-oxides **1a–e** but in low yields in the reactions of **1f–g**. The preferred attack of (1*S*)-(−)-β-pinene to the cyclic nitrone was shown to be *anti–endo*. The reaction of racemic nitrones (±)-**1f–g** with the homochiral β-pinene gave the adduct from the (*S*)-nitrone and the corresponding imidazole. The adducts **3** undergo retro-1,3-dipolar cycloaddition when heated in the condensed phase or in diphenyl ether to give the corresponding imidazole and β-pinene.

1. Introduction

1,3-Dipolar cycloadditions have become increasingly important in recent years, particularly those involving isoxazolidine cycloadducts, which can be elaborated to cyclic or acyclic compounds with complete control of the relative stereochemistry.[1] The use of enantiomerically pure cyclic nitrones in the synthesis of alkaloids,[2] and γ-aminoalcohols[3] is described in the literature. Recently we have reported the regio- and diastereoselective 1,3-dipolar cycloaddition of chiral nitrones **1** with achiral dipolarophiles such as aryl isocyanates,[4] styrene,[5] and DMAD.[6] Although the synthesis and use of chiral auxiliaries prepared from (−)-β-pinene are known,[7,8] no examples of acyclic and cyclic nitrone adducts with (*S*)-β-pinene have been reported.

Herein, we report on the regio- and diastereoselective 1,3-dipolar cycloaddition of achiral **1a–e** and racemic **1f** and **1g** imidazoline 3-oxides with (*S*)-(−)-β-pinene to give optically active spiro-compounds **3**. The adducts from these reactions were heated in the condensed phase under vacuum or in diphenyl ether at 200°C to give β-pinene and the corresponding imidazoles (not, unfortunately the 3-oxides).[9] It was hoped that the retro-dipolar cycloaddition of **3f** and **3g** would serve as a method for the preparation of homochiral 3-imidazoline 3-oxides but the reaction was not analogous with the cases of isocyanate[4] and styrene[5] adducts where regeneration of the nitrone is a highly efficient process.

2. Results and discussion

Cyclic nitrones **1** were heated for 72 h in xylene in the presence of a nine-fold excess of (*S*)-(−)-β-pinene. Homochiral adducts **3a–e** were isolated in high yields by recrystallization from ethanol or by column chromatography in some cases. The reaction of imidazoline 3-oxides **1f–g** with pinene in refluxing xylene led mainly to the formation of corresponding imidazole, therefore the reaction was performed in refluxing toluene for 18 days. The yields of **3f** and **3g** are drastically lower than in the cases of **1a–e** (see Table 1). Chromatographic assays during the reaction showed the presence of a single adduct with the unreacted nitrone and the corresponding imidazole. In one case we terminated the reaction after heating for 68 h and isolated the corresponding adduct and imidazoline 3-oxide expecting that the latter would be enriched by the enantiomer reacting slower with (*S*)-(−)-β-pinene. Unfortunately, the measurements showed the product to be optically inactive.

The structure of compounds **3** was determined on the basis of ^{1}H and ^{13}C and 2D NMR studies. Elemental analysis for all adducts and MS data for **3g** are in agreement with the proposed structure. Adducts **3a–g** have the same ^{1}H and ^{13}C NMR spectra related to their

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Table 1. Perhydroimidazo[1,5-*b*]isoxazoles **3**

	R	R^1	R^2	Yield (%)	Mp (°C)	Yield of recovered **1**	$[\alpha]_D^{20}$
a	4-MeC_6H_4	H	Ph	85	121.9–122.2	12	+212 ($CHCl_3$, 0.50)
b	4-$MeOC_6H_4$	H	Ph	80	138–139	15	+92 ($CHCl_3$, 0.50)
c	4-MeC_6H_4	H	3-$MeOC_6H_4$	60	168–170	24[b]	+137.5 ($CHCl_3$, 0.40)
d	4-$MeOC_6H_4$	H	3-$MeOC_6H_4$	47	115.5–117	41[b]	+93.75 ($CHCl_3$, 0.40)
e	4-$MeOC_6H_4$	H	4-$MeOC_6H_4$	55	140–140.7	20+20[b]	+100 ($CHCl_3$, 0.40)
f	4-MeC_6H_4	Ph	Ph	25[a]	194–195	70[b]	+80 ($CHCl_3$, 0.50)
g	4-$MeOC_6H_4$	Ph	Ph	24[a]	155.5–158	18+50[b]	+97.7 ($CHCl_3$, 0.44)

[a] The reaction was performed under reflux in toluene for 18 days.
[b] The yield of corresponding imidazole.

isoxazolidine and pinene components. In the ^1H NMR spectra of the adducts, the AB system near 2.45 ppm indicates the 2-spiro regioisomer shown in Scheme 1. The NOESY spectrum of **3g** shows correlations[10] (see Fig. 1) for C(6)H/C(b-3)H, C(a-4)H/C(3a)Ph, C(a-4)H/C(5)Ph, C(6)H/C(5)Ph which indicates that C(3a) and C(6) phenyls are *cis*-oriented.[6] The one proton triplet ($J=4.97$ Hz) at δ 2.11 ppm, was assigned to C(1′)H. Its NOESY correlations with C(6), C(b-3)H, C(b-4)H, C(7′) and C(8′) revealed that the preferred orientation of (*S*)-β-pinene in the 1,3-dipolar cycloaddition with cyclic nitrones **1** is *anti–endo*. The alternative *anti–exo* and *syn–exo* attacks (see Fig. 2) should give the adducts **3′** and **3″** having the bridgehead C(1′)H correlating with the aromatic *ortho* protons of the C(3a) phenyl group. The product of *syn–endo* attack **3″** would have the C(1′) proton correlating with C(6)H but the methyl groups are too far from C(3)H to give cross peaks.

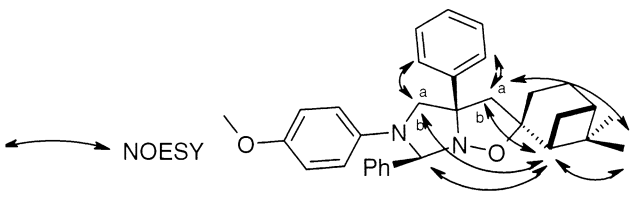


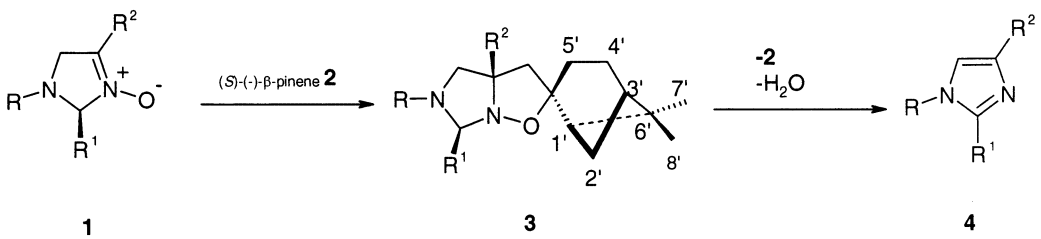
Figure 1. The stereochemistry of adduct **3g** based on selected NOESY correlations.

Molecular modelling studies revealed that the *syn–endo* adduct of the (*S*)-β-pinene should be more stable (the energy calculated for *syn–endo* adduct is 163.464 kJ/mol) than *anti–exo* adduct **3′**, E_{total} 168.748 kJ/mol. The s*yn–endo* adduct is seen to be more stable by 1 kJ/mol than adduct **3** but, in spite of this the (*R*)-3-oxide leading to **3″** seems to dehydrate more easily than the corresponding (*S*)-3-oxide. The *anti-endo* adduct is more stable than the *anti–exo* and *syn–exo* adducts by about 4 and 1 kJ/mol, respectively.

To prove that imidazoles in the reaction mixture are not products from the fragmentation of adducts **3** we refluxed adduct **3f** in xylene for a week but no conversion to imidazole was observed. Adducts **3f** and **3g** were heated in condensed phase at 200°C under vacuum for 1 h to give (*S*)-(−)-β-pinene and the corresponding imidazole. The same results were obtained when the adducts were heated in diphenyl ether. The products were determined by gas chromatography to be again β-pinene and the corresponding imidazole.

3. Experimental

Melting points were taken on an Electrothermal Digital melting point apparatus. Infrared spectra were recorded on a Mattson 1000 FTIR. Proton and ^{13}C magnetic resonance spectra were recorded on a Bruker Dpx 400 MHz spectrometer. All spectra were taken in deuteriochloroform. Mass spectrum of **3g** was recorded at 70 eV by electron impact on a Fisons VG Platform II instrument. The optical rotation of the adducts were measured on a WXG-4 disk polarimeter. Freshly prepared imidazoline 3-oxides were used after recrystallization from either ethanol or acetone. *1,4-Di-p-methoxyphenyl-Δ^3-imidazoline N-oxide is a new compound and was obtained according to the procedure reported by us.*[11] Yield 48%; mp 216.5–218°C; IR $\nu_{C=N}$ 1591 cm^{-1}; ^1H NMR ($CDCl_3$): δ ppm 3.71 (3H, s), 3.80 (3H, s), 4.65 (2H, t, $J=3.63$), 5.27 (2H, t, $J=3.63$), 6.50 (2H, d, $J=8.60$), 6.85 (2H, d, $J=8.60$), 6.94 (2H, d, $J=8.60$), 8.25 (2H, d, $J=8.60$). Anal. calcd for $C_{17}H_{20}N_2O_3$: C, 67.98; H, 6.71; N, 9.33. Found: C, 67.65; H, 6.19; N, 8.83%.



Scheme 1.

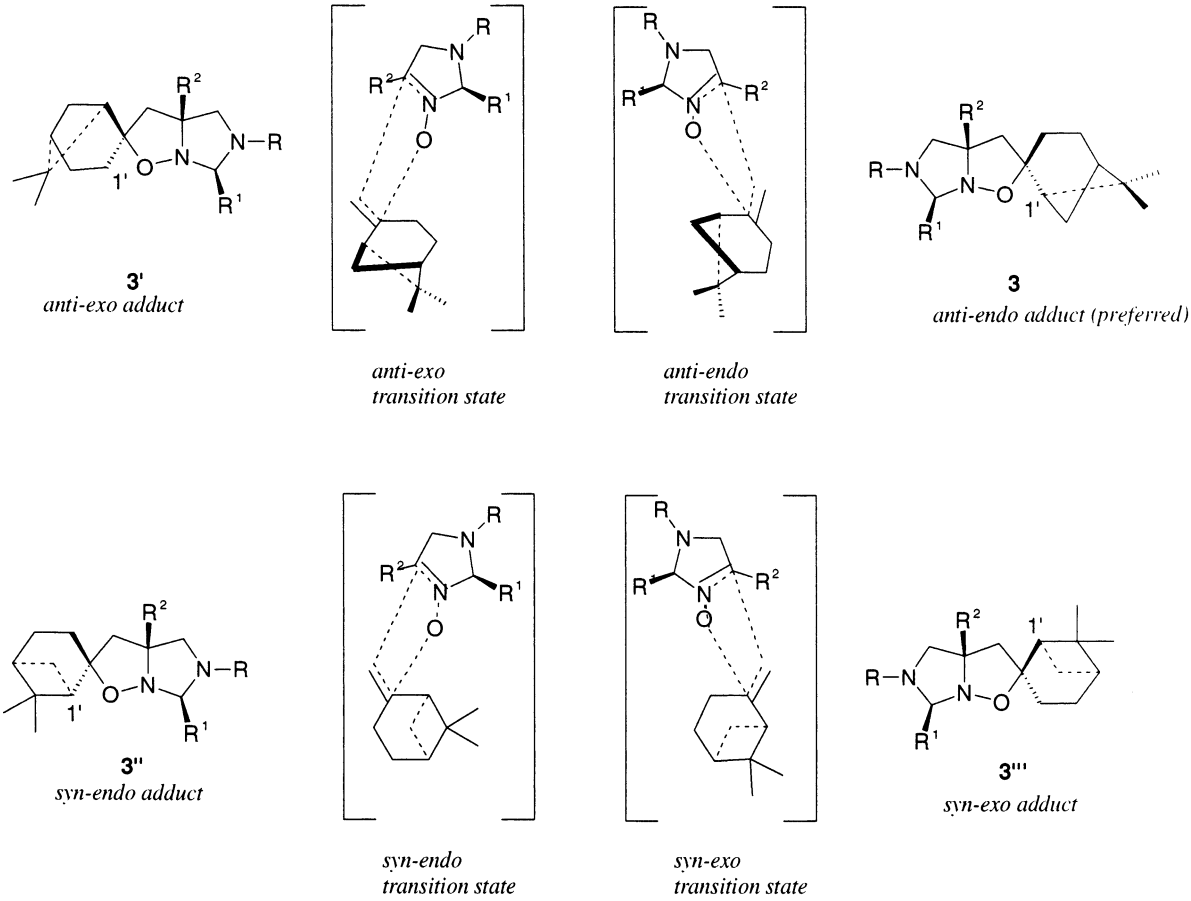


Figure 2. The possible four diastereomers of C(2) spiroisoxazolidines **3a–e** and four of the eight possible for **3f** and **3g** and the transition states leading to their formation.

3.1. Cycloaddition of imidazoline 3-oxides with (1*S*)-(−)-β-pinene. General procedure for the preparation of 3a–g

To a solution of imidazoline 3-oxide 1 (1 mmol) in xylene (*S*)-(−)-β-pinene (9 mmol) was added and the mixture was stirred under reflux for 72 h. The solvent and excess pinene were removed under vacuum and the residue triturated with ethanol. The solid formed was purified by recrystallization from ethanol.

3.2. Perhydroimidazoisoxazole 3a

Yield 82%; mp 121.9–122.2°C; $[\alpha]_D^{20}=+212$ ($c=0.50$, $CHCl_3$); ^1H NMR ($CDCl_3$): δ ppm 0.71 (3H, s), 1.10 (3H, s), 1.37 (1H, d, $J=10.25$), 1.61 (1H, m), 1.72 (1H, t, $J=2.84$), 1.76 (2H, m), 1.92 (1H, t, $J=3.52$), 2.05 (1H, m), 2.09 (3H, s), 2.27 (1H, m), 2.45 (2H, s), 3.52 (2H, s), 4.21 (1H, d, $J=9.90$), 4.44 (1H, d, $J=9.90$), 6.25 (2H, d, $J=8.44$), 6.84 (2H, d, $J=8.44$), 7.08 (1H, t, $J=8.00$), 7.18 (2H, t, $J=8.00$), 7.37 (2H, d, $J=8.00$). ^{13}C NMR ($CDCl_3$): δ ppm 20.77; 23.48; 24.86; 27.46; 27.50; 33.58; 39.04; 40.55; 53.44; 58.22; 58.46; 72.54; 78.58; 87.40; 112.93; 126.11; 126.26; 127.54; 128.96; 130.06; 143.66; 144.71. Anal. calcd for $C_{26}H_{32}N_2O$: C, 80.37; H, 8.30; N, 7.21. Found: C, 80.25; H, 8.25; N, 7.21%.

3.3. Perhydroimidazoisoxazole 3b

Yield 80%; mp 138–139°C; $[\alpha]_D^{20}=+92$ ($c=0.5$, $CHCl_3$); ^1H NMR ($CDCl_3$): δ ppm 0.78 (3H, s), 1.17 (3H, s), 1.46 (1H, d, $J=10.22$), 1.72 (1H, m), 1.79 (3H, m), 1.98 (1H, t, $J=5.42$), 2.11 (1H, m), 2.28 (1H, m), 2.51 (2H, $J_{AB}=12.36$), 3.54 (2H, $J_{AB}=8.52$), 3.65 (3H, s), 4.37 (2H, $J_{AB}=9.80$), 6.38 (2H, d, $J=8.90$), 7.12 (2H, d, $J=8.90$), 7.14 (1H, m), 7.24 (2H, t, $J=7.44$), 7.43 (2H, d, $J=7.45$). ^{13}C NMR ($CDCl_3$): δ ppm 23.48; 24.84; 27.46; 33.31; 39.02; 40.56; 53.35; 56.08; 58.46; 59.06; 73.31; 78.71; 87.45; 96.57; 114.03; 115.33; 126.08; 127.50; 128.96; 141.57; 143.92; 152.31. Anal. calcd for $C_{26}H_{32}N_2O_2$: C, 77.19; H, 7.97; N, 6.92. Found: C, 77.15; H, 8.10; N, 6.80%.

3.4. Perhydroimidazoisoxazole 3c

Yield 60%; mp 168–170°C; $[\alpha]_D^{20}=+137.5$ ($c=0.4$, $CHCl_3$); ^1H NMR ($CDCl_3$): δ ppm 0.84 (3H, s), 1.23 (3H, s), 1.48 (1H, d, $J=10.25$), 1.68–1.75 (1H, m), 1.81–1.90 (3H, m), 2.03 (1H, t, $J=5.43$), 2.14–2.19 (1H, m), 2.21 (3H, s), 2.31–2.43 (1H, m), 2.56 (2H, s), 3.61 (2H, $J_{AB}=8.61$), 3.79 (3H, s), 4.33 (1H, d, $J=9.86$), 4.56 (1H, d, $J=9.87$), 6.36 (2H, d, $J=8.41$), 6.72 (1H, dd, $J=10.26$, 2.14), 6.94 (2H, d, $J=8.26$), 6.99 (1H, d, $J=8.44$), 7.07 (1H, m), 7.18–7.24 (1H, m). ^{13}C NMR ($CDCl_3$): δ ppm 20.25; 22.95; 24.31; 26.93; 32.86; 38.46;

40.00; 52.82; 54.83; 57.98; 72.13; 77.96; 86.77; 111.51; 112.08; 112.48; 117.78; 125.71; 129.47; 129.36; 144.16; 145.02; 159.68. Anal. calcd for $C_{27}H_{34}N_2O_2$: C, 77.48; H, 8.19; N, 6.69. Found: C, 77.44; H, 8.23; N, 6.66%.

3.5. 4-*m*-Methoxyphenyl-1-*p*-tolyl-imidazole 4c

The compound was isolated from the mother liquor of compound **3c** as an oil. Yield 24%. IR (neat) $\nu_{C=N}$ 1625 cm^{-1}. ^1H NMR ($CDCl_3$): δ ppm 2.40 (3H, s), 3.86 (3H, s), 6.46 (1H, d, $J=8.35$), 7.29 (6H, m), 7.39 (1H, s), 7.49 (1H, s), 7.81 (1H, s). ^{13}C NMR ($CDCl_3$): δ ppm 21.43; 55.43; 110.25; 113.79; 117.69; 121.68; 129.85; 130.75; 137.64; 160.38.

3.6. Perhydroimidazoisoxazole 3d

The compound was purified by column chromatography using silica gel as adsorbent and ethyl acetate–petroleum ether (1:10) as eluent mixture. Yield 47%; mp 115.5–117°C; $[\alpha]_D^{20}=+93.75$ ($c=0.4$, $CHCl_3$); ^1H NMR ($CDCl_3$): δ ppm 0.84 (3H, s), 1.23 (3H, s), 1.50 (1H, d, $J=10.22$), 1.71–1.90 (4H, m), 2.04 (1H, t, $J=5.45$), 2.14 (1H, s), 2.19–2.17 (1H, m), 2.34–2.42 (1H, m), 2.52–2.66 (1H, m), 3.5 (1H d, $J=8.52$), 3.63 (1H d, $J=8.55$), 3.69 (3H, s), 3.80 (3H, s), 4.3 (1H d, $J=9.8$), 4.55 (1H, d, $J=9.77$), 6.43 (2H d, $J=8.88$), 6.71–6.77 (3H, m,), 6.94–7.03 (1H, m), 7.08–7.09 (1H, m), 7.17–7.25 (1H, m). ^{13}C NMR ($CDCl_3$): δ ppm 23.5; 24.84; 27.49; 31.12; 33.14; 38.99; 40.51; 53.26; 55.38; 55.97; 58.47; 59.46; 73.49; 78.67; 87.36; 111.94; 112.61; 114.21; 115.31; 118.31; 129.91; 141.48; 145.79; 152.44; 160.23. Anal. calcd for $C_{27}H_{34}N_2O_3$: C, 74.62; H, 7.89; N, 6.45. Found: C, 75.02; H, 7.78; N, 6.36%.

3.7. 1-*p*-Methoxyphenyl-4-*m*-methoxyphenylimidazole 4d

The product was isolated from the mother liquor of **3d** by flash column chromatography. Yields 41%. Recrystallized from ethanol; mp 82-82.5°C; IR (KBr) $\nu_{C=N}$ 1625 cm^{-1}; ^1H NMR ($CDCl_3$): δ ppm 3.82 (3H, s), 3.85 (3H, s), 6.78 (1H, d, $J=9.33$), 6.95 (2H, d, $J=8.75$), 7.24–7.35 (4H, m), 7.41 (1H, s), 7.44 (1H, s), 7.74 (1H, s). ^{13}C NMR ($CDCl_3$): δ ppm 55.54; 55.81; 110.27; 113.63; 114.83; 115.30; 117.72; 123.40; 129.93; 130.98; 135.68; 136.14; 143.24; 159.38; 160.40. Anal. calcd for $C_{17}H_{16}N_2O_2$: C, 72.84; H, 5.75; N, 9.99. Found: C, 71.93; H, 6.00; N, 9.85%.

3.8. Perhydroimidazoisoxazole 3e

Yield 55%; mp 140–140.7°C; $[\alpha]_D^{20}=+100$ ($c=0.4$, $CHCl_3$); ^1H NMR ($CDCl_3$): δ ppm 0.85 (3H, s), 1.23 (3H, s), 1.50 (1H, d, $J=10.24$), 1.69–1.76 (1H, m), 1.83–1.91 (2H, m), 2.03 (1H, t, $J=5.44$), 2.14 (3H, s), 2.36–2.43 (1H, m), 2.54 (2H, $J_{AB}=12.54$, 12.52), 3.57 (2H, $J_{AB}=8.57$, 8.52), 3.71 (3H, s), 3.77 (3H, s), 4.26 (1H, d, $J=9.85$), 4.51 (1H, d, $J=9.85$), 6.42 (2H, d, $J=8.81$), 6.74 (2H, d, $J=8.93$), 6.81 (2H, d, $J=8.77$), 7.34 (2H, d, 8.76). ^{13}C NMR ($CDCl_3$): δ ppm 23.50; 24.88; 27.50; 31.12; 33.48; 39.01; 40.55; 53.36; 55.40; 55.97; 58.47; 58.78; 73.10; 78.32; 87.24; 113.96; 114.29; 115.30; 127.22; 135.5; 159.07. Anal. calcd for $C_{27}H_{34}N_2O_3$: C, 74.62; H, 7.89; N, 6.45. Found: C, 74.50; H, 7.63; N, 6.45%.

3.9. 1,4-Di-*p*-methoxyphenylimidazole 4e

Obtained as the second product from the reaction mixture of **3e**. Yield 20%. Recrystallized from ethanol; mp 158°C; IR (KBr) $\nu_{C=N}$ 1625 cm^{-1}; ^1H NMR ($CDCl_3$): δ ppm 3.80 (3H, s), 3.84 (3H, s), 6.88 (2H, d, $J=8.69$), 6.95 (2H, d, $J=8.83$), 7.30 (1H, s), 7.33 (2H, s), 7.68 (1H, s), 7.70 (2H, s). ^{13}C NMR ($CDCl_3$): δ ppm 55.41, 55.75; 113.34; 114.37; 115.24; 123.35; 126.52; 127.17; 131.24; 135.87; 143.43; 159.20; 159.25.

3.10. Perhydroimidazoisoxazole 3f

The reaction mixture was stirred under reflux in toluene for 18 days. The product was purified by column chromatography using silica gel as adsorbent and ethyl acetate–petroleum ether as eluent. Yield 25%; mp 194–195°C; $[\alpha]_D^{20}=+80$ ($c=0.5$, $CHCl_3$);); ^1H NMR ($CDCl_3$): δ ppm 0.75 (3H, s), 1.17 (3H, s), 1.74 (1H, d, $J=10.14$), 1.64 (1H, m), 1.82 (3H, m), 2.00 (1H, t, $J=4.70$), 2.14 (3H, s), 2.45 (2H, $J_{AB}=12.51$), 3.95 (2H, $J_{AB}=8.68$), 5.50 (1H, s), 6.29 (2H, d, $J=8.20$), 6.91 (2H, d, $J=8.20$), 6.98 (6H, m), 7.08 (2H, m), 7.21 (2H, d, $J=5.76$). ^{13}C NMR ($CDCl_3$): δ ppm 22.73; 25.42; 26.86; 29.47; 36.11; 41.07; 42.51; 56.10; 60.56; 61.16; 87.86; 89.85; 98.58; 115.57; 128.45; 129.08; 129.62; 129.96; 130.07; 130.21; 131.92; 141.60; 144.95; 146.96. Anal. calcd for $C_{32}H_{36}N_2O$: C, 82.72; H, 7.81; N, 6.03. Found: C, 82.34; H, 7.56; N, 5.86%.

3.11. Perhydroimidazoisoxazole 3g

Purification as for **3f** 24%; mp 155.5–158°C; $[\alpha]_D^{20}=+97.7$ ($c=0.44$, $CHCl_3$); ^1H NMR ($CDCl_3$): δ ppm 0.81 (3H, s), 1.23 (3H, s), 1.56 (1H, d, $J=10.25$), 1.71 (1H, m), 1.85 (3H, m), 2.11 (1H, t, $J=4.97$), 2.20 (1H, m), 2.37 (1H, m) 2.44 (2H, $J_{AB}=12.55$), 3.69 (3H, s), 4.03 (2H, $J_{AB}=8.57$), 5.54 (1H, s), 6.39 (2H, d, $J=8.97$), 6.73 (2H, d, $J=8.97$), 6.72–7.00 (6H, m), 7.20 (2H, m), 7.33 (2H, m). Anal. calcd for $C_{32}H_{36}N_2O_2$: C, 79.96; H, 7.55; N, 5.83. Found: C, 79.84; H, 7.51; N, 5.85 m/z: 480.3 ($M^{+\bullet}$, 10.83), 327.2 (4.66), 262.1 (4.1), 225.1 (100), 211.1 (12.33), 193.1 (7.94), 165.1 (2.36), 134.1 (23.76), 121.1 (10.95), 104.1 (8.56), 77.0 (5.49), 69.1 (1.99), 41.0 (2.63).

3.12. Retro-cycloaddition of perhydroimidazoisoxazoles in condensed phase: general procedure for preparation of 3f and 3g

Compound **3** (0.1 mmol) was placed in a glass sample vial and heated in a vacuum oven at 200°C for 1 h at 1.3×10^{-3} mmHg and left to cool at room temperature. The obtained **4** was extracted with warm hexane (3×2 mL). The extract was concentrated and cooled. The formed crystals were collected by filtration. The identity of the obtained imidazoles was determined comparing their melting points as well as their IR spectra with those of the authentic samples.

3.13. Retro-cycloaddition of perhydroimidazoisoxazoles 3f and 3g in solution

Compound **3** (0.12 mmol.) was dissolved in diphenyl ether (3 mL) and heated for 1 h at 200°C. The yield of β-pinene was 65 and 60%, respectively, for **3f** and **3g** as determined by GC-FID.

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9. The imidazole forms from the thermal dehydration of the corresponding imidazoline 3-oxide.
10. The assignment for some of the protons of **3g** is as follows; 0.81 (3H, s, C-7′), 1.23 (3H, s, C-8′), 1.56 (1H, d, $J=10.25$), 1.71 (1H, m), 1.85 (3H, m), 2.11 (1H, t, $J=4.97$, C-1′), 2.20 (1H, m), 2.37 (1H, m) 2.44 (2H, $J_{AB}=12.55$, C-3), 3.69 (3H, s, OMe), 4.03 (2H, $J_{AB}=8.57$, C-4), 5.54 (1H, s, C-6), 6.39 (2H, d, $J=8.97$, *N*-Ph *ortho*), 6.73 (2H, d, $J=8.97$, *N*-phenyl *meta* H), 7.00 (6H, m, C-3a and C-6 phenyl *meta* and *para* H), 7.20 (2H, m, C-6 phenyl *ortho* H), 7.33 (2H, m C-3a phenyl *ortho* H)
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