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Diastereoselective Hetero-Diels-Alder Cycloaddition of a C-Nitroso Compound Prepared Starting from a Homochiral Imidazolidin-2-one

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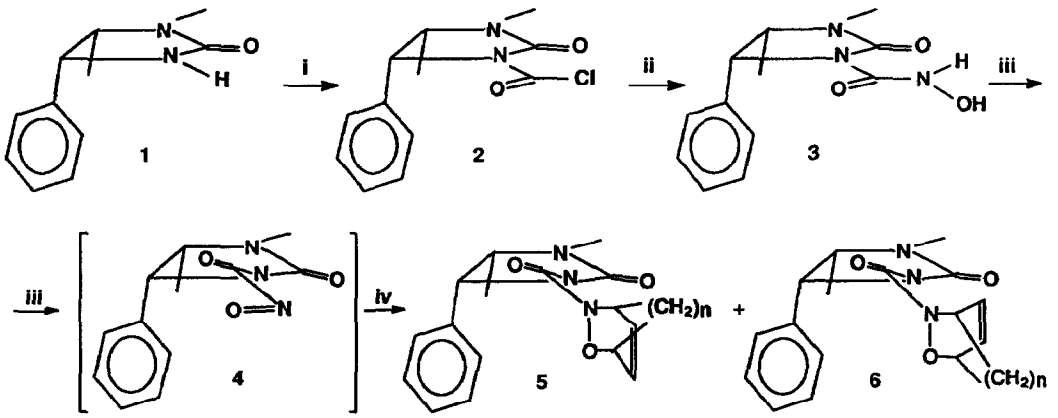
Abstract: By Swern oxidation of the hydroxamic acid **3**, prepared from the homochiral imidazolidin-2-one **1**, the transient C-nitroso derivative **4** is obtained, and its cycloaddition to either cyclohexadiene or cyclopentadiene proceeds with high diastereoselection, owing to the conformational stability of **4**. The stereochemical outcome of the reaction is determined from ^{1}H NMR data and further confirmed by the specific rotation value of **7**, obtained by cleavage of the major cycloadduct **5a**.

An appealing entry to 4-aminoalcohols in high enantiomeric excess involves the diastereoselective [4+2] hetero-Diels-Alder cycloaddition of 1,3-dienes with a nitroso group bonded to a chiral auxiliary. [1] In fact removal of the chiral auxiliary and reductive cleavage of the nitrogen-oxygen bond allow a convenient route to both aminoconduritols and aminoinositols, polyfunctionalised compounds with biological activity. [2]

In previous papers we have shown that imidazolidin-2-one **1** and its enantiomer, readily available from either (-)- or (+)-ephedrine, are versatile chiral auxiliaries in alkylation, [3] cyclofunctionalisation [4] and cycloaddition [5] reactions. C-nitroso compounds can be readily generated as transient dienophiles by oxidation of the corresponding hydroxamic acids and are trapped in situ by 1,3-dienes. We describe herein the results obtained by using the acyl nitroso derivative **4**, prepared starting from **1**, as a dienophile in hetero-Diels-Alder cycloadditions. The lithium anion of the imidazolidin-2-one **1** is treated at -78 °C with phosgene to give the corresponding chlorocarbonyl derivative **2** that can be stored indefinitely at 0 °C in dry medium. The hydroxamic acid **3** is then efficiently synthesized in good yield starting from **2** and hydroxylamine. [6,7]

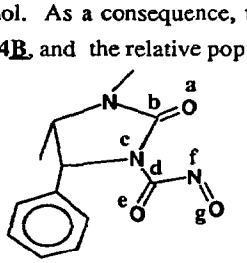
Initial efforts to obtain the C-nitroso compound **4** by oxidation performed with tetrabutylammonium periodate at -60 °C or -20 °C were unsuccessful. However, by using the Swern oxidising reagent [8] at -60 °C, the hydroxamic acid **3** is converted into the nitroso derivative **4** which, in the presence of either cyclohexadiene or cyclopentadiene, undergoes cycloaddition to give the cycloadducts **5** and **6**, respectively, in good yield and high diastereoselection, as reported in Scheme 1. The relative amounts of the diastereomers are easily determined by ^{1}H and ^{13}C NMR spectroscopy, since in the spectra of the crude reaction mixture two sets of a large number of peaks (pair of diastereomers) can be compared, for which the chemical environment is closely similar. In addition, by silica gel chromatography, the diastereomers are obtained pure as white solids.



a. n = 2, 73% yield, d.r. 93:7 b. n = 1, 64% yield, d.r. 87:13

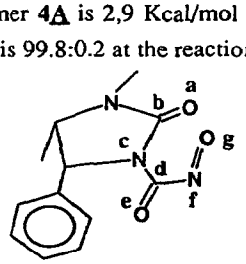
Scheme 1. *Reagents and conditions:* **i.** n-BuLi, -78 °C, then inverse addition to phosgene **ii.** NH_2OH, THF, r.t. **iii.** DMSO, $(COCl)_2$, CH_2Cl_2, -60 °C **iv.** Et_3N, cyclohexadiene or cyclopentadiene

In order to account for the observed asymmetric induction, the preferential conformations of compounds **2** and **3** were first examined. From conformational analysis performed by using MM+ force field,[9] it is possible to conclude that the carbonyl groups are *anti* each other in **2** and the dihedral angle between them is 172°. [10] On the other hand, the preferred conformation for the the hydroxamic acid **3** shows a planar disposition owing to a H-bond between N-H group and the oxygen of the carbonyl group at C-2. These conformations are in agreement with the observed deshielding of H-5 with respect to the observed value for H-4 in compound **1** (5.28 versus 4.78 δ), due to an almost co-planar disposition of the carbonyl group with the heterocyclic ring. An optimised structure for the C-nitroso derivative intermediate **4** using the MM+ force field by rotations about the bonds (N-1)-(C-1') and (C-1')-(N-2'), was then calculated. First, from these calculations the *anti* arrangement of the carbonyl groups results preferred over the *syn* one. The subsequent rotation about the bond (C-1')-(N-2') suggests that in the lowest energy conformation the two carbonyl groups and the nitroso group lie in the *anti,syn* arrangement, respectively (conformer **4A**). This is due to very high energy non-bonded interactions, leading to highly restricted rotation at the N-CO-N=O. In fact, the calculated value for the minor conformer **4B** is 13.3 Kcal/mol, whereas for the major one, **4A**, it is 8.5 Kcal/mol. As a consequence, the *anti,syn* conformer **4A** is 2,9 Kcal/mol more stable than the *anti,anti* conformer **4B**, and the relative population ratio **4A**:**4B** is 99.8:0.2 at the reaction temperature.



4A

abcd = - 3.29°
bcde = +174.6°
edfg = - 5.38°

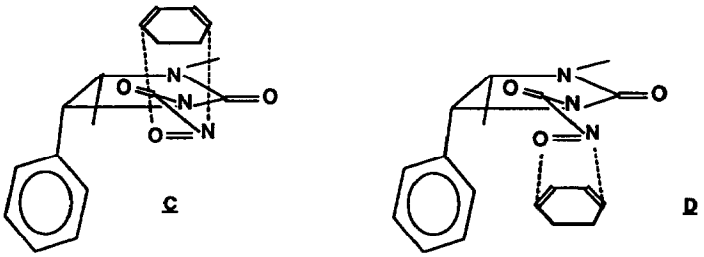


4B

abcd = - 6.38°
bcde = +167.99°
edfg = +132.98°

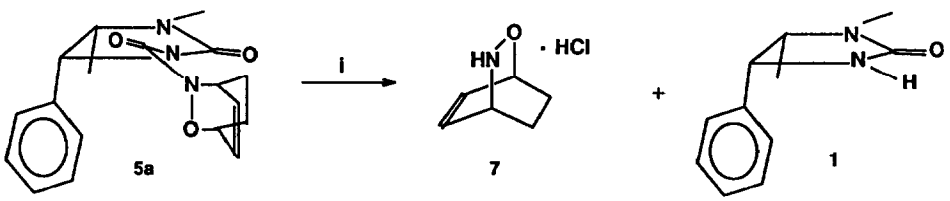
Thus, the geometry of the more stable conformer **4A** allows the high diastereoselection observed to be explained. In fact, since the diene approaches in an *endo* mode, the cycloaddition occurs mainly from the less

hindered diastereoface of the conformer 4A. On comparison of the transition states C and D, it results that the attack from the phenyl side in D is disfavoured, owing to steric interactions between the phenyl group and the approaching diene.



Besides mechanistic considerations, the configuration of the cycloadducts can be easily assigned on the basis of ^1H NMR data, which are confirmed by molecular modeling calculations performed with MM+. In fact, the more stable conformation for both the diastereomers **5a,b** and **6a,b** has been calculated and is reported in the Scheme 2. In the preferred conformation of the major diastereomers **5a,b**, the double bond lies on the opposite side of the carbonyl group of the imidazolidin-2-one moiety. The olefinic protons of **5a,b** resonate upfield, with respect to the minor diastereomer **6a,b**, in good agreement with the calculated more stable disposition, whereas the inverted trend is observed for the methylenic protons.

On the other hand, the structural assignment of **5a** was eventually confirmed by removal of the chiral auxiliary leading to the corresponding (1R,4S) 2-oxa-3-azabicyclo[2.2.2.]oct-5-ene hydrochloride **7**, whose configuration was determined on comparison with the specific rotation value reported in the literature. [13]



Scheme 2. *Reagents and conditions:* i. LiOH, H_2O_2, r.t., then HCl

These results confirm that by using the imidazolidin-2-one **1** as a chiral auxiliary in hetero-Diels-Alder cycloadditions, good levels of facial stereoselection can be obtained and the configuration of the products can be assigned on the basis of mechanistic considerations and ^1H NMR data. Applications of these findings to the preparation of conduramine A-1 will be reported in due course.

Experimental

General M.p.s were determined (uncorrected) on Electrothermal 910 melting point apparatus. IR spectra were measured on a Nicolet XS 20 FT-IR. ^1H and ^{13}C NMR spectra were recorded in $CDCl_3$, unless otherwise reported, on a Varian Gemini 200 spectrometer. Optical rotations were recorded with a Perkin-Elmer 241 polarimeter. All the solvents were distilled prior to use. Tetrahydrofuran (THF) was dried by distillation from sodium-benzophenone ketyl. Phosgene 1.9 M solution in toluene was purchased from Fluka.

(4R,5S)-1,5-Dimethyl-4-phenylimidazolidin-2-one 1

The title compound was prepared as described in the literature: [11] m.p. 177 °C (lit., [12] 177 - 179 °C); IR ($CHCl_3$) 3455, 1700 cm^{-1}; ^1H NMR : 0.71 (d, 3H, J = 6.5), 2.70 (s, 3H), 3.85 (dq, 1H, J = 9, 6.5), 4.78 (d, 1H, J = 9), 5.7 (bs, 1H, NH), 7.2 - 7.4 (m, 5 ArH); ^{13}C NMR: 14.3, 28.2, 57.8, 58.2, 127.6, 128.2, 128.8, 138.7, 163.2; $[\alpha]_D$ -44.2 (c 1, CH_3OH) (Lit., [12] -44.5 (c 3, CH_3OH)). Anal. Calcd. for $C_{11}H_{14}N_2O$: C, 69.45; H, 7.42. Found: C, 69.41; H, 7.40.

(4S,5R)-1-Chlorocarbonyl-3,4-dimethyl-5-phenylimidazolidin-2-one 2

To a solution of 1 (3.84 g; 20 mmol) in dry THF (30 ml), n-BuLi (20 mmol; 8 ml of a 2.5 M solution in hexanes) was added at 0 °C under argon atmosphere, and the mixture was stirred for 1 h at 0 °C. This solution was then slowly added dropwise to a solution of phosgene in toluene (10.5 ml of 1.9 M solution in toluene) cooled to -78 °C. After 1 h, the cooling bath was removed, the solvent evaporated in vacuo, and the residue was chromatographed on silica gel (cyclohexane:ethyl acetate 1:1), to give 2 (4.0 g; 80% yield) as a white solid that can be stored unchanged at 0 °C for several weeks; m.p. 140 °C; IR ($CHCl_3$) 1783, 1713 cm-1; ^1H NMR 0.71 (d, 3H, J = 6.5), 2.86 (s, 3H), 3.98 (dq, 1H, J = 9, 6.5), 5.28 (d, 1H, J = 9), 7.15 - 7.45 (m, 5 ArH); ^{13}C NMR: 15.3, 28.9, 54.1, 63.5, 127.6, 129.3, 145.6, 153.8; $[\alpha]_D$ +11.6 (c 1, $CHCl_3$). Anal. Calcd. for $C_{12}H_{13}N_2O_2Cl$: C, 57.13; H, 5.20. Found: C, 57.09; H, 5.16.

(4S,5R)-3,4-Dimethyl-5-phenylimidazolidin-2-one-1-carbohydroxamic acid 3

Hydroxylamine hydrochloride (4.2 g; 60 mmol) was dissolved in H_2O (10 ml) and then 3 M NaOH (20 ml) at 0 °C was added. Then a solution of compound 2 (5.0 g; 20 mmol) dissolved in THF (20 ml) was added, and the mixture was stirred for 12 h at r.t. After acidification to pH 3 with 1 M HCl and extraction with ethyl acetate, the solvent was removed under reduced pressure. The residue was chromatographed on silica gel (cyclohexane:ethyl acetate 3:7), to give 3 (4.2 g; 84% yield) as a white solid: m.p. 193 °C; IR ($CHCl_3$): 3289, 3216, 1717, 1671 cm^{-1}; ^1H NMR 0.73 (d, 3H, J = 6.5), 1.7 (bs, 2H, NHOH), 2.78 (s, 3H), 3.95 (dq, 1H, J = 9, 6.5), 5.25 (d, 1H, J = 9), 7.05 - 7.4 (m, 5 ArH); ^{13}C NMR: 15.1, 28.6, 54.1, 62.9, 127.4, 129.1, 144.8, 156.3; $[\alpha]_D$ +20.1 (c 1, CH_3OH). Anal. Calcd. for $C_{12}H_{15}N_3O_3$: C, 57.82; H, 6.07. Found: C, 57.79; H, 6.03.

Cycloaddition reaction: General Procedure

To a solution of oxalyl chloride (1.40 g; 11 mmol) in dichloromethane (15 ml) cooled to -60 °C under argon atmosphere, DMSO (1.25 g; 16 mmol) in dichloromethane was added, and the mixture was stirred for 20 min. Then 3 (2.5 g; 10 mmol) was slowly dropped, dissolved in dichloromethane (50 ml), followed by triethylamine (3.0 g; 30 mmol) and the appropriate cyclic 1,3-diene (20 mmol) in dichloromethane (20 ml). After 1 h at -60 °C, the cooling bath was removed and temperature raised to r.t. Water (15 ml) was added and, after extraction with ethyl acetate, the volatiles were removed under reduced pressure. The residue was purified by silica gel chromatography (cyclohexane:ethyl acetate 3:7), to give the pure cycloadducts, whose diastereomeric ratios were previously determined by ^1H and ^{13}C NMR spectroscopy.

(1R,4S)-3-[(4S,5R)-3,4-Dimethyl-5-phenylimidazolidin-2-one-1-carbonyl]-3-aza-2-oxabicyclo[2.2.2]oct-4-ene 5a and its (1S,4R)-Diastereomer 6a

By using cyclohexa-1,3-diene, the cycloadducts were obtained in 73% yield as white solids. Diastereomeric ratio 93:7. Major isomer 5a: m.p. 147 °C; IR ($CHCl_3$): 1712, 1686 cm^{-1}; ^1H NMR: 0.78 (d, 3H, J = 6.5), 1.35-1.55 (m, 2H), 2.18 - 2.5 (m, 2H), 2.83, s, 3H), 3.87 (dq, 1H, J = 9.0, 6.5), 4.75 (m, 1H), 4.97 (m, 1H), 5.45 (d, 1H, J = 9), 6.32 (ddd, 1H, J = 8.1, 6.0, 1.7), 6.45 (ddd, 1H, J = 8.1, 5.6, 1.7) 7.05 - 7.48 (m, % ArH);

^{13}C NMR: 15.7, 21.3, 23.7, 29.1, 52.9, 55.1, 60.2, 72.0, 128.3, 128.5, 128.9, 130.8, 132.3, 137.2, 154.0, 156.3; $[\alpha]_D$ -118.1 (c 1, $CHCl_3$). Minor isomer **6a**: ^1H NMR: 0.72 (d, 3H, J = 9, 6.5), 1.35 -1.6 (m, 2H), 1.98 - 2.25 (m, 2H), 2.83 (s, 3H), 4.85 (dq, 1H, J = 9, 6.5), 4.76 (m, 1H), 4.93 (m, 1H), 5.41 (d, 1H, J = 9), 6.48 (ddd, 1H, J = 8.0, 6.1, 1.6), 6.59 (ddd, 1H, J = 8.0, 5.5, 1.6), 7.03 - 7.45 (m, 5 ArH); ^{13}C NMR: 15.5, 21.3, 24.2, 29.0, 50.1, 55.3, 60.9, 72.3, 127.6, 128.4, 128.9, 131.7, 134.2, 137.4, 156.2, 157.8; $[\alpha]_D$ -64.2 (c 0.2; $CHCl_3$). Anal. Calcd. for $C_{17}H_{19}N_3O_3$: C 65.16; H, 6.11. Found: C, 65.11; H, 6.08.

(1R,4S)-3-[(4S,5R)-3,4-Dimethyl-5-phenylimidazolidin-2-one-1-carbonyl]-3-aza-2-oxabicyclo[2.2.1] hept-4-ene 5b and its (1S,4R)-Diastereomer 6b

By using cyclopenta-1,3-diene, the cycloadducts were obtained in 63% yield as white solids. Diastereomeric ratio 87:13. Major isomer **5b**: m.p. 160 °C; IR ($CHCl_3$): 1717, 1687 cm^{-1}; ^1H NMR 0.79 (d, 3H, J = 6.6), 1.69 (bd, 1H, J = 8.7), 2.11 (ddd, 1H, J = 8.7, 1.9. 1.9), 2.83 (s, 3H), 3.91 (dq, 1H, J = 9, 6.5), 5.22 (m, 1H), 5.38 (d, 1H, J = 9), 5.51 (m, 1H), 5.91 (ddd, 1H, J = 6.0, 1.9, 1.9), 6.21 (ddd, J = 6, 1.9, 1.9), 7.08 - 7.42 (m, 5 ArH); ^{13}C NMR: 15.6, 29.0, 48.6, 55.0, 58.6, 60.0, 67.3, 84.5, 127.4, 128.3, 128.9, 129.6, 133.0, 133.7, 155.3, 163.4; $[\alpha]_D$ -89.1 (c 1, $CHCl_3$). Minor Isomer **6b**: m.p. 151 °C; ^1H NMR: 0.81 (d, 3H, J = 6.6), 1.67 (bd, 1H, J = 8.7), 2.08 (ddd, 1H,. J = 8.7, 1.9, 1.9), 2.84 (s, 3H), 3.85 (dq, 1H, J = 9, 6.6), 5.15 (m, 1H), 5.26 (m, 1H), 6.35 (ddd, 1H, J = 5.9, 1.9, 1.9), 6.45 (ddd, 1H, J = 5.9, 1.9, 1.9), 7.05 - 7.45 (m, 5 ArH); ^{13}C NMR: 15.7, 28.6, 48.6, 54.9, 58.1, 60.8, 65.8, 84.5, 127.3, 128.1, 128.7, 130.6, 134.1, 137.2, 115.4, 163.3; $[\alpha]_D$ -70.6 (c 0.3, $CHCl_3$). Anal. Calcd. for $C_{18}H_{21}N_3O_3$: C, 66.04; H, 6.47. Found: C, 65.99; H, 6.44.

(1R,4S)-3-Aza-2-Oxabicyclo[2.2.2]oct-4-ene hydrochloride 7

To a solution of **5a** (980 mg; 3 mmol) in THF 85 ml) was added H_2O_2 (30% solution in water, 9 mmol, 0.9 ml) and then a solution of LiOH monohydrate (420 mg, 10 mmol) in H_2O (10 ml). After stirring at r.t for 24 h, $Na_2S_2O_4$ (1.6 g, 10 mmol) in H_2O (5 ml) was added and the mixture was extracted twice with ethyl acetate (15 ml). The organic phase was washed with 1 M HCl (8 ml), ethyl acetate containing **1** was removed and the aqueous phase was evaporated to give **7** (0.22 g; 51% yield) as a white solid: m.p. 162 °C (lit.,[1p] 163 °C); ^1H NMR (D_2O) 1.63 (m, 2H), 2.15 (m, 1H), 2.25 (m, 1H), 4.62 (ddd, 1H, J = 6.4, 3.5, 1.4), 5.04 (ddd, 1H, J = 5.7, 3.8, 1.4), 6.60 (ddd, 1H, J = 8.5, 5.7, 1.4), 6.91 (ddd, 1H, 8.5, 6.4, 1.4); ^{13}C NMR (D_2O) 19.1, 24.0, 51.5, 74.2, 130.5, 138.5; $[\alpha]_D$ -24.7 (c 1, $CHCl_3$) (lit.,[13] -25.2 (c 1.2, $CHCl_3$)). Anal. Calcd. for $C_6H_{10}NOCl$: C, 48.83; H, 6.83. Found: C, 48.79; H, 6.81.

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