

Diastereoselective Synthesis of Diamino 1,2-Diols from Homochiral α -Aminoacylsilanes

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Abstract: We have developed a new synthetic access to stereodefined diamino 1,2-diols starting from homochiral α -aminoacylsilanes. A [3+2] cycloaddition with benzo nitrile oxide of the vinylated adducts and a reductive ring opening constitute key steps of the reaction sequence.

Key words: α -aminoacylsilanes, vinylation, dipolar cycloaddition, aminopolyols

Stereodefined hydroxy amino alcohols are well recognised as key components for a variety of protease inhibitors and other new generation pharmaceuticals and as ligands for asymmetric catalysis.¹ We recently reported² the stereoselective synthesis of functionalized amino alcohol units and of statine analogs via three- and two-carbon elongations respectively, from α -aminoacylsilanes. The use of these synthetic equivalents of aldehydes, allows to overcome² the serious problems due to the easy racemization and avoids the special precautions necessary for the synthesis, handling and storing frequently encountered with α -amino aldehydes.³ In this paper we present a new approach to the two-carbon homologation of acylsilanes based on the delivery of a vinyl unit to a carbonyl group. The reaction under study is aimed at the synthesis of stereodefined aminopolyols possessing the core-unit present in dihydroxypropylamine derivatives capable of highly efficient renin⁴ and HIV-1 protease⁵ inhibitions (Fig.1).

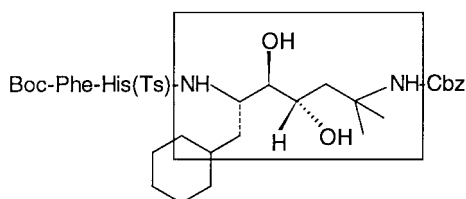
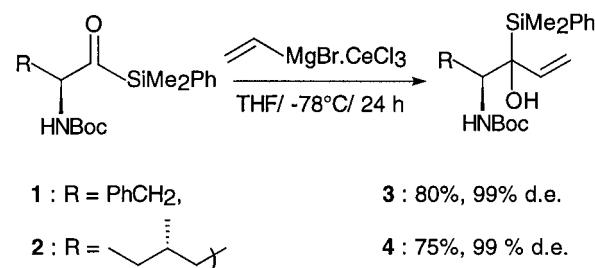


Figure 1

Our approach started with the homochiral aminoacylsilanes **1** and **2** derived from phenylalanine and isoleucine.² Since the addition of vinylmagnesium and lithium organometallics to acylsilanes studied by Kuwajima⁶ gives rise to extensive amounts of silyl enol ethers along with the expected allylic alcohols, in a previous paper⁷ we

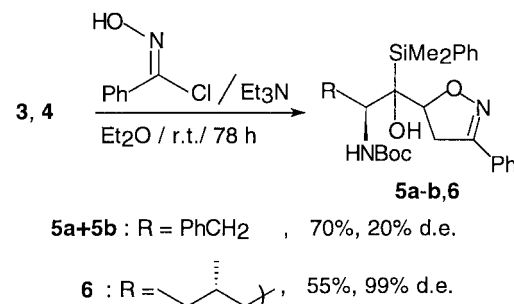
devised a convenient new entry to silylated allylic alcohols from acylsilanes by using the magnesium to cerium transmetallation technique.

Treatment of **1** and **2** with the vinylmagnesium bromide/CeCl₃ complex, led (Scheme 1) to the formation of adducts **3** and **4** in good isolated yields and high diastereoselectivities (99% d.e.).⁸



Scheme 1

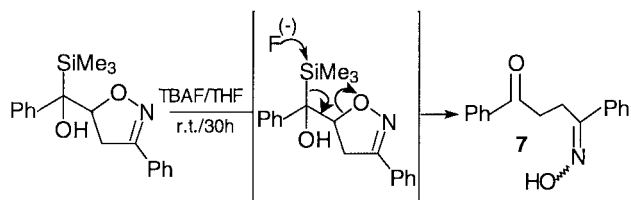
For introducing new hydroxy and amino functionalities in the vinyl moiety we envisioned the [3+2] cycloaddition with nitriloxides. Cycloadditions of this type are usually regarded as mildly electrophilic in character. Therefore (α -hydroxyallyl)silanes in which the preferred location of allylic substituents maximizes⁹ electron donation should act, as shown by Curran,¹⁰ as efficient dipolarophiles. When **3** and **4** were reacted with the in situ prepared benzo nitrile oxide, isoxazolines **5** and **6** were obtained in 70% and 55% yields. In contrast with the modest diastereomeric excess (20%) observed in the case of **5**, only one diastereoisomer (99% d.e.) could be detected and isolated in the case of **6**. The diastereomeric couple **5a,b** could be



Scheme 2

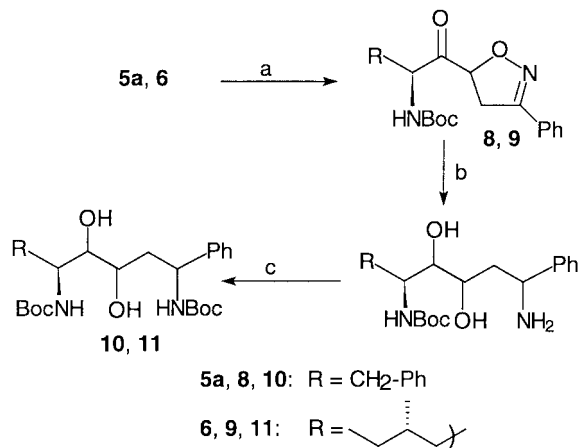
separated by column chromatography on silica gel and characterized.¹¹

The subsequent desilylation of the α -hydroxysilane cycloadducts carried out on a model compound, was disappointing. Under the action of bases the β -elimination of silanol led to ring opening with generation of an enolic form which upon rearrangement resulted in the formation of the desilylated hydroxylamino derivative **7** (Scheme 3).



Scheme 3

To prevent the occurrence of the above mentioned Peterson elimination reaction from the β -hydroxysilane moiety¹² in the presence of strongly basic reagents, isoxazolines **5a**, the major diastereoisomer, and **6** were subjected to an oxidative desilylation. This reaction, although already reported in the literature,¹³ has been however scarcely investigated. Since the oxidation with pyridinium dichromate (PDC) has found an useful and high yielding application in the synthesis of acylsilanes from 1,1-disilyl alcohols,^{13b} we applied this procedure to the desilylation of **5a** and **6**. Upon treatment with PDC at room temperature, these compounds underwent (Scheme 4) smooth desilylative oxidation to give **8** and **9** in high yields and virtually diastereomerically pure.¹⁴



Scheme 4 a) PDC, DMF, r.t., 18 h, 98% yield, 99% d.e. for both **8** and **9**; b) NaBH₄, NiCl₂·6H₂O, MeOH, -30 °C, 65 h; c) Boc₂O, MeOH, Et₃N, r.t., 4 days, 35% for **10** and 30% for **11**.

The reductive ring opening of the oxazolidine ring aimed at the formation of the new hydroxy and amino functionalities was then performed. To avoid reaction conditions able to promote an acidic proton abstraction, we exploited

the use of NaBH₄ in methanol in the presence of NiCl₂·6H₂O. Exposure of **8** and **9** to this reagent at -30 °C according to the literature¹⁵ led to heterocyclic ring opening and reduction of the carbonyl function and afforded after 65 h, the expected aminopolyols. These were directly converted to the corresponding bis-Boc derivatives: in both cases one diastereoisomer was found to largely prevail which was isolated by column chromatography and characterized.¹⁶ By this procedure N-Boc protected aminopolyols **10** and **11** are produced in 35% and 30% overall yields in three steps from oxazolidines **5a** and **6** respectively. Connectivity of diamino 1,2-diols **10** and **11** was achieved by homonuclear and heteronuclear correlation. The stereochemical assignment of the three unknown stereogenic centers of **11** was attempted on the basis of 2D-NOESY constraints and molecular modelling.¹⁷ The 15 observed constraints restricted the choice to two possible solutions: in both cases the three unknown centres are expected to display the same chirality and either configuration SSSSS or SSRRR, could be considered for **11** (Figure 2). To solve this ambiguity a single crystal X-ray diffraction¹⁸ was performed. The configuration was found to be SSSSS (Figure 2, left). Work is in progress to establish also the stereochemistry of compound **10**.

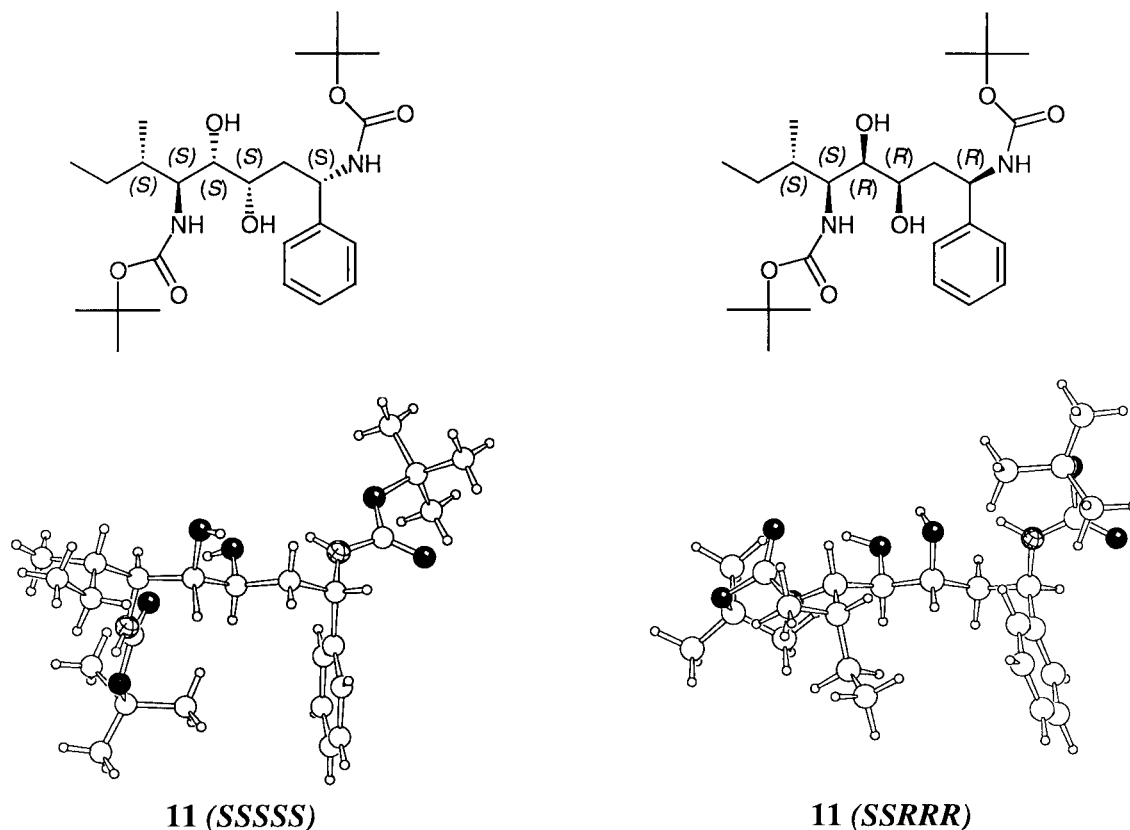
In conclusion we have established the viability of a new short route to diamino 1,2-diols starting from homochiral α -aminoacylsilanes. The importance of the target compounds highlights the significance of this new synthetic protocol in which in a single reductive-unfolding step, three new functional groups are generated.

Acknowledgement

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References and Notes

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- 3**: Yellowish oil: $[\alpha]_D^{20} = -49.3$ ($c = 2.0$, CHCl₃); ¹H NMR (200 MHz, CDCl₃) δ : 0.38 (s, 3H, Si-CH₃), 0.40 (s, 3H, Si-CH₃), 1.24 (s, 9H, C(CH₃)₃), 1.46 (s, 1H, OH), 2.54 (dd, 1H, $J = 13.6, 10.4$ Hz, CH₂-Ph), 2.90 (dd, 1H, $J = 13.6, 3.6$ Hz,



11 (SSSSS)

11 (SSRRR)

Figure 2 The two alternative structures of **11** obtained on the basis of 2D-NOESY constraints. (oxygens are represented as black, nitrogens as grid)

CH₂-Ph), 3.75-3.90 (m, 1H, CH-NH), 4.30 (d, 1H, *J* = 10.5 Hz, NH), 5.05-5.17 (m, 2H, CH₂=), 5.93-6.06 (m, 1H, CH=), 7.10-7.70 (m, 10H, Ar); ¹³C NMR (50.3 MHz, CDCl₃) δ: -4.79, -4.65, 28.11, 36.69, 57.78, 75.72, 79.06, 111.70, 125.95, 127.76, 128.12, 129.13, 129.40, 133.01, 134.57, 139.00, 139.92, 155.98. (**4**): white solid; m.p. = 65-66 °C. [α]_D²⁰ = -44.5 (*c* = 2.0, CHCl₃); ¹H NMR (300 MHz, CDCl₃) δ: 0.30 (s, 3H, SiMe), 0.33 (s, 3H, SiMe), 0.52 (t, 3H, *J* = 7.2 Hz, CH₃CH₂), 0.78-0.85 (m, 5H, CH₃CH, CH₂CH₃), 1.34 (s, 9H, C(CH₃)₃), 1.40-1.70 (m, 2H, CHCH₃, OH), 3.68, 3.78 (dd, 1H, *J* = 10.4, 1.9 Hz, CHNH), 4.50 (bd, 1H, *J* = 10.7 Hz), 4.78-5.04 (m, 2H, CH₂=), 5.88-6.05 (m, 1H, CH=), 7.25-7.55 (m, 5H, ArH); ¹³C NMR (75.46 MHz, CDCl₃) δ: -4.97, -4.34, 11.40, 17.94, 23.50, 28.34, 36.93, 59.68, 76.39, 78.78, 110.60, 127.67, 129.28, 134.60, 136.40, 141.25, 156.05.

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- (11) **Representative procedure:** preparation of (**5a-b**): Triethylamine (0.27 mL, 1.90 mmol) and **3** (600 mg, 1.52 mmol) were dissolved in Et₂O (15 mL). To this solution benzohydroxymoyl chloride (300 mg, 1.90 mmol) in Et₂O (10 mL) was slowly added (3.5 h) through a funnel. After the addition was completed the reaction was stirred for 48 h. The solvent was evaporated and the crude was purified by column chromatography on silica gel (petroleum ether : Et₂O/3:1) affording a 560 mg (70%) of two diastereoisomers in the ratio 60:40. *Major diastereoisomer (5a)*: R_f = 0.042 cm (petroleum

ether : Et₂O = 3:1). [α]_D²⁰ = +23.65 (*c* = 4.0, CHCl₃); ¹H NMR (300 MHz, CDCl₃) δ: 0.86 (s, 3H, Si-CH₃), 0.90 (s, 3H, Si-CH₃), 1.50 (s, 9H, C(CH₃)₃), 2.96 (dd, 1H, *J* = 12.6, 10.5 Hz, Ph-CH₂), 3.16 (s, 1H, OH), 3.32 (dd, 1H, *J* = 12.6, 3.1 Hz, Ph-CH₂), 3.42 (dd, 1H, *J* = 16.8, 10.5 Hz, CH₂-CH-O), 3.84 (dd, 1H, *J* = 16.8, 9.4 Hz, CH₂-CH-O), 4.10-4.25 (m, 1H, CH-N), 4.50 (d, 1H, *J* = 9.4 Hz, NH), 5.34 (t, 1H, *J* = 10.5 Hz, CH-O), 7.28-8.00 (m, 15H, Ar-H). ¹³C NMR (50.3 MHz, CDCl₃) δ: -2.83, -2.63, 28.02, 35.34, 36.83, 58.76, 73.72, 79.46, 85.47, 126.32, 126.64, 128.03, 128.33, 128.53, 128.83, 128.38, 129.54, 129.98, 134.77, 137.46, 138.21, 156.35, 157.62. HRMS (EI): *m/z* found: 517.2529 C₃₀H₂₇N₂O₄Si requires 517.2523. *Minor diastereoisomer (5b)*: R_f = 0.11 cm (petroleum ether : Et₂O/ 3:1). [α]_D²⁰ = -107.63 (*c* = 4.0, CHCl₃); ¹H NMR (300 MHz, CDCl₃) δ: 0.52 (s, 3H, Si-CH₃), 0.57 (s, 3H, Si-CH₃), 1.24 (s 9H, C(CH₃)₃), 2.56 (dd, 1H, *J* = 13.4, 8.6 Hz, CH₂-Ph.), 3.00 (dd, 1H, *J* = 16.1, 10.1 Hz, CH₂-CH-O), 3.22 (dd, 1H, *J* = 13.4, 2.2 Hz, CH₂-Ph), 3.30 (bs, 1H, OH), 3.38 (dd, 1H, *J* = 16.1, 11.2 Hz, CH₂-CH-O), 4.26 (d, 1H, *J* = 9.0, NH), 4.38-4.44 (m, 1H, CH-N), 5.40 (t, 1H, *J* = 10.1 Hz, CH-O), 7.05-7.54 (m, 15 H, Ar-H); ¹³C NMR (50.3 MHz, CDCl₃) δ: -3.55, -3.30, 28.09, 36.33, 37.36, 57.72, 73.64, 79.43, 85.02, 126.18, 126.53, 128.07, 128.44, 128.58, 129.07, 129.44, 129.68, 129.99, 133.35, 134.66, 136.81, 138.37, 156.78, 157.77. **6**: R_f = 0.8 cm (petroleum ether : Et₂O / 3:1); [α]_D²⁰ = -49.30 (*c* = 3.0, CHCl₃); ¹H NMR (CDCl₃, 200 MHz), δ: 0.55 (s, 6H, SiMe₂Ph), 0.71-0.92 (m, 8H, CH₂CH₂, CH₃CH), 1.35 (s, 9H, (CH₃)₃C), 1.55-1.75 (m, 1H, CHCH₃), 2.12 (bs, 1H, OH), 3.16 (dd, 1H, *J* = 16.5 Hz, 9.5 Hz, CH₂CHO), 3.46 (dd, 1H, *J* = 16.5 Hz, 10.0 Hz, CH₂CHO),

- 3.82 (m, 1H, CHNH), 4.76-4.98 (m, 2H, CH-O+NH), 7.19-7.39 (m, 6H, ArH), 7.42-7.58 (m, 4H, ArH); ^{13}C NMR (CDCl_3 , 50.3 MHz), δ : -3.39, -2.61, 11.73, 18.90, 24.70, 28.26, 36.05, 36.55, 59.77, 74.20, 79.07, 85.69, 126.59, 127.71, 127.84, 128.47, 129.34, 134.67, 129.95, 137.24, 155.98, 157.50. HRMS (EI): m/z found: 496.2748 $\text{C}_{28}\text{H}_{40}\text{N}_2\text{O}_4\text{Si}$ requires 496.2757.
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- (14) **Representative procedure:** preparation of **8**: To a solution of **5a** (503 mg, 0.77 mmol) in DMF (20 mL) pyridinium dichromate (PDC) (1.74 g, 4.36 mmol) was added under Ar and the mixture was stirred for 18 h. The reaction was quenched with water, the aqueous phase extracted twice with ethyl acetate, and the organic phases were combined, washed twice with water and dried over MgSO_4 . The drying agent was filtered and the solvent evaporated giving **8** (297 mg, 98%) as a yellow oil. From ^1H NMR the crude resulted pure enough for being used in the next step without any purification. **8**: $[\alpha]_{\text{D}} = +125.8$ ($c = 3.0$, CHCl_3); ^1H NMR (200 MHz, CDCl_3) δ : 1.31 (s, 9H, $\text{C}(\text{CH}_3)_3$), 2.83 (dd, 1H, $J = 13.8$, 8.9 Hz, CH_2 -Ph), 3.21 (dd, 1H, $J = 13.8$, 4.8 Hz, CH_2 -Ph), 3.47 (dd, 1H, $J = 17.2$, 11.6 Hz, CH_2 -CH-O), 3.76 (dd, 1H, $J = 17.2$, 6.0 Hz, CH_2 -CH-O), 4.82 (dd, 1H, $J = 13.4$, 8.3 Hz, CH-NH), 5.08 (d, 1H, $J = 6.5$ Hz, NH), 5.32 (dd, 1H, $J = 11.6$, 6.0 Hz, CH-O), 7.15-7.69 (m, 10 H, Ar-H); ^{13}C NMR (50.3 MHz, CDCl_3) δ : 28.18, 36.28, 36.84, 58.04, 80.22, 83.15, 126.93, 136.35, 155.35, 156.93, 206.39. HRMS (EI): m/z found: 394.1894 $\text{C}_{23}\text{H}_{26}\text{N}_2\text{O}_4$ requires 394.1892. **9** (98% yield): $R_{\text{f}} = 0.8$ (CH_2Cl_2 :MeOH / 10:1); $[\alpha]_{\text{D}} = +77.21$ ($c = 4.0$, CHCl_3); ^1H NMR (CDCl_3 , 200 MHz), δ : 0.69-1.11 (m, 8H, $\text{CH}_2\text{CH}_2 + \text{CH}_2\text{CH}$), 1.46 (s, 9H, $(\text{CH}_3)_3\text{C}$), 1.85-2.02 (m, 1H, CHCH_3), 3.58 (m, 2H, CH_2 -CH-O), 4.51 (dd, 1H, $J = 8.7$ Hz, $J = 5.2$ Hz, CHNH), 5.01 (d, 1H, $J = 8.7$ Hz, NH), 5.12-5.30 (m, 1H, CH-O), 7.29-7.42 (m, 3H, ArH), 7.58-7.66 (m, 2H, ArH); ^{13}C NMR (CDCl_3 , 50 MHz), δ : 11.30, 16.26, 24.03, 28.17, 35.69, 37.75, 61.98, 79.87, 83.25, 126.6, 128.46, 128.72, 130.49, 155.70, 156.47, 207.52. HRMS (EI): m/z found: 360.2052 $\text{C}_{20}\text{H}_{28}\text{N}_2\text{O}_4$ requires 360.2049.
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- (16) **Representative procedure:** preparation of **10**: To a solution of **8** (297 mg, 0.75 mmol) in methanol (19.3 mL) stirred under Ar, $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ (750 mg, 3.1 mmol) was added. The temperature was lowered to -30°C and NaBH_4 (290 mg, 7.7 mmol) was added in three portions. The reaction mixture was stirred at -30°C for 65 h in a cryostat, the solvent was evaporated and to the residue 25 mL of conc. NH_4OH were added. The aqueous phase was extracted twice with CH_2Cl_2 and the combined organic phases were dried over MgSO_4 , filtered and the solvent evaporated under vacuum. The crude, without any further purification, di-*tert*-butyl dicarbonate (188 mg, 0.86 mmol) and triethylamine (0.13 mL, 0.99 mmol) were sequentially dissolved in methanol (16 mL). The reaction mixture was stirred under Ar 4 days at room temperature. The solvent was evaporated and the crude purified by column chromatography over silica gel (petroleum ether: Et_2O / 1:1) giving 134 mg of the largely prevailing diastereoisomer **10** as a white solid (overall yield for three reaction steps 35%). **10**: mp 200-203 $^\circ\text{C}$; $[\alpha]_{\text{D}} = -13.3$ ($c = 1.5$, CHCl_3); ^1H NMR (200 MHz, CDCl_3) δ : 1.20 (s, 9H, $\text{C}(\text{CH}_3)_3$), 1.35 (s, 9H, $\text{C}(\text{CH}_3)_3$), 1.70 (bs, 1H, OH), 2.10 (bs, 1H, OH), 1.40-1.50 (m, 2H, CH_2CHPh), 2.40-2.50 (m, 2H, PhCH_2), 3.10 (d, 1H, $J = 9.9$ Hz, PhCH_2CHNH), 3.60-3.65 (m, 1H, PhCH_2CHCH), 3.70-3.75 (m, 1H, PhCH_2CH), 4.55-4.60 (m, 1H, $\text{PhCHCH}_2\text{CHOH}$), 4.90-5.00 (m, 1H, PhCHNH), 5.10 (d, 1H, $J = 10.1$ Hz, PhCHNH), 7.00-7.40 (m, ArH, 10H); ^{13}C NMR (50.3 MHz, CDCl_3) δ : 28.33, 28.46, 36.68, 39.08, 52.14, 53.04, 67.11, 75.26, 77.27, 79.64, 127.73-129.53, 137.68, 142.06, 156.05, 156.76. HRMS (EI): m/z found: 500.2894 $\text{C}_{20}\text{H}_{28}\text{N}_2\text{O}_4$ requires 500.2886. **11**: White solid: 30% overall yield for three reaction steps. M.p. 184-186. $^\circ\text{C}$; $[\alpha]_{\text{D}} = -11.46$ ($c = 3.0$, CHCl_3); ^1H NMR (200 MHz, CDCl_3) δ : 0.15-0.20 (m, 1H, CH_3CH_2), 0.60-0.80 (2s, 6H, CH_3 - $\text{CH}_2 + \text{CH}_3\text{CH}$), 1.00-1.10 (m, 1H, CH_3CH_2), 1.30-1.55 (m, 19H, $2\text{Me}_3\text{C}$, CH_2CHPh), 1.85-1.95 (m, 1H, CHCH_3), 1.95-2.00 (m, 1H, NHCHCHOH), 2.35-2.40 (m, 1H, CH_2CHPh), 2.80-2.85 (m, 1H, $\text{CH}(\text{OH})\text{CHNH}$), 3.40-3.45 (m, 1H, CHCHNH), 3.60-3.65 (m, 1H, CH_2CHOH), 4.00-4.10 (d, 1H, $J = 9.8$ Hz, CHCHNH), 4.60 (s, 1H, CH_2CHOH), 5.20-5.25 (m, 1H, PhCH), 6.10 (d, 1H, $J = 10.0$ Hz, PhCHNH), 7.10-7.40 (m, 5H, ArH); ^{13}C NMR (200 MHz, CDCl_3) δ : 12.05, 16.65, 22.93, 38.74, 28.10, 28.29, 38.74, 52.37, 57.39, 67.06, 73.30, 79.34, 80.07, 126.09, 128.20, 128.43, 142.25, 155.77, 157.68. HRMS (EI): m/z found: 466.3052 $\text{C}_{20}\text{H}_{28}\text{N}_2\text{O}_4$ requires 466.3043.
- (17) NMR spectra were recorded at 400 MHz in C_6D_6 . Homonuclear correlations were obtained by the gCOSY sequence. Heteronuclear correlations were obtained by edited-gHSQC sequence. 2D-NOESY spectra were recorded using mixing times of 1.5 s and 2.0 s. Conformational search was performed with PC-Spartan Pro v1.0.5, using the MMFF Force Field, and then the conformational minima were refined using PC-Model V7.5 and the MMX Force Field.
- (18) **Crystal Data of 11**: $\text{C}_{25}\text{H}_{42}\text{N}_2\text{O}_6$ (466.61), Orthorhombic, Space group $\text{P}2_12_12_1$, $Z = 4$, $a = 10.8904(6)$, $b = 11.2626(6)$, $c = 22.5995(12)$ \AA , $V = 2771.9(3)$ \AA^3 , $D_{\text{c}} = 1.118$ g cm^{-3} , $F(000) = 1016$, $\mu_{\text{Mo}} = 0.079$ cm^{-1} , $T = 298$ K. Data were collected using a graphite monochromated Mo-K α X-radiation ($\lambda = 0.71073$ \AA) in the range $3.25^\circ < \theta < 30.00^\circ$. Of 31346 reflections measured, 8104 were found to be independent ($R_{\text{int}} = 0.0847$), 3670 of which were considered as observed [$F_0 > 4\sigma(F_0)$], and were used in the refinement of 302 parameters leading to a final R_1 of 0.0546 and a R_{all} of 0.1141. The structure was solved by direct method and refined by full-matrix least squares on F^2 , using SHELXTL 97 program packages. In refinements were used weights according to the scheme $w = [\sigma^2(F_0^2) + (0.0895 P)^2 + 0.0000 P]^{-1}$ where $P = (F_0^2) + 2 F_c^2 / 3$. The hydrogen atoms were located by geometrical calculations and refined using a "riding" method; wR_2 was equal to 0.1447. The other atoms were anisotropically refined except the two *t*-butyl groups that were disordered and so splitted into two position and isotropically refined. The goodness of fit parameters S was 0.846. The largest difference between peak and hole was 0.572 and -0.272 $\text{e}\text{\AA}^{-3}$. Crystallographic data (excluding structure factors and including selected torsion angles) have been deposited with the Cambridge Crystallographic Data Center. CCDC-160358.

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