Evidence for Stereoelectronic Control in the OsO_4 Bis-hydroxylation of *trans*-Cyclohex-2-ene-1,4-diols. Synthesis of Differentially Protected *myo*-Inositol Derivatives

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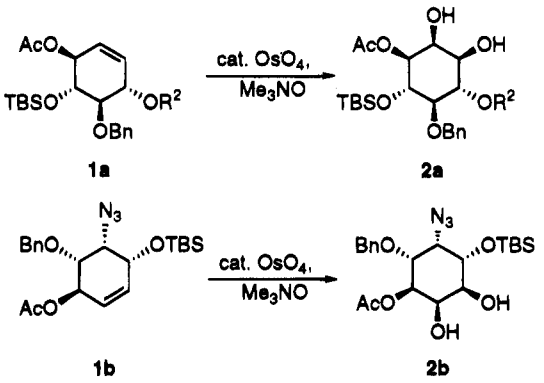
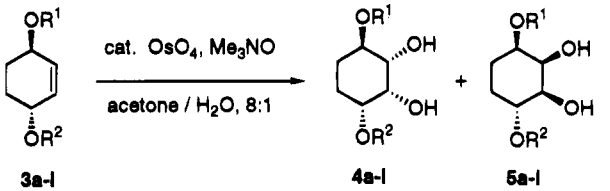
The importance of osmium tetraoxide-catalyzed *cis*-dihydroxylation of olefins is evident for the consideration of its use in the stereoselective synthesis of polyhydroxylated compounds.[1] In the case of the oxidation of allylic-substituted alkenes the origin of the diastereoselection can be attributed to steric,[2] stereoelectronic,[3] and conformational[4] effects. From the studies performed by Kishi,[5] an empiric rule established that osmylation of 1,2-disubstituted acyclic and cyclic olefins containing an allylic oxygen-bearing stereocenter affords diols with a stereochemically predictable outcome with the major product being the one arising from the *anti* attack to the oxygen function. If the change of the electronic properties of a stereogenic center controls the diastereoselectivity in the OsO_4 attack, this powerful tool should be employed in organic synthesis. In this context we have previously described short and efficient syntheses of the *myo*-inositol derivative **2a**[6a,b] and of the aminocyclitol moiety of the antibiotic hygromycin A (**2b**)[6c] from 7-oxanorbornenes[7] (Scheme 1).

Scheme 1



Scheme 2



In both cases, the key step in our synthetic strategy was the final, highly diastereoselective bis-hydroxylation of suitably protected systems **1a,b**. In these cases the empirical rule of Kishi is not operative because oxygen substituents are present in both allylic positions. Since the possible extension of this methodology to other synthetic procedures appears to be attractive, we decided to explore the factors that primarily control the process. This goal could be accomplished if a relationship between some "secondary" electronic characteristics of both allylic C–O bonds (for instance, the electron-withdrawing or electron-donating nature of the protecting groups attached to the allylic oxygens) and diastereoselectivity is observed for suitable models. The *trans*, sterically unbiased cyclohexene-1,4-diols **3** (Scheme 2) appear to be an appealing possibility.

The selectively protected model olefinic substrates **3a-l** were efficiently prepared using a previously described procedure[8] and subjected to catalytic osmium tetraoxide *cis*-dihydroxylation in an 8:1 acetone:water solvent system at room temperature for 16–40 h with trimethylamine *N*-oxide acting as the terminal oxidant,[9] leading to mixtures of dihydroconduritol C derivatives **4a-l** and **5a-l**. The results are gathered in Table 1. Diastereomeric ratios were taken from 300 MHz ^{1}H NMR spectra of crude mixtures of products. Selectivities ranging between 39:61 and 93:7 were obtained.

When it is considered that the presence of two *O*-substituents with the same electronic features but very different size ("bulky" TBS and "small" Me) did not show any selectivity (Table 1, entry 1), the steric effect associated with the *O*-substituent can be ruled out as the determining factor for the selectivity of the reaction. On the other hand, while a free hydroxy group increases the ratio to *ca.* 80:20 if an electron-donating group is attached to the other allylic oxygen (Table 1, entries 2 and 3), noteworthy reversal of diastereoselectivity occurs when

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(5) Cha, J. K.; Christ, W.; Kishi, Y. *Tetrahedron*, **1984**, *40*, 2247–2255.

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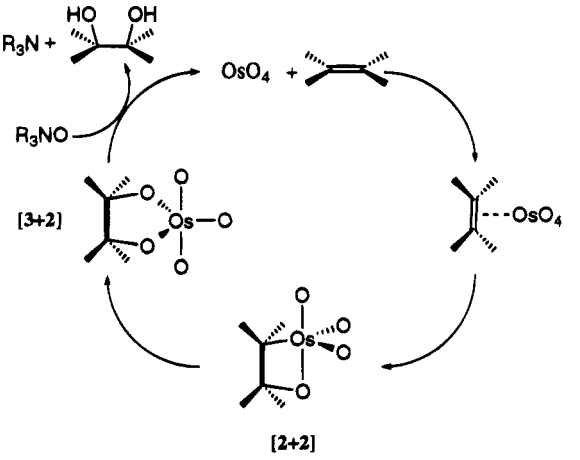
Table 1. OsO_4-Catalyzed Bis-hydroxylation of model *trans*-Cyclohexene-1,4-diol Derivatives 3a-l

entry	substrate	R^1	R^2	ratio **4**:**5**[a,b]
1	**3a**	TBS	Me	50:50
2	**3b**	TBS	H	80:20[c]
3	**3c**	Bn	H	78:22[c]
4	**3d**	Bz	H	39:61
5	**3e**	3,5-DNBz	H	44:56
6	**3f**	TBS	3,5-DNBz	93:7
7	**3g**	TBS	Bz	80:20
8	**3h**	TBS	*p*-MeOBz	76:24
9	**3i**	Bn	3,5-DNBz	79:21
10	**3j**	Bn	p-BrBz	77:23
11	**3k**	Bn	Bz	76:24
12	**3l**	*p*-MeOBn	Bz	81:19

[a] Determined by ^1H NMR (300 MHz) of the crude reaction mixture. [b] Variable amounts (*ca.* 20%) of product resulting from ester migration to the vicinal *syn*-hydroxyl group from **5d-e** and **4f-l** were detected. [c] Ratio obtained from the mixture of triacetate derivatives.

Scheme 3



R^1 is an electron-withdrawing group (Table 1, entries 4 and 5). The same trend was also observed for entries 6–12 in which R^1 is always an electron-donating group (TBS, Bn, *p*-MeOBn) and R^2 is an electron-withdrawing group (Bz, 3,5-$(NO_2)_2$Bz, *p*-BrBz, and *p*-MeOBz). The major product was always the diol *anti* to the allylic oxygen attached to the electron-donating group and *syn* to the the allylic oxygen bearing an electron-withdrawing group. However, no significant changes could be observed when tuning the electronic characteristics of the benzoate or benzyl moiety (Table 1, entries 4–12; compare with the previous results of Halterman and McEvoy[10]). In summary, an overall selectivity of *ca.* 3:1 is predictible for the combination benzyl ether–benzoate as can be deduced in the more simple example (Table 1, entry 11). The structures and stereochemistry of these products were determined by spectroscopic methods (^1H NMR and selective decouplings). For instance, ^1H NMR analyses of pure mixture of triols **4d** and **5d** (39:61) showed for the minor product a triplet of doublets at 5.22 ppm (J = 8.7 and 4.2 Hz, H-1) and for the major product a ddd at 4.89 ppm (J = 11.7, 4.5, and 2.1 Hz, H-1) indicating a *cis*-diol stereochemistry *anti* and *syn* to the benzoate, respectively. On the other hand, the mixture **4k** and **5k** (76:24) presented absorptions at 4.98 ppm (ddd, J = 10.7, 4.7, and 2.3 Hz, H-1, major product) and 5.24 ppm (td, J = 8.0 and 4.0 Hz, H-1, minor product) which ensures the proposed diol arrangement.

The results can be explained if we assume a product-like transition state[11] involving an irreversible [3 + 2] osmate adduct formation in the rate-determining step, i.e., the irreversible competitive attack of Os(VIII) on the two faces of the olefin (Scheme 3). In this way the controversial point about whether the mechanism is stepwise via a reversible [2 + 2] adduct[12] or not becomes irrelevant from the stereochemical point of view. On the other hand, this transition state is inherently electron deficient as can be deduced from the kinetic data which establish that electron-donating substituents accelerate the reaction.[13] Thus, the levels of diastereoselectivity due to variation in the electronic nature of the allylic C–O bonds can be explained by the Cieplak postulate of stereoelectronic control.[14]

Furthermore, the enhanced diastereoselection observed with an allylic free hydroxyl group can be explained by formation of hydrogen bond to an oxo group on osmium (Table 1, entries 2 and 3 *vs* entry 1). Sharpless has recently proposed this effect to explain enhanced regio- and enantioselection in asymmetric dihydroxylation of *cis*-allylic acyclic alcohols. The reversal of diastereoselectivity observed in entries 4 and 5 (Table 1, R_1: Bz, 3,5-DNBz; R_2: H) is due to the balance of the electron-donating characteristics of the OH group (favoring attack *anti* to this group) and the hydrogen bond formation (favoring attack *syn* to this group).

Now we turn our attention to our *myo*-inositol synthesis[6a,b] in an effort to achieve a complete diastereoselectivity when different conduritol B systems are osmylated. The route to these substrates is outlined in Scheme 4.

From racemic 6-*endo*-(benzyloxy)-5-*exo*-hydroxy-7-oxabicyclo[2.2.1]heptan-2-one prepared in three steps (65%, overall)[15] from 7-oxanorbornen-2-one and protected as TBS ether (100%) **6a** or benzyl ether (90%) **6b**, the corresponding enones **7a,b** were easily obtained using Vogel's ring opening conditions (TBSOTf / Et_3N system).[16] The selective 1,2 axial reduction of the carbonyl group using an excess of LiAl(*t*-BuO)$_3$H leads to diastereomerically pure conduritols B **8a,b**. Neither traces of the epimeric conduritols F from equatorial attack nor competitive 1,4 reduction products could be detected.[17]

Thus, this route allows for sequential protection after which **8a,b** were suitably functionalized and bis-hydrox-

(10) A Hammett correlation (ratio vs σ_p) with a moderate slope (ϱ = 0.46, $\Delta\Delta G^* \approx$ 1.1 Kcal/mol) indicating a moderate stereoelectronic control was obtained by Halterman (see ref 3d) by varying the electronic donation ability of aryl groups directly attached to the allylic position.

(11) Evans, D. A.; Kaldor, S. W. *J. Org. Chem. Soc.* **1990**, *55*, 1698–1700. See also refs 2c and 3d.

(12) The more accepted mechanism involves a stepwise process with an initial [2 + 2] cycloaddition with formation of a metalaoxetano which undergoes rearrangement to the [3 + 2] cycloadduct. See: (a) Sharpless, K. B.; Teranishi, A. Y.; Bäckvall, J. E. *J. Am. Chem. Soc.* **1977**, *99*, 3120–3128. (b) Tomioka, K.; Nakajima, M.; Iitaka, Y.; Koga, K. *Tetrahedron Lett.* **1988**, *29*, 573–576. (c) Tomioka, K.; Nakajima, M.; Koga, K. *Tetrahedron Lett.* **1990**, *31*, 1741–1742. (d) Jorgensen, K. A.; Schiott, B. *Chem. Rev.* **1990**, 1483–1506. See also: Jorgensen, K. A.; Hoffmann, R. *J. Am. Chem. Soc.* **1986**, *108*, 1867–1876.

(13) For kinetic studies on osmylation reactions, see: (a) Badger, G. M. *J. Chem. Soc.* **1949**, 456–463. (b) Badger, G. M.; Lynn, K. R. *J. Chem. Soc.* **1950**, 1726–1729. (c) Badger, G. M. *J. Chem. Soc.* **1950**, 1809–1814. (d) Henbest, H. B.; Jackson, W. R.; Robb, B. C. G. *J. Chem. Soc. B* **1966**, 803–807. (e) Sharpless, K. B.; Williams, D. R. *Tetrahedron Lett.* **1975**, 3045–3046. For more recent reports, see: Andersson, P. G.; Sharpless, K. B. *J. Am. Chem. Soc.* **1993**, *115*, 7047–7048.

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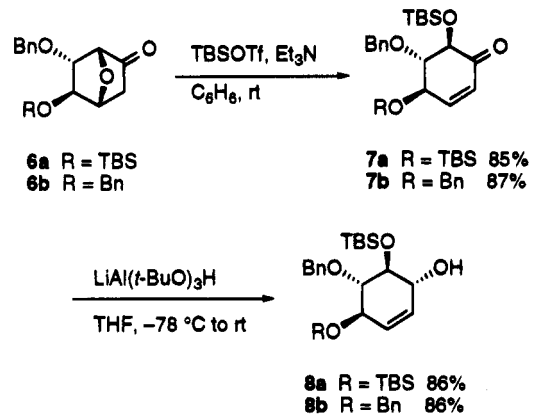
Scheme 4



Scheme 5

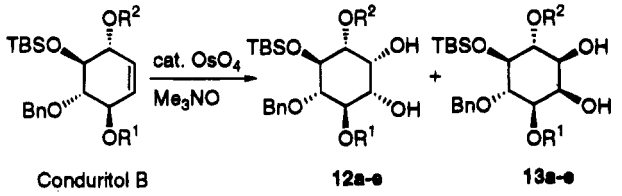


Table 2. Synthesis of *myo*-Inositol Derivatives via Diastereoselective Bis-hydroxylation of Conduritol B Systems

entry	conduritol B	R^1	R^2	ratio **12**:**13**	products[c]
1	**9a**	TBS	Ac	91:9[d]	12a:13a
2	**9b**	Bn	Ac	89:11	12b:13b
3	**8a**	TBS	H	91:9[d]	12c:13c
4	**10a**	TBS	3,5-DNBz	88:12	12d:13d
5	**11a**	TBS	Bn	52:48	12e:13e

[a] Determined by ^{1}H NMR (300 MHz) analyses of the crude reaction mixture. [b] Small amounts (<7%) of product resulting from ester migration to vicinal *syn* hydroxyl group in **12a,b** and **12d** were also detected. [c] Overall yields 80−95%. [d] Ratio obtained from the mixture of triacetate derivatives.

ylated. The results are summarized in Scheme 5 and Table 2. The diastereomeric ratios were determined,[18] and the major products **12a-d** (Table 2, entries 1−4) were isolated and fully characterized.

Once again, the electronic characteristics associated with the allylic substituents appear to be crucial to produce a synthetically useful result (compare entries 1 and 5, Table 2). The overall selectivity is now increased to *ca.* 9:1, and it does not depend on the specific electron-withdrawing group (Table 2, entries 1 and 4). Interestingly, the free hydroxyl group was found to direct the osmylation to the same extent as the acetyl group (Table 2, entries 1 and 3). For our cyclitols, the model compounds exhibit, in general, a moderate π-facial stereocontrol (Table 1), but for the more conformationally rigid conduritol B systems it is increased, probably due to inherent steric factors (*i.e.*, the specific substitution pattern (Table 2)). The overall chirality of the substrate must also play a role.

In conclusion, factors that affect stereoelectronic control in the OsO_4-catalyzed bis-hydroxylation of *trans*-cyclohex-2-ene-1,4-diols have been examined. The stereoselectivity can be tuned by the choice of electron-withdrawing or electron-donating protecting groups attached to the allylic oxygens. Complimentarity of both protecting groups appears to be necessary in order to obtain a high level of stereoselectivity. Moreover, the synthetic potential of this effect has been efficiently applied to the synthesis of biologically important *myo*-inositol derivatives.

Experimental Section

General Methods. Reagents and solvents were handled by using standard syringe techniques. Tetrahydrofuran was distilled from sodium and benzophenone; benzene from sodium; toluene, DMF, HMPA, dichloromethane, and triethylamine from calcium hydride; pyridine from KOH; and acetone from $KMnO_4$. All other solvents were reagent grade. Commercial osmium tetraoxide (2.5% weight solution in *t*-BuOH) was purchased from Aldrich. Flash chromatography was performed using Merck 230−400 mesh silica gel. Analytical TLC was carried out on Merck (Kiesegel 60F-254) silica gel plates with detection by UV light, acidic vanillin solution, or phosphomolybdic acid solution in ethanol. Melting points were determined on a Büchi 512 apparatus and are uncorrected. Elemental analyses were performed at the Universidad Complutense de Madrid.

Starting Materials. Compounds **3a-l** were prepared by a previously described method. See ref 8.

General Procedure for the Bis-hydroxylation with Osmium Tetraoxide. To a solution of 1 equiv of the alkene in a mixture of acetone and water (8:1; 20 mL per mmol) was added $Me_3NO{\cdot}2H_2O$ (2.0 equiv) and a solution (2.5% weight in *t*-BuOH) of a catalytic amount of OsO_4 (5 mol %). After being stirred at room temperature for 16−40 h (until total conversion of the starting materials), the reaction mixture was quenched with a few drops of a solution of 10% $NaHSO_3$, and the solvents were distilled in vacuo. The crude material was diluted with MeOH and filtrated through a short pad of silica gel. After removal of the solvent by distillation the crude product was purified by column chromatography on silica gel with the appropriate eluent.

(1*R,2*R**,3*S**,4*R**)-4-*O*-(*tert*-Butyldimethylsilyl)-1-*O*-(3,5-dinitrobenzoyl)cyclohexane-1,2,3,4-tetraol, 4f.** From **3f** (200 mg, 0.38 mmol), Me_3NO, and OsO_4 was obtained **4f** (190 mg, 90%) as a transparent syrup after chromatographic purification of the diastereomeric mixture of **4f**:**5f** (93:7). Data for **4f**: R_f = 0.14 (hexane/EtOAc, 3:1); ^{1}H NMR δ −0.05 (s, 3 H), 0.04 (s, 3 H), 0.73 (s, 9 H), 1.78−1.82 (m, 2 H), 1.89−1.97 (m, 2 H), 3.85 (td, 1 H, J = 8.9, 2.8 Hz), 4.16 (dd, 1 H, J = 8.7, 3.6 Hz), 4.20 (t, 1 H, J = 3.2 Hz), 5.00 (dd, 1 H, J = 8.6, 3.0 Hz), 9.20 (m, 3 H); ^{13}C NMR δ −4.8, −4.5, 17.7, 25.4, 25.7, 26.0, 67.7, 69.8, 70.8, 79.2, 122.4, 129.6, 133.8, 148.5, 161.9; IR (CCl_4) 3400, 1740 cm^{-1}. Anal. Calcd for $C_{19}H_{28}O_9N_2Si$: C, 49.87; H, 6.38. Found: C, 50.15; H, 6.10.

1-*O*-Acetyl-5-*O*-benzyl-4,6-bis-*O*-(*tert*-butyldimethylsilyl)-DL-*myo*-inositol, 12a. From conduritol (**9a**) (81 mg, 0.16 mmol), Me_3NO, and OsO_4 was obtained **12a** (81 mg, 95%) as a white solid after recrystallization from $CHCl_3$/hexane. Data for **12a**: R_f = 0.24 (hexane/EtOAc, 1:1); mp 118−119 °C; ^{1}H NMR δ −0.11 (s, 3 H), −0.06 (s, 3 H), 0.09 (s, 3 H), 0.12 (s, 3 H), 0.85 (s, 9 H), 0.80 (s, 9 H), 2.13 (s, 3 H), 2.26−2.41 (br s, 2 H), 3.22 (t, 1 H, J = 8.7 Hz), 3.55 (dd, 1 H, J = 8.8, 2.7 Hz), 3.94 (t, 1 H, J = 8.8 Hz), 4.16 (dd, 1 H, J = 9.3, 8.8 Hz), 4.19 (t, 1 H, J = 2.7 Hz), 4.76 (dd, 1 H, J = 9.4, 2.7 Hz), 4.85 (br s, 2 H), 7.26−7.30 (m, 5 H); ^{13}C NMR δ −4.7, −4.3, −4.3, −4.1, 17.8, 18.7, 21.3, 25.6, 25.8, 69.6, 71.2, 72.8, 74.1, 74.5, 74.9, 83.4, 125.9, 126.5, 127.7, 127.8, 138.7, 170.3; IR (KBr) 3460, 1730 cm^{-1}. Anal. Calcd for $C_{27}H_{48}O_7Si_2$: C, 59.96; H, 8.94. Found: C, 59.66; H, 9.02. ^{1}H NMR of the crude product showed a 91:9 ratio (**12a**:**13a**) of diastereomers. Selected ^{1}H NMR data for **13a**: 4.58 ppm (t, 1 H, J = 9.5 Hz).

(17) Reaction of **7a** under Luche's conditions ($NaBH_4/CeCl_3$, MeOH, 0 °C) provided a 94:6 mixture of epimers favoring conduritol B derivative **8a**, while the use of DIBAL, 9-BBN, or $NaBH_4$ led to mixture of 1,2 and 1,4 reduction products. There are controversial results in the literature about these reductions whose stereochemical outcome seems to depend on the specific substitution pattern for the cyclohexenone. For a general reference, see: Gemal, A. L.; Luche, J. L. *J. Am. Chem. Soc.* **1981**, *103*, 5454−5459.

(18) The osmilation products were unequivocally assigned using characteristic coupling constants (see Experimental Section). When acetates or benzoates were used as protecting groups the amount of migration product detected could not be avoided under a variety of reaction conditions.

1,2,3-Tri-*O*-acetyl-5-*O*-benzyl-4,6-bis-*O*-(*tert*-butyldimethylsilyl)-*myo*-inositol. From **12a** (100 mg, 0.18 mmol) was obtained *meso*-triacetate (110 mg, 100%) as a white solid after recrystallization from $CHCl_3$/hexane. Data: $R_f = 0.10$ (hexane/EtOAc, 10:1); mp 157 °C; ^1H NMR δ −0.12 (s, 6 H), 0.02 (s, 6 H), 0.76 (s, 18 H), 1.99 (s, 6 H), 2.12 (s, 3 H), 3.29 (t, 1 H, J = 9.1 Hz), 4.00 (t, 2 H, J = 9.5 Hz), 4.84 (dd, 2 H, J = 9.9, 2.9 Hz), 4.87 (br s, 2 H), 5.47 (t, 1 H, J = 2.9 Hz), 7.28 (m, 5 H); ^{13}C NMR δ −4.5, −4.0, 17.9, 20.6, 20.9, 25.6, 69.2, 71.7, 71.8, 75.5, 84.1, 126.0, 126.6, 127.8, 138.7, 169.7, 169.9; IR (KBr) 1760 cm^{-1}. Anal. Calcd for $C_{31}H_{52}O_9Si_2$: C, 59.58; H, 8.38. Found: C, 59.36; H, 8.60. The same triacetate was also obtained from osmylation of **8a** (that afforded a 91:9 mixture of diastereomeric triols **12c**:**13c**) and acetylation (82%, overall).

1-*O*-Acetyl-4,5-di-*O*-benzyl-6-*O*-(*tert*-butyldimethylsilyl)-*DL*-*myo*-inositol, 12b. From conduritol (**9b**) (250 mg, 0.52 mmol), Me_3NO, and OsO_4 was obtained **12b** (215 mg, 81%) as a white solid after recrystallization from $CHCl_3$/hexane. Data for **12b**: $R_f = 0.12$ (hexane/EtOAc, 2:1); mp 147−148 °C; ^1H NMR δ −0.04 (s, 3 H), 0.05 (s, 3 H), 0.81 (s, 9 H), 2.09 (s, 3 H), 2.58 (br s, 1 H), 2.83 (br s, 1 H), 3.30 (t, 1 H, J = 9.2 Hz), 3.54 (dd, 1 H, J = 9.6, 2.6 Hz), 3.73 (t, 1 H, J = 9.5 Hz), 4.09 (t, 1 H, J = 2.8 Hz), 4.11 (apparent t, 1 H, J = 9.3 Hz), 4.65 (dd, 1 H, J = 9.9, 2.8 Hz), 4.70 (AB system, 2 H), 4.83 (AB system, 2 H), 7.18−7.31 (m, 10 H); ^{13}C NMR δ −4.4, −4.0, 17.9, 21.3, 25.8, 69.8, 71.2, 71.5, 74.3, 75.3, 75.5, 81.3, 83.6, 126.8, 126.9, 127.1, 127.2, 127.4, 127.5, 127.8, 127.9, 128.1, 128.2, 128.4, 138.2, 138.5, 145.0, 170.3; IR (KBr) 3450, 3410, 1725 cm^{-1}. Anal. Calcd for $C_{28}H_{40}O_7Si$: C, 65.09; H, 7.80. Found: C, 64.83; H, 7.69. ^1H NMR of the crude product showed an 89:11 ratio (**12b**:**13b**) of diastereomers. Selected ^1H NMR data for **13b**: 5.15 ppm (t, 1 H, J = 9.4 Hz).

5-*O*-Benzyl-4,6-bis-*O*-(*tert*-butyldimethylsilyl)-1-*O*-(3,5-dinitrobenzoyl)-*DL*-*myo*-inositol, 12d. From **10a** (75 mg, 0.11 mmol), Me_3NO, and OsO_4 was obtained **12d** (61 mg, 80%) as a transparent syrup. Data for **12d**: $R_f = 0.17$ (hexane/EtOAc, 3:1); ^1H NMR δ −0.10 (s, 3 H), −0.09 (s, 3 H), −0.04 (s, 3 H), 0.10 (s, 3 H), 0.67 (s, 9 H), 0.87 (s, 9 H), 2.40−2.50 (br s, 2 H), 3.35 (t, 1 H, J = 8.2 Hz), 3.70 (dd, 1 H, J = 8.2, 3.0 Hz), 4.05 (t, 1 H, J = 8.5 Hz), 4.29 (t, 1 H, J = 2.9 Hz), 4.38 (t, 1 H, J = 8.4 Hz), 4.86 (AB system, 2 H), 5.18 (dd, 1 H, J = 9.0, 3.0 Hz), 7.27 (m, 5 H), 9.19 (d, 2 H, J = 2.1 Hz), 9.23 (t, 1 H, J = 2.1 Hz); ^{13}C NMR δ −4.2, −4.1, −4.0, −3.9, 17.9, 18.3, 25.7, 26.0, 69.3, 71.4, 73.1, 73.9, 75.0, 77.1, 83.2, 122.7, 126.2, 126.9, 128.0, 129.7, 129.8, 133.8, 138.5, 148.7, 162.3; IR (CCl_4) 3580, 3500, 1735 cm^{-1}. Anal. Calcd for $C_{32}H_{48}N_2O_{11}Si_2$: C, 55.47; H, 6.98. Found: C, 55.82; H, 6.60. ^1H NMR of the crude product showed an 88:12 ratio (**12d**:**13d**) of diastereomers. Selected ^1H NMR data for **13d**: 5.67 ppm (t, 1 H, J = 9.4 Hz).

1,3-Di-*O*-benzyl-2,4-bis-*O*-(*tert*-butyldimethylsilyl)-*DL*-*myo*-inositol, 12e. From **11a** (120 mg, 0.21 mmol), Me_3NO, and OsO_4 were obtained **12e**:**13e** (105 mg, 75%) as a transparent syrup of unseparable mixture of diastereomers in a ratio of 52:48 (**12e**:**13e**). Data for **12e**:**13e**: $R_f = 0.11$ (hexane/EtOAc, 10:1); ^1H NMR δ −0.04 (s, 3 H), 0.02 (s, 3 H), 0.05 (s, 3 H), 0.06 (s, 6 H), 0.08 (s, 3 H), 0.09 (s, 3 H), 0.10 (s, 3 H), 0.70 (s, 9 H), 0.77 (s, 9 H), 0.81 (s, 9 H), 0.86 (s, 9 H), 3.41−3.70 (m, 8H), 3.71 (m, 4H), 3.90 (m, 2H), 4.03 (m, 2H), 4.58−4.87 (m, 4H), 4.65 (brs, 2H), 4.80 (brs, 2H), 7.18−7.35 (m, 20H).

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Supporting Information Available: Experimental and spectroscopic data for compounds **3a-l**, **4a-l**, **5a-l**, **7a,b**, **8a**, **9a,b**, **10a**, and **11a** (6 pages). This material is contained in libraries on microfiche, inmediately follows this article in the microfilm version of the journal, and can be ordered from the ACS; see any current masthead page for ordering information.

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