Novel Ring Enlargement of Lactams via Quinazolinone Annelation. A Facile Route to Benzoannelated Large-Membered Cyclic 1,5-Diamines

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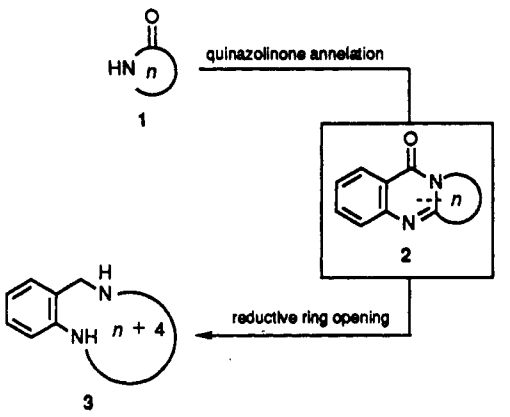
A novel route to benzoannelated large-membered cyclic 1,5-diamines from lactams is described. Thus, *n*-membered lactams **1** were N-acylated by *o*-azidobenzoyl chloride **5** to afford the corresponding imides **4**. These were treated with tributylphosphine and underwent an intramolecular aza-Wittig reaction to give *n*-membered ring-fused quinazolinones **2** in 84–96% yield. By reductive cleavage of **2** with BH_3·THF, (*n* + 4)-membered cyclic diamines **3** were obtained in 52–95% yield.

Introduction

The aza-Wittig reaction, in particular its intramolecular version, has drawn considerable attention recently because of its high potential for the synthesis of nitrogen heterocycles.[1] We demonstrated recently[2] that imide carbonyls are reactive in the intramolecular aza-Wittig reaction and that imidazolinones and quinazolinones can be formed under mild conditions. When lactams are used as starting materials, the corresponding fused-ring quinazolinone derivatives can be readily prepared. Furthermore, ring enlargement of the quinazolinone moiety can provide novel benzoannelated large-membered cyclic 1,5-diamines. We now report the synthesis of a number of *n*-membered ring-fused quinazolinones **2** and their reductive ring enlargement to (*n* + 4)-membered cyclic diamines **3** (Scheme I).[3]

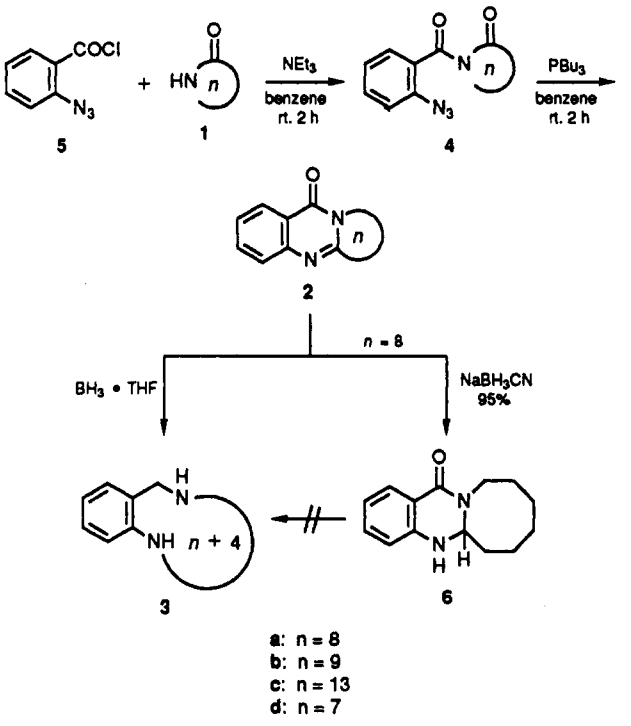
Scheme I



Results and Discussion

First studied was the 8-membered lactam **1a**, which was expected to yield a product **2a** that could be cleaved more easily, because of its higher ring strain than, say, a 12-membered quinazolinone.[4] The azide **4a** was obtained in 88% yield by *o*-azidobenzoylation of **1a** with **5**[5] (Scheme II). The cyclization of **4a** to **2a** with triphenylphosphine, via the corresponding iminophosphorane, required heating at 140 °C for 5 h, presumably because of steric hindrance. However, with tributylphosphine in benzene at 25 °C, the reaction proceeded smoothly to give the aza-Wittig product **2a** in 96% yield. This demonstrated clearly that fused-ring quinazolinones could be prepared under mild conditions. Therefore, the 9-, 13-, and 7-membered lactams **1b–d** were converted similarly to the corresponding fused-ring quinazolinones **2b–d**, via the azides **4b–d**, in good yield (Table I).

Scheme II



a: n = 8
b: n = 9
c: n = 13
d: n = 7

The reductive cleavage of the central C–N bond of the quinazolinone moiety (at the dotted line of **2**, Scheme I) was first attempted with sodium cyanoborohydride. The cleavage of the C–N bond of some acyl amidines with that reagent has been reported.[6] However, when **2a** in ethanol was treated with a 3-fold molar excess of sodium cyanoborohydride for 48 h at 20–25 °C under acidic conditions, only the carbon–nitrogen doube bond was reduced, to afford **6** (95% yield). Further reductive ring cleavage did not occur, even in refluxing solution, or with acetic acid as solvent. Because of its high reactivity as a hydride

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Table I. Summary of the Reductive Ring Enlargement of Lactams via Quinazolinone Annelation

lactam	n^a	azido deriv	yield,[b] %	quinazolinone	yield,[b] %	cyclic diamine	yield,[b] %
1a	8	**4a**	88	**2a**	96	**3a**	95
1b	9	**4b**	93	**2b**	96	**3b**	88
1c	13	**4c**	51	**2c**	84	**3c**	89
1d	7	**4d**	98	**2d**	91	**3d**	52[c]

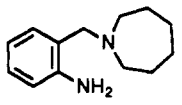
[a] Ring size of lactam. [b] Isolated yield. [c] Compound **7** was also obtained, in 30% yield.

Table II. Reaction of 2a with Diborane–Tetrahydrofuran Complex

entry	BH_3·THF, mol equiv	reaction time,[a] h	yield of **3a**,[b] %
1	3.0	6.0	87
2	5.0	4.0	85
3	10.0	2.0	95

[a] In refluxing THF. [b] Isolated yield.

transfer agent, the borane–tetrahydrofuran complex (BH_3·THF) was next employed as the reducing agent.[7] Thus, with 3 molar equiv of BH_3·THF in refluxing THF, reductive ring opening of **2a** was complete after 6 h, affording the 12-membered cyclic diamine **3a** in 87% yield. With increased amounts of BH_3·THF reagent, reductive ring opening proceeded more rapidly. Thus, reaction of **2a** with 5 and 10 molar equiv of BH_3·THF afforded **3a** in 85% and 95% yield after 4 and 2 h, respectively (Table II). Reductive ring opening was also attempted with the fused-ring quinazolinones **2b–d**. Treatment of **2b** and **2c** with 10 molar equiv of BH_3·THF afforded the 13- and 17-membered cyclic diamines **3b** and **3c**, respectively, in excellent yield (Table I). The reaction with **2d** gave the corresponding 11-membered cyclic diamine **3d**, but in a moderate yield (52%), along with an isomer diamine **7** (30%). The two were easily separated by column chromatography. The formation of **7** can perhaps be explained in terms of the relief of ring strain in **2d**: An 11-membered ring, such as **3d**, generally has a larger ring strain than a 7-membered ring, such as **7**.[4a] Therefore, cleavage of the central C–N bond (to form **3d**) competed with cleavage of the neighboring C–N bond (to form **7**).



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In summary, a novel and convenient route to benzoannelated large-membered cyclic 1,5-diamines from lactams via quinazolinone annelation and reductive ring-cleavage has been developed. The β-lactam route of Wasserman and co-workers[8] and the organoaluminium reagent method of Yamamoto and co-workers[9] are useful for the synthesis of nitrogen macrocycles. The present method provides a new and efficient route to some large-membered cyclic 1,5-diamines.[10]

Experimental Section

Melting points were determined with a Yanagimoto micro melting point hot stage apparatus and are uncorrected. IR spectra were recorded with a JASCO IRA-1 spectrometer. ^1H NMR spectra of $CDCl_3$ solutions were recorded with a Varian Gemini 200 instrument at 200 MHz. Chemical shifts are reported in parts per million (δ) relative to Me_4Si as an internal standard. Electron impact mass spectra (EIMS) were recorded with an ESCO EMD-05B or a JEOL JMS-HX110 spectrometer at 70 eV. Microanalyses were performed with a Perkin-Elmer 240B elemental analyzer. Fuji-Davison BW-300 silica gel was used for column chromatography.

Preparation of Azide Derivatives (4). General Procedure. A mixture of *o*-azidobenzoic acid (1.00 mmol) and thionyl chloride (0.82 mL, 1.33 g, 11.2 mmol) was heated at 80 °C for 2 h under nitrogen.[5] The mixture was cooled to room temperature, and the solvent was evaporated under reduced pressure. The residue was dissolved in dry THF (5.0 mL). To this solution were added the lactam (1.00 mmol) and triethylamine (2.00 mmol) under nitrogen. The mixture was stirred at room temperature (20–25 °C) for 3 h. The mixture was filtered to remove precipitated solids, and the filtrate was evaporated under reduced pressure to afford the crude azide. The crude product was purified by column chromatography on silica gel (ethyl acetate/hexane, 1:8–2:1) to give the pure azide **4** as a colorless solid or an oil.

***N*-(*o*-Azidobenzoyl)-2-azacyclooctanone (4a)**: mp 90 °C dec; IR ($CHCl_3$) 2130, 1690, 1670 cm^{-1}; ^1H NMR δ 7.17–7.11 (m, 4 H), 4.60–4.04 (m, 2 H), 2.70–2.64 (m, 2 H), 1.93–1.81 (m, 4 H), 1.72–1.51 (m, 4 H). A satisfactory elemental analysis was not obtained, but ion peaks m/z 272 (M^+) and m/z 230 ($M^+ - N_3$) corresponding to formula $C_{14}H_{16}N_4O_2$ appeared in the EIMS.

***N*-(*o*-Azidobenzoyl)-2-azacyclononanone (4b)**: mp 54–56 °C; IR ($CHCl_3$) 2930, 2130, 1700, 1675. 1660 cm^{-1}; ^1H NMR δ 7.49–7.36 (m, 2 H), 7.23–7.14 (m, 2 H), 4.02 (t, 2 H, J = 6.0 Hz), 2.55–2.49 (m, 2 H), 1.92–1.43 (m, 10 H). Anal. Calcd for $C_{15}H_{18}N_4O_2$: C, 62.92; H, 6.34; N, 19.57. Found: C, 62.83; H, 6.41; N, 19.58.

***N*-(*o*-Azidobenzoyl)-2-azacyclotridecanone (4c)**: oil; IR ($CHCl_3$) 2130, 1690 cm^{-1}; ^1H NMR δ 7.52–7.16 (m, 4 H), 3.84 (dd, 2 H, J = 6.4, 6.8 Hz), 2.49 (dd, 2 H, J = 6.6, 7.2 Hz), 1.76–1.66 (m, 4 H), 1.41–1.27 (m, 14 H). A satisfactory elemental analysis was not obtained, and only an ion peak at m/z 300 ($M^+ - N_3$, for $C_{19}H_{26}N_4O_2$) was observed in the EIMS.

***N*-(*o*-Azidobenzoyl)-2-azacycloheptanone (4d)**: mp 59–61 °C; IR ($CHCl_3$) 2130, 1715, 1680, 1600 cm^{-1}; ^1H NMR δ 7.47–7.12 (m, 4 H), 4.05–4.02 (m, 2 H), 2.72–2.67 (m, 2 H), 1.84 (br s, 6 H). Anal. Calcd for $C_{13}H_{14}N_4O_2$: C, 60.46; H, 5.46; N, 21.69. Found: C, 60.56; H, 5.77; N, 21.47.

Synthesis of Fused-Ring Quinazolinones (2). General Procedure. To a solution of *o*-azidobenzoyl lactam **4** (1.00 mmol) in benzene (5.0 mL) was added tributylphosphine (1.10 mmol) under nitrogen. The mixture was then stirred for 2 h at room temperature. After removal of the solvent under reduced pressure, the residue was purified by column chromatography on silica gel (ethyl acetate/hexane, 1:6–2:1) to afford the quinazolinone **2** as colorless crystals.

Azacyclooctano[2,1-*b*]-4(3*H*)-quinazolinone (2a): mp 109–110 °C; IR ($CHCl_3$) 1665, 1610, 1585, 1570 cm^{-1}; ^1H NMR δ 8.27 (dd, 1 H, J = 1.0, 8.0 Hz), 7.74 (ddd, 1 H, J = 1.6, 6.8, 8.2 Hz), 7.64 (dd, 1 H, J = 1.0, 8.2 Hz), 7.44 (ddd, 1 H, J = 1.6, 6.8, 8.0 Hz), 4.35 (dd, 1 H, J = 5.6, 6.0 Hz), 3.08–3.02 (m, 2 H), 2.05–1.38 (m, 8 H). Anal. Calcd for $C_{14}H_{16}N_2O$: C, 73.66; H, 7.06; N, 12.27. Found: C, 73.92; H, 7.21; N, 12.05.

Azacyclononano[2,1-*b*]-4(3*H*)-quinazolinone (2b): mp 99–110 °C; IR ($CHCl_3$) 1675, 1610, 1580, 1565 cm^{-1}; ^1H NMR δ 8.26 (dd, 1 H, J = 1.5, 8.0 Hz), 7.73 (ddd, 1 H, J = 1.5, 6.8, 8.2 Hz), 7.64 (dd, 1 H, J = 1.4, 8.2 Hz), 7.44 (ddd, 1 H, J = 1.4, 6.8, 8.0 Hz), 4.32 (br s, 2 H), 3.09–3.02 (m, 2 H), 2.08–1.34 (m, 10 H). Anal. Calcd for $C_{15}H_{18}N_2O$: C, 74.35; H, 7.49; N, 11.56. Found: C, 74.65; H, 7.58; N, 11.39.

Azacyclotridecano[2,1-*b*]-4(3*H*)-quinazolinone (2c): mp 107–108 °C; IR ($CHCl_3$) 1680, 1595, 1570 cm^{-1}; ^1H NMR δ 8.26 (ddd, 1 H, J = 0.6, 1.6, 8.0 Hz), 7.72 (ddd, 1 H, J = 1.6, 6.2, 8.2

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Hz), 7.63 (ddd, 1 H, J = 0.6, 1.5, 8.2 Hz), 7.43 (ddd, 1 H, J = 1.5, 6.8, 8.0 Hz), 2.87-2.79 (m, 2 H), 2.02-1.39 (m, 18 H). Anal. Calcd for $C_{19}H_{26}N_2O$: C, 76.47; H, 8.78; N, 9.39. Found: C, 76.50; H, 8.79; N, 9.39.

Azacycloheptano[2,1-*b*]-4(3*H*)-quinazolinone (2d): mp 95-97 °C; IR ($CHCl_3$) 1680, 1615, 1600 cm^{-1}; ^1H NMR δ 8.27 (ddd, 1 H, J = 0.6, 1.6, 8.0 Hz), 7.73 (ddd, 1 H, J = 1.6, 6.8, 8.2 Hz), 7.62 (ddd, 1 H, J = 0.6, 1.4, 8.2 Hz), 7.44 (ddd, 1 H, J = 1.4, 6.8, 8.0 Hz), 4.43-4.38 (m, 2 H), 3.11-3.06 (m, 2 H), 1.86 (br s, 6 H). Anal. Calcd for $C_{13}H_{14}N_2O$: C, 72.87; H, 6.59; N, 13.07. Found: C, 72.83; H, 6.85; N, 12.86.

Synthesis of Cyclic Diamines 3 by Reductive Ring Opening of 2. General Procedure. To a stirred solution of quinazolinone **2** (1.00 mmol) in anhydrous THF (10 mL) was added a solution of BH_3·THF (10 mL of 1.0 M solution in THF, 10.0 mmol) under nitrogen. Stirring was continued for 2 h at room temperature, and then the mixture was heated at reflux for several hours, during which time the reaction was monitored by TLC. The mixture was cooled to room temperature, and water (ca. 5 mL) was added drop-by-drop to the stirred mixture. After 50% aqueous NaOH (ca. 2 mL) was added, stirring was continued for 1 h. The mixture was then extracted with dichloromethane (5 × 30 mL). The combined extracts were dried (Na_2SO_4) and were evaporated under reduced pressure to give the crude product, which was purified by column chromatography on silica gel (ethyl acetate/hexane, 1:6-1:2) to afford cyclic diamine **3**.

1,5-Diazabenzo[*b*]cyclododecane (3a): colorless oil; IR ($CHCl_3$) 3280, 1610, 1590 cm^{-1}; ^1H NMR δ 7.21-7.12 (m, 1 H), 6.99-6.94 (m, 1 H), 6.63-6.54 (m, 2 H), 3.96 (s, 2 H), 3.16-3.11 (m, 2 H), 2.64-2.59 (m, 2 H), 1.85-1.37 (m, 12 H, 5 CH_2 and 2 NH). Anal. Calcd for $C_{14}H_{22}N_2$: C, 77.01; H, 10.16; N, 12.83. Found: C, 76.92; H, 10.14; N, 12.67.

1,5-Diazabenzo[*b*]cyclotridecane (3b): colorless solid; mp 38.0-39.5 °C; IR ($CHCl_3$) 3300, 1610, 1585 cm^{-1}; ^1H NMR δ 7.22-7.14 (m, 1 H), 7.04-6.99 (m, 1 H), 6.64-6.57 (m, 2 H), 3.80 (s, 2 H), 3.11-3.06 (m, 2 H), 2.72-2.67 (m, 2 H), 1.74-1.33 (m, 14 H, 6 CH_2 and 2 NH). Anal. Calcd for $C_{15}H_{24}N_2$: C, 77.53; H, 10.41; N, 12.06. Found: C, 77.24; H, 10.40; N, 12.36.

1,5-Diazabenzo[*b*]cycloheptadecane (3c): colorless oil; IR ($CHCl_3$) 3300, 1605, 1585 cm^{-1}; ^1H NMR δ 7.22-7.13 (m, 1 H), 7.04-7.00 (m, 1 H), 6.65-6.57 (m, 2 H), 3.77 (s, 2 H), 3.14 (t, 2 H, J = 5.9 Hz), 2.66 (t, 2 H, J = 5.8 Hz), 1.71-1.33 (m, 20 H, 9 CH_2 and 2 NH). Anal. Calcd for $C_{19}H_{32}N_2$: C, 79.11; H, 11.18; N, 9.71. Found: C, 79.30; H, 10.90; N, 9.52.

1,5-Diazabenzo[*b*]cycloundecane (3d): colorless oil; IR ($CHCl_3$) 3250, 1595 cm^{-1}; ^1H NMR δ 7.19-7.10 (m, 1 H), 7.00-6.88 (m, 1 H), 6.70-6.56 (m, 2 H), 4.04 (s, 2 H), 3.5-2.8 (br s, 2 H, 2 NH), 3.37-3.31 (m, 2 H), 2.66-2.60 (m, 2 H), 1.72-1.28 (m, 8 H). Anal. Calcd for $C_{13}H_{20}N_2$: C, 76.42; H, 9.87; N, 13.71. Found: C, 76.55; H, 9.60; N, 13.52.

***N*-(*o*-Aminobenzyl)azepane (7)**: colorless oil; IR ($CHCl_3$) 3440, 3280, 1615, 1600 cm^{-1}; ^1H NMR δ 7.13-6.94 (m, 2 H), 6.70-6.62 (m, 2 H), 4.94 (br s, 2 H, 2 NH), 3.60 (s, 2 H), 2.58 (br s, 4 H), 1.59 (br s, 8 H). Anal. Calcd for $C_{13}H_{20}N_2$: C, 76.42; H, 9.87; N, 13.71. Found: C, 76.44; H, 9.61; N, 13.82.

1,2-Dihydroazacyclooctano[2,1-*b*]-4(3*H*)-quinazolinone (6). To a solution of $NaBH_3CN$ (80 mg, 1.27 mmol) in ethanol was added quinazolinone **2a** (100 mg, 0.44 mmol) and bromocresol green indicator (2 mg). The mixture was stirred under acidic conditions to bromocresol green for 2 days at room temperature. A solution of 1 M ethanolic HCl was added as necessary to maintain acidity. The mixture was then diluted with water and was extracted with dichloromethane (3 × 50 mL). The combined extracts were dried (Na_2SO_4) and evaporated under reduced pressure to give the crude product, which was purified by preparative TLC (Merck aluminum oxide 60PF_{254}, type E, development with $CHCl_3$) to afford dihydroquinazolinone **6** as a colorless solid (96 mg, 95%): mp 160-163 °C; IR ($CHCl_3$) 1640, 1615 cm^{-1}; ^1H NMR δ 7.90 (dd, 1 H, J = 1.6, 7.8 Hz), 7.27 (ddd, 1 H, J = 1.6, 7.4, 8.0 Hz), 6.84 (ddd, 1 H, J = 1.2, 7.4, 7.8 Hz), 6.62 (dd, 1 H, J = 1.2, 8.0 Hz), 4.92 (ddd, 1 H, J = 1.6, 2.8, 7.0 Hz), 4.31 (ddd, 1 H, J = 4.4, 5.2, 14.2 Hz), 4.11 (br s, 1 H, NH), 3.02 (ddd, 1 H, J = 3.4, 10.0, 14.2 Hz), 2.15-1.38 (m, 10 H); high-resolution EIMS calcd for $C_{14}H_{18}N_2O$ 230.1420, found 230.1409.

Registry No. **1a**, 673-66-5; **1b**, 935-30-8; **1c**, 947-04-6; **1d**, 105-60-2; **2a**, 58314-97-9; **2b**, 60811-55-4; **2c**, 131043-45-3; **2d**, 4425-23-4; **3a**, 131043-46-4; **3b**, 131043-47-5; **3c**, 131043-48-6; **3d**, 131043-49-7; **4a**, 131043-41-9; **4b**, 131043-42-0; **4c**, 131043-43-1; **4d**, 131043-44-2; **5**, 31162-13-7; **6**, 131043-50-0; **7**, 19668-00-9.

Molecular Recognition: α-Cyclodextrin and Penicillin V Inclusion Complexation

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Specific molecular recognition between α-cyclodextrin (1) and a β-lactam antibiotic, penicillin V (2), was systematically determined. A stable 1:1 inclusion complex was established in the solid state, gaseous phase, and solution. The distinct structure of this inclusion complex was rigorously elucidated by FT-IR, FAB-MS, CP/MAS solid-state ^{13}C NMR, and 500-MHz ^1H NMR. Based on strictly determined ^1H NMR data, a time-averaged conformation of the α-CD-penicillin V inclusion complex was proposed, which was supported by CPK model studies and the intermolecular NOE results. Moreover, α-cyclodextrin exhibited significant catalytic activity toward the hydrolysis of penicillin V in weakly alkaline solution. These findings imply that the initial molecular recognition and the concomitant molecular association are essential in a biomimetic process.

Introduction

Cyclodextrins (cycloamyloses, CD) are naturally occurring cyclic oligosaccharides composed of 6-8 α-(1→4)-linked D-glucosyl residues. The shape of a CD molecule is a toroidal, hollow, truncated cone with primary and secondary hydroxyl groups crowning the narrower and the wider rims, respectively. The ability of CDs to admit a variety of guest molecules into their hydrophobic cavities without any covalent bond being formed[2,3] has rendered them very useful models for studying topochemistry and

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