Facile Synthesis of Model 1,4-Dihydro-1*H*-1,3,4-benzotriazepin-5-ones

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2-Aminobenzoic acid reacts readily, in the presence of triethylamine, with hydrazonoyl chlorides (**5a-c**) (precursors of the reactive nitrile imine 1,3-dipolar species) to afford high yields of the corresponding acyclic amidrazone adducts (**6a-c**). The latter adducts undergo, in THF in presence of 1,1'-carbonyldiimidazole, smooth intramolecular cyclization involving the activated carboxyl and the NH- termini to deliver unequivocally the respective dihydro-1,3,4-benzotriazepin-5-ones (**7a-c**).

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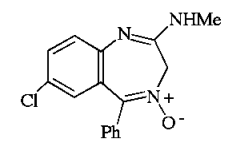
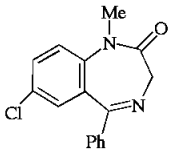
Introduction.

1,4-benzodiazepines, exemplified by chlorbenzdiazepoxide (Librium, **1**) and diazepam (Valium, **2**) (Chart 1), constitute a unique class of CNS drugs that are widely used as tranquilizers in several anxiety states [1]. Extensive research in the field of benzodiazepines has stimulated interest in the synthesis and pharmacologic action of their aza-homologues, the benzotriazepines. Amongst these, the 1,3,4-benzotriazepin-5-ones (*e.g.* **3,** Chart 1) [2-7] have been attended to, but research efforts in this area were comparatively modest. Their preparation was reported at large *via* the reaction of 2-aminobenzoylhydrazides with orthoesters (**3a-d**) [2], or *via* the reaction of 3,1-benzoxazin-4-ones with hydrazine (**3e,f**) [3], and once from the reaction of isatoic anhydride with 2-pyridylamidrazone and cyclizing the product with acid (**3g**) [4]. Some 1,3,4-benzotriazepin-6-ones (**4**) (Chart 1) were reported as useful antihypertensives, cardiotonics and fungicides [5].

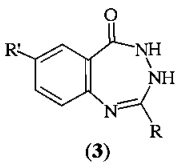
Chart 1



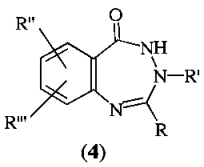
(**1**)



(**2**)



(**3**)



(**4**)

a: R = Ph (R'= H)
b: R = Me (R'= H)
c: R =R'= H
d: R = H; Me; Et; Bu (R'= H; Me; Cl; Br; NO_2)
e: R' = H; R = C_6H_4-X(*p*) (X = Me; OMe; Cl; NO_2)
f: R' = H; R = COX (X = allylamino)
g: R = 2-Pyridyl (R' = H)

R = NH_2; alkylthio
R'= Furoyl
R'' and R''' = H; alkoxy

It is worth mentioning that considerable confusion has surrounded these literature methods, which could have easily generated varying amounts of the isomeric 3-aminoquinazolin-4-ones and/or 1,3,4-oxadiazoles. In fact, several of the earlier reported syntheses of presumed 1,3,4-benzotriazepines [6] were later proven erroneous, and the alternate isomeric structures established [2a,4,7]. The formation of those intervening isomeric five- and six-membered heterocycles was occasionally suppressed by the proper choice, adjustment of, and monitoring of the reaction conditions [2c], and was excluded by the use of 1-(2-aminobenzoyl)-1-methylhydrazides [8,9].

In light of these uncertainties, we thought it was worthwhile to explore alternate preparative avenues starting from easily accessible compounds. Herein we wish to report on a convenient and versatile synthetic route toward new 1,3,4-benzotriazepin-5-ones (**7**), utilizing hydrazonoyl chlorides (**5**) (Scheme 1).

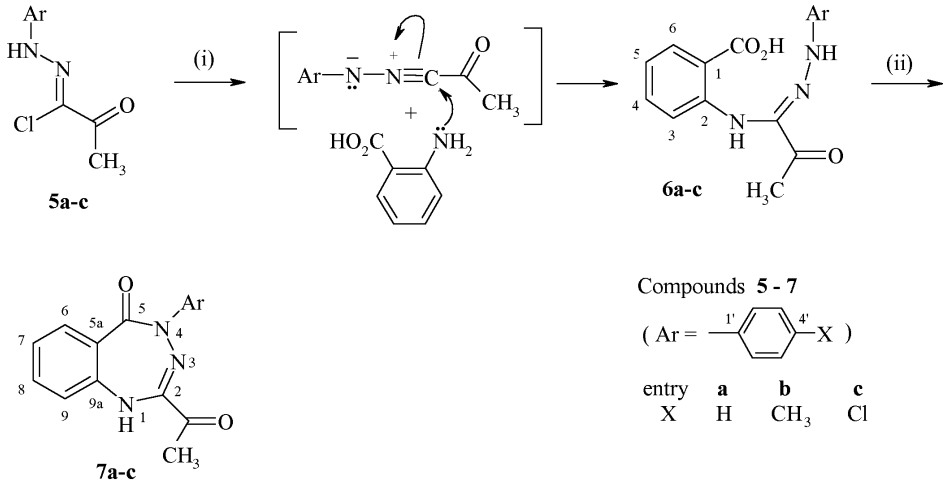
Results and Discussion.

Synthesis.

2-Aminobenzoic acid, acting as nitrogen nucleophile in basic medium, adds readily onto nitrile imines (the reactive 1,3-dipolar intermediates, generated *in situ* from their hydrazonyl chloride precursors **5a-c** in presence of triethylamine) to furnish the corresponding amidrazone adducts **6** (Scheme 1). This mode of nucleophilic addition reaction of various nucleophiles onto 1,3-dipoles is well-documented, and several amidrazone adducts, related to **6**, were obtained from the reaction of primary and secondary amines with hydrazonoyl chlorides (such as **5**) [10,11]. The required hydrazonoyl chlorides **5a** [12-14], **5b** [12,13] and **5c** [12-14] were previously characterized, and are made accessible *via* the Japp-Klingemann reaction [12,15,16] involving coupling of the appropriate arenediazonium chloride with 3-chloro-2,4-pentanedione in aqueous pyridine.

In a subsequent step, these acyclic adducts **6** in tetrahydrofuran in presence of the coupling reagent, 1,1'-carbonyldiimidazole, underwent smooth cyclocondensation involving the activated carboxyl group and the hydrazone-NH terminus to deliver the requisite benzotriazepinones **7** (Scheme 1).

Scheme 1



(i) aq. MeOH, THF, NEt_3 / 0 ^{0}C ⟶ rt, 2 - 3 hours
(ii) 1,1'-carbonyldiimidazole, THF / rt, 1 - 2 hours

Spectral Data.

The ir, ms, and nmr spectral data and microanalyses of the new compounds **6a-c** and **7a-c** conform to the suggested structures, and are given in the experimental section. Thus, their ms spectra display the correct molecular ions for which the measured high-resolution data are in good agreement with the calculated values. ^{1}H-signal assignments are straightforward, and ^{13}C-assignments follow from DEPT and 2D (COSY, HMQC and HMBC) experiments. Thus, long range correlation is observed between H-6 and each of C-9a and C-5 in HMBC experiments for compounds **7a-c**. Likewise, H-8 is correlated with C-5a, while H-3'/H-5' show correlation with C-1'.

In conclusion, a new synthetic route toward 1,3,4-benzotriazepin-5-ones, described in the present work, utilizes readily available and inexpensive reactants (anthranilic acid and hydrazonoyl chlorides). This versatile and efficient route involves two-step reactions that are conveniently conducted in a short time, at or below room temperature, to give unequivocally the desired benzotriazepinones in good overall yield and high purity. In essence, this viable method competes favorably with ambiguous reported methods which also suffered from inherent drawbacks (*vide supra*).

EXPERIMENTAL

Melting points (uncorrected) were determined on an electrothermal Mel-Temp. Apparatus. ^{1}H- and ^{13}C nmr spectra were measured on a Bruker DPX-300 instrument with TMS as internal reference. Electron-impact mass spectra were obtained using a Finnigan MAT TSQ-70 spectrometer at 70 eV; ion source temperature = 200 °C. Ir spectra were recorded as KBr discs on a Nicolet Impact-400 FT-IR spectrophotometer. Microanalyses were preformed at the Microanalytical Laboratory-Inorganic Chemistry Department, Tübingen University, Germany.

1-Arylhydrazono-1-chloropropanones (**5a-c**).

These hydrazonoyl chlorides were prepared *via* direct coupling of the appropriate arenediazonium chloride with 3-chloro-2,4-pentanedione in aqueous pyridine, following standard procedures [12,16].

2-[(2-Oxo-1-phenylhydrazonopropan-1-yl)amino]benzoic acid (**6a**).

A homogeneous solution of 2-aminobenzoic acid (1.65 g, 12 mmol) in aqueous methanol (70%, 20 ml) and triethylamine (3 ml) was added dropwise to a stirred and cooled (0 °C) solution of 1-phenylhydrazono-1-chloropropanone (**5a**) (2.16 g, 11 mmol) in tetrahydrofuran (30 ml). Additional triethylamine (3 ml) in tetrahydrofuran (5 ml) was then introduced dropwise into the reaction mixture, which was further stirred at 0 °C for 15-20 minutes, and then at room temperature for 2-3 hours. The organic solvents were then removed *in vacuo* from the reaction mixture, and the residual aqueous solution was directly acidified with glacial acetic acid (3 ml). The resulting crude solid product was collected, dried and recrystallized from hot chloroform/methanol (5:1 v/v) as fine yellow needles. Yield of **6a** = 82%; mp 210-211 °C; ir (potassium bromide): 3480 (OH), 3310 (NH), 3246 (NH), 1690 (CO), 1603, 1584, 1516, 1491, 1232 cm^{-1}; ms: m/z (% rel. int.): 297 (M^+, 100), 279 (8), 236 (18), 208 (4), 174 (18), 148 (49), 118 (60), 108 (87); hrms: Calcd for $C_{16}H_{15}N_3O_3$: 297.111316. Found: 297.108719; ^{1}H nmr (300 MHz, DMSO-d_6): δ 2.46 (s, 3H, COC*H*$_3$), 6.10 (dd, 1H, J = 8.3 Hz, 0.7 Hz, H-3), 6.78 (m, 1H, H-5), 6.89 (m, 1H, H-4'), 7.22-7.33 (m, 5H, H-2'/ H-6', H-3'/ H-5' and H-4), 7.88 (dd, 1H, J = 7.9 Hz, 1.6 Hz, H-6), 9.25 (s, 1H, C_2-N*H*), 10.02 (s, 1H, $C_{1'}$-N*H*), 13.10 (s, 1H, CO_2*H*); ^{13}C nmr (75 MHz, DMSO-d_6): δ 25.1 (CO*C*H$_3$), 114.2 (C-1), 114.7 (C-2'/C-6'), 115.8 (C-3), 118.5 (C-5), 121.9 (C-4'), 129.5 (C-3'/C-5'), 131.9 (C-6), 134.4 (C-4), 134.9 (NH-*C*=N-), 144.2 (C-1'), 145.7 (C-2), 170.5 (*C*O$_2$H), 193.3 (CH$_3$*C*=O).

Anal. Calcd. for $C_{16}H_{15}N_3O_3$ (297.32): C, 64.64; H, 5.09; N, 14.13. Found: C, 64.35; H, 5.03; N, 14.11.

2-{[2-Oxo-1-(4-methylphenylhydrazonopropan-1-yl)]amino}-benzoic Acid (**6b**).

This compound was prepared from 2-aminobenzoic acid (1.65 g, 12 mmol) and 1-chloro-1-(4'-methylphenylhydrazono)-propanone (**5b**) (2.30 g, 11 mmol) by following the same procedure and experimental conditions described above for obtaining **6a**. The product was recrystallized from chloroform/methanol as pale yellow prisms. Yield of **6b** = 86%; mp 214-215 °C; ir (potassium bromide): 3466 (OH), 3332 (NH), 3305 (NH), 1674 (CO), 1574, 1555, 1524, 1501, 1244 cm^{-1}; ms: m/z (% rel. int.): 311 (M^+, 84), 293 (13), 250 (7), 222 (4), 189 (26), 174 (13), 148 (26), 146 (28), 132 (100), 106 (74); hrms: Calcd for $C_{17}H_{17}N_3O_3$: 311.1270. Found: 311.1250; ^{1}H nmr (300 MHz, DMSO-d_6): δ 2.20 (s, 3H, $C_{4'}$-CH_3), 2.44 (s, 3H, COCH_3), 6.08 (d, 1H, J = 8.3 Hz, H-3), 6.77 (dd, 1H, J = 7.8 Hz, 7.5 Hz, H-5), 7.06 (d, 2H, J = 8.3 Hz, H-3'/H-5'), 7.22 (d, 2H, J = 8.3 Hz, H-2'/H-6'), 7.29 (m, 1H, H-4), 7.87 (dd, 1H, J = 7.8 Hz, 1.3 Hz, H-6), 9.21 (br s, 1H, C_2-N*H*), 9.97 (s, 1H, $C_{1'}$-N*H*), 13.09 (br s, 1H, CO_2*H*); ^{13}C nmr (75 MHz, DMSO-d_6): δ 20.8 ($C_{4'}$-*C*H$_3$), 25.0 (CO*C*H$_3$), 114.1 (C-1), 114.8 (C-2'/C-6'), 115.7 (C-3), 118.4 (C-5), 130.0 (C-3'/C-5'), 130.8 (C-6), 131.8 (C-4), 134.3 (C-4'), 134.4 (NH-*C*=N-), 141.9 (C-1'), 145.8 (C-2), 170.3 (*C*O$_2$H), 193.1 (CH$_3$-*C*=O).

Anal. Calcd. for $C_{17}H_{17}N_3O_3$ (311.34): C, 65.58; H, 5.50; N, 13.50. Found: C, 65.44; H, 5.52; N, 13.38.

2-{[2-Oxo-1-(4-chlorophenylhydrazonopropan-1-yl)]amino}-benzoic Acid (**6c**).

This compound was prepared from 2-aminobenzoic acid (1.65 g, 12 mmol) and 1-chloro-1-(4'-chlorophenylhydrazono)-propanone (**5c**) (2.31 g, 11 mmol) by following the same procedure and experimental conditions described above for obtaining **6a**. Recrystallization from chloroform/methanol produced light orange granules. Yield of **6c** = 88%; mp 234-235 °C; ir (potassium bromide): 3450 (OH), 3348 (NH), 3244 (NH), 1695 (CO), 1652, 1586, 1567, 1502, 1490, 1360, 1281, 1240, 1088, 1028 cm^{-1} ; ms: m/z (% rel. int.): 331 (M^+, 100), 313 (30), 270 (11), 235 (13), 174 (28), 148 (95), 126 (97), 125 (88); hrms: Calcd for $C_{16}H_{14}ClN_3O_3$: 331.0723. Found: 331.0692; ^{1}H nmr (300 MHz, DMSO-d_6): δ 2.46 (s, 3H, COCH_3), 6.10 (dd, 1H, J = 8.3 Hz, 0.7 Hz, H-3), 6.79 (ddd, 1H, J = 8.1 Hz, 7.2 Hz, 1.0 Hz, H-5), 7.27-7.34 (m, 5H, H-2'/ H-6', H-3' / H-5', H-4), 7.89 (dd, 1H, J = 7.2 Hz, 1.5 Hz, H-6), 9.29 (s, 1H, C_2-N*H*), 10.10 (s, 1H, $C_{1'}$-N*H*), 13.09 (br s, 1H, CO_2*H*); ^{13}C nmr (75 MHz, DMSO-d_6): δ 25.11 (CO*C*H$_3$), 114.3 (C-1), 115.9 (C-3), 116.2 (C-2'/C-6'), 118.6 (C-5), 125.4 (C-4'), 129.4 (C-3'/C-5'), 131.9 (C-6), 134.4 (C-4), 135.3 (NH-*C*=N-), 143.2 (C-1'), 145.4 (C-2), 170.3 (*C*O$_2$H), 193.4 (CH$_3$-*C*=O).

Anal. Calcd. for $C_{16}H_{14}ClN_3O_3$ (331.76): C, 57.93; H, 4.25; Cl, 10.69; N, 12.67. Found: C, 57.68; H, 4.15; Cl, 10.53; N, 12.58.

2-Acetyl-4-phenyl-1,4-dihydro-1*H*-1,3,4-benzotriazepin-5-one (**7a**).

To a mixture of 1,1'-carbonyldiimidazole (2.0 g, 12.5 mmol) and compound **6a** (3.0 g, 10 mmol) was added dry tetrahydrofuran (50 ml), and the resulting solution was stirred at room temperature for 1-2 hours. The solvent was then removed *in vacuo* and the residue was immediately treated with water (40 ml) and extracted with chloroform (2 x 40 ml). The combined organic extracts were dried (Na_2SO_4), the solvent chloroform was evaporated and the residual solid product was recrystallized from $CHCl_3$/petroleum ether (bp 40-60 °C) forming beige fine crystals. Yield of **7a** = 78 %; mp 152-153 °C; ir (potassium bromide): 3319 (NH), 1711 (CO), 1590, 1500, 1465, 1366, 1294, 1270, 1166, 1011 cm^{-1}; ms: m/z (% rel. int.): 279 (M^+, 100), 250 (10), 236 (44), 220 (10), 174 (9), 146 (22), 132 (64), 130 (17), 105 (24); hrms : Calcd for $C_{16}H_{13}N_3O_2$: 279.1008. Found : 279.0975; ^{1}H nmr (300 MHz, $CDCl_3$): δ 2.53 (s, 3H, COCH_3), 6.72 (dd, 2H, J = 8.5 Hz, 1.1 Hz, H-2'/ H-6'), 6.92 (m, 1H, H-4'), 7.16 (m, 2H, H-3'/ H-5'), 7.46 (br s, 1H, N_1-H), 7.43 (m, 1H, H-7), 7.74 (m, 2H, H-8 / H-9), 8.16 (ddd, 1H, J = 7.6 Hz, 1.9Hz, 1.0Hz, H-6); ^{13}C nmr (75 MHz, $CDCl_3$): δ 28.7 (CO*C*H$_3$), 114.5 (C-2'/C-6'), 122.4 (C-5a), 122.8 (C-4'), 127.0 (C-6), 128.1 (C-7), 128.2 (C-9), 129.4 (C-3'/C-5'), 135.2 (C-8), 145.7 (C-1'), 146.6 (C-9a), 153.7 (C-2), 160.6 (O=*C*$_5$-N), 194.2 (CH$_3$*C*=O).

Anal. Calcld. for $C_{16}H_{13}N_3O_2$ (279.3): C, 68.81; H, 4.69; N, 15.04. Found: C, 68.63; H, 4.55; N, 14.86.

2-Acetyl-4-(4-methylphenyl)-1,4-dihydro-1*H*-1,3,4-benzotriazepin-5-one (**7b**).

This compound was prepared from **6b** (3.1 g, 10 mmol) and 1,1'-carbonyldiimidazole (2.0 g, 12.5 mmol) by following the same procedure and experimental conditions described above for **7a**. The product was recrystallized from dichloromethane/*n*-hexane as yellow prisms. Yield of **7b** = 81%; mp 155-156 °C; ir (potassium bromide): 3268 (NH), 1705 (CO), 1626, 1606, 1523, 1510, 1344, 1286, 1234, 1130 cm^{-1}; ms: m/z (% rel. int.): 293 (M^+, 45), 279 (9), 250 (4), 189 (21), 167 (28), 149 (97), 132 (19), 119 (22), 91 (100); hrms: Calcd for $C_{17}H_{15}N_3O_2$: 293.1164. Found: 293.1117; ^{1}H nmr (300 MHz, $CDCl_3$): δ 2.38 (s, 3H, $C_{4'}$-CH_3), 2.43 (s, 3H, COCH_3), 6.69 (d, 1H, J = 7.9 Hz, H-8), 6.97 (dd, 1H, J = 7.4 Hz, 7.7 Hz, H-7), 7.23 (d, 2H, J = 8.3 Hz, H-3'/ H-5'), 7.36 (d, 2H, J =8.3 Hz, H-2'/ H-6'), 7.33 (m, 1H, H-9, overlapped with H-2'/ H-6'), 7.30 (br s, 1H, N_1-H), 7.96 (d, 1H, J = 7.7 Hz, H-6); ^{13}C nmr (75 MHz, $CDCl_3$): δ 21.2 (C4'-*C*H$_3$), 24.1 (CO*C*H$_3$), 118.3 (C-8), 121.9 (C-5a), 122.7 (C-7), 126.0 (C-2'/C-6'), 129.4 (C-3'/C-5'), 134.0 (C-6), 134.6 (C-9), 137.5 (C-4'), 141.5 (C-1'), 143.6 (C-9a), 145.8 (C-2), 165.6 (O=*C*$_5$-N), 194.0 (CH$_3$*C*=O).

Anal. Calcd. for $C_{17}H_{15}N_3O_2$(293.33): C, 69.61; H, 5.15; N, 14.33. Found: C, 69.37; H, 5.02; N, 14.15.

2-Aetyl-4-(4-chlorophenyl)-1,4-dihydro-1*H*-1,3,4-benzotriazepin-5-one (**7c**).

This compound was prepared from **6c** (3.3 g, 10 mmol) and 1,1'-carbonyldiimidazole (2.0 g, 12.5 mmol) by following the same procedure and experimental conditions described above for **7a**. Recrystallization from dichloromethane/*n*-hexane gave pale yellow granules. Yield of **7c** = 83 %; mp 181-182 °C; ir (potassium bromide): 3313 (NH), 1732 (CO), 1701 (CO), 1608, 1595, 1491, 1467, 1413, 1374, 1168, 1097 cm^{-1}; ms: m/z (% rel. int.): 313 (M^+, 94), 278 (3), 270 (21), 254 (10), 235 (22), 189 (10), 174 (13), 146 (25), 132 (58), 126 (84), 125 (100), 111 (63); hrms: Calcd for $C_{16}H_{12}ClN_3O_2$: 313.0618. Found: 313.0592; ^{1}H nmr (300 MHz, $CDCl_3$): δ 2.57 (s, 3H, COCH_3), 6.69 (m, 2H, H-2'/ H-6'), 7.16 (m, 2H, H-3'/ H-5'), 7.20 (br s, 1H, N_1-H), 7.54 (ddd, 1H, J = 8.0 Hz, 6.3 Hz, 1.5 Hz, H-7), 7.79 (m, 2H, H-8 / H-9), 8.22 (dd, 1H, J = 8.0 Hz, 1.5 Hz, H-6); ^{13}C nmr (75 MHz, $CDCl_3$): δ 28.6 (CO*C*H$_3$), 116.0 (C-2'/ C-6'), 122.3 (C-5a), 127.2 (C-6), 128.1 (C-4'), 128.3 (C-9), 128.4 (C-7), 129.4 (C-3'/ C-5'), 135.3 (C-8), 144.3 (C-1'), 146.4 (C-9a), 152.9 (C-2), 160.5 (O=*C*$_5$-N), 193.9 (CH$_3$*C*=O).

Anal. Calcd. for $C_{16}H_{12}ClN_3O_2$ (313.75): C, 61.25; H, 3.86; Cl, 11.30; N, 13.39. Found: C, 61.12; H, 3.81; Cl, 11.18; N, 13.35.

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