Synthesis of 4*H*-Tetrazolo[1,5-*a*][1]benzazepines from the Baylis-Hillman Adducts of 2-Azidobenzaldehyde

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Novel heterocycles, 4*H*-tetrazolo[1,5-*a*][1]benzazepines **6** were prepared by the intramolecular 1,3-dipolar cycloaddition reaction of azidophenylcyanomethyl compounds **5**. The latter were readily obtained from 2-azidobenzaldehyde through the Baylis-Hillman adducts **3** followed by acetylation to compounds **4** and nucleophilic substitution by cyanide to compounds **5**.

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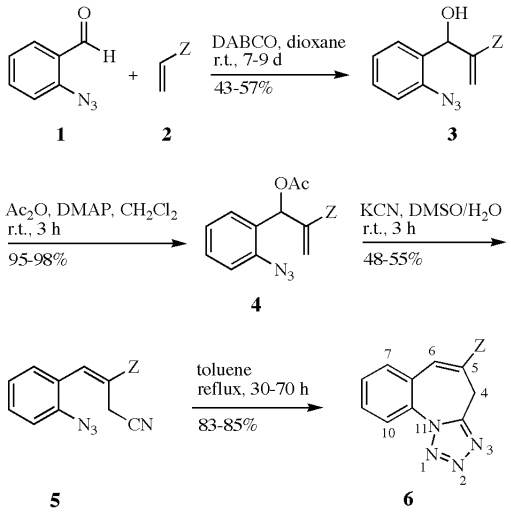
Tetrazole and tetrazole containing heterocycles are known to exhibit a wide range of biological activities such as analeptic [1], anti-inflammartory [1,2], antimicrobial [1,3], antilipemic [4] and antiallergic [1,2] properties. They can serve as metabolically stable surrogates for a carboxylic acid group [5], and as simple lipophilic spacers. The most common method for the preparation of tetrazoles is the 1,3-dipolar cycloaddition reaction of azides with nitriles [6].

The Baylis-Hillman reaction is one of the most powerful carbon-carbon bond forming reactions in organic chemistry [7]. Most Baylis-Hillman reactions involve the reaction of activated alkenes and carbonyl compounds to produce the Baylis-Hillman adducts *viz* allylic alcohol derivatives [7]. Besides the usefulness of these adducts themselves, further derivatization with various nucleophilic reagents toward synthetically useful compounds has been studied [8]. Among them, adducts have been applied to provide convenient access to benzannulated or other heterocyclic systems. These include indolizines [9], quinolines [10], chromenes [11], thiochromenes [12], indenes [13], pyridopyrimidones [14], 1,4-oxazepin-7-ones [15], coumarins [16], isobenzofuranones [17] and isoindoles [18]. We herein describe a facile synthesis of 4*H*-tetrazolo-[1,5-*a*][1]benzazepines *via* the treatment of the acetates of Baylis-Hillman adducts of 2-azidobenzaldehyde with cyanide followed by intramolecular 1,3-dipolar cycloaddition reaction.

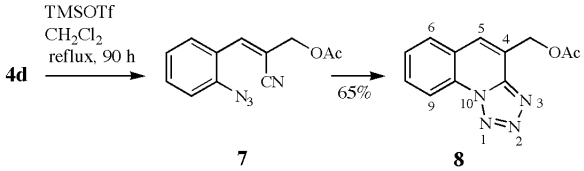
Scheme I



2, **3**, **4**, **5**, **6**	Z
a	CO_2Me
b	CO_2Et
c	COMe
d	CN

The reaction of 2-azidobenzaldehyde (**1**) with 3.0 molar equivalents of methyl acrylate in the presence of 0.1 molar equivalents of 1,4-diazabicyclo[2,2,2]octane (DABCO) in dioxane at room temperature afforded the adduct, methyl 3-(2-azidophenyl)-3-hydroxy-2-methylenepropanoate (**3a**) in 57% yield. Similarly, the treatment of **1** with ethyl acrylate, methyl vinyl ketone and acrylonitrile in the presence of DABCO gave the corresponding adducts **3b**-**d** in 43-53% yields. In the cases of acrolein, acrylamide, and phenyl vinyl sulfone, adducts were not produced. Generally, the reactions were very slow (7-9 days) at room temperature whereas at reflux temperature the conversions were unsuccessful. The acetates of adducts **4a**-**d** were produced by the reaction of **3a**-**d** with acetic anhydride in the presence of a catalytic amount of *N*,*N*-dimethylaminopyridine (DMAP) in dichloromethane at room temperature for 3 hours in excellent yields (95-98%). Nucleophilic substitution reaction of the acetates **4a**-**c** with potassium cyanide in aqueous dimethyl sulfoxide at room temperature for 3 hours occurred in a S_N2' fashion to give the required key intermediates azidophenylcyanomethyl com-

Scheme II



pounds **5a-c** in 100% (*E*)-selectivity as evidenced by the ^{1}H nmr spectral analysis in comparison with literature values [19,20] in moderate yields (48-55%). Unfortunately, **4d** did not undergo nucleophilic substitution at room or elevated temperature. The 1,3-dipolar cycloaddition of azidophenylcyanomethyl compounds **5a-c** in refluxing toluene produced excellent yields (85-88%) of the corresponding 4*H*-tetrazolo[1,5-*a*][1]benzazepines **6a-c** . The infrared spectra of **6a-c** showed the disappearance of absorption for azide (2124-2132 cm^{-1}) and cyano (2248-2260 cm^{-1}) bands. To our knowledge, this is the first example of the synthesis of the 4*H*-tetrazolo[1,5-*a*][1]benzazepine ring system.

With a view to study the application of Baylis-Hillman acetate **4d**, we finally examined the isomerization of **4d** with a catalytic amount of trimethylsilyl trifluoromethanesulfonate (TMSOTf) [20]. This reaction provided a simple and convenient direct synthesis of the 4-acetoxymethyltetrazolo[1,5-*a*]quinoline (**8**) *via* the intermediate 2-(acetoxymethyl)-3-(2-azidophenyl)-2-propenenitrile (**7**) followed by cycloaddition of the azido group to the neighboring nitrile by known method [21].

In summary, outlined here is a new route to 4*H*-tetrazolo[1,5-*a*][1]benzazepines by an intramolecular 1,3-dipolar cycloaddition of readily available azidophenylcyanomethyl compounds from the Baylis-Hillman adducts of 2-azidobenzaldehyde.

EXPERIMENTAL

All reagents and solvents were reagent grade or were purified by standard methods before use. Silica gel 60 (70-230 mesh ASTM) used for column chromatography was supplied by E. Merck. Analytical thin layer chromatography (tlc) was performed on silica gel with fluorescent indicator coated on aluminium sheets. Melting points were taken using an Electrothermal melting point apparatus and are uncorrected. Microanalyses were obtained using a Carlo Erba EA 1180 element analyzer. Mass spectra were obtained using a ThermoQuest Polaris Q mass spectrometer operating at 70 eV. Infrared spectra were recorded on a Nicolet Magna 550 FTIR spectrometer. The ^{1}H and ^{13}C nmr spectra were measured on a Gemini 300 spectrometer. All chemical shifts are reported in parts per million (δ) relative to tetramethylsilane.

The 2-azidobenzaldehyde (**1**) was prepared following the literature procedure [22].

Methyl 3-(2-Azidophenyl)-3-hydroxy-2-methylenepropanoate (**3a**).

To a stirred solution of 2-azidobenzaldehyde (**1**) (1.0 g, 6.80 mmoles) in dioxane (10 ml) was added methyl acrylate (**2a**) (1.84 ml, 20.40 mmoles) and DABCO (76 mg, 0.68 mmoles) at room temperature. After stirring at same temperature for 7 days the reaction mixture was diluted with water (10 ml) and extracted with dichloromethane (3 x 20 ml). The combined organic layers were dried over anhydrous magnesium sulfate and the solvent was evaporated *in vacuo*. The resulting mixture was chromatographed on silica gel eluting with hexane/ethyl acetate (7 :1) to afford 904 mg (57%) of **3a** as an oil; ir (neat): 3452, 2108, 1704, 1634, 1588 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 3.22 (d, 1H, J = 5.5 Hz), 3.76 (s, 3H), 5.68 (s, 1H), 5.82 (d, 1H, J = 5.5 Hz), 6.33 (s, 1H), 7.15-7.19 (m, 2H), 7.26-7.38 (m, 1H), 7.45-7.48 (m, 1H); ^{13}C nmr (deuteriochloroform): δ 52.0, 67.8, 117.9, 124.9, 126.4, 128.0, 129.0, 131.9, 137.2, 140.9, 166.8; ms: m/z (%) 233 (M$^+$, 4), 206 (26), 205 (39), 175 (39), 144 (100).

Anal. Calcd. for $C_{11}H_{11}N_3O_3$: C, 56.65; H, 4.75; N, 18.02. Found: C, 56.41; H, 4.57; N, 17.79.

Ethyl 3-(2-Azidophenyl)-3-hydroxy-2-methylenepropanoate (**3b**).

The procedure was the same as described above except using ethyl acrylate (**2b**) (2.21 ml, 20.40 mmoles) instead of methyl acrylate. Yield: 874 mg (52%); oil; ir (neat): 3436, 2124, 1712, 1627, 1580 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 1.27 (t, 3H, J = 7.0 Hz), 3.27 (d, 1H, J = 5.5 Hz), 4.21 (q, 2H, J = 7.0 Hz), 5.67 (s, 1H), 5.81 (d, 1H, J = 5.5 Hz), 6.33 (s, 1H), 7.14-7.19 (m, 2H), 7.32-7.38 (m, 1H), 7.45-7.48 (m, 1H); ^{13}C nmr (deuteriochloroform): δ 14.0, 61.0, 67.9, 117.9, 124.9, 126.1, 128.0, 129.1, 132.0, 137.2, 141.2, 166.4; ms: m/z (%) 247 (M$^+$, 6), 220 (12), 219 (57), 190 (38), 162 (13), 146 (40), 118 (100), 91 (33).

Anal. Calcd. for $C_{12}H_{13}N_3O_3$: C, 58.29; H, 5.30; N, 17.00. Found: C, 58.12; H, 5.11; N, 16.73.

3-[(2-Azidophenyl)hydroxymethyl]-3-buten-2-one (**3c**).

The procedure was the same as described in **3a** except using methyl vinyl ketone (**2c**) (1.70 ml, 20.40 mmoles) instead of methyl acrylate. Yield: 635 mg (43%); mp 54-56°; ir (neat): 3324, 2135, 1677 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 2.38 (s, 3H), 3.36 (d, 1H, J = 5.5 Hz), 5.80 (s, 1H), 5.84 (d, 1H, J = 5.5 Hz), 6.18 (s, 1H), 7.14-7.19 (m, 2H), 7.32-7.37 (m, 1H), 7.44-7.47 (m, 1H); ^{13}C nmr (deuteriochloroform): δ 26.4, 67.6, 118.0, 124.9, 127.1, 128.0, 128.9, 132.2, 137.1, 148.9, 200.4; ms: m/z (%) 189 (M$^+$-N$_2$, 41), 147 (100), 118 (88), 104 (95), 91 (36).

Anal. Calcd. for $C_{11}H_{11}N_3O_2$: C, 60.82; H, 5.10; N, 19.34. Found: C, 60.68; H, 4.80; N, 19.03.

3-(2-Azidophenyl)-3-hydroxy-2-methylenepropanenitrile (**3d**).

The procedure was the same as described in **3a** except using acrylonitrile (**2d**) (1.34 ml, 20.40 mmoles) instead of methyl acrylate and stirring for 9 days. Yield: 722 mg (53%); mp 67-69°; ir (potassium bromide): 3456, 2236, 2132, 1584 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 2.75 (d, 1H, J = 5.3 Hz), 5.55 (d, 1H, J = 5.3 Hz), 6.06 (d, 1H, J = 1.3 Hz), 6.09 (d, 1H, J = 1.3 Hz), 7.18-7.26 (m, 2H), 7.39-7.51 (m, 2H); ^{13}C nmr (deuteriochloroform): δ 69.2, 116.8, 118.2, 125.1, 125.3, 127.9, 129.9, 130.0, 130.6, 137.2; ms: m/z (%) 172 (M$^+$-N$_2$, 100), 143 (84), 117 (99), 89 (34).

Anal. Calcd. for $C_{10}H_8N_4O$: C, 59.99; H, 4.03; N, 27.99. Found: C, 59.76; H, 4.28; N, 27.71.

Methyl 3-Acetoxy-3-(2-azidophenyl)-2-methylenepropanoate (**4a**).

To a stirred solution of **3a** (933 mg, 4.0 mmoles) in dichloromethane (10 ml) was added acetic anhydride (0.57 ml, 6.0 mmoles) and *N*,*N*-dimethylaminopyridine (98 mg, 0.8 mmoles) at room temperature. After stirring at same temperature for 3 hours the reaction mixture was diluted with water (10 ml) and extracted with dichloromethane (3 x 10 ml). The combined organic layers were dried over anhydrous magnesium sulfate and the solvent was evaporated *in vacuo* to afford 1.08 g (98%) of **4a** as an oil; ir (neat): 2132, 1751, 1724, 1634, 1584 cm^{-1}; ^{1}H nmr

(deuteriochloroform): δ 2.11 (s, 3H), 3.74 (s, 3H), 5.65 (s, 1H), 6.44 (s, 1H), 6.91 (s, 1H), 7.11-7.19 (m, 2H), 7.30-7.40 (m, 2H); ^{13}C nmr (deuteriochloroform): δ 20.9, 52.0, 68.2, 118.3, 124.7, 127.4, 128.2, 128.7, 129.6, 138.0, 138.5, 165.3, 169.2; ms: m/z (%) 275 (M^+, 0.3), 248 (47), 247 (40), 205 (100), 188 (22), 146 (34), 145 (51).

Anal. Calcd. for $C_{13}H_{13}N_3O_4$: C, 56.72; H, 4.76; N, 15.27. Found: C, 56.50; H, 4.58; N, 15.17.

Ethyl 3-Acetoxy-3-(2-azidophenyl)-2-methylenepropanoate (**4b**).

The procedure was the same as described in **4a** using **3b** (989 mg, 4.0 mmoles). Yield: 1.12 g (97%); oil; ir (neat): 2128, 1751, 1720, 1638, 1584 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 1.24 (t, 3H, J = 7.0 Hz), 2.11 (s, 3H), 4.18 (q, 2H, J = 7.0 Hz), 5.62 (s, 1H), 6.43 (s, 1H), 6.92 (s, 1H), 7.11-7.19 (m, 2H), 7.30-7.40 (m, 2H); ^{13}C nmr (deuteriochloroform): δ 14.0, 20.9, 61.0, 68.2, 118.2, 124.7, 127.1, 128.4, 128.8, 129.7, 138.0, 138.8, 164.9, 169.2; ms: m/z (%) 261 (M^+-N_2, 23), 219 (100), 173 (18), 146 (58), 145 (67), 117 (10).

Anal. Calcd. for $C_{14}H_{15}N_3O_4$: C, 58.13; H, 5.23; N, 14.53. Found: C, 58.35; H, 5.02; N, 14.18.

3-[(2-Azidophenyl)acetoxymethyl]-3-buten-2-one (**4c**).

The procedure was the same as described in **4a** using **3c** (869 mg, 4.0 mmoles). Yield: 1.02 g (98%); oil; ir (neat): 2124, 1743, 1685 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 2.10 (s, 3H), 2.36 (s, 3H), 5.84 (s, 1H), 6.27 (s, 1H), 6.94 (s, 1H), 7.09-7.18 (m, 2H), 7.26-7.38 (m, 2H); ^{13}C nmr (deuteriochloroform): δ 21.0, 26.2, 67.8, 118.4, 124.7, 126.9, 128.1, 129.0, 129.5, 138.0, 146.4, 169.3, 197.1; ms: m/z (%) 231 (M^+-N_2, 12), 189 (50), 147 (11), 146 (100), 118 (10).

Anal. Calcd. for $C_{13}H_{13}N_3O_3$: C, 60.22; H, 5.05; N, 16.21. Found: C, 60.38; H, 4.81; N, 15.93.

3-Acetoxy-3-(2-azidophenyl)-2-methylenepropanenitrile (**4d**).

The procedure was the same as described in **4a** using **3d** (801 mg, 4.0 mmoles). Yield: 920 mg (95%); yellow solid after crystallization from hexane; mp 76-78°; ir (potassium bromide): 2229, 2135, 1755, 1584 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 2.18 (s, 3H), 6.06 (d, 1H, J = 1.1 Hz), 6.07 (d, 1H, J = 1.1 Hz), 6.59 (s, 1H), 7.18-7.25 (m, 2H), 7.40-7.45 (m, 1H), 7.51-7.54 (m, 1H); ^{13}C nmr (deuteriochloroform): δ 20.8, 69.3, 115.9, 118.2, 121.9, 125.2, 126.8, 127.5, 130.3, 132.8, 137.4, 169.0; ms: m/z (%) 214 (M^+-N_2, 2), 155 (43), 154 (100), 127 (29).

Anal. Calcd. for $C_{12}H_{10}N_4O_2$: C, 59.50; H, 4.16; N, 23.13. Found: C, 59.26; H, 3.89; N, 22.85.

Methyl 3-(2-Azidophenyl)-2-cyanomethyl-2-propenoate (**5a**).

To a stirred solution of **4a** (826 mg, 3.0 mmoles) in dimethyl sulfoxide (40 ml) and water (20 ml) was added potassium cyanide (293 mg, 4.5 mmoles) at room temperature. After stirring at same temperature for 3 hours the reaction mixture was diluted with water (20 ml) and extracted with dichloromethane (3 x 30 ml). The combined organic layers were dried over anhydrous magnesium sulfate and the solvent was evaporated *in vacuo*. The resulting mixture was chromatographed on silica gel eluting with hexane/ethyl acetate (7:1) to afford 363 mg (51%) of **5a** as a solid after crystallization from hexane; mp 67-69°; ir (potassium bromide): 2248, 2124, 1716, 1634, 1592 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 3.42 (s, 2H), 3.91 (s, 3H), 7.24-7.27 (m, 2H), 7.33-7.35 (m, 1H), 7.45-7.50 (m, 1H), 7.95 (s, 1H); ^{13}C nmr (deuteriochloroform): δ 17.2, 52.9, 117.1, 118.7, 123.3, 125.0, 125.2, 129.8, 131.1, 139.2, 139.7, 165.8; ms: m/z (%) 214 (M^+-N_2, 28), 199 (100), 155 (14), 128 (28).

Anal. Calcd. for $C_{12}H_{10}N_4O_2$: C, 59.50; H, 4.16; N, 23.13. Found: C, 59.21; H, 3.98; N, 22.89.

Ethyl 3-(2-Azidophenyl)-2-cyanomethyl-2-propenoate (**5b**).

The procedure was the same as described in **5a** using **4b** (868 mg, 3.0 mmoles). Yield: 370 mg (48%); mp 69-70°; ir (potassium bromide): 2260, 2132, 1712, 1642, 1596 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 1.40 (t, 3H, J = 7.0 Hz), 3.41 (s, 2H), 4.36 (q, 2H, J = 7.0 Hz), 7.24-7.26 (m, 2H), 7.32-7.35 (m, 1H), 7.44-7.50 (m, 1H), 7.94 (s, 1H); ^{13}C nmr (deuteriochloroform): δ 14.2, 17.2, 61.9, 117.2, 118.6, 123.6, 124.9, 125.3, 129.8, 131.0, 139.1, 139.4, 165.2; ms: m/z (%) 228 (M^+-N_2, 15), 199 (100), 155 (8), 128 (17).

Anal. Calcd. for $C_{13}H_{12}N_4O_2$: C, 60.93; H, 4.72; N, 21.86. Found: C, 60.80; H, 4.52; N, 21.63.

4-(2-Azidophenyl)-3-cyanomethyl-3-buten-2-one (**5c**).

The procedure was the same as described in **5a** using **4c** (778 mg, 3.0 mmoles). Yield: 373 mg (55%); mp 96-98°; ir (potassium bromide): 2256, 2128, 1673, 1634, 1592 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 2.54 (s, 3H), 3.39 (s, 2H), 7.27-7.29 (m, 2H), 7.40-7.42 (m, 1H), 7.48-7.53 (m, 1H), 7.81 (s, 1H); ^{13}C nmr (deuteriochloroform): δ 15.6, 25.3, 117.3, 118.6, 125.2, 129.9, 131.4, 132.4, 139.1, 139.7, 139.9, 196.8; ms: m/z (%) 198 (M^+-N_2, 86), 183 (100), 158 (42), 156 (46), 128 (52).

Anal. Calcd. for $C_{12}H_{10}N_4O$: C, 63.71; H, 4.46; N, 24.77. Found: C, 63.90; H, 4.58; N, 24.51.

5-Carbomethoxy-4*H*-tetrazolo[1,5-*a*][1]benzazepine (**6a**).

A stirred solution of **5a** (242 mg, 1.0 mmoles) in 20 ml of toluene was heated at reflux temperature for 30 hours and the solvent was evaporated *in vacuo*. The residue was crystallized from hexane/diethyl ether to give 201 mg (83%) of **6a** as a solid; mp 137-139°; ir (potassium bromide): 1708, 1631, 1603 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 3.89 (s, 3H), 4.12 (s, 2H), 7.57-7.69 (m, 3H), 7.83 (s, 1H), 8.11 (d, 1H, J = 7.9 Hz); ^{13}C nmr (deuteriochloroform): δ 21.2, 52.9, 123.5, 125.9, 128.8, 129.3, 131.2, 132.2, 137.8, 137.9, 153.3, 165.1; ms: m/z (%) 242 (M^+, <1), 214 (M^+-N_2, 30), 199 (100), 155 (9), 128 (24), 101 (6).

Anal. Calcd. for $C_{12}H_{10}N_4O_2$: C, 59.50; H, 4.16; N, 23.13. Found: C, 59.29; H, 4.02; N, 23.03.

5-Carbethoxy-4*H*-tetrazolo[1,5-*a*][1]benzazepine (**6b**).

The procedure was the same as described in **6a** using **5b** (256 mg, 1.0 mmoles). Yield: 218 mg (85%); mp 166-168°; ir (potassium bromide): 1700, 1638, 1603 cm^{-1}; ^{1}H nmr (deuteriochloroform): δ 1.39 (t, 3H, J = 7.0 Hz), 4.12 (s, 2H), 4.34 (q, 2H, J = 7.0 Hz), 7.27-7.67 (m, 3H), 7.82 (s, 1H), 8.11 (d, 1H, J = 7.9 Hz); ^{13}C nmr (deuteriochloroform): δ 14.2, 21.2, 62.1, 123.5, 126.0, 129.2, 129.3, 131.1, 132.2, 137.5, 137.6, 153.4, 164.7; ms: m/z (%) 256 (M^+, <1), 228 (M^+-N_2, 16), 199 (100), 155 (10), 128 (19).

Anal. Calcd. for $C_{13}H_{12}N_4O_2$: C, 60.93; H, 4.72; N, 21.86. Found: C, 60.68; H, 4.51; N, 21.53.

5-Acetyl-4*H*-tetrazolo[1,5-*a*][1]benzazepine (**6c**).

The procedure was the same as described in **6a** using **5c** (226 mg, 1.0 mmoles) except reaction time (70 hours). Yield: 192 mg

(85%); mp 151-152°; ir (potassium bromide): 1669, 1631 cm^{-1}; ^1H nmr (deuteriochloroform): δ 2.53 (s, 3H), 4.08 (s, 2H), 7.59-7.71 (m, 4H), 8.13 (d, 1H, J = 7.9 Hz); ^{13}C nmr (deuteriochloroform): δ 19.3, 25.4, 123.6, 126.0, 129.3, 131.4, 132.0, 132.3, 137.6, 137.7, 153.5, 195.0; ms: m/z (%) 198 (M^+-N_2, 100), 183 (13), 155 (84), 128 (37).

Anal. Calcd. for $C_{12}H_{10}N_4O$: C, 63.71; H, 4.46; N, 24.77. Found: C, 63.50; H, 4.19; N, 24.52.

4-Acetoxymethyltetrazolo[1,5-*a*]quinoline (**8**).

To a stirred solution of **4d** (242 mg, 1.0 mmoles) in dichloromethane (5 ml), was added trimethylsilyl trifluoromethanesulfonate (222 mg, 1.0 mmoles) at room temperature. After 90 hours at reflux temperature, the reaction mixture was diluted with water (5 ml) and extracted with dichloromethane (3 x 10 ml). The combined organic layer was dried ($MgSO_4$), the solvent was evaporated and the crude thus obtained was crystallized from hexane/diethyl ether to provide 157 mg (65%) of **8** as a pure solid; mp 186-187°; ir (potassium bromide): 1740, 1617, 1535 cm^{-1}; ^1H nmr (deuteriochloroform): δ 2.20 (s, 3H), 5.64 (s, 2H), 7.71-7.77 (m, 1H), 7.87-7.93 (m, 1H), 7.97(s, 1H), 7.99 (d, 1H, J = 8.0 Hz), 8.70 (d, 1H, J = 8.0 Hz); ^{13}C nmr (deuteriochloroform): δ 20.9, 61.1, 116.8, 121.5, 123.7, 128.2, 129.1, 130.3, 131.2, 131.5, 146.6, 170.6; ms: m/z (%) 242 (M^+, <1), 216 (82), 173 (100), 156 (38), 128 (48).

Anal. Calcd. for $C_{12}H_{10}N_4O_2$: C, 59.50; H, 4.16; N, 23.13. Found: C, 59.27; H, 3.89; N, 22.85.

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