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Synthesis of a Tritium Labeled Tetrafluoro-Substituted Aryl Azide Photoaffinity Labeling Agent for Chloride Channels. Application of [^{3}H]-Sodium Borohydride-Cobalt Chloride to Tritium Labeling.

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SUMMARY

5-Nitro-2-[N-3-(4-azido-2,3,5,6-tetrafluorophenyl)-propylamino]-benzoic acid (FAzNPPB), a photoaffinity analog of the potent epithelial chloride channel blocker 5-nitro-2-(3-phenylpropylamino)-benzoic acid (NPPB) has been prepared in five steps from commercially available 4-amino-2,3,5,6-tetrafluorobenzonitrile. The main feature of this synthesis was the use of $NaBH_4$-$CoCl_2$ to convert an aryl-substituted alkenyl nitrile precursor to the corresponding alkyl amine. The feasibility of this approach and the stoichiometry were developed by model work with cinnamonitrile. Using sodium borotritide-cobalt chloride, [^{3}H]-FAzNPPB (specific activity 13.9 mCi/mmol, radiochemical purity >99%) was prepared in three steps from (E)-4-amino-2,3,5,6-tetrafluoro-cinnamonitrile.

Key Words: [^{3}H]-Sodium borohydride, cobalt chloride, azide, photoaffinity, 5-nitro-2-(3-phenylpropylamino)-benzoic acid (NPPB).

INTRODUCTION

AzNPPB (5-nitro-2-[N-3-(4-azidophenyl)-propylamino]-benzoic acid, Figure 1), a photoaffinity analog of the potent epithelial chloride channel blocker 5-nitro-2-(3-phenyl-propylamino)-benzoic acid NPPB (Figure 1), has been synthesized (1). Preliminary characterization of AzNPPB with human erythrocyte ghost membranes suggested (1) that the aryl azide should be a useful structural probe for chloride channels in a variety of cells in which NPPB is an effective blocker (2). However, before attempting to characterize the membrane site(s) of AzNPPB incorporation, we prepared compound I (FAzNPPB, Figure 1),

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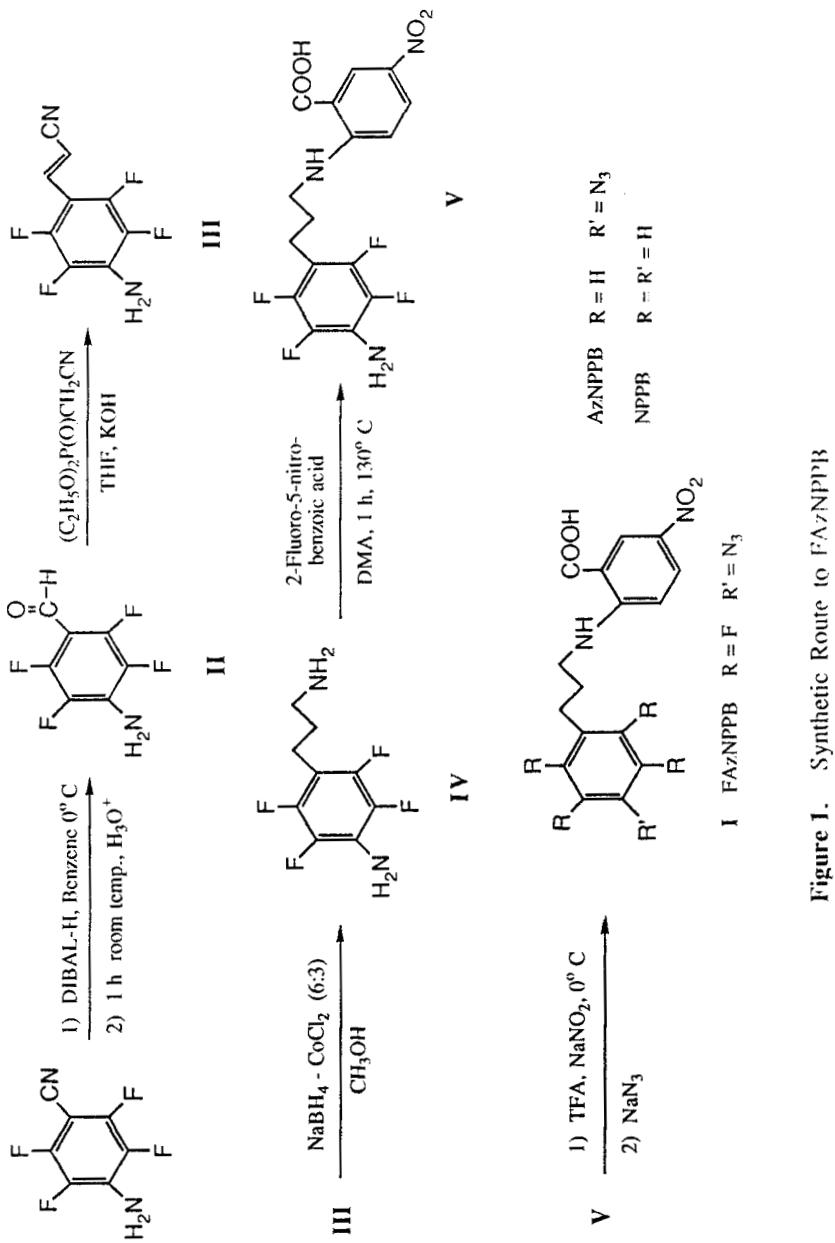

Figure 1. Synthetic Route to FAzNPPB

a tetrafluoro-substituted aryl azide analog of AzNPPB. Our rationale was based on reports by Platz and coworkers (3) that photolysis of polyfluorinated aryl azides often results in more efficient carbon-hydrogen insertion chemistry than is observed with the corresponding aryl azides. Indeed, preliminary evaluation of FAzNPPB with ghost membranes revealed that the tetrafluoro compound is a much more efficient

photoinactivating agent than AzNPPB. For example, the concentrations required to reduce chloride efflux rates by 50% for FAzNPPB and AzNPPB were 50 μM and 150 μM, respectively.

A synthetic route to FAzNPPB (Figure 1) was developed with the eventual aim of preparing tritium-labeled analogs for subsequent membrane characterization experiments. The main feature of this synthesis is the extension of appropriate $NaBH_4$-$CoCl_2$ chemistry (4-6) to the preparation of substituted alkyl amines from the corresponding alkenyl nitriles. This enabled the preparation of FAzNPPB and the corresponding model deuterium- and tritium-containing compounds.

RESULTS AND DISCUSSION

The synthetic scheme used to prepare unlabeled and labeled FAzNPPB is shown in Figure 1. Commercially available 4-amino-2,3,5,6-tetrafluorobenzonitrile was converted into unlabeled compound **I** in five steps in 7% overall isolated chemical yield. It was not necessary to protect the primary aromatic amino group which was converted into the azido functionality of **I** in the final step (Figure 1). The FAzNPPB structure was confirmed by ^{1}H- and ^{19}F-NMR, IR, UV and high resolution MS.

Prior to undertaking the radiochemical synthesis, a model synthesis was carried out using $NaBD_4$. Accordingly, [^{2}H]-FAzNPPB was prepared with partial ^{2}H substitution in all positions of the propyl moiety. Using ^{1}H-NMR, it was determined that approximately 2.4 D atoms/molecule were incorporated and by mass spectral analysis of [^{2}H]-compound **V** the distribution was 6.0/13.4/29.0/33.3/18.2/1.3/0 ($d_0/d_1/d_2/d_3/d_4/d_5/d_6$). A sample of [^{3}H]-FAzNPPB (specific activity 13.9 mCi/mmol, radiochemical purity >99%) was prepared in three steps (Figure 1) beginning with compound **III** and $NaBT_4$ (specific activity 40 mCi/mmol).

In our original synthesis of unlabeled FAzNPPB, we converted the aryl-substituted alkenyl nitrile **III** to alkylamine **IV** (as the HCl salt) in moderate yield by catalytic hydrogenation in alcohol with 3 equivalents of EtOH-HCl (data not shown). Though it would have been possible to prepare [^{3}H]-FAzNPPB by carrying out this reduction with $^3H_2$ gas, we sought an alternative reagent that would avoid special equipment requirements. Satoh had reported (4) that a 1:10:2 mol ratio of substrate/sodium borohydride/cobalt

chloride in alcohol could be used to reduce several nitriles to the corresponding amines. Battersby and coworkers employed this method (4) using borotritide-cobalt to reduce indolylacetonitrile and *p*-benzyloxybenzyl cyanide to the corresponding [^{3}H]-amines (7); however, no experimental details were provided. Additionally, Chung used a 1:2:1 mol ratio of substrate/borohydride/cobalt chloride to convert several alkenes to the corresponding alkanes in high yield (5). Since the cobalt-borohydride reducing agent had been conveniently employed using simple solution chemistry to reduce alkenes or nitriles, we determined the stoichiometry to simultaneously reduce both functionalities of III. Model work with cinnamonitrile, a simple aryl substituted alkenyl nitrile, demonstrated that a substrate/$NaBH_4$/$CoCl_2$ ratio of 1/4/2 was required for complete reduction to hydrocinnamylamine (Table I). With a lower substrate/borohydride/metal salt ratio (1/2/1), a small amount of the alkyl amine was produced and unreacted starting material remained. Additional products were isolated which indicated that only the alkene (hydrocinnamonitrile) or only the nitrile (cinnamylamine) had been reduced (Table I). With

Table I. Reduction[a] of Alkenyl Nitriles by $NaBH_4$-$CoCl_2$

Substrate	Mol Ratios Substrate/$NaBH_4$/$CoCl_2$	Product	(% yield)[b]
1. Cinnamonitrile	1/2/1	Hydrocinnamonitrile	(44)
		Cinnamylamine	(15)
		Hydrocinnamylamine	(11)
		Cinnamonitrile	(30)
2. Cinnamonitrile	1/4/2	Cinnamylamine	(99)
		Cinnamonitrile	(1)
3. Compound III	1/4/2	Compound IV	(52)
		4'-Amino-2',3',5',6'-tetrafluoro-hydrocinnamonitrile	(26)
		4'-Amino-2',3',5',6'-tetrafluoro-cinnamylamine	(21)
		Compound I	(1)
4. Compound III	1/6/3	Compound IV	(99)
		Compound III	(1)

[a]All reductions were carried out according to the general procedure described in the Experimental Section for compound III.
[b]Yields are based on ^{1}H-NMR analysis and are expressed as per cent of total isolated products.

compound **III**, it was necessary to increase the substrate/borohydride/metal salt ratio to 1/6/3 (Table I), since a 1/4/2 ratio was not sufficient to completely reduce both functional groups (Table I). The increased amount of metal-borohydride reagent was necessary because the reagent reacted with the aromatic amino group of **III** thereby lowering the effective reducing equivalent. The hydrolytic workup restored the amino functionality, so it is not necessary to protect it.

Using low specific activity sodium borotritide-cobalt chloride to convert **III** to [^{3}H]-compound **IV**, we prepared [^{3}H]-FAzNPPB in two additional steps using simple solution chemistry throughout. The specific activity of compound I was 58% of that predicted from the model deuterium synthesis. According to a mechanistic study of $CoCl_2$-$NaBH_4$ reactions (6), it is likely that the reagents react to form Co_2B which then catalyzes hydride transfer from residual $NaBH_4$. The lower than expected [^{3}H]-incorporation we observed may have resulted from the addition of an inhomogeneous solid mixture of borotritide:borohydride (1:6) and subsequent disproportionally higher loss of $NaBT_4$ during the formation of Co_2B. This problem might be alleviated by adding the diluted borotritide as a methanolic solution.

It should be possible to use the cobalt-borotritide reagent to prepare high specific activity tritium-labeled substituted alkyl amines from alkenyl nitriles simply by using readily available high specific activity [^{3}H]-$NaBH_4$. For substituted alkenyl amines without interfering substituents or with appropriately protected ones, a 4-fold molar excess of [^{3}H]-$NaBH_4$ would be required. This stoichiometry is not optimal and could be problematic for some applications. However, in view of the proposed mechanism (6) for the reduction, it may be possible to significantly lower the required molar excess of [^{3}H]-$NaBH_4$ by adding borotritide after Co_2B has been prepared *in situ* from unlabeled borohydride and $CoCl_2$. Whether or not this proves to be feasible, our results document the usefulness of the cobalt-borotritide reagent in radiochemical synthesis.

EXPERIMENTAL

Materials and Methods

Cinnamonitrile, sodium borohydride, sodium borodeuteride, cobalt (II) chloride hexahydrate, 4-amino-2,3,5,6-tetrafluorobenzonitrile, diisobutylaluminum hydride (DIBAL-H, 1.5 M in toluene), diethyl cyanomethylphosphonate, potassium hydroxide (semiconductor grade), N,N-dimethyl acetamide (DMA), and trifluoroacetic acid were purchased from Aldrich Chemical Co. [^{3}H]- Sodium borohydride (500 mCi/mmol, 93.8%

radiochemical purity) was obtained from Amersham. 2-Fluoro-5-nitrobenzoic acid was prepared by a literature procedure (8).

Infrared and UV-visible spectra were taken with Perkin-Elmer Model 1600 FTIR and Lambda 4A spectrometers, respectively. ^{1}H- and ^{19}F-NMR spectra were measured with a Bruker AC-250 instrument, and values are reported relative to TMS or C_6F_6 (-162.9 δ). High resolution mass spectra were obtained with a VG 70/250S at Pfizer Central Research. Hplc analyses were performed with an LDC/Milton Roy CM4000 system using a Rainin Dynamax-60A C18 reverse-phase column (10 x 250 mm) and mobile phase containing 92% methanol and 8% aqueous acetic acid (2%) at a flow rate of 3.0 mL/min. Chromatography was monitored at 240 nm and 366 nm. For radiochemical purity determinations, fractions were collected for liquid scintillation counting. Radioactivity was measured on samples diluted with OptiPhase "Hi Safe" II (LKB) using an LKB-Wallac model 1214 liquid scintillation counter. Melting points (uncorrected) were obtained with a Mel-Temp apparatus.

Synthesis

4-Amino-2,3,5,6-tetrafluorobenzaldhyde (II). To a solution of 4-amino-2,3,5,6-tetrafluorobenzonitrile (2.85 g, 15.0 mmol) in benzene (190 mL) under N_2 at 0°C, DIBAL-H (1.5 M in toluene, 30 mL, 45.0 mmol) was added dropwise with stirring over 10 min. The cooling bath was removed and the mixture was allowed to warm to room temperature. After 1 h, the mixture was cooled to 0°C and methanol (14 mL) was added. Aqueous H_2SO_4 (10%) was added dropwise until the aqueous layer remained acidic to litmus and stirring was continued for 30 min without cooling. The mixture was extracted with ether (3 x 50 mL) and the combined organic layers were washed with sat'd $NaHCO_3$ (2 x 50 mL), sat'd NaCl (2 x 50 mL), and water (50 mL) and dried over anhydrous Na_2SO_4. Solvents were removed by flash evaporation and crude aldehyde was obtained as a yellow solid (2.226 g, 11.7 mmol, 78%) with mp 103-105°C. After sublimation, pure white product was obtained (74% recovery) with mp 110.5-112.5°C, lit. (9) mp 110-111°C. IR (KBr, cm^{-1}) 3437, 3340, 3229, 2912 and 1650; ^{1}H-NMR ($CDCl_3$) δ 10.11 (m, 1H, ArCHO) and 4.71 (br s, ~2H, $ArNH_2$); and ^{19}F-NMR ($CDCl_3$) δ -163.19 (m, 2F, ArF *ortho* to NH_2) and -148.18 (m, 2F, ArF *ortho* to CHO). HRMS (EI) calcd. for $C_7H_3F_4NO$ 193.0151; found 193.0151.

(E)-4-Amino-2,3,5,6-tetrafluorocinnamonitrile (III). A solution of II (1.0 g, 5.2 mmol) and diethyl cyanomethylphosphonate (0.86 mL, 5.2 mmol) in THF (13 mL) was added to a suspension of KOH (0.58 g, 10.4 mmol) in THF (18 mL) and stirred under N_2 for 0.5 h. The mixture was filtered and, after removal of the solvent by flash evaporation, a yellow solid was obtained. Water (10 mL) was added to the solid and the mixture was extracted with chloroform (5 x 10 mL). The pooled organic layers were dried over anhydrous Na_2SO_4, and a moist yellow solid was obtained after flash evaporation of the solvent. Recrystallization from ethyl acetate--cyclohexane afforded white product (0.66 g, 3.1 mmol, 59%) with mp = 159-160°C. Pure product also could be obtained by flash chromatography (silica gel,

hexanes:acetone 8:2, v/v). IR (KBr, cm^{-1}) 3469, 3346, 3219, 2225, 1676 and 968; ^{1}H-NMR ($CDCl_3$) δ 7.33 (d, 1H, J = 17.0 Hz, CH α to Ar), 6.07 (d, 1H, J = 17.0 Hz, CH α to CN) and 4.42 (br s, ~2H, $ArNH_2$); ^{19}F-NMR ($CDCl_3$) δ –163.60 (m, 2F, ArF *ortho* to NH_2) and -144.13 (m, 2F, ArF *meta* to NH_2). HRMS (EI), calcd. for $C_9H_4F_4N_2$ 216.0311; found 216.0312.

3-(4-Amino-2,3,5,6-tetrafluorophenyl)-1-aminopropane (IV). To a stirred solution of III (108 mg, 0.50 mmol) in methanol under N_2 at 0°C was added $CoCl_2 \cdot 6H_2O$ (357 mg, 1.5 mmol). Sodium borohydride (114 mg, 3.0 mmol) was added in small portions over 0.5 h. The purple solution turned black and a black precipitate formed as gas (H_2) evolved. Methanol (2 mL) was added, the mixture was allowed to warm to room temperature, and stirring was continued under N_2 for 20 h. The black precipitate was dissolved by addition of 3 M HCl (3 mL) and the resulting red solution was flash evaporated to remove methanol. The resulting purple solution was extracted with ether (3 x 5 mL) and the organic extracts were discarded. The pH of the purple aqueous layer was adjusted to 10 (litmus) with concentrated NH_4OH and the resulting reddish-brown solution was extracted with ether (5 x 10 mL). The combined organic layers were washed with sat'd NaCl, dried over anhydrous Na_2SO_4, and flash evaporated to yield an orange crystalline solid (78 mg, 0.351 mmol, 70%). After sublimation, a white crystalline solid was obtained (68% recovery) with mp 77.5-78.5°C. IR (KBr, cm^{-1}) 3472, 3334, 3106, 2922 and 1313; UV (CH_3OH) λ_{max} 231 nm (ε = 18,216); ^{1}H-NMR ($CDCl_3$) δ 3.84 (br s, ~2H, $ArNH_2$), 2.66 (m, 4H, CH_2 α to Ar and CH_2 α to NH_2), 1.68 (dt, 2H, CH_2 β to NH_2) and 1.51 (s, ~2H, CH_2NH_2); ^{19}F-NMR ($CDCl_3$) δ -163.53 (m, 2F, ArF *ortho* to NH_2) and -148.72 (m, 2F, ArF *meta* to NH_2); and hplc R_T = 4.6 min. HRMS (FAB) MH^+ calcd. for $C_9H_{11}F_4N_2$ 223.0859; found 223.0858.

[^{2}H]-Compound IV. Using the procedure described above substituting sodium borodeuteride for sodium borohydride [^{2}H]-compound **IV** was obtained in 78% yield as a pale yellow crystalline solid with mp 165-170°C. This material was used without further purification.

[^{3}H]-Compound IV. Using the procedure described above for protio **IV**, 1.0 mmol of III was reacted with 3.0 mmol $CoCl_2 \cdot 6H_2O$ and 6.1 mmol [^{3}H]-$NaBH_4$ (244 mCi, specific activity 40 mCi/mmol). [^{3}H]-Compound **IV** was obtained in 49% chemical yield as an off-white solid [7.8 mCi, specific activity 15.9 mCi/mmol, radiochemical purity 84%, radiochemical yield 3% (based on total amount of borotritide)].

5-Nitro-2-[N-3-(4-amino-2,3,5,6-tetrafluorophenyl)-propylamino]-benzoic Acid (V). Compound **IV** (45 mg, 0.20 mmol) and 2-fluoro-5-nitrobenzoic acid (41 mg, 0.22 mmol) were dissolved in DMA (2 mL) with stirring under N_2. The mixture was heated at 130°C for 1.5 h and then poured over ice (10 g). The resulting suspension was extracted with EtOAc (4 x 15 mL) and the combined organic layers were dried over anhydrous Na_2SO_4 and flash evaporated. A yellow oil was obtained which crystallized after drying overnight *in vacuo* yielding a yellow solid product (76 mg, 0.196 mmol, 99%) with mp 250-255°C (dec.).

Occasionally, it was necessary to use flash chromatography (silica gel, hexanes:acetone, 1:1 v/v) to obtain pure product. IR (KBr, cm^{-1}) 3494, 3346, 3286, 2962, 3000-2500 (br), 1672, 1523 and 1320; UV (CH_3OH) λ_{max} 228 nm (ε = 21,220) and 380 nm (ε = 13,160); ^{1}H-NMR (acetone-d_6) δ 8.95 (s, 1H, COOH), 8.83 (d, 1H, J = 2.6 Hz, ArH *ortho* to COOH), 8.20 (dd, 1H, J = 2.6 and 9.5 Hz, ArH *para* to COOH), 6.94 (d, 1H, J = 9.5 Hz, ArH *meta* to COOH), 5.20 (br s, ~2H, NH_2), 3.52 (m, 2H, CH_2 α to NH), 2.80 (m, 2H, CH_2 γ to NH) and 1.95 (m, 2H, CH_2 β to NH); ^{19}F-NMR (acetone-d_6) δ -162.18 (m, 2F, ArF *ortho* to NH_2) and -147.71 (m, 2F, ArF *meta* to NH_2); and hplc R_T = 5.7 min. HRMS (EI) calcd. for $C_{16}H_{13}F_4N_3O_4$ 387.0843; found 387.0842.

[^{2}H]-Compound V. Using the procedure described above substituting [^{2}H]-compound IV for the protio compound, [^{2}H]-compound V was obtained in 60% yield after flash chromatography. Incorporation of ^{2}H was 83% based on ^{1}H-NMR integration data (assuming theoretical maximum incorporation to be 3.0). ^{1}H-NMR (DMSO-d_6 and acetone d_6) 3.50 (br m, 0.8H, CH_2 α to NH), 2.68 (br m, 1.4H, CH_2 γ to NH), and 1.88 (br m, 1.3 H CH_2 β to NH).

[^{3}H]-Compound V. Using the procedure described above for protio V, 0.45 mmol of [^{3}H]-compound IV (7.15 mCi, specific activity 15.9 mCi/mmol) was converted in 99% chemical yield into [^{3}H]-compound V which was obtained as an orange solid (5.32 mCi, specific activity 14.6 mCi/mmol, radiochemical purity 86%, radiochemical yield 74%).

5-Nitro-2-[N-3-(4-azido-2,3,5,6-tetrafluorophenyl)-propylamino]benzoic Acid (I). All procedures were carried out in the dark. A solution of V (126 mg, 0.325 mmol) in TFA (2.0 mL) was stirred under N_2 with cooling (ice/salt bath). Sodium nitrite (56 mg, 0.81 mmol) was added and stirring was continued with cooling for 40 min. Sodium azide (528 mg, 8.1 mmol) was added, the mixture was layered with ether (2.5 mL), and stirring was continued with cooling for 1 h. The cooling bath was removed and after 0.5 h water (4.0 mL) was added. The mixture was transferred to a separatory funnel and extracted with ether (3 x 5 mL). The combined organic layers were washed with sat'd NaCl (15 mL), dried over anhydrous Na_2SO_4, and flash evaporated to remove the ether. Hexanes (1 mL) were added to the residue and the resulting solid was collected by filtration, washed with hexanes, and dried *in vacuo*. Pure product was obtained as a bright yellow solid (61 mg, 0.15 mmol, 46%) with mp = 149-151°C (dec.). IR (KBr, cm^{-1}) 3340, 3000-2500 (br), 2150, 2120, 1672, 1533 and 1329; UV (CH_3OH) λ_{max} 248 nm (ε = 19,700) and 368 nm (ε = 18,820); ^{1}H-NMR (DMSO-d_6) δ 8.78 (br s, 1H, COOH), 8.62 (d, 1H, J = 2.6 Hz, ArH *ortho* to COOH), 8.16 (dd, 1H, J = 2.7 Hz and 9.5 Hz, ArH *para* to COOH), 6.90 (d, 1H, J = 9.5 Hz, ArH *meta* to COOH), 3.39 (m, 2H, CH_2 α to NH), 2.78 (t, 2H, CH_2 γ to NH) and 1.90 (m, 2H, β to NH); ^{19}F-NMR (DMSO-d_6) δ -145.11 (m, 2F, ArF *meta* to N_3) and -153.21 (m, 2F, ArF *ortho* to N_3); and hplc R_T = 8.3 min. HRMS (EI) calcd. for $C_{16}H_{11}F_4N_5O_4$ 413.0748; found 413.0747.

[^{2}H]-Compound I. Using the procedure described above substituting [^{2}H]-compound V for the protio material, [^{2}H]-compound I was obtained in 46% yield. Incorporation of deuterium was 80% based on ^{1}H-NMR integration data (assuming theoretical maximum

incorporation to be 3.0). ^{1}H-NMR (DMSO-d_6 and acetone d_6) δ 3.52 (br m, 0.8H, CH_2 α to NH), 2.90 (br m, 1.4H, CH γ to NH) and 1.88 (br m, 1.4H, CH β to NH).

[^{3}H]-Compound I. Using the procedure described above for protio **I**, 0.28 mmol of [^{3}H]-compound **V** (4.09 mCi, specific activity 14.6 mCi/mmol) was converted in 56% chemical yield into [^{3}H]-compound **I**, which was obtained as a bright yellow solid (2.2 mCi, specific activity 13.9 mCi/mmol, radiochemical purity >99%, radiochemical yield 53%) with mp 148-150°C (dec.).

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