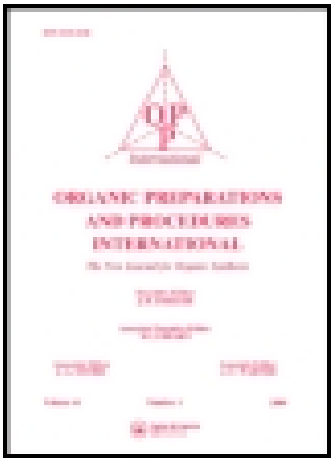

Organic Preparations and Procedures International: The New Journal for Organic Synthesis

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/uopp20

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Published online: 11 Feb 2009.

To cite this article: Carmen Balo , Franco Fernández , Xerardo García-Mera & Carmen López (2000) SYNTHESIS OF FLUORESCENCE PROBES WITH A 2,6-AMINONAPHTHALENE-CARBONYL CHROMOPHORE, Organic Preparations and Procedures International: The New Journal for Organic Synthesis, 32:4, 367-372, DOI: 10.1080/00304940009355936

To link to this article: http://dx.doi.org/10.1080/00304940009355936

ORGANIC PREPARATIONS AND PROCEDURES INT., 32 (4), 367-372 (2000)

SYNTHESIS OF FLUORESCENCE PROBES WITH A 2,6-AMINONAPHTHALENE-CARBONYL CHROMOPHORE

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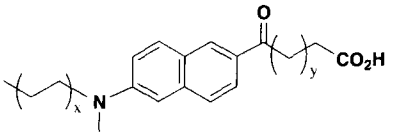
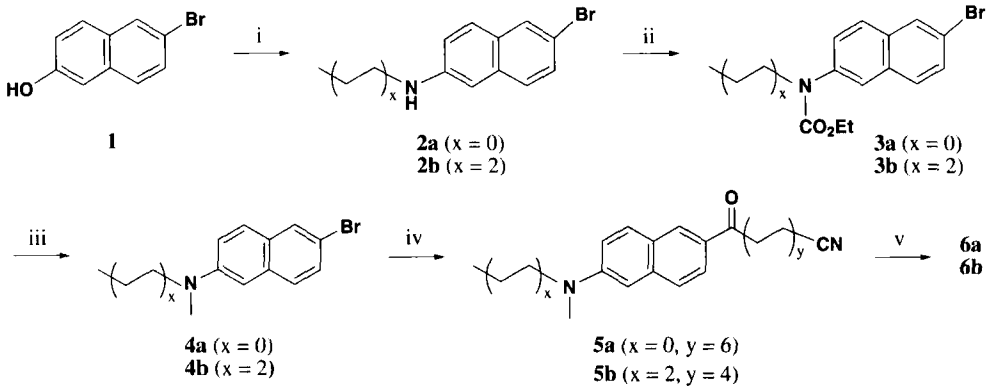
Compounds such as **6a** and **6b** have been shown to be suitable for use as fluorescent probes to study lipid bilayers in biological membranes or artificial model systems.[1] Both **6a** and **6b** are straight-chain fatty acids of 18 Å length (roughly half the mean depth of natural lipid bilayers);[2] they incorporate a 6-dialkylamino-2-naphthoyl fluorophore similar to that of PRODAN,[3] which is highly sensitive to changes in the polarity of its environment.[1] The compounds differ in the number of ethylene units between the fluorophore and the polar carboxylic acid group, thus allowing variations of the polarity and dynamics (mobility, phase transitions) of lipid bilayer at different depths inside the membranes to be studied. We present here a straightforward synthesis of **6a** and **6b**.



a) x = 0, y = 6
b) x = 2, y = 4

6

Easily available 6-bromo-2-naphthol (**1**) seemed to be a good starting material, as it should be feasible to transform its substituents into the desired groups of the target molecules. Having in mind the use of an organometallic derivative to generate the ketone, by coupling the aromatic moiety to an alkyl chain bearing a precursor of the carboxylic acid group, we chose to introduce first the dialkylamino group to avoid the use of protection and deprotection processes related to the initial hydroxy group, as illustrated in *Scheme 1*. The monoalkylamino group was introduced by means of a Bucherer reaction[4] of **1** with the appropriate amine, using aqueous sodium bisulfite as catalyst and reaction medium. Treatment of the resulting secondary amines **2** with ethyl chloroformate in dry pyridine gave high yields of the corresponding carbamates **3**, which were reduced with lithium aluminium hydride to afford the desired tertiary amines **4**.



i) $Na_2S_2O_5$, H_2O, CH_3NH_2•HCl, NaOH, 140° 16 h (**2a**) or $Na_2S_2O_5$, H_2O, $CH_3(CH_2)_4NH_2$, reflux 70 h (**2b**); ii) $ClCO_2Et$, pyr, PhMe, 70° 30 min; iii) $LiAlH_4$, Et_2O, reflux 2 h, then H_2O; iv) BuLi, Et_2O, tetradecanedinitrile, then H_2O/HOAc (**5a**) or Li, Et_2O, decanedinitrile, then H_2O/HOAc (**5b**); v) KOH, H_2O/EtOH, reflux 20 h, then $HCO_2H/NaCO_2H$ buffer.

Scheme 1

Compound **4a** was initially lithiated with *n*-butyllithium and the crude organolithium reagent was treated with tetradecanedinitrile (**7a**), with the usual order of addition reversed to minimize nucleophilic attack at both nitrile groups of **7a.** Chromatography of the crude products obtained after mild hydrolysis afforded a 14% yield of the desired **5a,** together with unreacted **4a** and **7a** and an aliphatic ketonitrile that was identified by spectroscopic analysis as 14-oxo-octadecanenitrile, formed by reaction of residual butyllithium with **7a.** To avoid the formation of any undesired aliphatic ketonitrile, lithiation of the amino bromides **4a** and **4b** was performed with lithium metal instead of butyllithium. Addition of the corresponding naphthyllithium to decanedinitrile (**7b**) as above, followed by usual work-up, lead to a 36% yield of **5b**. However, the yield of isolated **5a** using this procedure rose only to 18% (compared to 14%, see above), while requiring a two-stage chromatography. Alkaline hydrolysis of aminoketonitriles **5a** and **5b** was performed with ethanolic potassium hydroxide under standard conditions. The low basicity of aromatic amines with a conjugated electron-withdrawing substituent,[5] led us to isolate the desired compounds **6** in non-ionic form; thus, protonation of the amino group in the work-up was avoided by adjusting the pH of the reaction mixture to 4, with a formic acid-formate buffer.

EXPERIMENTAL SECTION

Melting points (uncorrected): Reichert Kofler Thermopan. - GLC: Hewlett Packard 5710 A. - IR (in KBr discs, for solids, or films between NaCl plates, for oils): Perkin Elmer FT-IR 1640. - UV: Hewlett Packard 8452 A. - ^1H NMR (in $CDCl_3$ with TMS as internal standard): Bruker AMX-300 (300 MHz). - HRMS: Micromass Autospec (EI, at 70 eV). - Microanalyses: Perkin-Elmer 240B Elemental Analyser. - Silica gel 60 (70-230 mesh) for chromatography was from Merck. - Reagents and solvents were of commercial grade and were supplied by Aldrich Chemical Co.

6-Bromo-*N*-methyl-2-naphthylamine (2a).- A mixture of 10.0 g (44.8 mmol) of **1**, 16.5 g (86.8 mmol) of $Na_2S_2O_5$, 9.0 g (225 mmol) of NaOH and 40 mL of H_2O was placed in a pressure reactor. Then 15.0 g (222 mmol) of methylamine hydrochloride were quickly mixed in with a stirring rod, and the reactor was sealed and heated at 140° for 96 h. After cooling, the reaction mixture was poured into 100 mL of 2 N NaOH, and the precipitate was collected, washed with water and air-dried to constant mass, affording a dark solid that gave a single peak under GLC analysis (10.3 g; 97%). The product was recrystallized twice from MeOH to give a white solid, mp 106-107°. IR (KBr): 3400 (NH), 1610 and 1580 (aromatic C=C) cm^{-1}. ^1H NMR: δ 2.82 (s, 3 H, CH_3), 3.92 (br s, 1 H, NH), 6.84 (d, J = 2.2 Hz, 1 H, 1-H), 6.93 (dd, J = 8.8, 2.2 Hz, 1 H, 3-H), 7.42 (dd, J = 8.9, 1.9 Hz, 1 H, 7-H), 7.48 (d, J = 8.9 Hz, 1 H, 8-H), 7.54 (d, J = 8.8 Hz, 1 H, 4-H), 7.81 (d, J = 1.9 Hz, 1 H, 5-H).

Anal. Calcd. for $C_{11}H_{10}BrN$: C, 55.96; H, 4.27; N, 5.93. Found: C, 55.71; H, 4.35; N, 6.21

6-Bromo-*N*-pentyl-2-naphthylamine (2b).- A suspension of 10.0 g (44.8 mmol) of **1**, 11.7 mL (101 mmol) of pentylamine and 11.2 g (59 mmol) of $Na_2S_2O_5$ in 66 mL of H_2O was heated at reflux for 70 h. Once cold, the reaction mixture was poured into 100 mL of 2 N NaOH, and the precipitate was collected, washed with water and air-dried to afford 7.80 g of **2b** (60%). Acidification of the filtrate allowed partial recovery of **1** (0.93 g). For **2b**: mp 49-50° (EtOH). IR (KBr): 3390 (NH), 1620 and 1580 (aromatic C=C) cm^{-1}. ^1H NMR: δ 0.94 (t, J = 7.1 Hz, 3 H, CH_3), 1.36-1.46 (m, 4 H, $N(CH_2)_2(C\underline{H}_2)_2$), 1.69 (quint, J = 7.2 Hz, 2 H, $NCH_2C\underline{H}_2$), 3.20 (t, J = 7.1 Hz, 2 H, NCH_2), 3.96 (br s, 1 H, NH), 6.76 (d, J = 2.2 Hz, 1 H, 1-H), 6.88 (dd, J = 8.8, 2.2 Hz, 1 H, 3-H), 7.40 (dd, J = 8.9, 1.8 Hz, 1 H, 7-H), 7.47 (d, J = 8.9 Hz, 1 H, 8-H), 7.52 (d, J = 8.8 Hz, 1 H, 4-H), 7.80 (d, J = 1.8 Hz, 1 H, 5-H).

Anal. Calcd. for $C_{15}H_{18}BrN$: C, 61.65; H, 6.21; N, 4.79. Found: C, 61.57; H, 6.38; N, 5.01.

Ethyl *N*-(6-Bromo-2-naphthyl)-*N*-methylcarbamate (3a).- A solution of 10.0 g (42.3 mmol) of **2a** in 90 mL of toluene was carefully added to a mixture of 8.0 mL (83.7 mmol) of ethyl chloroformate and 4.5 mL (55.6 mmol) of dry pyridine. The mixture was heated at 70° until the reaction was complete (30 min, GLC monitoring), then it was cooled and acidified to pH 4 with 2 N HCl. The organic phase was separated, washed with water, dried (Na_2SO_4), and the solvent was removed *in vacuo* to afford 8.20 g of **3a** (63%) as a dark viscous oil. IR (film): 1710 (C=O), 1590 (aromatic C=C) cm^{-1}. ^1H NMR: δ 1.24 (t, J = 7.1 Hz, 3 H, $CH_2C\underline{H}_3$), 3.38 (s, 3 H, NCH_3), 4.20 (q, J = 7.1 Hz, 2 H, OCH_2), 7.43 (dd, J = 8.8, 1.9 Hz, 1 H, 7-H), 7.50 (dd, J = 8.8, 1.8 Hz, 1 H, 3-H), 7.62-7.69 (m, 3 H, 1-H + 4-H + 8-H), 7.94 (d, J = 1.9 Hz, 1 H, 5-H). HRMS: Calcd. for $C_{14}H_{14}BrNO_2$, m/z = 307.0208. Found, 307.0212.

Ethyl *N*-(6-Bromo-2-naphthyl)-*N*-pentylcarbamate (3b).- This compound was prepared by the method described for the preparation of **3a**, from 6.0 g (20.5 mmol) of **2b** in 90 mL of toluene and 5.8 mL of ethyl chloroformate (60.8 mmol) in 3.2 mL of dry pyridine (39.6 mmol). Work-up afforded 7.37 g of **3b** (99%), as a viscous liquid. IR (film): 1700 (C=O), 1585 (aromatic C=C) cm^{-1}. ^1H NMR: δ 0.85 (t, J = 6.7 Hz, 3 H, $N(CH_2)_4C\underline{H}_3$), 1.18-1.30 (m, 7 H, $N(CH_2)_2(C\underline{H}_2)_2$ + $OCH_2C\underline{H}_3$), 1.53-1.60 (m, 2 H, $NCH_2C\underline{H}_2$), 3.74 (t, J = 7.5 Hz, 2 H, NCH_2), 4.16 (q, J = 7.1 Hz, 2 H, OCH_2), 7.36 (dd, J =

8.8, 1.7 Hz, 1 H, 7-H), 7.54 (dd, *J* = 8.8, 1.8 Hz, 1 H, 3-H), 7.61 (d, *J* = 1.8 Hz, 1 H, 1-H), 7.67 (d, *J* = 8.8 Hz, 1 H, 4-H), 7.73 (d, *J* = 8.8 Hz, 1 H, 8-H), 7.99 (d, *J* = 1.7 Hz, 1 H, 5-H). HRMS: Calcd. for $C_{18}H_{22}BrNO_2$, *m/z* = 363.0834. Found, 363.0828.

6-Bromo-*N,N*-Dimethyl-2-naphthylamine (4a).- A solution of 8.0 g (26 mmol) of **3a** in 65 mL of dry Et_2O was slowly added to a stirred mixture of 1.0 g (26 mmol) of $LiAlH_4$ and 23 mL of dry Et_2O maintained at 0° by means of an ice-bath. The mixture was refluxed for 2 h and then allowed to cool to rt and treated with 50 mL of wet Et_2O. The solids were filtered out and washed repeatedly with diethyl ether. The pooled ethereal filtrates were washed with water, dried (Na_2SO_4) and concentrated *in vacuo* to afford 4.78 g of a solid. This solid was chromatographed on silica gel, with toluene as eluent, to afford 4.10 g of **4a** (63%), as a white solid from MeOH, mp 129-130°. IR (KBr): 1620 (aromatic C=C) cm^{-1}. ^1H NMR: δ 3.05 (s, 6 H, $N(CH_3)_2$), 6.89 (d, *J* = 2.3 Hz, 1 H, 1-H), 7.18 (dd, *J* = 9.1, 2.3 Hz, 1 H, 3-H), 7.42 (dd, *J* = 8.8, 1.8 Hz, 1 H, 7-H), 7.52 (d, *J* = 8.8 Hz, 1 H, 8-H), 7.61 (d, *J* = 9.1 Hz, 1 H, 4-H), 7.83 (s, 1 H, 5-H).

Anal. Calcd. for $C_{12}H_{12}BrN$: C, 57.62; H, 4.84; N, 5.60. Found: C; 57.54; H, 4.98; N, 5.76

6-Bromo-*N*-methyl-*N*-pentyl-2-naphthylamine (4b).- This compound, mp. 39-40° (MeOH), was prepared in 64% yield from **3b** by the method described for **4a**. IR (KBr): 1610 and 1580 (aromatic C=C) cm^{-1}. ^1H NMR: δ 0.90 (t, *J* = 6.7 Hz, 3 H, $CH_2C\underline{H}_3$), 1.31-1.34 (m, 4 H, $N(CH_2)_2(C\underline{H}_2)_2$), 1.59 (quint, *J* = 7.4 Hz, 2 H, $NCH_2C\underline{H}_2$), 3.03 (s, 3 H, NCH_3), 3.40 (t, *J* = 7.6 Hz, 2 H, NCH_2), 6.81 (s, 1 H, 1-H), 7.14 (d, *J* = 9.3 Hz, 1 H, 3-H), 7.40 (d, *J* = 8.6 Hz, 1 H, 7-H), 7.51 (d, *J* = 8.6 Hz, 1 H, 8-H), 7.60 (d, *J* = 9.3 Hz, 1 H, 4-H), 7.82 (s, 1 H, 5-H).

Anal. Calcd. for $C_{16}H_{20}BrN$: C, 62.75; H, 6.58; N, 4.57. Found: C, 62.54; H, 6.68; N, 4.76

14-[6-(Dimethylamino)-2-naphthyl]-14-oxotetradecanenitrile (5a).- To a solution of 2.95 g (11.8 mmol) of dried (*in vacuo*, over P_2O_5) **4a** in 20 mL of anhydrous Et_2O kept under Ar, 7.5 mL of 1.6 M BuLi in hexane (12 mmol) was added dropwise, and the mixture was stirred at rt for 30 min and then slowly added via syringe to a cold (0°), vigorously stirred solution of 2.61 g (11.8 mmol) of **7a** in 20 mL of anhydrous Et_2O under Ar. After the addition was complete, the mixture was refluxed for 1 h, then treated with 10 mL of 1:1 water/acetic acid and refluxed for a further 1 h. The reaction mixture was diluted with Et_2O, washed with water, 1 N NaOH and water, then dried (Na_2SO_4). Evaporation of the ether *in vacuo* left 4.69 g of a residue which was chromatographed on silica gel, with CH_2Cl_2 as eluent to give in order of elution, 1.11 g of **4a**, 0.03 g of 14-oxooctadecanenitrile, 0.93 g of **7a** and 0.65 g of a foam, shown (^1H NMR) to contain >96% **5a** (*ca.* 14%). IR (KBr): 2242 (CN), 1669 (C=O) cm^{-1}. ^1H NMR: δ 1.25-1.45 (m, 16 H, central $(CH_2)_8$), 1.65 (quint, *J* = 7.3 Hz, 2 H, $C\underline{H}_2CH_2CN$), 1.78 (quint, *J* = 7.3 Hz, 2 H, $C\underline{H}_2CH_2CO$), 2.33 (t, *J* = 7.1 Hz, 2 H, CH_2CN), 3.04 (t, *J* = 7.4 Hz, 2 H, CH_2CO), 3.11 (s, 6 H, $N(CH_3)_2$), 6.91 (d, *J* = 2.2 Hz, 1 H, 5-H), 7.19 (dd, *J* = 9.1, 2.2 Hz, 1 H, 7-H), 7.65 (d, *J* = 8.7 Hz, 1 H, 4-H), 7.81 (d, *J* = 9.1 Hz, 1 H, 8-H), 7.93 (dd, *J* = 8.7, 1.6 Hz, 1 H, 3-H), 8.33 (s, 1 H, 1-H). HRMS: Calcd. for $C_{26}H_{36}N_2O$, *m/z* = 392.2829. Found: 392.2833.

10-[6-(*N*-Methyl-*N*-pentylamino)-2-naphthyl]-10-oxodecanenitrile (5b).- A solution of 1.27 g (4.15 mmol) of **4b** in 3 mL of anhydrous Et_2O was added to a stirred mixture of 87 mg (12.5 mmol) of

Li in 20 mL of anhydrous Et_2O and refluxed for 5 h (formation of the aryllithium reagent was confirmed by the method of Gilman[6]). This reaction mixture was then slowly added *via* syringe to a solution of 0.70 g (4.26 mmol) of **7b** in 5 mL of anhydrous Et_2O. The mixture was refluxed for 1 h, then treated with 4 mL of 1:1 water/acetic acid and refluxed for a further 1 h. Work-up as for **5a** led to 1.05 g of a residue, which was chromatographed on silica gel with 4:1 toluene/Et_2O as eluent, to give 0.62 g of a foam, shown (^{1}H NMR) to contain >95% of **5b** (*ca.* 36%). IR (KBr): 2243 (CN), 1671 (C=O) cm^{-1}. ^{1}H NMR: δ 0.92 (t, *J* = 6.7 Hz, 3 H, $CH_2C\underline{H}_3$), 1.26-1.43 (m, 12 H, $CH_3(C\underline{H}_2)_2$ + central $(CH_2)_4$), 1.58-1.67 (m, 4 H, $C\underline{H}_2CH_2N$ + $C\underline{H}_2CH_2CN$), 1.76 (quint, *J* = 7.2 Hz, 2 H, $C\underline{H}_2CH_2CO$), 2.33 (t, *J* = 7.0 Hz, 2 H, CH_2CN), 3.02 (t, *J* = 7.2 Hz, 2 H, CH_2CO), 3.06 (s, 3 H, NCH_3), 3.44 (t, *J* = 7.5 Hz, 2 H, CH_2N), 6.82 (s, 1 H, 5-H), 7.14 (d, *J* = 9.2 Hz, 1 H, 7-H), 7.60 (d, *J* = 8.7 Hz, 1 H, 4-H), 7.78 (d, *J* = 9.2 Hz, 1 H, 8-H), 7.89 (dd, *J* = 8.7, 1.5 Hz, 1 H, 3-H), 8.31 (s, 1 H, 1-H). HRMS: Calcd. for $C_{26}H_{36}N_2O$, *m/z*, 392.2829. Found: 392.2834.

14-[6-(Dimethylamino)-2-naphthyl]-14-oxotetradecanoic Acid (6a).- Compound **5a** (200 mg, 0.51 mmol) was added to a solution of 100 mg (1.78 mmol) KOH in 0.15 mL of water and 1 mL of EtOH, and the mixture was refluxed for 20 h. Once cool, the reaction mixture was extracted with Et_2O, and the aqueous layer was adjusted to pH 4 by addition of a formic acid/sodium formate buffer. The precipitated solid was collected, washed with water and vacuum dried to constant mass to afford 163 mg of **6a** (78%) as white plates from cyclohexane, mp 97-98°. IR (KBr): 1709 (carboxylic acid C=O), 1668 (ketone C=O), 1626 and 1508 (aromatic C=C) cm^{-1}. UV/Vis (EtOH): λ_{max} (log e) = 362 nm (4.95). ^{1}H NMR: δ 1.26-1.41 (m, 16 H, central $(CH_2)_8$), 1.66 (quint, *J* = 7.5 Hz, 2 H, $C\underline{H}_2CH_2CO_2H$), 1.77 (quint, *J* = 7.3 Hz, 2 H, $C\underline{H}_2CH_2CO$), 2.35 (t, *J* = 7.5 Hz, 2 H, CH_2CO_2H), 3.03 (t, *J* = 7.5 Hz, 2 H, CH_2CO), 3.12 (s, 6 H, $N(CH_3)_2$), 6.83 (d, *J* = 2.4 Hz, 1 H, 5-H), 7.14 (dd, *J* = 9.2, 2.4 Hz, 1 H, 7-H), 7.61 (d, *J* = 8.7 Hz, 1 H, 4-H), 7.77 (d, *J* = 9.2 Hz, 1 H, 8-H), 7.91 (dd, *J* = 8.7, 1.7 Hz, 1 H, 3-H), 8.30 (s, 1 H, 1-H), 11.98 (br s, 1 H, CO_2H).

Anal. Calcd. for $C_{26}H_{37}NO_3$: C, 75.87; H, 9.06; N, 3.40. Found: C, 76.01; H, 9.20; N, 3.35

10-[6-(*N*-Methyl-*N*-pentylamino)-2-naphthyl]-10-oxodecanoic Acid (6b).- Following the procedure described above for the preparation of **6a**, nitrile **5b** was hydrolysed to **6b** in 58% yield as white needles from EtOH, mp 92-93°. IR (KBr): 1705 (carboxylic acid C=O), 1670 (ketone C=O), 1624 (aromatic C=C) cm^{-1}. UV/Vis (EtOH): λ_{max} (log e) = 374 nm (4.94). ^{1}H NMR: δ 0.88 (t, *J* = 6.7 Hz, 3 H, $CH_2C\underline{H}_3$), 1.25-1.36 (m, 12 H, $CH_3(C\underline{H}_2)_2$ + central $(CH_2)_4$), 1.61-1.66 (m, 4 H, $C\underline{H}_2CH_2N$ + $C\underline{H}_2CH_2CO_2H$), 1.74-1.79 (m, 2 H, $C\underline{H}_2CH_2CO$), 2.35 (t, *J* = 7.4 Hz, 2 H, $C\underline{H}_2CO_2H$), 3.00-3.07 (m, 5 H, CH_2CO + NCH_3), 3.45 (t, *J* = 7.5 Hz, 2 H, NCH_2), 6.82 (d, *J* = 2.4 Hz, 1 H, 5-H), 7.13 (dd, *J* = 9.1, 2.4 Hz, 1 H, 7-H), 7.61 (d, *J* = 8.7 Hz, 1 H, 4-H), 7.77 (d, *J* = 9.1 Hz, 1 H, 8-H), 7.91 (dd, *J* = 8.7, 1.5 Hz, 1 H, 3-H), 8.30 (d, *J* = 1.5 Hz, 1 H, 1-H), 12.22 (br s, 1 H, CO_2H).

Anal. Calcd. for $C_{26}H_{37}NO_3$: C, 75.87; H, 9.06; N 3.40. Found: C, 75.98; H, 9.28; N, 3.45

Acknowledgements.- The authors thank the *Xunta de Galicia* (XUGA 20309B98) for financially supporting this work.

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(Received March 11, 2000; in final form June 20, 2000)