[Chem. Pharm. Bull.]
30(12)4378-4383(1982)

Thiazole Analogs of Benzomorphans. I. Synthesis of Novel Thiazolo[4,5-*f*]morphans

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(Received July 6, 1982)

The synthesis and analgesic properties of thiazolo[4,5-*f*]morphans (XIIa—c) are described. Monothiazolization of ethyl 2-methyl-1,3-dioxo-2-cyclohexaneacetate (I) afforded the 2-aminothiazole derivative (II), which was deaminated to ethyl 4,5,6,7-tetrahydro-4-methyl-5-oxo-4-benzothiazoleacetate (V) by means of the Sandmeyer reaction and subsequent hydrogenolysis of the resulting chloride (IVa). In several steps, V was converted to 2,5-dimethyl-9-oxothiazolo[4,5-*f*]morphan (X). Thiazolo[4,5-*f*]morphan derivatives (XIIa—c) were synthesized from this key intermediate (X).

Keywords—analgesic activity; 2-aminothiazole; 4,5,5a,6,7,8a-hexahydro-8*H*-thiazolo[4,5-*e*]indole; 9-oxothiazolo[4,5-*f*]morphan; Wittig reaction; Wolff-Kishner reduction; thiazolo[4,5-*f*]morphan

It has been proposed that a benzene moiety in an analgesic molecule binds to an analgesic receptor with a plane hydrophobic site.¹⁾ It was also claimed that a *meta* (2'-position of 6,7-benzomorphan) hydroxyl group on the benzene ring interacts with a dipolar site on the receptor by forming a hydrogen bond.²⁾ This dipolar interaction, specific to the *meta* position, aids the molecule in binding to the receptor and enhances the analgesic effect.

In view of the structure-activity relationship for the benzene moiety, much attention has been paid to the analgesic and antagonist activities of benzomorphan analogs in which the benzene ring is replaced by a heteroaromatic ring. For this reason, pyridomorphans,³⁾ thienomorphans,⁴⁾ and more recently, pyrrolomorphans⁵⁾ have been synthesized.

In general, thiophenes possess chemical, physical, and in certain cases, biological properties similar to those of their benzene analogs. In spite of the similarities between the pyridine and thiazole series, the structure of thiazole, which has both electron-rich and electron-poor sites and also has a π -excess character as a whole, is still unique in these aromatic systems. Therefore, we attempted to synthesize thiazolomorphans, in order to investigate the interaction between thiazole and the dipolar site on the analgesic receptor.

In a previous communication,⁶⁾ we briefly described the synthesis of novel thiazolo[4,5-*f*]morphans, following the method described for the synthesis of 9-oxobenzomorphan by Kavadias,⁷⁾ and full details are presented in this paper.

In our synthetic approach, the first key intermediate (V), a thiazole analog of tetralone, was prepared as follows. Bromination of ethyl 2-methyl-1,3-dioxo-2-cyclohexaneacetate (I)⁸⁾ with one equivalent of bromine in acetic acid followed by treatment of the resulting material with thiourea in tetrahydrofuran (THF) afforded 2-aminothiazole derivatives, II and IIIa, in 63% and 11% yields, respectively. The use of pyridinium hydrobromide perbromide instead of bromine in this thiazolization gave the same result. The by-product (IIIa), which was insoluble in usual solvents, was acetylated to confirm the structure. The proton nuclear magnetic resonance (¹H-NMR) spectrum of the acetate (IIIb) exhibited signals due to two methyl groups of the acetamido function at δ 2.13 as a 6H singlet, and the signal due to a methylene group located between two thiazole rings at δ 3.97 as a 2H singlet. The molecular ion (M⁺) peak in the mass spectrum (MS) of IIIb was detected at *m/e* 408. These spectral data and the elemental analysis demonstrated that compound (IIIb) was ethyl

Chart 1

2,6-diacetamino-4,8-dihydro-4-methyl-4-benzo[1,2-*d*; 5,4-*d'*]dithiazoleacetate.

Deamination of II was successfully carried out by means of the Sandmeyer reaction and subsequent hydrogenolysis. The diazonium salt, which was prepared from II by treatment with sodium nitrite in 40% sulfuric acid, was treated with a solution of cupric sulfate and sodium chloride. The product was, however, the mixture of the desired compound (IVa) and the corresponding acid (IVb), which was presumably formed by the hydrolysis of IVa by sulfuric acid. Hence the crude mixture, without purification, was esterified with ethanolic hydrogen chloride solution and IVa was obtained as a single product in 64% yield. Hydrogenolysis of IVa over palladium-carbon in the presence of triethylamine afforded ethyl 4,5,6,7-tetrahydro-4-methyl-5-oxo-4-benzothiazoleacetate (V) quantitatively. A characteristic proton of the 2-position in the thiazole ring appeared at δ 8.65 as a singlet in the ^1H -NMR spectrum of V. On the other hand, direct deamination of II, by decomposition of the corresponding diazonium salt with calcium hypophosphite, resulted in the formation of a low yield of V.

Chart 2

The synthetic sequence to the second intermediate, 2,5-dimethyl-9-oxothiazolo[4,5-*f*]-morphinan (X), is illustrated in Chart 2. Treatment of V with methanolic methylamine solution afforded VI. Its infrared (IR) spectrum has bands at 3320 and 1680 cm^{-1} , and these indicate the formation of the five-membered lactam ring. Azeotropic dehydration of VI with *p*-toluene sulfonic acid in toluene gave VII. Its ^1H -NMR spectrum showed a doublet (2H, $J=4$ Hz)

Experimental

Melting points were measured on a Yanaco PM-S3 apparatus (a hot stage type) and are uncorrected. IR spectra were determined on a JASCO IRA-1 spectrometer and MS were recorded on a Shimadzu LKB-9000 spectrometer. ^1H -NMR spectra were taken on JEOL PMX-60 and PS-100 spectrometers in deuteriochloroform using tetramethylsilane as an internal standard unless otherwise stated, and assignments of NH and OH protons were confirmed by exchange with D_2O . All organic extracts were dried over anhydrous magnesium sulfate. Concentration of all organic solutions was carried out by evaporation under reduced pressure. Column chromatography was performed with Waco silica gel C-200.

Ethyl 2-Amino-4,5,6,7-tetrahydro-4-methyl-5-oxo-4-benzothiazoleacetate (II) and Ethyl 2,6-Diamino-4,8-dihydro-4-methyl-4-benzo[1,2-*d*; 5,4-*d'*]dithiazoleacetate (IIIa)—A solution of Br_2 (14.0 g, 87.5 mmol) in AcOH (50 ml) was added dropwise with stirring to an ice-cooled solution of I (19.7 g, 92.8 mmol) in AcOH (200 ml) containing a few drops of 48% HBr. After being stirred for 30 min, the reaction mixture was concentrated to half the initial volume, poured into ice-water, and extracted with ether. The extract was washed successively with NaHCO_3 solution and water, dried, and concentrated. The residual material and thiourea (8.4 g, 110 mmol) were dissolved in THF (300 ml) and, after being refluxed for 4 h, the solution was concentrated. The residue was dissolved in dil. HCl, washed with ether, made basic with NaHCO_3 solution, and mixed with CHCl_3 . The mixture was stirred for 1.5 h, then the insoluble material was filtered off, washed successively with water and CHCl_3 , and dried to yield 3.2 g (11%) of IIIa as an amber powder, mp $>350^\circ\text{C}$. IR (Nujol): 3430, 3260, 1720 cm^{-1} . MS m/e : 324 (M^+).

The filtrates and the washings were combined and the CHCl_3 layer was separated, washed with water, dried, and concentrated. The residue was crystallized from benzene-AcOEt to give 15.6 g (63%) of II as yellow prisms, mp $160\text{--}162^\circ\text{C}$. Anal. Calcd for $\text{C}_{12}\text{H}_{16}\text{N}_2\text{O}_3\text{S}$: C, 53.71; H, 6.01; N, 10.44. Found: C, 53.49; H, 5.91; N, 10.21. IR (CHCl_3): 3500, 3400, 1720 cm^{-1} . ^1H -NMR δ : 1.13 (3H, t, $J=7$ Hz, OCH_2CH_3), 1.30 (3H, s, 4- CH_3), 2.63–3.20 (6H, m), 4.00 (2H, q, $J=7$ Hz, OCH_2CH_3), 5.37 (2H, br s, $-\text{NH}_2$). MS m/e : 268 (M^+).

Acetylation of IIIa—A mixture of IIIa (9.2 g, 28.5 mmol), Ac_2O (6.5 ml) and dry pyridine (150 ml) was stirred for 2 d. The reaction mixture was poured into ice-water, made acidic with dil. HCl, and extracted with $\text{CHCl}_3\text{--CH}_3\text{OH}$ (4:1). The extract was washed successively with NaHCO_3 solution and water, dried, and concentrated. The residue was crystallized from CH_3OH to give 4.95 g (43%) of IIb as amber needles, mp $>350^\circ\text{C}$. Anal. Calcd for $\text{C}_{17}\text{H}_{20}\text{N}_4\text{O}_4\text{S}_2$: C, 49.99; H, 4.94; N, 13.72. Found: C, 49.75; H, 4.82; N, 13.71. IR (Nujol): 3260, 1720, 1670 cm^{-1} . ^1H -NMR ($\text{DMSO-}d_6$) δ : 0.83 (3H, t, $J=7$ Hz, OCH_2CH_3), 1.53 (3H, s, 4- CH_3), 2.13 (6H, s, $2 \times \text{COCH}_3$), 2.93 (2H, br s, $\text{CH}_2\text{CO}_2\text{Et}$), 3.70 (2H, q, $J=7$ Hz, OCH_2CH_3), 3.97 (2H, s, 8-H), 12.15 (2H, s, NH). MS m/e : 408 (M^+).

Ethyl 2-Chloro-4,5,6,7-tetrahydro-4-methyl-5-oxo-4-benzothiazoleacetate (IVa)—a) A solution of NaNO_2 (0.31 g, 4.6 mmol) in 4 ml of water was added dropwise to a stirred solution of II (1.0 g, 3.7 mmol) in 40% H_2SO_4 (15 ml) at $-12 \pm 2^\circ\text{C}$. The mixture was stirred for 2 h at the same temperature, then added to a solution of $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ (7.5 g) and NaCl (7.5 g) in water (15 ml) at $-2 \pm 2^\circ\text{C}$ with vigorous stirring. The reaction mixture was stirred for 2 h at room temperature and extracted with CHCl_3 . The extract was washed with water, dried, and concentrated. The residue was chromatographed on a silica gel column. Elution with benzene gave 170 mg (16%) of IVa. Recrystallization from ether gave colorless prisms, mp $63.5\text{--}64.5^\circ\text{C}$. Anal. Calcd for $\text{C}_{12}\text{H}_{14}\text{ClNO}_3\text{S}$: C, 50.09; H, 4.90; N, 4.89. Found: C, 50.07; H, 4.80; N, 4.91. IR (CHCl_3): 1720 cm^{-1} . ^1H -NMR δ : 1.13 (3H, t, $J=7$ Hz, OCH_2CH_3), 1.40 (3H, s, 4- CH_3), 2.65–3.30 (6H, m), 3.97 (2H, q, $J=7$ Hz, OCH_2CH_3). MS m/e : 287, 289 (M^+).

Elution with benzene-AcOEt (4:1) gave 350 mg (36%) of IVb. Recrystallization from ether gave pale yellow needles, mp $147\text{--}149^\circ\text{C}$. Anal. Calcd for $\text{C}_{10}\text{H}_{10}\text{ClNO}_3\text{S}$: C, 46.25; H, 3.88; N, 5.39. Found: C, 46.07; H, 3.68; N, 5.29. IR (CHCl_3): 1780, 1720 cm^{-1} . ^1H -NMR δ : 1.50 (3H, s, 4- CH_3), 2.10–3.20 (4H, m), 2.91 (2H, s, $\text{CH}_2\text{CO}_2\text{Et}$). MS m/e : 259, 261 (M^+).

An ice-cooled solution of IVb (190 mg) in abs. EtOH (15 ml) was saturated with dry hydrogen chloride. After standing overnight, the reaction mixture was poured into ice-water and extracted with CHCl_3 . The extract was washed successively with NaHCO_3 solution and water, dried, and concentrated. The residual oil (206 mg, 98%) was shown to be identical with IVa by comparison of IR and ^1H -NMR spectra.

b) Following the procedure mentioned above, 30.2 g (113 mmol) of II in 40% H_2SO_4 (390 ml) was diazotized with NaNO_2 (11.6 g, 168 mmol), and the resulting diazonium solution was treated with a solution of $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ (220 g) and NaCl (220 g) in water (390 ml). The crude residue, which was obtained by concentration of the CHCl_3 extract, was treated with a saturated solution of dry hydrogen chloride in abs. EtOH (190 ml). After standing overnight, the reaction mixture was concentrated to half the initial volume, poured into ice-water, and extracted with CHCl_3 . The extract was washed successively with NaHCO_3 solution and water, dried, and concentrated. The residual oil was distilled and 20.7 g (64%) of IVa was obtained as a colorless oil, bp $120\text{--}125^\circ\text{C}/0.20$ mmHg, which was identified by comparison of IR and ^1H -NMR spectra with those of an authentic sample.

Ethyl 4,5,6,7-Tetrahydro-4-methyl-5-oxo-4-benzothiazoleacetate (V)—a) A solution of IVa (8.15 g,

28.4 mmol) and triethylamine (4 ml) in EtOH (150 ml) was shaken with H_2 in the presence of 10% Pd-C (0.85 g) under atmospheric pressure. After removal of the catalyst and the solvent, the residue was dissolved in $CHCl_3$. The solution was washed successively with dil. HCl and water, dried, and concentrated. Distillation of the residual oil gave 7.1 g (99%) of V as a colorless oil, bp 113–114°C/0.15 mmHg. *Anal.* Calcd for $C_{12}H_{15}NO_3S$: C, 56.90; H, 5.97; N, 5.53. Found: C, 57.00; H, 5.95; N, 5.54. IR ($CHCl_3$): 1720 cm^{-1} . 1H -NMR δ : 1.10 (3H, t, $J=7$ Hz, OCH_2CH_3), 1.45 (3H, s, 4- CH_3), 2.67–3.40 (6H, m), 3.94 (2H, q, $J=7$ Hz, OCH_2CH_3), 8.65 (1H, s, 2-H). MS m/e : 253 (M^+).

b) A solution of $NaNO_2$ (6.0 g, 86.9 mmol) in water (40 ml) was added dropwise to a stirred solution of II (11.6 g, 43.3 mmol) in 40% H_2SO_4 (230 ml) at $-8^\circ C$. After being stirred for 30 min, the diazotized solution was added to a suspension of $Ca(H_2PO_4)_2$ (133 g) in water (500 ml) at the same temperature with vigorous stirring. The mixture was stirred for 30 min at $0^\circ C$ then for 1.5 h at room temperature, filtered, and washed well with $CHCl_3$. The filtrates and the washings were combined and the organic layer was separated, washed with water, dried, and concentrated. Distillation of the residual oil gave 2.58 g (24%) of a colorless oil, bp 110–119°C/0.20 mmHg, which was shown to be identical with V by comparison of IR and 1H -NMR spectra.

4,5,5a,6,7,8a-Hexahydro-5a-hydroxy-6,8a-dimethyl-7-oxo-8H-thiazolo[4,5-e]indole (VI)—A solution of V (2.37 g, 9.4 mmol) in 30% methanolic methylamine (3.0 ml) was left overnight. Colorless prisms formed and were collected to give 1.73 g (78%) of VI, mp 217.5–219°C. *Anal.* Calcd for $C_{11}H_{14}N_2O_2S$: C, 55.44; H, 5.92; N, 11.76. Found: C, 55.55; H, 5.76; N, 11.64. IR (Nujol): 3320, 1680 cm^{-1} . 1H -NMR ($CDCl_3 + CD_3OD=4:1$) δ : 1.50 (3H, s, 8a- CH_3), 2.05–3.00 (4H, m), 2.73 (2H, s, 8-H), 2.85 (3H, s, N- CH_3), 3.57 (1H, br s, -OH), 8.63 (1H, s, 2-H). MS m/e : 238 (M^+).

4,6,7,8a-Tetrahydro-6,8a-dimethyl-7-oxo-8H-thiazolo[4,5-e]indole (VII)—A mixture of VI (8.05 g, 33.8 mmol) and *p*-TsOH (20 mg) in toluene (100 ml) was refluxed for 4 h with azeotropic removal of water. After cooling, the reaction mixture was washed with water, dried, and concentrated. Recrystallization of the residual solid from benzene-ether gave 6.58 g (88%) of VII as colorless needles, mp 156–161°C. *Anal.* Calcd for $C_{11}H_{12}N_2OS$: C, 59.98; H, 5.49; N, 12.72. Found: C, 59.84; H, 5.50; N, 12.77. IR ($CHCl_3$): 1720, 1670 cm^{-1} . 1H -NMR δ : 1.40 (3H, s, 8a- CH_3), 2.83 (2H, s, 8-H), 3.05 (3H, s, N- CH_3), 3.60 (2H, d, $J=4$ Hz, 4-H), 5.15 (1H, t, $J=4$ Hz, 5-H), 8.70 (1H, s, 2-H). MS m/e : 220 (M^+).

5-Bromo-4,5,7,8a-tetrahydro-8a-methyl-8H-thiazolo[4,5-e]indole Methobromide (IX)— $LiAlH_4$ (2.42 g, 63.8 mmol) was added portionwise to an ice-cooled solution of VII (9.35 g, 42.5 mmol) in dry ether (340 ml) with stirring. The mixture was refluxed for 2.5 h under argon. After cooling, the reaction mixture was treated with Rochelle salt solution, and the precipitates were filtered off and washed with $CHCl_3$. Concentration of the combined filtrates and washings gave 8.12 g of crude product (VIII) as a pale reddish oil. IR ($CHCl_3$): 1660 cm^{-1} . 1H -NMR δ : 1.33 (3H, s, 8a- CH_3), 2.00–2.45 (2H, m, 8-H), 2.68 (3H, s, N- CH_3), 2.80–3.80 (4H, m), 4.25 (1H, m, 5-H), 8.65 (1H, s, 2-H).

A solution of the crude oil (VIII) in CH_2Cl_2 (50 ml) was added in one portion to a stirred solution of Br_2 (6.82 g, 42.6 mmol) in CH_2Cl_2 (420 ml) at $-60^\circ C$ under argon. After being stirred for 10 min at the same temperature and for 20 min at room temperature, the reaction mixture was concentrated to give 14.9 g (96%) of IX as a dark reddish solid. To obtain an analytical sample, the perchlorate of IX was recrystallized from CH_3OH as yellow needles, mp 150–152.5°C. *Anal.* Calcd for $C_{11}H_{14}BrClN_2O_4S$: C, 34.26; H, 3.66; N, 7.26. Found: C, 33.98; H, 3.48; N, 7.29. IR (Nujol): 1680 cm^{-1} .

2,5-Dimethyl-9-oxothiazolo[4,5-f]morphinan (X)—A solution of ammonium bicarbonate (3.4 g, 42.5 mmol) in water (17 ml) was added dropwise to a stirred solution of IX (14.9 g, 40.7 mmol) in 95% EtOH (390 ml) at $-10^\circ C$ under argon. The reaction mixture was stirred for 2 h at $-10^\circ C$ and for 19 h at room temperature, then concentrated to half the initial volume, made basic with $NaHCO_3$ solution, and extracted with $CHCl_3$. The extract was washed with sat. NaCl solution and dried. Removal of the solvent gave 6.50 g (72%) of X as an amber solid. Recrystallization from ether gave pale yellow prisms, mp 122.5–125.5°C. *Anal.* Calcd for $C_{11}H_{14}N_2OS$: C, 59.43; H, 6.35; N, 12.60. Found: C, 59.33; H, 6.20; N, 12.65. IR ($CHCl_3$): 1735 cm^{-1} . 1H -NMR δ : 1.58 (3H, s, 5- CH_3), 1.95–3.80 (7H, m), 2.45 (3H, s, N- CH_3), 8.72 (1H, s, 2'-H). MS m/e : 222 (M^+).

2,5-Dimethylthiazolo[4,5-f]morphinan (XIIa)—A mixture of X (877 mg, 4.0 mmol), 95% hydrazine hydrate (2.0 ml), KOH (2.0 g) and diethylene glycol (15 ml) was heated at $170^\circ C$ for 15 h under argon. After cooling, the reaction mixture was poured into water and extracted with $CHCl_3$. The extract was washed with water, dried, and concentrated. The residue was chromatographed on a silica gel column. Elution with 2% $CH_3OH-CHCl_3$ gave 403 mg (49%) of XIIa as a yellow oil. 1H -NMR δ : 1.48 (3H, s, 5- CH_3), 2.40 (3H, s, N- CH_3), 8.58 (1H, s, 2'-H). MS m/e : 208 (M^+).

The oxalate of XIIa was crystallized from EtOH to give colorless needles, mp 233.5–234°C (dec.). *Anal.* Calcd for $C_{18}H_{18}N_2O_4S$: C, 52.33; H, 6.08; N, 9.39. Found: C, 52.17; H, 5.89; N, 9.28.

2,5-Dimethyl-9-methylenethiazolo[4,5-f]morphinan (XI)—A 15% solution of *n*-BuLi in hexane (27 ml, 42.1 mmol) was added dropwise to a stirred suspension of methyltriphenyl phosphonium iodide (16.7 g, 41.4 mmol) in dry ether (180 ml) at $0^\circ C$ under argon, and the mixture was stirred at room temperature for 4 h to complete the formation of the ylide. A solution of X (6.1 g, 27.6 mmol) in dry THF (70 ml) was added dropwise to the stirred suspension of the ylide, and the mixture was stirred for 16 h at room tempera-

ture then refluxed for 2 h. After cooling, the reaction mixture was treated with water (1 ml) and filtered. The filtrate was concentrated and the residue was dissolved in dil. HCl, washed with CHCl_3 , made basic with NaHCO_3 solution, and extracted with CHCl_3 . The extract was washed with sat. NaCl solution and dried. Removal of the solvent gave 5.05 g (83%) of XI as a colorless oil. $^1\text{H-NMR}$ δ : 1.67 (3H, s, 5- CH_3), 3.43 (3H, s, N- CH_3), 4.94 (2H, s, $\text{C}=\text{CH}_2$), 8.62 (1H, s, 2'-H). MS m/e : 220 (25%, M^+), 219 (58%, M^+-1), 205 (100%, M^+-CH_3).

The oxalate of XI was crystallized from CH_3OH to give colorless prisms, mp 243.5–244°C (dec.). *Anal.* Calcd for $\text{C}_{14}\text{H}_{18}\text{N}_2\text{O}_4\text{S}$: C, 54.18; H, 5.84; N, 9.02. Found: C, 54.08; H, 5.68; N, 8.92.

2,5,9 α -Trimethylthiazolo[4,5-*f*]morphan (XIb**) and 2,5,9 β -Trimethylthiazolo[4,5-*f*]morphan (XI**c**)**—A mixture of XI (210 mg), 10% Pd-C (140 mg) and EtOH (20 ml) was shaken under 5 atmospheres pressure of H_2 in a Skita apparatus. After removal of the catalyst and the solvent, the residual oil was chromatographed on a silica gel column. Elution with 2% $\text{CH}_3\text{OH}-\text{CHCl}_3$ gave 20 mg (9%) of XI**c** as a colorless oil. $^1\text{H-NMR}$ δ : 1.24 (3H, d, $J=7$ Hz, 9 β - CH_3), 1.45 (3H, s, 5- CH_3), 2.33 (3H, s, N- CH_3), 8.53 (1H, s, 2'-H). MS m/e : 222 (26%, M^+), 207 (40%, M^+-CH_3), 84 (100%).

The oxalate of XI**c** was crystallized from EtOH to give colorless needles, mp 213.5–215.5°C. *Anal.* Calcd for $\text{C}_{14}\text{H}_{20}\text{N}_2\text{O}_4\text{S}$: C, 53.83; H, 6.45; N, 8.97. Found: C, 53.67; H, 6.21; N, 8.73.

Elution with 5% $\text{CH}_3\text{OH}-\text{CHCl}_3$ gave 158 mg (75%) of XI**b** as a colorless oil. $^1\text{H-NMR}$ δ : 0.85 (3H, d, $J=7$ Hz, 9 α - CH_3), 1.45 (3H, s, 5- CH_3), 2.37 (3H, s, N- CH_3), 8.58 (1H, s, 2'-H). MS m/e : 222 (8%, M^+), 207 (9%, M^+-CH_3), 84 (100%).

The oxalate of XI**b** was crystallized from EtOH to give colorless needles, mp 213–214°C. *Anal.* Calcd for $\text{C}_{14}\text{H}_{20}\text{N}_2\text{O}_4\text{S}$: C, 53.83; H, 6.45; N, 8.97. Found: C, 53.90; H, 6.37; N, 8.98.

Acknowledgement The authors are grateful to Profs. S. Sakai and N. Aimi of Chiba University and Prof. S. Shiotani of Toyama University for valuable advice and discussions. They are also grateful to Prof. A. Tanaka of this university for his encouragement during this work, and to Dr. N. Muto of this university for his advice and cooperation in the pharmacological testing.

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