Transformations of Isomeric Naphthopyranones. The Synthesis of Substituted β-Naphthyl-α,β-dehydro-α-amino Acid Derivatives

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The opening of the pyranone ring in 2*H*-naphtho[1,2-*b*]pyran-2-one derivative (**1**) and 3*H*-naphtho[2,1-*b*]-pyran-3-one derivatives **8** and **20** with nucleophiles afforded 3-(naphthyl-1)- and 3-(naphthyl-2)propenoates (substituted β-naphthyl-α,β-dehydro-α-amino acid derivatives) **7**, **13**, **14**, **15**, **24**, and **35**.

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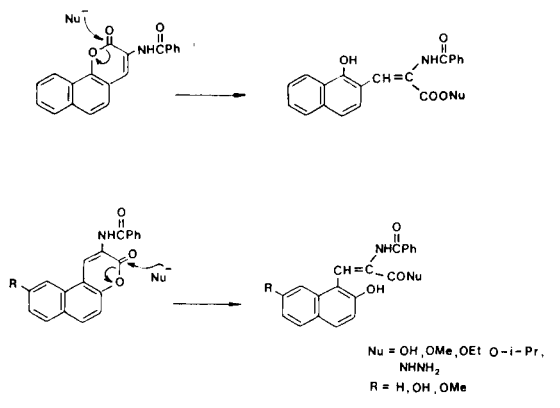
Methyl (*Z*)-2-benzoylamino-3-dimethylaminopropenoate [1,2] has been used as a reagent which gives with aliphatic or cyclic active methylene compounds or potentially active methylene compounds with two hydroxy or potentially hydroxy groups in 1,3-position 3-benzoylamino-2*H*-pyran-2-ones [3] and the corresponding derivatives of condensed systems, such as 2*H*-1-benzopyran-2-ones, pyranopyrazoles, and pyranopyrimidines [4]. With heterocyclic active methylene compounds methyl 2-benzoylamino-3-heteroarylpropenoates (methyl β-heteroaryl-α,β-dehydro-α-aminopropenoates) as intermediates have been isolated [4,5] and in some instances it has been shown either by chemical transformations or by X-ray analysis that they exist in (*Z*)-form [6].

Recently, benzoylamino derivatives of isomeric naphthopyranones and naphthodipyranones have been prepared from mono and dihydroxynaphthalenes and methyl 2-benzoylamino-3-dimethylaminopropenoate in a single step procedure [7]. Since, under the employed reaction conditions the corresponding methyl 2-benzoylamino-3-(naphthyl-1)- or 2-benzoylamino-3-(naphthyl-2)-propenoates, as representatives of β-aryl-α,β-dehydro-α-amino acids, could not be isolated, we tried to prepare this type of compounds by opening of the pyranone ring in the fused systems with nucleophiles attacking the polarized carbonyl group (Scheme 1).

Scheme 1

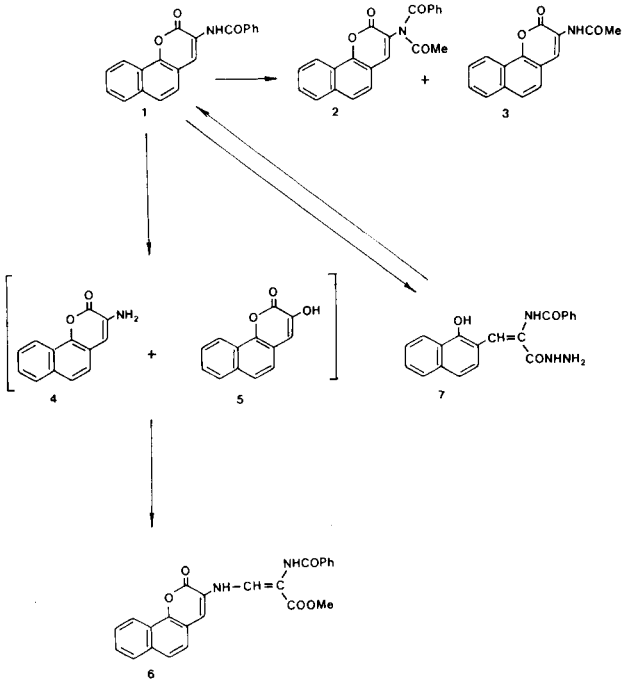


In this connection the following derivatives of isomeric naphthopyranones were selected: 3-benzoylamino-2*H*-naphtho[1,2-*b*]pyran-2-one (**1**) [7], 2-benzoylamino-3*H*-naphtho[2,1-*b*]pyran-3-one (**8**) [7] and its 9-hydroxy **16** [7] and 9-methoxy **20** derivatives. Acetylation of **1** with acetyl chloride in pyridine gave a mixture of *N*-acetyl-*N*-benzoyl derivative **2** and *N*-acetyl derivative **3**, while **8** produced only *N*-acetyl-*N*-benzoyl derivative **9**. Hydrolysis of the benzoylamino group was achieved only under vigorous reaction conditions by heating the compounds **1**, **8**, and **16** in a mixture of glacial acetic acid and aqueous hydrochloric acid (37%) in the presence of zinc chloride. In all cases a mixture of the corresponding amino compounds and hydroxy compounds as a result of the hydrolysis of the amino group were formed. Thus, the compounds **1**, **8**, **16** and **20** gave the corresponding pairs of **4** and **5**, **10** and **11**, **17** and **18**, and **21** and **22**, respectively. Since these pairs were inseparable by the usual chromatographic methods, the mixtures were treated with methyl 2-benzoylamino-3-dimethylaminopropenoate in order to convert the amino compounds into *N*-substituted methyl 2-benzoylamino-3-aminopropenoates **6**, **12**, **13** and **24**, respectively. These compounds were easily separated from the contaminating hydroxy compounds in boiling 2-propanol, in which the hydroxy compounds **5**, **11**, **18**, and **22** are soluble.

The hydrolysis of **8** with ethanolic potassium hydroxide solution gave substituted propenoic acid **13**, while **16** was, due to its insolubility, first transformed with *N*,*N*-dimethylformamide dimethyl acetal (DMFDMA) into its 9-methoxy derivative **20**, which afforded with ethanolic potassium hydroxide solution the propenoic acid derivative **24**. The acid derivative **13** and **24** were esterified with lower alcohols to give the corresponding esters **14a,b** and **25a-c**, respectively. Treatment of **1** and **8** with hydrazine hydrate yield the corresponding hydrazides **7** and **15**. We observed, that the acid **13** and its esters **14a,b** cyclize by heating at melting point for several hours into naphthopyranones **8**, and esters **25a,b** into naphthopyranone **20**, while the hydrazide **7** cyclize by heating in acetic acid for 6 hours into naphthopyranone **1** (Schemes 2-4). Ammonia,

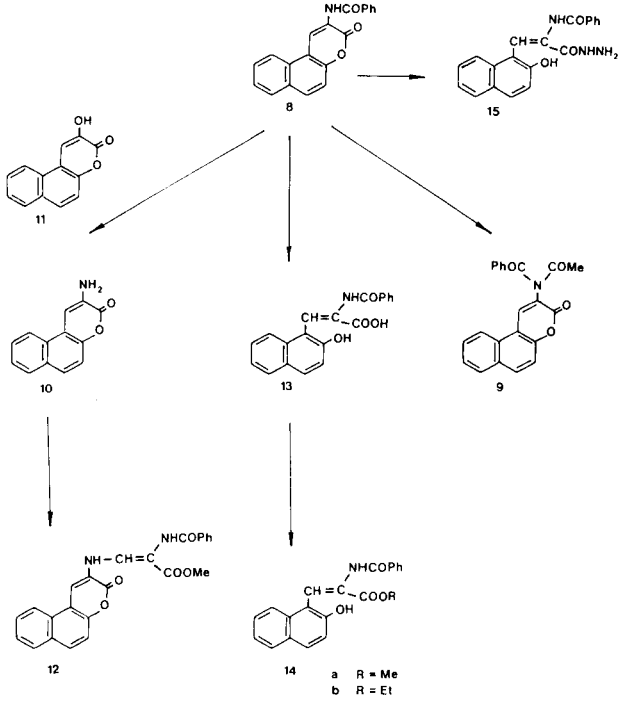
hydroxylamino and sodium glycinate as nucleophiles do not react with isomeric naphthopyranone derivatives.

Scheme 2

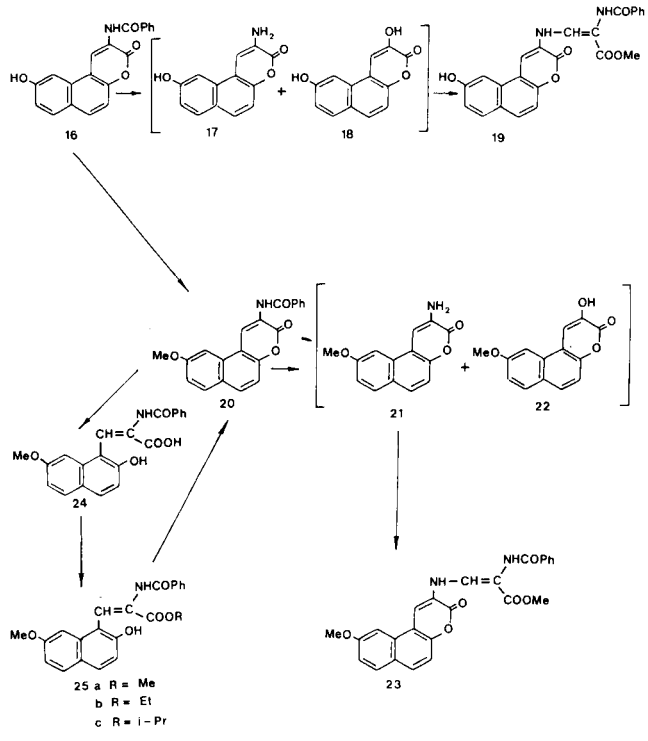


The structure of the new compounds were determined on the basis of elemental analyses for C, H, and N, and the ^{1}H nmr spectra.

Scheme 3



Scheme 4



EXPERIMENTAL

Melting points were determined on a Kofler micro hot stage. The ^{1}H nmr spectra were recorded on a JEOL FX 90Q FT spectrometer with TMS as internal standard. Elemental analyses for C, H, and N were obtained on a PERKIN-ELMER CHN Analyser 2400.

The following compounds were prepared according to the procedures described in the literature: methyl 2-benzoylamino-3-dimethylaminopropenoate [2], 3-benzoylamino-2*H*-naphtho[1,2-*b*]pyran-2-one (**1**) [3], 2-benzoylamino-3*H*-naphtho[2,1-*b*]pyran-3-one (**8**) [3] and 2-benzoylamino-9-hydroxy-3*H*-naphtho[2,1-*b*]pyran-3-one (**16**) [7].

3-(*N*-Acetyl-*N*-benzoyl)amino-2*H*-naphtho[1,2-*b*]pyran-2-one (**2**) and 3-Acetylamino-2*H*-naphtho[1,2-*b*]pyran-2-one (**3**).

A suspension of **2** (315 mg, 0.001 mole) in a mixture of pyridine (2.5 ml) and acetic anhydride (2.5 ml) was heated under reflux for 7 hours. The volatile components were evaporated *in vacuo*, methanol (1 ml) was added to the residue and the solid collected by filtration to give a mixture of **2** and **3** (322 mg), which was separated by chromatothrone (silica gel and chloroform as eluent) to give **2** as the first fraction, yield 75 mg (21%), mp 196-197° (from methanol); ^{1}H nmr (DMSO-d_6): δ 2.53 (s, 3H, COMe), 7.37-8.47 (m, 11H, H_5, H_6, H_7, H_8, H_9, H_{10}, PhCO), 8.50 (s, 1H, H_4).

Anal. Calcd. for $C_{22}H_{15}NO_4$: C, 73.94; H, 4.23; N, 3.91. Found: C, 73.92; H, 4.28; N, 3.91.

The second fraction gave, after evaporation of chloroform *in vacuo*, **3** (59 mg, 23%), mp 263-265° (from methanol); H nmr (DMSO-d_6): δ 2.23 (s, 3H) and 2.40 (s, 3H) (2 x COMe), 7.60-8.40 (m, 6H, H_5, H_6, H_7, H_8, H_9, H_{10}), 8.43 (s, 1H) and 8.80 (s, 1H).

Anal. Calcd. for $C_{15}H_{11}NO_3$: C, 71.14; H, 4.38; N, 5.53. Found: C, 70.93; H, 4.54; N, 5.29.

Methyl 2-Benzoylamino-3-(2-oxo-2*H*-naphtho[1,2-*b*]pyranyl-3)-aminopropenoate (**6**).

A mixture of **1** (315 mg, 0.001 mole), zinc chloride (500 mg) in acetic acid (20 ml) and hydrochloric acid (36%, 20 ml) was heated under reflux for 10 hours. The volume was reduced *in vacuo* to 10 ml, refrigerated and the precipitate was collected by filtration to give a mixture of **4** and **5** (206 mg in a ratio 1:2). Attempts to separate this mixture by chromatography were unsuccessful. The mixture was used in further transformation into **6**.

The mixture obtained above (206 mg) and methyl 2-benzoylamino-3-dimethylaminopropenoate (248 mg, 0.001 mole) dissolved in acetic acid (2 ml) was heated under reflux for 2 hours. Ethanol (1 ml) and water (1 ml) were added to the reaction mixture after cooling to room temperature. The precipitate was collected by filtration to give **6** (144 mg, 35%), mp 237-239° (from ethanol); ^{1}H nmr (DMSO-d_6): δ 3.70 (s, 3H, OCOMe), 7.47-8.33 (m, 13H, H_4, H_5, H_6, H_7, H_8, H_9, H_{10}, CH=C, PhCO), 9.57 (br s, 1H, NHCO).

Anal. Calcd. for $C_{24}H_{18}N_2O_5$: C, 69.56; H, 4.38; N, 6.77. Found: C, 69.47; H, 4.46; N, 6.64.

2-Benzoylamino-3-(1-hydroxynaphthyl-2)propenoic Acid Hydrazide (**7**).

To a suspension of **1** (315 mg, 0.001 mole) in ethanol (15 ml) hydrazine hydrate (80%, 2.5 ml) was added and the mixture was heated under reflux for 6 hours. The precipitate was, after cooling to room temperature, collected by filtration to give **7** (151 mg, 44%), mp 249-250° (from a mixture of DMF and ethanol); ^{1}H nmr (DMSO-d_6): δ 5.0 (br s, 2H, N*H*$_2$NH), 7.37-8.40 (m, 12H, H_3, H_4, H_5, H_6, H_7, H_8, CH=C, PhCO), 9.00 (br s, 1H, NH$_2$N*H*), 9.80 (br s, 1H, NHCO).

Anal. Calcd. for $C_{20}H_{17}N_3O_3$: C, 69.15; H, 4.93; N, 12.10. Found: C, 69.06; H, 5.06; N, 12.06.

2-(*N*-Acetyl-*N*-benzoyl)amino-3*H*-naphtho[2,1-*b*]pyran-3-one (**9**).

A mixture of **8** (315 mg, 0.001 mole), acetic anhydride (2.5 ml) and pyridine (2.5 ml) was heated under reflux for 7 hours. The volatile components were evaporated *in vacuo*, methanol (1 ml) was added to the residue and the solid residue was collected by filtration and purified by chromatothrone (silica gel, and chloroform as eluent) to give **9** (72 mg, 23%), mp 175-179° (from methanol); ^{1}H nmr (DMSO-d_6): δ 2.50 (s, 3H, COMe), 7.40-8.63 (m, 11H, H_5, H_6, H_7, H_8, H_9, H_{10}, PhCO), 9.33 (s, 1H, H_1).

Anal. Calcd. for $C_{22}H_{15}NO_4$: C, 73.94; H, 4.23; N, 3.91. Found: C, 73.61; H, 4.36; N, 3.96.

2-Amino-3*H*-naphtho[2,1-*b*]pyran-3-one (**10**) and 2-Hydroxy-3*H*-naphtho[2,1-*b*]pyran-3-one (**11**).

A mixture of **8** (315 mg, 0.001 mole), hydrochloric acid (36%, 25 ml), acetic acid (25 ml) and zinc chloride (500 mg) was heated under reflux for 10 hours. The volume of the reaction mixture was reduced *in vacuo* to 10 ml and cooled to 0°. The precipitate was then collected by filtration to give a mixture of **10** and **11**. Several crystallizations from 2-propanol gave pure **11** (71 mg, 33%), mp 245-247°; ^{1}H nmr (DMSO-d_6): δ 7.73-8.40 (m, 7H, H_1, H_5, H_6, H_7, H_8, H_9, H_{10}).

Anal. Calcd. for $C_{13}H_8O_3$: C, 73.58; H, 3.80. Found: C, 73.50; H, 3.71.

Evaporation of combined filtrates obtained by crystallization of **11** gave crude **10** (212 mg). Since this compound was not possible to obtain in pure form, it was used directly in the preparation of **12**.

Methyl 2-Benzoylamino-3-(3-oxo-3*H*-naphtho[2,1-*b*]pyranyl-2)-aminopropenoate (**12**).

A mixture of the crude **10** (212 mg) and methyl 2-benzoylamino-3-dimethylaminopropenoate (248 mg, 0.001 mole) in acetic acid (1.5 ml) was heated under reflux for 1.5 hours. The precipitate, formed after cooling to room temperature, gave a mixture of **11** and **12**. The mixture was suspended in boiling 2-propanol (10 ml) in which **11** is soluble. The solid residue was collected by filtration to give pure **12** (106 mg, 26%), mp 282-284° (from a mixture of ethanol and chloroform); ^{1}H nmr (DMSO-d_6): δ 3.80 (s, 3H, OCOMe), 7.50-8.87 (m, 13H, H_1, H_5, H_6, H_7, H_8, H_9, H_{10}, CH=C, PhCO), 9.60 (s, 1H, NHCO).

Anal. Calcd. for $C_{24}H_{18}N_2O_5$: C, 69.56; H, 4.38; N, 6.77. Found: C, 69.13; H, 4.57; N, 6.54.

2-Benzoylamino-3-(2-hydroxynaphthyl-1)propenoic Acid (**13**).

A mixture of **8** (315 mg, 0.001 mole) and potassium hydroxide (240 mg) in anhydrous ethanol (40 ml) was heated under reflux for 6 hours. The solvent was evaporated *in vacuo*, water (5 ml) was added to the residue and the solution was cooled to 0° and adjusted with hydrochloric acid to *p*H 3. The precipitate was collected by filtration to give **13** (295 mg, 88%), mp 201-202° (from a mixture of DMF and ethanol); ^{1}H nmr (DMSO-d_6): δ 7.17-7.90 (m, 12H, H_3, H_4, H_5, H_6, H_7, H_8, CH=C, PhCO), 9.50 (br s, 1H, NHCO).

Anal. Calcd. for $C_{20}H_{15}NO_4$: C, 72.06; H, 4.53; N, 4.20. Found: C, 72.23; H, 4.56; N, 4.09.

Methyl 2-Benzoylamino-3-(2-hydroxynaphthyl-1)propenoate (**14a**).

To a suspension of **13** (333 mg, 0.001 mole) in anhydrous methanol hydrogen chloride gas was bubbled until all the starting material was dissolved, followed by heating for 2 hours at 50°. After cooling to 0° the precipitate was formed, which was collected by filtration to give **14a** (176 mg, 73%). The compound cyclized by heating above 190° into **24**; ^{1}H nmr (DMSO-d_6): δ 3.80 (s, 3H, OCOMe), 7.17-7.92 (m, 12H, H_3, H_4, H_5, H_6, H_7, H_8, CH=C, PhCO), 9.53 (br s, 1H, NHCO), 10.63 (br s, 1H, OH).

Anal. Calcd. for $C_{21}H_{17}NO_4$: C, 72.61; H, 4.93; N, 4.03. Found: C, 72.53; H, 5.04; N, 3.97.

In the same manner the following compound was prepared:

Ethyl 2-Benzoylamino-3-(2-hydroxynaphthyl-1)propenoate (**14b**).

This compound was prepared from **13** and ethanol in 61% yield, mp 196-198° dec (from chloroform); ^{1}H nmr (DMSO-d_6): δ 1.30 (t, 3H, MeCH$_2$), 4.27 (q, 2H, MeC*H*$_2$), 7.10-7.87 (m, 12H, H_3, H_4, H_5, H_6, H_7, H_8, CH=C, PhCO), 9.50 (br s, 1H, NHCO).

Anal. Calcd. for $C_{22}H_{19}NO_4$: C, 73.12; H, 5.29; N, 3.87. Found: C, 73.09; H, 5.30; N, 3.75.

2-Benzoylamino-3-(2-hydroxynaphthyl-1)propenoic Acid Hydrazide (**15**).

To a suspension of **8** (315 mg, 0.001 mole) in ethanol (25 ml) and DMF (5 ml) hydrazine hydrate (80%, 5 ml) was added and the mixture was heated under reflux for 8 hours. The volatile components were evaporated *in vacuo*. Chloroform (15 ml) was added to the residue and the suspension was stirred at room temperature for two hours in order to remove the starting material. The solid was collected by filtration to give **15** (127 mg, 37%), mp 235-237° (from a mixture of DMF and ethanol); ^{1}H nmr (DMSO-d_6): δ 5.53 (br s, 2H, N*H*$_2$NH), 7.07-8.02 (m, 12H, H_3, H_4, H_5, H_6, H_7, H_8, CH=C, PhCO), 8.67 (br s) and 8.80 (br s) (1H, NH$_2$N*H*), 9.63 (br s, 1H, NHCO).

Anal. Calcd. for $C_{20}H_{17}N_3O_3$: C, 69.15; H, 4.93; N, 12.10. Found: C, 69.25; H, 4.96; N, 11.82.

Methyl 2-Benzoylamino-3-(9-hydroxy-3-oxo-3*H*-naphtho[2,1-*b*]-pyranyl-2)propenoate (**19**).

A mixture of **16** (331 mg, 0.001 mole), zinc chloride (500 mg) in hydrochloric acid (36%, 30 ml) and acetic acid (30 ml) was heated under reflux for 10 hours. The unreacted starting material was filtered off and the volume of the filtrate was reduced by evaporation *in vacuo* to 10 ml. The precipitate was, after cooling to 0°, collected by filtration to give a mixture of **17** and **18** (157 mg) in a ratio 4:1. The mixture was not possible to separate by chromatography and was used in the preparation of **19**.

The above mixture (227 mg) and methyl 2-benzoylamino-3-dimethylaminopropenoate (248 mg, 0.001 mole), dissolved in acetic acid (2.5 ml), was heated under reflux for 1 hour. The precipitate was, after cooling to room temperature, collected by filtration to give **19** (143 mg, 33%), mp 250° dec (from ethanol); ^{1}H nmr (DMSO-d_6): δ 3.83 (s, 3H, MeOCO), 7.07-8.60 (m, 12H, H_1, H_5, H_6, H_7, H_8, H_{10}, CH=C, PhCO), 9.57 (s, 1H, NHCO), 10.33 (br s, 1H, OH).

Anal. Calcd. for $C_{24}H_{18}N_2O_6$: C, 69.56; H, 4.38; N, 6.76. Found: C, 69.35; H, 4.40; N, 6.56.

2-Benzoylamino-9-methoxy-3*H*-naphtho[2,1-*b*]pyran-3-one (**20**).

A mixture of **16** (993 mg, 0.03 mole) and DMFDMA (7.14 g, 0.06 mole) in toluene (200 ml) was heated under reflux for 10 hours. The precipitate was, after cooling to room temperature, collected by filtration and extracted with boiling chloroform (3 times, 100 ml each time). The combined extracts were evaporated *in vacuo* to give **20** (3.78 g, 37%), mp 262-264° (from a mixture of DMF and ethanol); ^{1}H nmr (DMSO-d_6): δ 3.99 (s, 3H, MeO), 7.19-8.03 (m, 10H, H_5, H_6, H_7, H_8, H_{10}, PhCO), 9.26 (br s, 2H, H_1, NHCO).

Anal. Calcd. for $C_{21}H_{15}NO_5$: C, 73.04; H, 4.37; N, 4.06. Found: C, 72.87; H, 4.35; N, 3.92.

Methyl 2-Benzoylamino-3-(9-methoxy-3-oxo-3*H*-naphtho[2,1-*b*]-pyranyl-2)aminopropenoate (**23**).

To a suspension of **20** (345 mg, 0.001 mole) in a mixture of hydrochloric acid (36%, 30 ml) and acetic acid (30 ml) zinc chloride (500 mg) was added and the reaction mixture was heated under reflux for 10 hours. The solid residue was, after cooling, filtered off. The volume of the filtrate was reduced *in vacuo* to 10 ml and the precipitate, formed after cooling to 0°, was collected by filtration to give a mixture of **21** and **22** (141 mg). The mixture was without separation used in transformation into **23**.

The above mixture (240 mg) and methyl 2-benzoylamino-3-dimethylaminopropenoate (248 mg, 0.001 mole) acetic acid (2 ml) was heated under reflux for 30 minutes. The precipitate was, after cooling to room temperature, collected by filtration to give **23** (148 mg, 33%), mp 267-270° (from a mixture of ethanol and chloroform); ^{1}H nmr (DMSO-d_6): δ 3.68 (s, 3H, OCOMe), 3.97 (s, 3H, MeO), 7.10-8.10 (m, 10H, H_5, H_6, H_7, H_8, H_{10}, PhCO), 8.20 (d, 1H, NHC*H*), 8.47 (s, 1H, H_1), 9.60 (s, 1H, NHCO), 11.3 (d, 1H, NHCH), J_{NHCH} = 13.0 Hz.

Anal. Calcd. for $C_{25}H_{20}N_2O_6$: C, 67.56; H, 4.53; N, 6.30. Found: C, 67.77; H, 4.56; N, 6.23.

2-Benzoylamino-3-(2-hydroxy-7-methoxynaphthyl-1)propenoic Acid (**24**).

A mixture of **20** (345 mg, 0.001 mole) and potassium hydroxide (240 mg) in ethanol was heated under reflux for 4 hours. The solvent was evaporated *in vacuo*, water (20 ml) was added to the residue and resulting solution, cooled to 0°, was neutralized with hydrochloric acid. The precipitate was collected by filtration and washed with chloroform in order to remove the starting material to give **24** (174 mg, 48%), mp 210° dec (from a mixture of DMF and water); ^{1}H nmr (DMSO-d_6): δ 3.83 (s, 3H, MeO), 6.87-7.83 (m, 11H, H_3, H_4, H_5, H_6, H_8, CH=C, PhCO), 9.60 (br s, 1H, NHCO).

Anal. Calcd. for $C_{21}H_{17}NO_5 \cdot H_2O$: C, 66.13; H, 5.02; N, 3.67. Found: C, 66.40; H, 4.66; N, 3.77.

Methyl 2-Benzoylamino-3-(2-hydroxy-7-methoxynaphthyl-1)propenoate (**25a**).

To a suspension of **24** (363 mg, 0.001 mole) in anhydrous methanol (25 ml) hydrogen chloride was bubbled until all the starting material was dissolved. The reaction mixture was then heated for 2 hours at 50°. The solvent was evaporated *in vacuo*. Methanol (3 ml) was added to the residue and the precipitate was collected by filtration to give **25a** (184 mg, 49%), the compound cyclizes by heating into **20** (from chloroform); ^{1}H nmr (DMSO-d_6): δ 3.77 (s, 6H, MeO, MeOCO), 6.77-7.83 (m, 11H, H_3, H_4, H_5, H_6, H_8, CH=C, PhCO), 9.67 (br s, 1H, NHCO).

Anal. Calcd. for $C_{22}H_{19}NO_5$: C, 70.02; H, 5.07; N, 3.71. Found: C, 69.88; H, 5.21; N, 3.54.

In the same manner the following compounds were obtained:

Ethyl 2-Benzoylamino-3-(2-hydroxy-7-methoxynaphthyl-1)propenoate (**25b**).

This compound was obtained from **24** and ethanol in 45% yield, the compound cyclizes by heating into **20** (from a mixture of chloroform and cyclohexane); ^{1}H nmr (DMSO-d_6): δ 1.27 (t, 3H, *Me*CH_2), 3.77 (s, 3H, MeO), 4.30 (q, 2H, MeC*H*$_2$), 6.73-7.83 (m, 11H, H_3, H_4, H_5, H_6, H_8, CH=C, PhCO), 9.57 (br, 1H, NHCO).

Anal. Calcd. for $C_{23}H_{21}NO_5$: C, 70.58; H, 5.41; N, 3.58. Found: C, 70.79; H, 5.58; N, 3.49.

2-Propyl 2-Benzoylamino-3-(2-hydroxy-7-methoxynaphthyl-1)-propenoate (**25c**).

This compound was prepared from **24** and 2-propanol in 36% yield, mp 255-257° (from chloroform); ^{1}H nmr (DMSO-d_6): δ 1.23 (d, 6H, *Me*$_2$CH), 3.77 (s, 3H, MeO), 5.05 (hept, 1H, Me$_2$C*H*), 6.77-7.80 (m, 11H, H_3, H_4, H_5, H_6, H_8, CH=C, PhCO), 9.60 (br s, NHCO).

Anal. Calcd. for $C_{24}H_{23}NO_5 \cdot H_2O$: C, 68.07; H, 5.95; N, 3.30. Found: C, 68.44; H, 5.57; N, 3.58.

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