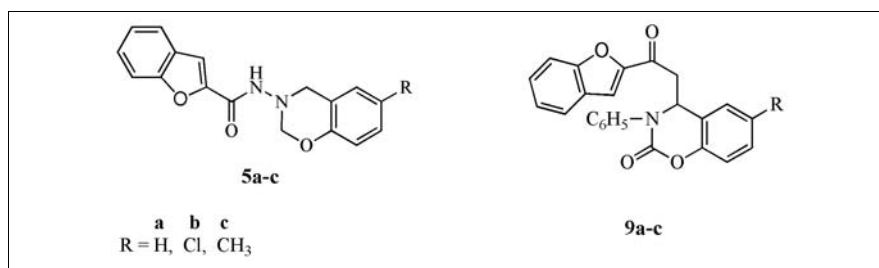


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New benzofuranyl-1,3-benzoxazines and 1,3-benzoxazin-2-ones are synthesized in which benzofuran is coupled with 1,3-benzoxazines and 1,3-benzoxazin-2-ones through -CONH- and -COCH₂- bridges, respectively. The antimicrobial activity of these compounds is reported.

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Introduction.

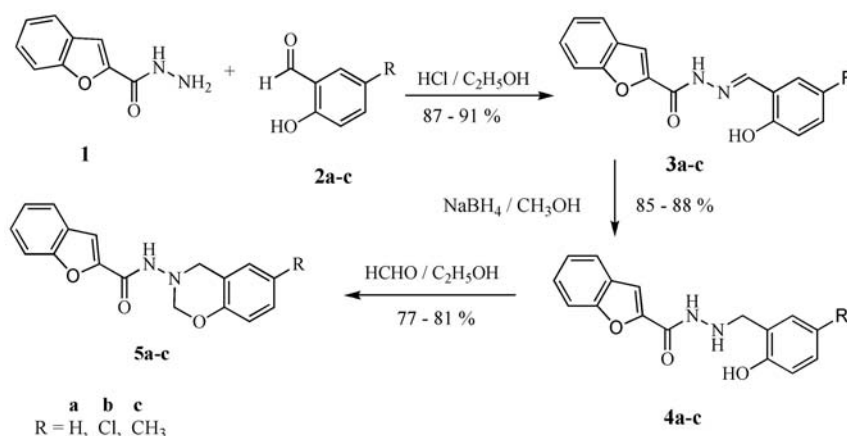
Among a wide variety of heterocycles that have been explored for developing pharmaceutically important molecules, benzofurans, 1,3-benzoxazines and 1,3-benzoxazinones have played an important role in medicinal chemistry. Benzofuran derivatives have acquired a special place in the heterocyclic field because of their broad spectrum of biological activities [1]. Many 1,3-benzoxazine derivatives are reported to possess antibacterial [2,3], antifungal [4,5], analgesic [6], anti-inflammatory [6], smooth muscle relaxant and spermicidal activities [7]. A number of 1,3-benzoxazinone derivatives have been synthesized and tested for their antimicrobial [8,9] and analgesic properties [10]. Furthermore, few furodihydrobenzoxazines have found the inhibitory effect on some of the gram positive and gram negative bacteria and fungi [11]. Prompted by the varied biological activities of 1,3-

benzoxazines, and 1,3-benzoxazinones and in continuation of our investigation on pharmacologically active benzofuran derivatives [12-14], it was thought of interest to synthesize a new series of 2*H*-3-(benzofuran-2-carboxamidylyl)-3,4-dihydro-1,3-benzoxazines **5a-c** and 3-phenyl-3,4-dihydro-4-(benzofuran-2-acetyl)-2*H*-1,3-benzoxazin-2-ones **9a-c** and to screen them for antimicrobial properties.

Results and Discussion.

Benzofuran-2-carbohydrazide [15] **1** was used as a key starting material for the synthesis of compounds **5a-c** (Scheme 1). First, benzofuran-2-carbohydrazide **1** was reacted with salicylaldehyde **2a** in ethanol in presence of catalytic amount of hydrochloric acid to give 2-(benzofuran-2-carboxamidylimino)methylphenol **3a**. Then the reduction of **3a** using sodium borohydride in methanol gave 2-(benzofuran-2-carboxamidylamino)methylphenol **4a**. The treatment of **4a** with formaldehyde in ethanol underwent

Scheme 1



internal Mannich reaction to give **5a**. Benzofuran-2-carbohydrazide **1** was similarly reacted with 5-chloro and 5-methyl salicylaldehydes in ethanol to get **3b** and **3c**. The resulting compounds **3b** and **3c** were reduced to **4b** and **4c** using sodium borohydride and finally subjected to internal Mannich reaction using formaldehyde to get **5b** and **5c**. The structures of all the products were in good agreement with spectral data and elemental analysis.

The above described approach was suitable for coupling benzofuran with 1,3-benzoxazine moiety through -CONH- bridge. An alternative approach which led to similar coupling of these two heterocycles through -COCH₂- bridge instead of -CONH- is also successfully accomplished (Scheme 2). This method required benzofuran analogues of chalcone as the starting materials. Hence such chalcone analogues **7a-c** were prepared from 2-acetylbenzofuran [16] and salicylaldehydes **2a-c** under the conditions of Claisen-Schmidt reaction. The structures of 3-(2-hydroxyphenyl)-1-benzofuran-2-yl-2-propen-1-ones **7a-c** were confirmed by spectroscopic data and elemental analysis. The chalcone analogues **7a-c** when reacted with phenylisocyanate in benzene and in presence of catalytic amount of potassium hydroxide gave 3-phenyl-3,4-dihydro-4-(benzofuran-2-acetyl)-2H-1,3-benzoxazin-2-ones **9a-c**, presumably through the intermediate open chain carbamate **8a-c**. The structures of the products **9a-c** were inferred from elemental analysis and spectral data. The IR spectra (potassium bromide) of **9a-c** which lack ν_{NH} excluded the possible open chain carbamate structures **8a-c**. IR spectra of **9a-c** contained two distinct bands at 1666-1673 and 1718-1725 cm⁻¹ region due to ketonic and carbomoyloxy carbonyls, respectively. The structures of all the products were in good agreement with spectral data and elemental analysis.

In summary, new benzofuran-yl-1,3-benzoxazines **5a-c** and 1,3-benzoxazin-2-ones **9a-c** are synthesized in minimum number of steps wherein benzofuran is coupled with 1,3-

benzoxazines and 1,3-benzoxazin-2-ones through -CONH- and -COCH₂- bridges, respectively. These coupled heterocycles demonstrated some promising antimicrobial activity.

Antimicrobial Activity.

Compounds **5a-c** and **9a-c** were evaluated *in vitro* for their antibacterial activity against *E. coli*, *P. aeruginosa*, *S. aureus* and *B. subtilis* using dimethyl formamide (DMF) as a solvent by cup-plate method [17]. All compounds were dissolved in DMF for antimicrobial activity. The activity was compared with Gentamycin and Ciprofloxacin. The results are presented in Table 1. Antifungal activity of compounds **5a-c** and **9a-c** were evaluated against *A. niger* and *C. albicans* using dimethyl formamide as a solvent by cup-plate method [17]. The activity was compared with Griseofulvin and Fluconazole. The results are presented in Table 2. The zone of inhibition was measured after 24 h of incubation at 37 °C. The zone of inhibition developed, if any, was then accurately measured and recorded. Each zone of inhibition recorded was average of six measurements. Zone of inhibition for DMF was done separately and found that there was no activity.

Table 1
Antibacterial Activity (zone of inhibition in mm)

Compd.	R	<i>E.coli</i>	<i>P. aeruginosa</i>	<i>S. aureus</i>	<i>B. subtilis</i>
5a	H	16	17	12	16
5b	Cl	15	19	18	19
5c	CH ₃	16	16	14	13
9a	H	16	18	16	14
9b	Cl	18	17	19	19
9c	CH ₃	15	14	18	12
Standards:					
Gentamycin		20	23	22	22
Ciprofloxacin		28	26	24	32

Scheme 2

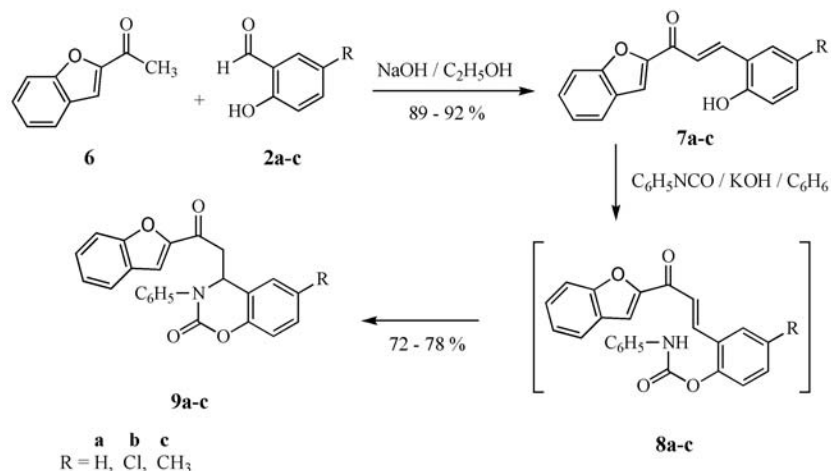


Table 2
Antifungal Activity (zone of inhibition in mm)

Compd.	R	<i>A. niger</i>	<i>C. albicans</i>
5a	H	12	14
5b	Cl	13	13
5c	CH ₃	-	14
9a	H	10	-
9b	Cl	13	12
9c	CH ₃	11	-
Standards:			
Griseofulvin		16	18
Fluconazole		18	21

EXPERIMENTAL

Melting points were determined in open capillary tubes and are uncorrected. IR spectra (potassium bromide) were recorded on Perkin Elmer FTIR 1615 double beam spectrophotometer ($\nu_{\max}$ in cm^{-1}), ^1H nmr spectra were recorded at 300 MHz (on a Bruker AM-300) instrument using dimethyl sulfoxide- d_6 as a solvent (chemical shifts in δ , ppm downfield from TMS as internal reference), and mass spectra on a Varian MAT CH-5 and CH-7 instruments at 70 eV.

Preparation of Compounds **3a-c**.

General Procedure.

Benzofuran-2-carbohydrazide **1** (1.76 g, 0.01 mol) and salicylaldehyde/5-substituted salicylaldehyde (0.01 mol) were refluxed in ethanol (50 mL) containing two drops of concentrated hydrochloric acid for 45 minutes. Crystalline solid which separated on cooling was collected and crystallized from the suitable solvent.

2-(Benzofuran-2-carboxamidylimino)methylphenol (**3a**).

This compound was obtained in 87 % yield; pale yellow crystals from ethanol; mp 218-220 °C; ir (potassium bromide): 3400 (OH), 3250 (NH), 1660 (CO), 1625 (CN) cm^{-1} ; ^1H nmr (dimethyl sulfoxide- d_6): δ 6.87-7.80 (m, 9H, aromatic protons), 8.75 (s, 1H, CH), 11.22 (s, 1H, OH, deuterium oxide-exchangeable), 12.48 (s, 1H, NH, deuterium oxide-exchangeable); MS (EI, 70 eV): m/z 280.

Anal. Calcd. for $\text{C}_{16}\text{H}_{12}\text{N}_2\text{O}_3$: C, 68.56; H, 4.32; N, 9.99 %. Found: C, 68.43; H, 4.58; N, 10.06.

2-(Benzofuran-2-carboxamidylimino)-5-chloro-methylphenol (**3b**).

This compound was obtained in 91 % yield; pale yellow crystals from ethanol; mp 293-295 °C; ir (potassium bromide): 3416 (OH), 3273 (NH), 1662 (CO), 1621 (CN) cm^{-1} ; ^1H nmr (dimethyl sulfoxide- d_6): δ 6.93-7.78 (m, 8H, aromatic protons), 8.48 (s, 1H, CH), 10.05 (s, 1H, OH, deuterium oxide-exchangeable), 11.23 (s, 1H, NH, deuterium oxide-exchangeable); MS (EI, 70 eV): m/z 314, 316.

Anal. Calcd. for $\text{C}_{16}\text{H}_{11}\text{ClN}_2\text{O}_3$: C, 61.06; H, 3.52; N, 8.90 %. Found: C, 61.27; H, 3.43; N, 8.95.

2-(Benzofuran-2-carboxamidylimino)-5-methyl-methylphenol (**3c**).

This compound was obtained in 89 % yield; pale yellow crystals from ethanol; mp 246-248 °C; ir (potassium bromide):

3423 (OH), 3326 (NH), 1664 (CO), 1618 (CN) cm^{-1} ; ^1H nmr (dimethylsulfoxide- d_6): δ 2.18 (s, 3H, CH₃), 6.80-7.60 (m, 8H, aromatic protons), 8.70 (s, 1H, CH), 9.40 (s, 1H, OH, deuterium oxide-exchangeable), 10.40 (s, 1H, NH, deuterium oxide-exchangeable); MS (EI, 70 eV): m/z 294.

Anal. Calcd. for $\text{C}_{17}\text{H}_{14}\text{N}_2\text{O}_3$: C, 69.38; H, 3.52; N, 9.52 %. Found: C, 69.54; H, 4.66; N, 9.48.

Preparation of Compounds **4a-c**.

General Procedure.

Sodium borohydride (0.3 g, 0.088 mol) was added to a solution of 2-(benzofuran-2-carboxamidylimino)methylphenols **3a-c** (0.005 mol) in methanol (20 mL) and the reaction mixture was stirred for 30 minutes. Then it was poured into cold water (50 mL). The product, which separated as a solid, was collected by filtration after washing with water and crystallized from proper solvent.

2-(Benzofuran-2-carboxamidylamino)methylphenol (**4a**).

This compound was obtained in 90 % yield; white crystals from ethanol; mp 175-177 °C; ir (potassium bromide): 3390 (OH), 3225 (NH), 3120 (NH), 1645 (CO) cm^{-1} ; ^1H nmr (dimethyl sulfoxide- d_6): δ 4.03 (s, 2H, CH₂), 5.7 (s, 1H, OH, deuterium oxide-exchangeable), 6.75-7.78 (m, 9H, aromatic protons), and 9.69 (s, 1H, NH, deuterium oxide-exchangeable), 10.45 (s, 1H, NH, deuterium oxide-exchangeable); MS (EI, 70 eV): m/z 282.

Anal. Calcd. for $\text{C}_{16}\text{H}_{14}\text{N}_2\text{O}_3$: C, 68.07; H, 5.00; N, 9.92 %. Found: C, 67.90; H, 5.18; N, 10.11.

2-(Benzofuran-2-carboxamidylamino)-5-chloro-methylphenol (**4b**).

This compound was obtained in 91 % yield; white crystals from ethanol; mp 293-295 °C; ir (potassium bromide): 3405 (OH), 3227 (NH), 3175 (NH), 1643 (CO) cm^{-1} ; ^1H nmr (dimethyl sulfoxide- d_6): δ 4.25 (s, 2H, CH₂), 6.79-7.89 (m, 8H, aromatic protons), 8.94 (s, 1H, OH, deuterium oxide-exchangeable), 9.94 (s, 1H, NH, deuterium oxide-exchangeable), 11.12 (s, 1H, NH, deuterium oxide-exchangeable); MS (EI, 70 eV): m/z 316, 318.

Anal. Calcd. for $\text{C}_{16}\text{H}_{13}\text{ClN}_2\text{O}_3$: C, 60.67; H, 4.14; N, 8.84 %. Found: C, 60.78; H, 4.25; N, 8.67.

2-(Benzofuran-2-carboxamidylamino)-5-methyl-methylphenol (**4c**).

This compound was obtained in 89 % yield; white crystals from ethanol; mp 246-248 °C; ir (potassium bromide): 3418 (OH), 3310 (NH), 3187 (NH), 1651 (CO) cm^{-1} ; ^1H nmr (dimethyl sulfoxide- d_6): δ 2.05 (s, 3H, CH₃), 4.17 (s, 2H, CH₂), 6.58-7.15 (m, 8H, aromatic protons), 7.49 (s, 1H, OH, deuterium oxide-exchangeable), 7.73 (s, 1H, NH, deuterium oxide-exchangeable), 8.89 (s, 1H, NH, deuterium oxide-exchangeable); MS (EI, 70 eV): m/z 296.

Anal. Calcd. for $\text{C}_{17}\text{H}_{16}\text{N}_2\text{O}_3$: C, 68.91; H, 5.44; N, 9.45 %. Found: C, 68.76; H, 5.67; N, 9.13.

Preparation of Compounds **5a-c**.

General Procedure.

Compounds **4a-c** (0.002 mol) and formalin (1 mL, 37%) were refluxed in ethanol (15 mL) for five hours. The reaction mixture was poured onto ice cold water (50 mL) and the product that separated was collected and crystallized from suitable solvent.

2*H*-3-(Benzofuran-2-carboxamidyl)-3,4-dihydro-1,3-benzoxazine (**5a**).

This compound was obtained in 77 % yield; white tiny crystals from ethanol; mp 156-158 °C; ir (potassium bromide): 1660 (CO), 1150 (C-O-C) cm^{-1} ; ^1H nmr (dimethyl sulfoxide- d_6): δ 4.45 (s, 2H, N-CH₂-C), 5.16 (N-CH₂-O), 8.43 (s, 1H, NH, deuterium oxide-exchangeable), 6.95-7.75 (m, 9H, aromatic protons); MS (EI, 70 eV): m/z 294.

Anal. Calcd. for C₁₇H₁₄N₂O₃: C, 69.38; H, 4.79; N, 9.52 %. Found: C, 69.55; H, 4.83; N, 9.28.

2*H*-3-(Benzofuran-2-carboxamidyl)-6-chloro-3,4-dihydro-1,3-benzoxazine (**5b**).

This compound was obtained in 81 % yield; white tiny crystals from ethanol; mp 197-199 °C; ir (potassium bromide): 1667 (CO), 1155 (C-O-C) cm^{-1} ; ^1H nmr (dimethyl sulfoxide- d_6): δ 4.40 (s, 2H, N-CH₂-C), 5.10 (N-CH₂-O), 8.40 (s, 1H, NH, deuterium oxide-exchangeable), 6.80-7.70 (m, 8H, aromatic protons); MS (EI, 70 eV): m/z 328, 330.

Anal. Calcd. for C₁₇H₁₃ClN₂O₃: C, 62.11; H, 3.99; N, 8.52 %. Found: C, 62.45; H, 4.18; N, 8.43.

2*H*-3-(Benzofuran-2-carboxamidyl)-6-methyl-3,4-dihydro-1,3-benzoxazine (**5c**).

This compound was obtained in 79 % yield; white tiny crystals from ethanol; mp 185-187 °C; ir (potassium bromide): 1670 (CO), 1147 (C-O-C) cm^{-1} ; ^1H nmr (dimethyl sulfoxide- d_6): δ 2.10 (s, 3H, CH₃), 4.30 (s, 2H, N-CH₂-C), 5.30 (N-CH₂-O), 6.95-7.70 (m, 8H, aromatic protons), 8.40 (s, 1H, NH, deuterium oxide-exchangeable); MS (EI, 70 eV): m/z 308.

Anal. Calcd. for C₁₈H₁₆N₂O₃: C, 70.12; H, 5.23; N, 9.09 %. Found: C, 70.43; H, 5.44; N, 9.17.

Preparation of Compounds **7a-c**.

General Procedure.

A solution of 2-acetylbenzofuran **6** (1.60 g, 0.01 mol) and salicylaldehyde/5-substituted salicylaldehyde (0.01 mol) in ethanol (25 mL) at ice cold temperature, was treated with aqueous solution of sodium hydroxide (70 %, 5 mL) in portions with constant stirring. The reaction mixture was further stirred for two hours at the same temperature and left overnight at room temperature. It was then poured into cold water (100 mL) and carefully acidified to pH 6.5 with 2 *N* hydrochloric acid. The separated solid was collected and crystallized from suitable solvent.

3-(2-Hydroxyphenyl)-1-benzofuran-2-yl-2-propen-1-one (**7a**).

This compound was obtained in 92 % yield; yellow silky crystals from aqueous ethanol; mp 178-181 °C; ir (potassium bromide): 3425 (OH), 1640 (CO) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 10.10 (s, 1H, OH, deuterium oxide-exchangeable), 6.90-8.05 (m, 11H, -COCH=CH- and aromatic protons); MS (EI, 70 eV): m/z 264.

Anal. Calcd. for C₁₇H₁₂O₃: C, 72.26; H, 4.58 %. Found: C, 72.02; H, 4.35.

3-(5-Chloro-2-hydroxyphenyl)-1-benzofuran-2-yl-2-propen-1-one (**7b**).

This compound was obtained in 91 % yield; yellow silky crystals from aqueous ethanol; mp 196-199 °C; ir (potassium bromide): 3410 (OH), 1646 (CO) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 6.23 (s,

1H, OH, deuterium oxide-exchangeable), 6.80-8.15 (m, 10H, -COCH=CH- and aromatic protons); MS (EI, 70 eV): m/z 298, 300.

Anal. Calcd. for C₁₇H₁₁ClO₃: C, 68.35; H, 3.71 %. Found: C, 68.11; H, 3.96.

3-(2-Hydroxy-5-methylphenyl)-1-benzofuran-2-yl-2-propen-1-one (**7c**).

This compound was obtained in 89 % yield; yellow silky crystals from aqueous ethanol; mp 182-185 °C; ir (potassium bromide): 3425 (OH), 1651 (CO) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 2.20 (s, 3H, CH₃), 6.10 (s, 1H, OH, deuterium oxide-exchangeable), 6.70-8.10 (m, 10H, -COCH=CH- and aromatic protons); MS (EI, 70 eV): m/z 278.

Anal. Calcd. for C₁₈H₁₄O₃: C, 77.68; H, 5.07 %. Found: C, 77.49; H, 5.26.

Preparation of Compounds **9a-c**.

General Procedure.

A mixture of chalcone derivatives **7a-c** (0.001 mol) and phenylisocyanate (0.0015 mol) in anhydrous benzene (15 mL) containing a catalytic amount of potassium hydroxide (10 mg) was refluxed for 30 minutes and the reaction mixture was allowed to cool. The solid that separated was collected by filtration, washed with pet ether and crystallized from suitable solvent.

3-Phenyl-3,4-dihydro-4-(benzofuran-2-acetyl)-2*H*-1,3-benzoxazin-2-one (**9a**).

This compound was obtained in 78 % yield; red crystals from benzene-pet ether; mp 224-227 °C; ir (potassium bromide): 1720 (COO), 1670 (CO) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 3.40 (d, *J* = 6.3 Hz, 2H, CH₂), 5.50 (t, *J* = 5.9 Hz, 1H, CH), 6.80-7.70 (m, 14H, aromatic protons); MS (EI, 70 eV): m/z 383.

Anal. Calcd. for C₂₄H₁₇NO₄: C, 75.19; H, 4.47; N, 3.65 %. Found: C, 75.40; H, 4.38; N, 3.37.

6-Chloro-3-phenyl-3,4-dihydro-4-(benzofuran-2-acetyl)-2*H*-1,3-benzoxazin-2-one (**9b**).

This compound was obtained in 75 % yield; brown crystals from ethyl acetate-pet ether; mp 218-220 °C; ir (potassium bromide): 1725 (COO), 1666 (CO) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 3.54 (d, *J* = 6.4 Hz, 2H, CH₂), 5.48 (t, *J* = 5.7 Hz, 1H, CH), 6.70-7.68 (m, 13H, aromatic protons); MS (EI, 70 eV): m/z 417, 419.

Anal. Calcd. for C₂₄H₁₆ClNO₄: C, 68.99; H, 3.86; N, 3.35 %. Found: C, 68.91; H, 3.94; N, 3.29.

6-Methyl-3-phenyl-3,4-dihydro-4-(benzofuran-2-acetyl)-2*H*-1,3-benzoxazin-2-one (**9c**).

This compound was obtained in 72 % yield; brown crystals from ethyl acetate-pet ether; mp 230-232 °C; ir (potassium bromide): 1718 (COO), 1673 (CO) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 2.10 (s, 1H, CH₃), 3.48 (d, *J* = 6.3 Hz, 2H, CH₂), 5.50 (t, *J* = 5.9 Hz, 1H, CH), 6.80-7.70 (m, 13H, aromatic protons); MS (EI, 70 eV): m/z 397.

Anal. Calcd. for C₂₅H₁₉NO₄: C, 75.55; H, 4.82; N, 3.35 %. Found: C, 68.91; H, 3.94; N, 3.29.

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