# Synthesis of Some Furo-Thiazolidine Derivatives Starting from Aldimines

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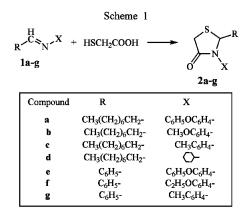
Alkyl and aryl substituted furothiazolidine derivatives have been synthesized by the reaction of Wittig reagent with benzylidene derivatives of 4-thiazolidinones obtained from aldimines and thioglicolic acid.

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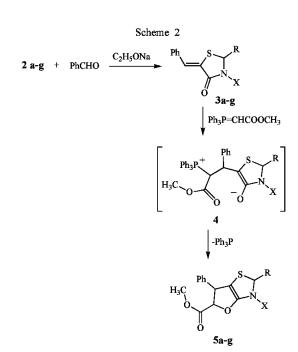
In recent years, interests of researchers have been focused on the heterocyclic systems which contain various heteroatoms such as nitrogen, sulphur and oxygen, because of their biological importance. As a part of our ongoing research on the preparations and reactions of 4-thiazolidinones, which show some biological activities such as antibiotic, diuretic, organoleptic, tuberculostatic, antileukemic and antiparasitical [1,2], we have reported recently the synthesis, transformations and some biological properties of 2-heteroaryl substituted 4-thiazolidinones [3-5].

Here we report the synthesis of some alkyl and aryl substituted furothiazolidine derivatives *via* the reactions of Wittig reagent with 5-benzylidene-4-thiazolidinones obtained from a series of nonaldimines and benzaldimines. Aldimines are used in the synthesis of nitrogen containing heterocycles [6,7], so the synthesis of 4-thiazolidinones have been accomplished starting from imines. Compounds **1a-d** are new compounds which were prepared from condensation of nonanal with *p*-phenoxyaniline, *p*-methoxyaniline, *p*-methylaniline and cyclohexylamine as described in experimental section.

The 3-aryl-2-nonyl- or phenyl-4-thiazolidinones (**2a-g**) (Scheme 1) were obtained by refluxing equimolar amounts of imines and thioglycolic acid in dry benzene. After purification, these compounds were subjected to aldol condensation with benzaldehyde in the presence of sodium ethoxide to give 5-benzylidene derivatives (**3a-g**) [4]. These were refluxed with methoxycarbonylmethylenet-riphenylphosphorane in ethyl acetate containing triethyl-



amine. Chromatographic separation produced successively two different substances. In every case, the first eluent contained triphenylphosphine, the second products were 5a-g (Scheme 2). This reaction applied for the first time to substituted thiazolidinone derivatives by our group to provide a new way for synthesizing furothiazolidines [5]. In this report, we chose aliphatic and aromatic substituted thiazolidinone derivatives to investigate the effect of substituent on the cyclisation reaction. Obviously, the dipolar indermediate 4 formed from the initial attack of the carbanion centre in the Wittig reagent on the active exocyclic electrophilic carbon atom of , -unsaturated system in **3a-g** undergoes *O*-alkylation with triphenylphosphine elimination to give 6-methoxycarbonyl-3substituted phenyl-7-phenyl-2-(nonyl or phenyl)dihydro-<sup>4</sup>-furo-[2,3-*d*]thiazolidines, **5a-g**.



The structures of the new compounds were firmly established on the basis of their ftir, <sup>1</sup>H-nmr, mass spectra. Elemental analyses results also supported the structures of **5a-g**. The <sup>1</sup>H-nmr spectra of **2a-d** displayed doublets at 3.66-3.78 due to  $H_A$  and  $H_B$  systems in addition to other signals. In the condensation products **3a-d**, these AB systems were absent confirming that an aldol condensation had taken place. In the <sup>1</sup>H nmr spectra of **5a-g**, 6-CH and 7-CH protons of the dihydrofuran nucleus appared as two dublets at 4.23-4.93 ppm and 3.75-3.95 ppm, respectively.

In the ftir spectra of the thiazolidinones, **2a-g** and **3a-g**, the characteristic C=O bands appeared in the region of 1651-1676 cm<sup>-1</sup>. The disappearance of the strong sharp bands at 1620-1605 cm<sup>-1</sup> of the initial azomethines was the most characteristic evidence of the cyclocondensation. Regarding compounds **5a-g** amide carbonyl bands were absent which clearly confirmed that a cyclocondensation with Wittig reagent had taken place.

In conclusion, we have prepared a new series of alkyl and aryl substituted fused thiazolidine derivatives. In light of all these results, the cyclocondensation reactions of benzylidene derivatives of thiazolidinones took place with aromatic and heteroaromatic substituted thiazolidinones in high yield, because of electronic effects of aromatic rings. Besides, we observed that the reaction with aliphatic derivatives were very difficult because of steric effects resulting from the long chain. The biological activity measurements of the new compounds are under investigations.

#### **EXPERIMENTAL**

Melting points are uncorrected and were measured in open capillaries with an Electrothermal IA 9100 melting point apparatus. Infrared spectra were recorded on a Mattson 1000 ftir spectrometer as KBr pellets. Proton nmr spectra were determined on a Bruker DPX 400 spectrometer in CDCl<sub>3</sub> with tetramethylsilane as internal standard. Chemical shifts ( ) were measured in ppm. Mass spectra were obtained with a Shimadzu GS/MS QP 2000 A spectrometer with 70 eV electron impact ionization. Elemental analyses were performed on a Leco CHN-600 analyzer. Column chromatography was performed with silica gel 60 (70-230 mesh) purchased from E. Merck AG. The purity of the obtained substances and the composition of the reaction mixtures were monitoring by thin layer chromatography on silica gel sheets with fluorescent indicator (Merck No. 5554). Solvents and common reagents obtained from Merck and Aldrich, were of reagent grade. Thiazolidinones 2e-g and benzylidene derivative 3g were prepared by literature procedures [8,9] and the spectral data are in agreement with that given in the literature.

## Nonylidene-(4-phenoxyphenyl)amine (1a).

#### General Procedure for 1a-d.

Nonanal (1.42 g, 0.01 mol) and *p*-phenoxyaniline (1.85 g, 0.01 mol) were heated for 8 hours without solvent then 15 mL toluene was added to this solution and refluxed for an additional 15 hours. Toluene was evaporated and the crude Schiff-base was purified by column chromatography on silica gel eluted with toluene to give yellow oil, 85%. <sup>1</sup>H nmr (deuteriochloroform): 0.78-1.39 (m, 15H, CH<sub>2</sub> and CH<sub>3</sub>), 2.25-2.36 (m, 2H, CH<sub>2</sub>), 6.50-7.12 (m, 9H,

aromatic), 7.92 (t, J = 6.6 Hz,1H, =CH); ftir (potassium bromide): 1600 (C=N) cm<sup>-1</sup>; ms: m/z 309 (molecular ion).

## Nonylidene-(4-methoxyphenyl)amine (1b).

Purification by column chromatography on silica gel eluted with toluene gave yellow oil, 77%; <sup>1</sup>H nmr: 0.79-1.42 (m, 15H, CH<sub>2</sub> and CH<sub>3</sub>), 2.33-2.40 (m, 2H, CH<sub>2</sub>), 3.58 (s, 3H, OCH<sub>3</sub>), 6.46 (d, J = 8.7 Hz, 2H, aromatic), 7.05 (d, J = 8.7 Hz, 2H, aromatic), 7.05 (d, J = 8.7 Hz, 2H, aromatic), 7.83 (t, J = 6.6 Hz, 1H, =CH); ftir (potassium bromide); 1600 (C=N) cm<sup>-1</sup>; ms: m/z 247 (molecular ion).

## Nonylidene-(p-tolyl)amine (1c).

The crude product was subjected to column chromatography on silica gel eluted with toluene to yield yellow oil, 68%; <sup>1</sup>H nmr: 0.75-1.33 (m, 15H, CH<sub>2</sub> and CH<sub>3</sub>), 2.31 (s, 3H, CH<sub>3</sub>), 2.35-2.42 (m, 2H, CH<sub>2</sub>), 6.56 (d, J = 7.2 Hz, 2H, aromatic), 6.79 (d, J = 7.2 Hz, 2H, aromatic), 7.85 (d, J = 6.6 Hz, 1H, =CH); ftir (potassium bromide): 1605 (C=N) cm<sup>-1</sup>; ms: m/z 231 (molecular ion).

## Nonylidenecyclohexylamine (1d).

The crude product subjected to column chromatography with toluene/ethyl acetate (4:1) gave yellow oil, 54%; <sup>1</sup>H nmr: 0.76-1.35 (m, 25H, CH<sub>2</sub> and CH<sub>3</sub>), 2.20-2.25 (m, 2H, CH<sub>2</sub>), 3.25-3.28 (m, 1H, CH), 7.16 (t, J = 6.6 Hz, 1H, =CH); ftir (potassium bromide): 1603 (C=N) cm<sup>-1</sup>; ms: m/z 223 (molecular ion).

## 3-(4-Phenoxyphenyl)-2-octyl-4-thiazolidinone (2a).

#### General Procedure for **2a-d**.

To a solution of the Schiff-base (**1a**) (3.09 g, 0.01 mol) derived from nonanal and *p*-phenoxyaniline in 15 ml dry benzene, thioglicolic acid (0.92 g, 0.01 mol) was added. The mixture was refluxed on a water bath for 18 h, then cooled and poured into water. The upper organic layer was washed with NaHCO<sub>3</sub> solution (15 ml, 10%) and then with H<sub>2</sub>O, dried (Na<sub>2</sub>SO<sub>4</sub>), and the benzene was distilled off. Upon crystallization of the residue from petroleum ether (40-60 °C)/ethanol (1:1), the thiazolidinone was obtained as yellow crystals, yield 69%, mp 121-123 °C; <sup>1</sup>H nmr: 0.93-1.92 (m, 17H, CH<sub>2</sub> and CH<sub>3</sub>), 3.67 (d, J = 16.5 Hz, 1H, CH<sub>2</sub>), 3.71 (d, J = 16.5 Hz, 1H, CH<sub>2</sub>), 4.92 (t, J = 7.0 Hz, 1H, CH), 6.93-7.65 (m, 9H, aromatic); ftir (potassium bromide): 1676 (C=O) cm<sup>-1</sup>; ms: m/z 383 (molecular ion).

## 3-(4-Methoxyphenyl)-2-octyl-4-thiazolidinone (2b).

Reaction was completed after 6 h reflux, then the crude product was recrystallized from petroleum ether (40-60 °C)/ether (1:1), to yield yellow crystals, yield 73%, mp 118-119 °C; <sup>1</sup>H nmr: 0.76-1.75 (m, 17H, CH<sub>2</sub> and CH<sub>3</sub>), 3.65 (s, 3H, CH<sub>3</sub>), 3.72 (d, J = 16.5 Hz, 1H, CH<sub>2</sub>), 3.77 (d, J = 16.5 Hz, 1H, CH<sub>2</sub>), 4.55 (t, J = 7.0 Hz, 1H, CH), 6.60 (d, J = 9 Hz, 2H, aromatic), 7.28 (d, J = 9 Hz, 2H, aromatic); ftir (potassium bromide): 1676 (C=O) cm<sup>-1</sup>; ms: m/z 321 (molecular ion).

## 2-Octyl-3-(*p*-tolyl)-4-thiazolidinone (2c).

Reaction was completed after 8 h reflux, then the crude product was recrystallized from petroleum ether (40-60 °C)/ethanol (1:1), the thiazolidinone was obtained as yellow crystals, yield 72%, mp 96-98 °C; <sup>1</sup>H nmr: 0.87-1.99 (m, 17H, CH<sub>2</sub> and CH<sub>3</sub>), 2.11 (s, 3H, CH<sub>3</sub>), 3.68 (d, J = 16.5 Hz, 1H, CH<sub>2</sub>), 3.74 (d, J =16.5 Hz, 1H, CH<sub>2</sub>), 4.76 (t, J = 7.0 Hz, 1H, CH), 7.49 (d, J = 8.3Hz, 2H, aromatic), 7.10 (d, J = 8.3 Hz, 2H, aromatic); ftir (potassium bromide): 1651 cm<sup>-1</sup>; ms: m/z 305 (molecular ion). Jul-Aug 2003

3-Cyclohexyl-2-octyl-4-thiazolidinone (2d).

Reaction was completed after 9 h reflux, then the crude product was purified with column chromatography on silica gel eluted with ethyl acetate/hexane (1:4), the product was obtained as yellow crystals, yield 65%, mp 109-110 °C; <sup>1</sup>H nmr: 0.82-1.65 (m, 27H, CH<sub>2</sub>), 3.25-3.29 (m,1H, CH), 3.69 (d, J = 16.5 Hz, 1H, CH<sub>2</sub>), 3.71 (d, J = 16.5 Hz, 1H, CH<sub>2</sub>), 4.35 (t, J = 7.0 Hz, 1H, CH); ftir (potassium bromide): 1676 (C=O) cm<sup>-1</sup>; ms: m/z 297 (molecular ion).

5-Benzylidene-3-(4-phenoxyphenyl)-2-octyl-4-thiazolidinone (**3a**).

General Procedure for 3a-f.

Equimolar solutions of **2a** (3.83 g, 0.01 mol) and benzaldehyde (1.06 g, 0.01 mol) in dry benzene (25 mL) in presence of sodium ethoxide were refluxed for about 10-12 h, cooled and poured into ice cold water, and acidified with glacial acetic acid. The solution was extracted, the benzene layer separated, dried (CaCl<sub>2</sub>) and evaporated *in vacuo*. The crude product was crystallized from petroleum ether: diethyl ether (1:1) to give yellow crystals, yield 61 %, mp 153-155 °C; <sup>1</sup>H nmr: 0.82-1.83 (m, 17H, CH<sub>2</sub> and CH<sub>3</sub>), 5.53 (t, J = 7.0 Hz, 1H, CH), 6.62-7.73 (m, 15H, aromatic and =CH); ftir (potassium bromide): 1676 (C=O) cm<sup>-1</sup>; ms: m/z 471 (molecular ion).

5-Benzylidene-3-(4-methoxyphenyl)-2-octyl-4-thiazolidinone (**3b**).

The crude product was crystallized from petroleum ether: diethyl ether (1:1) to give yellow crystals, yield 62 %, mp 147-148 °C; <sup>1</sup>H nmr: 0.83-1.68 (m, 17H, CH<sub>2</sub> and CH<sub>3</sub>), 3.83 (s, 3H, OCH<sub>3</sub>), 5.25 (t, J = 7.0 Hz, 1H, CH), 6.99-7.68 (m, 10H, aromatic and =CH); ftir (potassium bromide): 1676 (C=O) cm<sup>-1</sup>; ms: m/z 409 (molecular ion).

#### 5-Benzylidene-2-octyl-3-(*p*-tolyl)-4-thiazolidinone (3c).

The crude product was crystallized from benzene:diethyl ether (1:1) to give yellow crystals, yield 59 %, mp 169-171 °C; <sup>1</sup>H nmr: 0.75-1.56 (m, 17H, CH<sub>2</sub> and CH<sub>3</sub>), 2.16 (s, 3H, CH<sub>3</sub>), 5.22 (t, J = 7.0 Hz, 1H, CH), 6.93-7.56 (m, 10H, aromatic and =CH); ftir (potassium bromide): 1676 (C=O) cm<sup>-1</sup>; ms: m/z 393 (molecular ion).

#### 5-Benzylidene-3-cyclohexyl-2-octyl-4-thiazolidinone (3d).

The crude product was crystallized from benzene:diethyl ether (1:1) to give yellow crystals, yield 68 %, mp 156-157 °C; <sup>1</sup>H nmr: 0.79-1.72 (m, 27H, CH<sub>2</sub> and CH<sub>3</sub>), 3.36-3.41 (m, 1H, CH), 5.15 (t, J = 7.0 Hz, 1H, CH), 7.24-7.83 (m, 6H, aromatic and =CH); ftir (potassium bromide): 1651 (C=O) cm<sup>-1</sup>; ms: m/z (molecular ion).

5-Benzylidene-3-(4-phenoxyphenyl)-2-phenyl-4-thiazolidinone (**3e**).

The crude product was crystallized from benzene: diethyl ether (1:1) to give yellow crystals, yield 51 %, mp 148-150 °C; <sup>1</sup>H nmr: 6.42 (s, 1H, CH), 6.94-7.43 (m, 20H, aromatic and =CH); ftir (potassium bromide): 1673 (C=O) cmr<sup>1</sup>; ms: m/z 435 (molecular ion).

5-Benzylidene-3-(4-ethoxyphenyl)-2-phenyl-4-thiazolidinone (3f).

The crude product was crystallized from benzene:diethyl ether

(1:1) to give yellow crystals, yield 69 %, mp 153-154 °C; <sup>1</sup>H nmr: 1.35 (t, J = 7.0 Hz, 3H, CH<sub>3</sub>), 3.94 (q, J = 7.0 Hz, 2H, CH<sub>2</sub>), 6.73 (s, 1H, CH), 6.89-7.77 (m, 15H, aromatic and =CH); ftir (potassium bromide): 1675 (C=O) cmr<sup>1</sup>; ms: m/z 387 (molecular ion).

6-Methoxycarbonyl-3-(4-phenoxyphenyl)-7-phenyl-2-octyl-dihydro-4-furo[2,3-d]thiazolidine (5a).

### General Procedure for **5a-g**.

A solution of **3a** (4.71 g, 0.01 mol) and methoxycarbonylmethylenetriphenylphosphorane [10] (5.01 g, 0.015 mol) in ethyl acetate (50 mL) was refluxed for 24 h in the presence of triethylamine (1.51 g, 0.015 mol). After evaporation of the solvent, the remainder was subjected to column chromatography on silica gel eluted with petroleum ether/acetone (9:1) with increasing amounts of acetone (up to 3:1). The two products obtained were triphenylphosphine and **5a** (26%) which was isolated as orange crystals and identified, mp 236-237 °C; <sup>1</sup>H nmr: 0.79-1.87 (m, 17H, CH<sub>2</sub> and CH<sub>3</sub>), 3.62 (s, 3H, OCH<sub>3</sub>), 3.83 (d, J = 5.7 Hz, 1H, CH), 4.93 (d, J = 5.7 Hz, 1H, CH), 5.01 (t, J = 7.0 Hz, 1H, CH), 6.83-7.22 (m, 14H, aromatic); ftir (potassium bromide): 1723 (C=O) cm<sup>-1</sup>; ms: m/z 543 (molecular ion).

*Anal.* Calcd. for C<sub>33</sub>H<sub>37</sub>NO<sub>4</sub>S: C, 72.79; H, 6.85; N, 2.57; S, 5.88. Found: C, 72.72; H, 6.87; N, 2.60; S, 5.91.

6-Methoxycarbonyl-3-(4-methoxyphenyl)-7-phenyl-2-octyldihydro-<sup>4</sup>-furo[2,3-*d*]thiazolidine (**5**b).

Dark yellow crystals were obtained after purification with column chromatography, yield 32%, mp 229-230 °C; <sup>1</sup>H nmr: 0.84-1.92 (m, 17H, CH<sub>2</sub> and CH<sub>3</sub>), 3.66 (s, 3H, OCH<sub>3</sub>), 3.71 (s, 3H, OCH<sub>3</sub>), 3.75 (d, J = 5.7 Hz, 1H, CH), 4.75 (d, J = 5.7 Hz, 1H, CH), 4.98 (t, J = 7.0 Hz, 1H, CH), 6.92-7.33 (m, 9H, aromatic); ftir (potassium bromide): 1720 (C=O) cm<sup>-1</sup>; ms: m/z 481 (molecular ion).

*Anal.* Calcd. for C<sub>28</sub>H<sub>35</sub>NO<sub>4</sub>S: C, 69.81; H, 7.32; N, 2.90; S, 6.65. Found: C, 69.79; H, 7.35; N, 2.89; S, 6.62.

6-Methoxycarbonyl-3-(4-methylphenyl)-7-phenyl-2-octyl-dihydro- <sup>4</sup>-furo[2,3-*d*]thiazolidine (**5c**).

Yellow crystals were obtained after purification with column chromatography, yield 23%, mp 267-268 °C; <sup>1</sup>H nmr: 0.75-1.92 (m, 17H, CH<sub>2</sub> and CH<sub>3</sub>), 2.27 (s, 3H, CH<sub>3</sub>), 3.52 (s, 3H, OCH<sub>3</sub>), 3.80 (d, J = 5.7 Hz, 1H, CH), 4.85 (d, J = 5.7 Hz, 1H, CH), 4.96 (t, J = 7.0 Hz, 1H, CH), 6.87-7.12 (m, 9H, aromatic); ftir (potassium bromide): 1727 (C=O) cmr<sup>1</sup>; ms: m/z 465 (molecular ion).

*Anal.* Calcd. for C<sub>28</sub>H<sub>35</sub>NO<sub>3</sub>S: C, 72.21; H, 7.57; N, 3.00; S, 6.88. Found: C, 72.25; H, 7.58; N, 2.97; S, 6.86.

6-Methoxycarbonyl-3-cyclohexyl-7-phenyl-2-octyl-dihydro-<sup>4</sup>furo[2,3-*d*]thiazolidine (**5d**).

Orange crystals were obtained after purification with column chromatography, yield 28%, mp 243-244 °C; <sup>1</sup>H nmr: 0.85-1.62 (m, 27H, CH<sub>2</sub> and CH<sub>3</sub>), 3.52 (s, 3H, OCH<sub>3</sub>), 3.32-3.38 (m, 1H, CH), 3.95 (d, J = 5.7 Hz, 1H, CH), 4.23 (d, J = 5.7 Hz, 1H, CH), 4.83 (t, J = 7.0 Hz, 1H, CH), 7.03-7.32 (m, 5H, aromatic); ftir (potassium bromide): 1723 (C=O) cm<sup>-1</sup>; ms: m/z 457 (molecular ion).

*Anal.* Calcd. for C<sub>27</sub>H<sub>39</sub>NO<sub>3</sub>S: C, 70.85; H, 8.58; N, 3.06; S, 7.00. Found: C, 70.89; H, 8.61; N, 3.10; S, 7.01.

6-Methoxycarbonyl-3-(4-phenoxyphenyl)-2,7-diphenyl-dihydro-<sup>4</sup>-furo[2,3-*d*]thiazolidine (**5e**).

Yellow crystals were obtained after purification with column chromatography, yield 49%, mp 239-241 °C; <sup>1</sup>H nmr: 3.58 (s, 3H, OCH<sub>3</sub>), 3.69 (d, J = 5.7 Hz, 1H, CH), 4.97 (d, J = 5.7 Hz, 1H, CH), 6.28 (s, 1H, CH), 7.12-7.44 (m, 19H, aromatic); ftir (potassium bromide): 1725 (C=O) cm<sup>-1</sup>; ms: m/z 507 (molecular ion).

*Anal.* Calcd. for C<sub>31</sub>H<sub>25</sub>NO<sub>4</sub>S: C, 73.34; H, 4.96; N, 2.75; S, 6.31. Found: C, 73.41; H, 4.91; N, 2.69; S, 6.32.

6-Methoxycarbonyl-3-(4-ethoxyphenyl)-2,7-diphenyl-dihydro-<sup>4</sup>-furo[2,3-*d*]thiazolidine (**5f**).

Yellow crystals were obtained after purification with column chromatography, yield 52%, mp 217-219 °C; <sup>1</sup>H nmr: 1.44 (t, J = 7.0 Hz, 3H, CH<sub>3</sub>), 3.72 (s, 3H, OCH<sub>3</sub>), 3.66 (d, J = 5.7 Hz, 1H, CH), 3.98 (q, J = 7.0 Hz, 2H, CH<sub>2</sub>), 5.12 (d, J = 5.7 Hz, 1H, CH), 6.28 (s, 1H, CH), 7.08-7.29 (m, 14H, aromatic); ir (potassium bromide): 1720 (C=O) cm<sup>-1</sup>; ms: m/z 459 (molecular ion).

*Anal.* Calcd. for C<sub>27</sub>H<sub>25</sub>NO<sub>4</sub>S: C, 70.56; H, 5.48; N, 3.04; S, 6.97. Found: C, 70.57; H, 5.45; N, 3.03; S, 6.95.

6-Methoxycarbonyl-3-(4-methylphenyl)-2,7-diphenyl-dihydro-<sup>4</sup>-furo[2,3-*d*]thiazolidine (**5g**).

Yellow crystals were obtained after purification with column chromatography, yield 59%, mp 226-228 °C. <sup>1</sup>H nmr: 2.33 (s, 3H, CH<sub>3</sub>), 3.68 (s, 3H, OCH<sub>3</sub>), 3.71 (d, J = 5.7 Hz, 1H, CH), 5.02 (d, J = 5.7 Hz, 1H, CH), 6.32 (s, 1H, CH), 7.04-7.33 (m,

14H, aromatic); ir (potassium bromide): 1725 (C=O) cm<sup>-1</sup>; ms: m/z 429 (molecular ion).

*Anal.* Calcd. for C<sub>26</sub>H<sub>23</sub>NO<sub>3</sub>S: C, 72.69; H, 5.39; N, 3.26; S, 7.46. Found: C, 72.73; H, 5.42; N, 3.21; S, 7.42.

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