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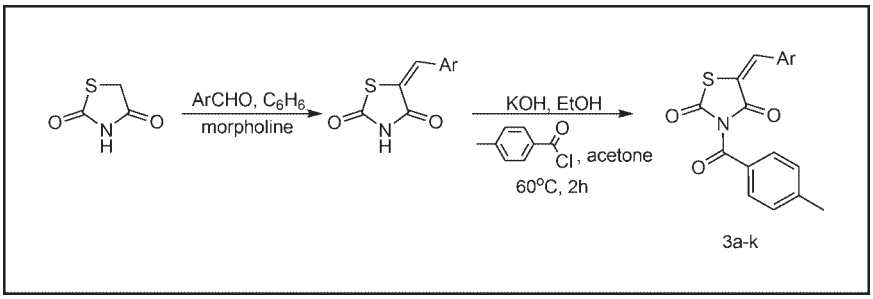

The derivatives of 5-substituted-2,4-thiazolidinedione have a broad spectrum of biological activities. In this article, new 5-(arylidene)-3-(4-methylbenzoyl)thiayolidine-2,4-diones **3a–k,** with arylidene groups such as 4-phenylbenzylidene **3a,** 3,4-dimethoxybenzylidene **3b,** 2-hydroxybenzylidene **3c,** 4-ethoxybenzylidene **3d,** 5-methyl-2-furfurylidene **3e**, 4-dimethylaminobenzylidene **3f,** 1-naphthylidene **3g,** 3,4-methylenedioxybenzylidene **3h,** 4-benzyloxybenzylidene **3i**, benzylidene **3j**, and 4-methoxybenzylidene **3k,** were synthesized by direct acylation of alkali metal salts of 5-arylidene-2,4-thiazolidinediones with 4-methylbenzoylchloride. Their structures were confirmed by elemental analysis, IR, ^{1}H NMR and MS spectroscopy. In addition, crystal structure of the compound **3d** was determined using single-crystal X-ray diffraction data.

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INTRODUCTION

Thiazolidinone derivates are reported to show variety of biological activities. Depending on the substituents, especially thiazolidinediones can produce different pharmacological activities such as antibacterial, antifungal [1], anticonvulsant [2], antidiabetic [3], cyclooxygenase and lipogenase inhibitory [4], antioxidant [5], and antiproliferative [6] activity. Moreover, the importance of 5-substituted-2,4-thiazolidinedione and their derivatives is due to their biological activities including antimicrobial [1] and fungicidal activity [7], as well as their utilization in a variety of therapeutic areas [8–14].

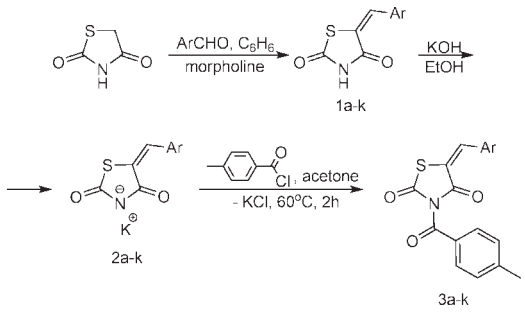
In some heterocyclylbenzenes [15,16] and substituted pyrazoles [17], which have herbicidal and defoliant characteristic, there is a 3-methylbenzoyl substructure. This study is focused on the link of 5-arylidene-2,4-thiazolidinediones and bioactive structural unit, 3-methylbenzoyl group, in order to find novel potential herbicides and defoliants. Crystal structure of the selected compound **3d** was also determined using single-crystal X-ray diffraction data.

RESULTS AND DISCUSSION

The reactive methylene group of the 2,4-thiazolidinedione has previously been successfully condensed with aldehydes, forming respectively 5-arylidene-2,4-thiazolidinediones **1a–k** [18]. These compounds were later transformed in their potassium salts **2a–k** [19]. By direct acylation of solid alkali metal salts of 5-arylidene-2,4-thiazolidinediones with 4-methylbenzoylchloride in refluxing dry acetone the corresponding 5-(arylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-diones **3a–k** were prepared. The obtained compounds **3a–k** were synthesized as yellow crystals in high yields (56.2–99.4%). The synthesis route is shown in Scheme 1.

All the newly synthesized compounds were characterized by elemental analysis, IR, ^{1}H NMR, and MS (see

Scheme 1



Ar = $C_{12}H_9$ (a); 3,4-$(MeO)_2C_6H_3$ (b); 2-OHC_6H_4 (c); 4-$EtOC_6H_4$ (d); 5-MeC_4H_2O (e); 4-$Me_2NC_6H_4$ (f); 1-$C_{10}H_7$ (g); 3,4-$OCH_2OC_6H_3$ (h); 4-$PhCH_2OC_6H_4$ (i); C_6H_5 (j); 4-$MeOC_6H_4$ (k)

''Experimental'' section). The elemental analysis and MS of 5-(arylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-diones **3a–k**, agreed with the molecular formula of these compounds. The structure of synthesized compounds was confirmed by ^{1}H NMR and IR spectroscopy. ^{1}H NMR spectra 5-(arylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-diones **3a–k,** clearly showed presence of benzoyl protons ($\delta \sim 7.3$ and 7.8 ppm) and methyl protons ($\delta \sim 2.45$ ppm) in comparison with spectra of 5-arylidene-2,4-thiazolidinediones **1a–k** [18]. IR spectra of all newly synthesized compounds **3a–k,** contain characteristic bands attributed to the methyl-benzoyl (C═O, about 1760 cm^{-1}) and benzoyl (about 1450 and 1600 cm^{-1}) vibrations that are used in the structural characterization of this type of compounds.

Crystal structure of the compound 3d. The molecular structure of compound **3d** is shown in Figure 1, and selected bond lengths and bond angles are listed in Table 1. Bond lengths and angles are in well agreement with some parent and similar compounds [20–22]. The main part of molecule, including the side ethoxy group, is almost planar with dihedral angle between benzylidene and thiazolidine-2,4-dione rings of only 5.8°. This is as expected, since practically the same angles (5.4°–5.9°) are found in some similar compounds [20,21]. The slight deviation from planarity can be attributed to the short repulsive S···H14–C14 contact with S···C14 distance of only 3.260(2) Å. On the other hand, dihedral angle between planes of thiazolidine-2,4-dione and methylbenzylidene rings is 67.1°.

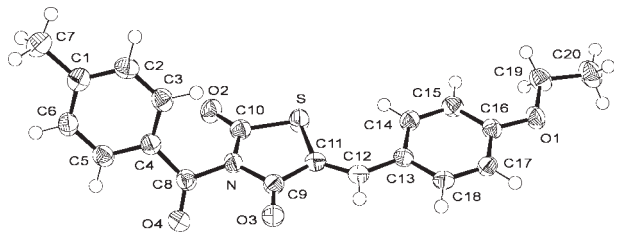


Figure 1. The molecular structure of compound **3d.**

The characteristic feature in crystal packing is the stacking of molecules in columns running along *b*-axis (Fig. 2). These columns further make a regular grid parallel to the planes (1 0 1) and (1 0 0) [Fig. 2(a)]. Within the columns there are stacking π−π interactions, which alternatively involve two benzylidene rings or pairs of both benzylidene and thiazolidine-2,4-dione rings [Fig. 2(b)]. As, for example, distances between planes of two benzylidene rings are only 3.32 and 3.33 Å the stacking π–π interactions can be described as very strong. In addition to van der Waals contacts, between the neighboring columns there are several C—H···O interactions involving mainly C atoms from terminal CH_3 groups, as well as few C atoms from 4-methylbenzoyl and benzylidene rings. In this way, the columns are only loosely connected to each other.

On the basis of presented crystal structure of **3d** compound it can be assumed that: (a) in all compounds (**3a–k**) the main part of molecule, including benzylidene and thiazolidine-2,4-dione rings, is almost planar; (b) π–π interactions between aromatic rings have a predominant role in molecular packing; (c) in all derivatives the C11—C12 bond is longer than double (C═C) bond, while the C12—C13 bond is shorter than the expected value for a single bond, probably because of delocalization of π-electrons through the whole substructure (C11═CH—C_{arom}). This delocalization is a reason why the attempts to carry out some addition reactions, which are inherent for the double bond, were unsuccessful.

EXPERIMENTAL

Materials and methods. The solvent and all reagents used in this study were purchased from commercial suppliers and were used as received. The melting points were obtained with

Table 1

Selected bond lengths (Å) and bond angles (°) for **3d**.

Bond lenghts (Å)			
S—C11	1.7586(16)	N—C9	1.414(2)
S—C10	1.7620(18)	N—C8	1.459(2)
O1—C16	1.360(2)	C1—C7	1.502(3)
O1—C19	1.450(2)	C4—C8	1.471(2)
O2—C10	1.210(2)	C9—C11	1.480(2)
O3—C9	1.206(2)	C11—C12	1.342(2)
O4—C8	1.200(2)	C12—C13	1.450(2)
N—C10	1.397(2)	C19—C20	1.501(3)
Bond angles (°)			
C11—S—C10	92.30(18)	O2—C10—N	125.19(16)
C16—O1—C19	117.49(13)	O2—C10—S	123.99(14)
C10—N—C9	116.01(14)	N—C10—S	110.78(12)
C10—N—C8	121.01(14)	C12—C11—C9	120.63(15)
C9—N—C8	122.44(13)	C12—C11—S	128.61(14)
O4—C8—N	118.55(16)	C9—C11—S	110.75(12)
O4—C8—C4	124.94(16)	C11—C12—C13	131.73(16)
N—C8—C4	116.50(15)	O1—C16—C15	124.91(15)
O3—C9—N	123.22(16)	O1—C16—C17	115.92(15)
O3—C9—C11	126.70(16)	O1—C19—C20	107.02(16)
N—C9—C11	110.04(14)		

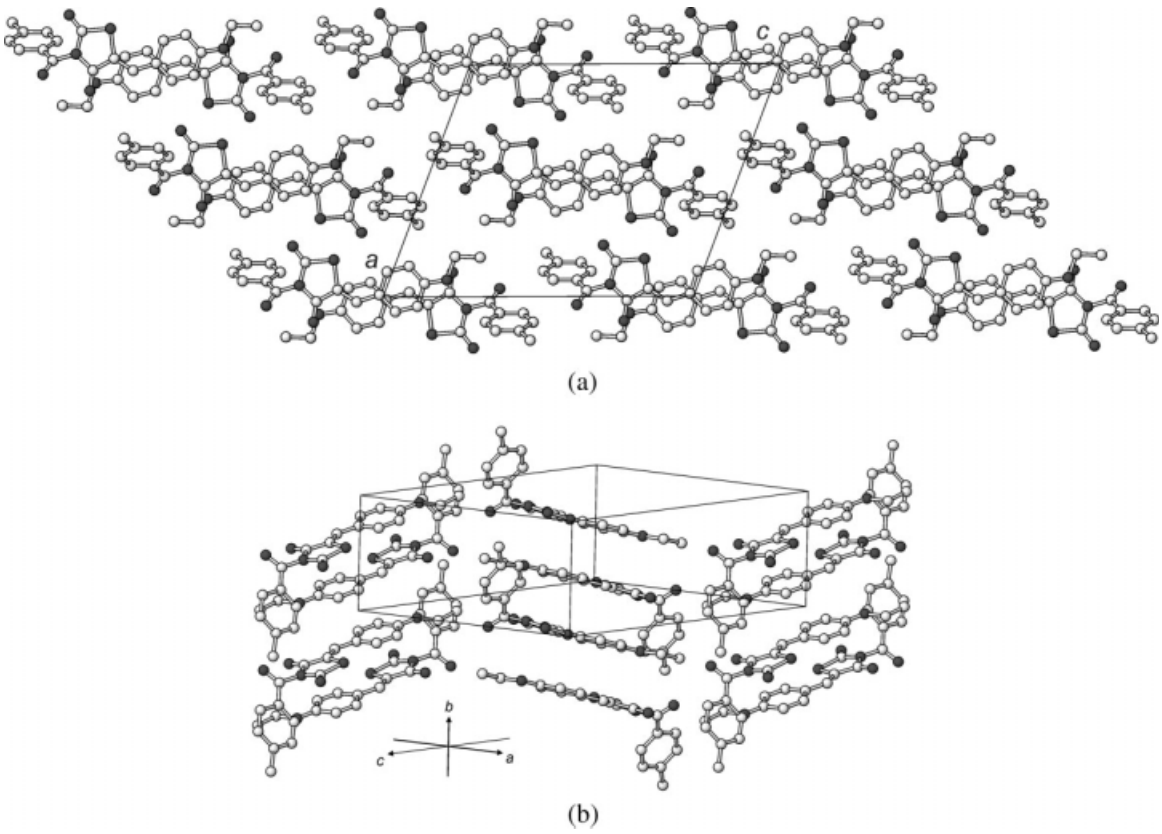


Figure 2. The packing of molecules **3d** (heteroatoms are dark, hydrogen atoms are omitted for clarity): (a) projection onto *ac*-plane and (b) projection approximately parallel to the (101) plane.

STUART SMP 10 melting point apparatus. Infrared spectra (ν in cm^{-1}) were recorded on a Perkin Elmer FTIR 1725 X spectrophotometer using KBr disks. The ^1H NMR spectra were recorded on a Varian Gemini 200 (200 MHz) instrument; chemical shifts (δ) are given relative to tetramethylsilane (TMS). The mass spectra were obtained on Finnigan MAT–8230 BE spectrometer with EI–CI source at 200°C, EI: 70 eV, 0.5 mA; CI: 1 m Torr of isobutane, 150 eV, 0.2 mA.

Single-crystals of the compound **3d** were obtained by recrystallization from EtOH. X-ray diffraction data were collected at 150 K on a Nonius Kappa CCD diffractometer using Mo Kα radiation. The structure was solved by direct methods and refined by a full-matrix least-squares procedure based on F^2 using the programs from WinGX suite [23]. All nonhydrogen atoms were refined anisotropically, while the hydrogen atoms were found in ΔF maps and were refined isotropically with no constraints. Selected crystal data and refinement results are listed in Table 2.

Syntheses. ***General procedure for the preparation of the 5-(arylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-diones 3a–k.*** 5-Arylidene-2,4-thiazolidinedione potassium salts (1 mmol) were suspended in dry acetone (20 mL) and 4-methylbenzoylchloride (1 mmol) was added at room temperature. The reaction mixtures were stirred at 60°C for 2 h and cooled to room temperature. Finally, the reaction mixtures were filtered and acetone was removed to give the crystalline products. The products were recrystallized from absolute EtOH.

Table 2

Crystal data and refinement details for **3d**.

Empirical formula	$C_{20}H_{17}NO_4S$
Formula weight	367.41
Crystal system	monoclinic
Space group	$P2_1/n$
a (Å)	14.4880(5)
b (Å)	6.9780(3)
c (Å)	18.0600(7)
α (°)	90
β (°)	110.7290(10)
γ (°)	90
V (Å^3)	1707.62(12)
Z	4
Calculated density (g cm^{-3})	1.429
Absorption coefficient (mm^{-1})	0.216 mm
F(000)	768
Crystal size (mm)	0.75 × 0.70 × 0.05
Θ range (°)	3.16–26.37
Limiting indices	$-17 \le h \le 18$, $-8 \le k \le 8$, $-22 \le l \le 22$
Reflections collected/unique	6383/3453 ($R_{int} = 0.0313$)
Data/restraints/parameters	3453/0/304
Goodness-of-fit on F^2	1.030
Final R indices [$I > 2\sigma(I)$]	$R_1 = 0.0378$, $wR_2 = 0.0786$
R indices (all data)	$R_1 = 0.0592$, $wR_2 = 0.0874$

5-(4-Phenylbenzylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-dione (3a). This compound was obtained as yellow crystals (ethanol), yield 69.3%, mp 183°C; IR, ν, cm^{-1}: 3035 (=C—H), 2925, 2794 (C—H), 1762 (C=O), 1716 (C=O), 1693 (C=O), 1603, 1515, 1448 (C=C), 1408 (C—N), 1258, 1177, 1063, 837, 718, 691; ^{1}H NMR ($CDCl_3$), δ, ppm (*J*, Hz): 2.46 (s, 3H, CH_3), 7.34 (d, $2H_{arom}$, $J = 8.0$), 7.41–7.73 (m, $9H_{arom}$), 7. 87 (d, $2H_{arom}$, $J = 8.4$), 7.99 (s, 1H, =CH); *m/z* (CIMS): 398 (M^+). Anal. Calcd. for $C_{24}H_{17}NO_3S$ (399.5): C, 72.16; H, 4.29; N, 3.51; S, 8.03. Found: C, 72.01; H, 4.21; N, 3.59; S, 8.05.

5-(3,4-Dimethoxybenzylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-dione (3b). This compound was obtained as yellow crystals (ethanol), yield 56.2%, mp 228°C; IR, ν, cm^{-1}: 3007 (=C—H), 2960, 2838 (C—H), 1751 (C=O), 1712 (C=O), 1686 (C=O), 1591, 1512, 1447 (C=C), 1418 (C—N), 1272, 1242, 1180, 1074 (C—O), 857, 730, 678; ^{1}H NMR ($CDCl_3$), δ, ppm (*J*, Hz): 2.45 (s, 3H, CH_3), 3.95 (s, 6H, 2 CH_3), 6.96–7.20 (m, $3H_{arom}$), 7.31 (d, $2H_{arom}$, $J = 8.0$), 7.83 (d, $2H_{arom}$, $J = 8.4$), 7.89 (s, 1H, =CH); *m/z* (CIMS): 383 (M^+). Anal. Calcd. for $C_{20}H_{17}NO_5S$ (383.2): C, 62.66; H, 4.74; N, 3.65; S, 8.37. Found: C, 62.50; H, 4.71; N, 3.68; S, 8.57.

5-(2-Hydroxybenzylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-dione (3c). This compound was obtained as yellow crystals (ethanol), yield 83.4%, mp 233°C; IR, ν, cm^{-1}: 3415 (O—H), 3043 (=C—H), 2764 (C—H), 1745 (C=O), 1701 (C=O), 1681 (C=O), 1602, 1510, 1455 (C=C), 1412 (C—N), 1293, 1265, 1152 (C—O), 838, 750, 684; ^{1}H NMR ($CDCl_3$), δ, ppm (*J*, Hz): 2.47 (s, 3H, CH_3), 7.33–7.61 (m, $4H_{arom}$), 7.80 (d, 2 H_{arom}, $J = 8.2$), 7.99 (s, 1H, =CH), 8.11 (d, 2 H_{arom}, $J = 8.0$); *m/z* (CIMS): 339 (M^+). Anal. Calcd. for $C_{18}H_{13}NO_4S$ (339.4): C, 63.77; H, 3.86; N, 4.13; S, 9.46. Found: C, 63.47; H, 3.76; N, 3.83; S, 9.55.

5-(4-Ethoxybenzylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-dione (3d). This compound was obtained as yellow crystals (ethanol), yield 97.4%, mp 178°C; IR, ν, cm^{-1}: 3041 (=C—H), 2936, 2883 (C—H), 1755 (C=O), 1716 (C=O), 1690 (C=O), 1596, 1509, 1448 (C=C), 1398 (C—N), 1281, 1176, 1143, 1072 (C—O), 840, 729, 690; ^{1}H NMR ($CDCl_3$), δ, ppm (*J*, Hz): 1.46 (t, 3H, CH_3, $J = 7.0$), 2.45 (s, 3H, CH_3), 4.11 (q, 2H, CH_2, $J = 7.0$), 7.00 (2 H_{arom}, *m AA′* $J_1 = 6.8$, $J_2 = 1.99$), 7.32 (d, $2H_{arom}$, $J = 8.0$), 7.47 (2 H_{arom}, *m BB′* $J_1 = 6.8$, $J_2 = 1.8$), 7.84 (2 H_{arom}, *m BB′* $J_1 = 8.4$, $J_2 = 1.8$), 7.89 (s, 1H, =CH); *m/z* (CIMS): 367 (M^+). Anal. Calcd. for $C_{20}H_{17}NO_4S$ (367.4): C, 65.38; H, 4.66; N, 3.81; S, 8.73. Found: C, 65.18; H, 4.62; N, 3.83; S, 8.81.

5-(5-Methyl-2-furfurylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-dione (3e). This compound was obtained as yellow crystals (ethanol), yield 95.2%, mp 168°C (decomp.); IR, ν, cm^{-1}: 3038 (=C—H), 2920 (C—H), 1758 (C=O), 1711 (C=O), 1681 (C=O), 1611, 1513, 1438 (C=C), 1412 (C—N), 1253, 1159, 1081 (C—O), 864, 731, 690; ^{1}H NMR ($CDCl_3$), δ, ppm (*J*, Hz): 2.45 (s, 3H, CH_3), 6.24 (d, $1H_{furyl}$, $J = 3.5$), 6.78 (d, $1H_{furyl}$, $J = 3.5$), 7.32 (d, 2 H_{arom}, $J = 8.0$), 7.60 (s, 1H, =CH), 7.82 (d, $2H_{arom}$, $J = 8.4$); *m/z* (CIMS): 327 (M^+). Anal. Calcd. for $C_{17}H_{13}NO_4S$ (327.4): C, 62.37; H, 4.00; N, 4.28; S, 9.79. Found: C, 62.09; H, 3.90; N, 4.25; S, 9.88.

5-(4-Dimethylaminobenzylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-dione (3f). This compound was obtained as yellow crystals (ethanol), yield 91.8%, mp 208°C (decomp.); IR, ν, cm^{-1}: 3032 (=C—H), 2911, 2884 (C—H), 1751 (C=O), 1714 (C=O), 1680 (C=O), 1585, 1530, 1441 (C=C), 1379 (C—N), 1296, 1197, 884, 769, 659; ^{1}H NMR ($CDCl_3$), δ, ppm (*J*, Hz): 2.45 (s, 3H, CH_3), 3.09 (s, 6H, N $(CH_3)_2$), 6.75 ($2H_{arom}$, *m AA′* $J_1 = 7.0$, $J_2 = 1.99$), 7.33 ($2H_{arom}$, *m AA′* $J_1 = 7.99$, $J_2 = 0.4$), 7.44 ($2H_{arom}$, *m BB′* $J_1 = 7.2$, $J_2 = 1.99$), 7.83 ($2H_{arom}$, *m BB′* $J_1 = 8.4$, $J_2 = 1.99$), 7.85 (s, 1H, =CH); *m/z* (CIMS): 366 (M^+). Anal. Calcd. for $C_{20}H_{18}N_2O_3S$ (366.4): C, 65.49; H, 4.95; N, 7.64; S, 8.75. Found: C, 65.29; H, 4.83; N, 7.59; S, 8.83.

5-(1-Naphthylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-dione (3g). This compound was obtained as yellow crystals (ethanol), yield 99.4%, mp 147°C; IR, ν, cm^{-1}: 3046 (=C—H), 2909 (C—H), 1762 (C=O), 1717 (C=O), 1690 (C=O), 1603, 1572, 1448 (C=C), 1397 (C—N), 1298, 1181, 891, 736, 641; ^{1}H NMR ($CDCl_3$), δ, ppm (*J*, Hz): 2.47 (s, 3H, CH_3), 7.33 (d, $2H_{arom}$, $J = 8.2$), 7.55–8.15 (m, $9H_{arom}$), 8.69 (s, 1H, =CH); *m/z* (CIMS): 373 (M^+). Anal. Calcd. for $C_{22}H_{15}NO_3S$ (373.4): C, 70.76; H, 4.05; N, 3.75; S, 8.59. Found: C, 70.57; H, 4.02; N, 3.77; S, 8.66.

5-(3,4-Methylenedioxybenzylidene)-3-(4-methylbenzoul)thiazolidine-2,4-dione (3h). This compound was obtained as yellow crystals (ethanol), yield 71.2%, mp 152°C; IR, ν, cm^{-1}: 3051 (=C—H), 2997, 2908 (C—H), 1752 (C=O), 1713 (C=O), 1689 (C=O), 1607, 1590, 1449 (C=C), 1365 (C—N), 1263, 1066 (C—O), 862, 726, 658; ^{1}H NMR ($CDCl_3$), δ, ppm (*J*, Hz): 2.46 (s, 3H, CH_3), 6.09 (s 2H, —OCH_2O—), 7.33 (d, $2H_{arom}$, $J = 8.2$), 6.92–7.12 (m, $3H_{arom}$), 7.81 (d, $2H_{arom}$, $J = 7.8$), 8.85 (s, 1H, =CH); *m/z* (CIMS): 367 (M^+). Anal. Calcd. for $C_{19}H_{13}NO_5S$ (367.4): C, 62.12; H, 3.57; N, 3.81; S, 8.73. Found: C, 61.94; H, 3.50; N, 3.77; S, 8.80.

5-(4-Benzyloxybenzylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-dione (3i). This compound was obtained as yellow crystals (ethanol), yield 96.3%, mp 144°C; IR, ν, cm^{-1}: 3034 (=C—H), 2923, 2882 (C—H), 1763 (C=O), 1716 (C=O), 1687 (C=O), 1593, 1511, 1453 (C=C), 1386 (C—N), 1292, 1149 (C—O), 831, 783, 698; ^{1}H NMR ($CDCl_3$), δ, ppm (*J*, Hz): 2.45 (s, 3H, CH_3), 5.15 (s, 2H, CH_2O), 7.07–7.53 (m, $11H_{arom}$), 7.82 ($2H_{arom}$, *m BB′* $J_1 = 6.6$, $J_2 = 1.6$), 7.89 (s, 1H, =CH); *m/z* (CIMS): 429 (M^+). Anal. Calcd. for $C_{25}H_{19}NO_4S$ (429.5): C, 69.91; H, 4.46; N, 3.26; S, 7.47. Found: C, 69.74; H, 4.37; N, 3.23; S, 7.52.

5-Benzylidene-3-(4-methylbenzoyl)thiazolidine-2,4-dione (3j). This compound was obtained as yellow crystals (ethanol), yield 86.3%, mp 118°C; IR, ν, cm^{-1}: 3056 (=C—H), 2917 (C—H), 1758 (C=O), 1717 (C=O), 1692 (C=O), 1606, 1492 (C=C), 1373 (C—N), 1254, 1182, 883, 686; ^{1}H NMR ($CDCl_3$), δ, ppm (*J*, Hz): 2.46 (s, 3H, CH_3), 7.33 (d, $2H_{arom}$, $J = 8.0$), 7.48–7.55 (m, $5H_{arom}$), 7.84 ($2H_{arom}$, *m BB′* $J_1 = 6.5$, $J_2 = 1.99$), 7.95 (s, 1H, =CH); *m/z* (CIMS): 323 (M^+). Anal. Calcd. for $C_{18}H_{13}NO_3S$ (323.4): C, 66.86; H, 4.05; N, 4.33; S, 9.92. Found: C, 66.66; H, 3.94; N, 4.27; S, 9.98.

5-(4-Methoxybenzylidene)-3-(4-methylbenzoyl)thiazolidine-2,4-dione (3k). This compound was obtained as yellow crystals (ethanol), yield 90.3%, mp 154°C; IR, ν, cm^{-1}: 3069 (=C—H), 2844 (C—H), 1766 (C=O), 1720 (C=O), 1689 (C=O), 1594, 1463 (C=C), 1374 (C—N), 1290, 1151 (C—O), 878, 715, 689; ^{1}H NMR ($CDCl_3$), δ, ppm (*J*, Hz): 2.45 (s, 3H, CH_3), 3.89 (s, 3H, —OCH_3), 7.01 ($2H_{arom}$, *m AA′* $J_1 = 6.8$, $J_2 = 1.99$), 7.32 (d, $2H_{arom}$, $J = 8.4$), 7.50 ($2H_{arom}$, *m BB′* $J_1 = 7.0$, $J_2 = 1.99$), 7.83 (d, $2H_{arom}$, $J = 8.2$), 7.89 (s, 1H, =CH); *m/z* (CIMS): 353 (M^+). Anal. Calcd. for $C_{19}H_{15}NO4S$ (353.4): C, 64.57; H, 4.28; N, 3.96; S, 9.07. Found: C, 64.29; H, 4.18; N, 3.94; S, 9.108.

SUPPLEMENTARY DATA

CCDC 733926 contains the supplementary crystallographic data for this article. These data can be obtained free of charge via www.ccdc.cam.ac.uk/conts/retrieving.html (or from the Cambridge Crystallographic Data Centre, 12, Union Road, Cambridge CB2 1EZ, UK; fax: +44 1223 336033; or deposit@ccdc.cam.ac.uk).

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