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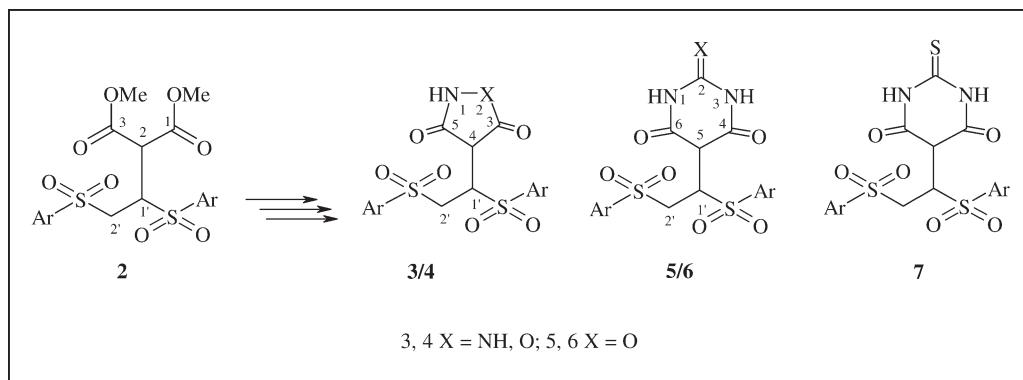
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A new class of pyrazolidinediones, isoxazolidinediones, pyrimidinetriones, and thioxopyrimidinediones were synthesized by the reaction of Michael adduct, dimethyl 2-(1',2'-diarylsulfonyl)ethylmalonate with different nucleophiles, hydrazine hydrate, hydroxylamine hydrochloride, and urea derivatives.

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INTRODUCTION

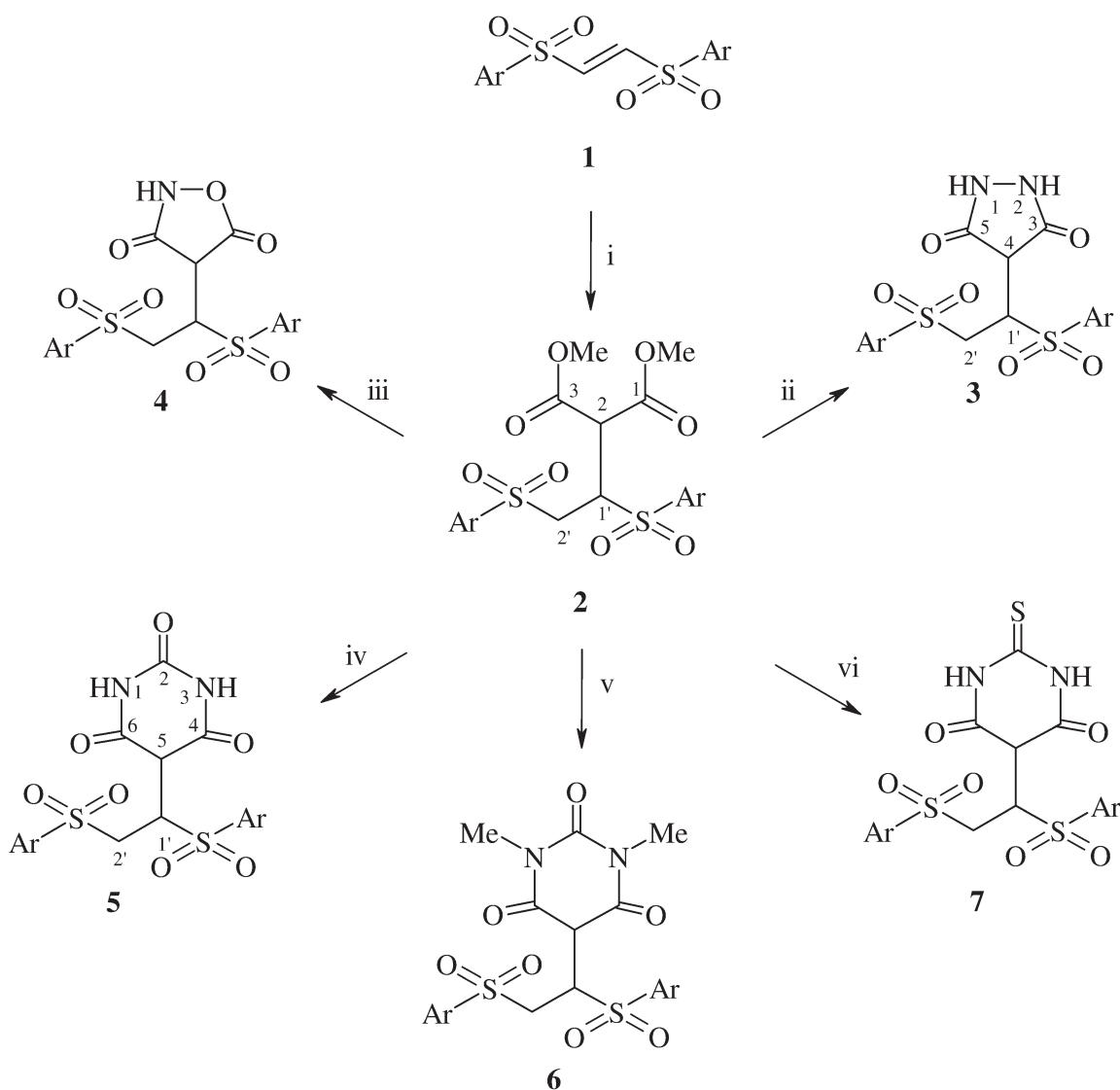
α,β -Unsaturated sulfur compounds have been used as important reagents in synthetic organic chemistry [1]. For example, vinyl sulfides as carbonyl synthons [2] and vinyl sulfonium salts as precursors of cyclopropanes [3]. Moreover, these are valuable intermediates in a variety of synthetic transformations and useful as building blocks in the synthesis of biologically potent heterocycles [4]. The biological properties of pyrazoles, isoxazoles, pyrimidines, thioxopyrimidines, and their derivatives have been reviewed extensively [5–16]. In our continued interest on the Michael addition reactions [17–20], we have examined the reactivity of α,β -unsaturated sulfones towards the synthesis of bioactive heterocycles. Based on these, herein we wish to report a new class of pyrazolidinediones, isoxazolidinediones, pyrimidinetriones, and thioxopyrimidinediones from the Michael acceptor, *E*-1,2-diarylsulfonylethene.

RESULTS AND DISCUSSION

The Michael addition of dimethyl malonate in the presence of anhydrous potassium carbonate in methyl ethyl ketone to *E*-1,2-diarylsulfonylethene (**1**) produced the Michael adduct, dimethyl 2-(1',2'-diarylsulfonyl)ethylmalonate (**2**) (Scheme 1 and Table 1). The IR

spectra of compounds **2** displayed absorption bands in the region 1120–1130 and 1338–1341 (SO₂), 1730–1735 cm^{−1} (CO₂Me) (Table 2). The ¹H NMR spectrum of **2a** showed two double doublets at δ 3.47, 3.56 (C_{2'}—H), a doublet at 4.05 (C₂—H), and a multiplet at 4.22–4.28 (C_{1'}—H). In addition, two singlets were observed at 3.61, 3.69 ppm due to methoxy protons of carbomethoxy group. The downfield shift was assigned to the one present towards arylsulfonyl moiety. This may be due to the deshielding effect exerted by this group (Table 3). The ¹³C NMR spectrum of **2a** exhibited signals at δ 39.7 (C-1'), 52.8, 53.6 (OCH₃), 60.4 (C-2), 54.9 (C-2'), 168.2, 168.9 (CO₂Me) (Table 3). The cyclocondensation of **2** with hydrazine hydrate and hydroxylamine hydrochloride furnished 4-(1',2'-diarylsulfonylethyl)pyrazolidine-3,5-dione (**3**) and 4-(1',2'-diarylsulfonylethyl)isoxazolidine-3,5-dione (**4**), respectively. Similar reaction of **2** with urea, *N,N*'-dimethylurea and thiourea afforded 5-(1',2'-diarylsulfonylethyl)pyrimidine-2,4,6-trione (**5**), 5-(1',2'-diarylsulfonylethyl)-1,3-dimethylpyrimidine-2,4,6-trione (**6**), and 5-(1',2'-diarylsulfonylethyl)-2-thioxopyrimidine-4,6-dione (**7**), respectively (Scheme 1 and Table 1). The absence of a band due to ester moiety in the IR spectra of the compounds **3–7** and the presence of absorption bands in the regions 1655–1680 (CO—N), 1115–1140 and 1330–1345 cm^{−1} (SO₂) indicated their formation. All the compounds except **6** also displayed

Scheme 1



- (i) $\text{CH}_2(\text{CO}_2\text{Me})_2 / \text{K}_2\text{CO}_3 / \text{MEK}$
- (ii) $\text{NH}_2\text{NH}_2\cdot\text{H}_2\text{O} / \text{NaOMe} / \text{MeOH}$
- (iii) $\text{NH}_2\text{OH}\cdot\text{HCl} / \text{NaOMe} / \text{MeOH}$
- (iv) $\text{NH}_2\text{CONH}_2 / \text{NaOMe} / \text{MeOH}$
- (v) $\text{MeNHCONHMe} / \text{NaOMe} / \text{MeOH}$
- (vi) $\text{NH}_2\text{CSNH}_2 / \text{NaOMe} / \text{MeOH}$

- a) $\text{Ar} = \text{C}_6\text{H}_5$
- b) $\text{Ar} = p\text{-CH}_3\text{C}_6\text{H}_4$
- c) $\text{Ar} = p\text{-ClC}_6\text{H}_4$

an absorption band at $3306\text{--}3331\text{ cm}^{-1}$ due to NH. Further, the compound **4** showed a band at $1736\text{--}1748\text{ (CO--O)}$, whereas **7** exhibited a band at $1488\text{--}1497\text{ cm}^{-1}$ (C=S) (Table 2). The ^1H NMR spectra of **3a** and **4a** showed two double doublets at δ 3.12, 3.85 and 3.14, 3.78 (C_{2'}-H), a multiplet at 4.34–4.42, 4.31–4.38 (C_{1'}-H), and a doublet at 4.49, 4.48 (C₄-H) ppm besides signals of aromatic protons. Apart from these, a

broad singlet was observed at δ 9.11 in **3a** and at 10.10 ppm in **4a** due to NH which disappeared on deuteration (Table 3). The ^{13}C NMR spectrum of **3a** displayed signals at δ 51.6 (C-2'), 55.4 (C-1'), 63.1 (C-4), 171.6 (C-3 and C-5), whereas **4a** at 50.7 (C-2'), 55.1 (C-1'), 62.9 (C-4), 172.6 (C-3), 178.7 ppm (C-5) in addition to signals of aromatic carbons (Table 3). The ^1H NMR spectra of **5a**, **6a** and **7a** also exhibited two double doublets

Table 1
Physical and analytical data of compounds 2–7.

Compound	M_p (°C)	Yield (%)	Ar	Molecular Formula	Analysis % Calcd./Found		
					C	H	N
2a	105–107	75	C ₆ H ₅	C ₁₉ H ₂₀ O ₈ S ₂ (440.49)	51.81 51.90	4.58 4.55	—
2b	154–156	71	<i>P</i> -CH ₃ C ₆ H ₄	C ₂₁ H ₂₄ O ₈ S ₂ (468.54)	53.83 53.88	5.16 5.18	—
2c	185–187	78	<i>P</i> -ClC ₆ H ₄	C ₁₉ H ₁₈ Cl ₂ O ₈ S ₂ (509.38)	44.80 44.75	3.56 3.60	—
3a	198–200	74	C ₆ H ₅	C ₁₇ H ₁₆ N ₂ O ₆ S ₂ (408.45)	49.99 49.94	3.95 3.93	6.86 6.92
3b	207–209	76	<i>P</i> -CH ₃ C ₆ H ₄	C ₁₉ H ₂₀ N ₂ O ₆ S ₂ (436.50)	52.28 52.32	4.62 4.66	6.42 6.45
3c	222–224	79	<i>P</i> -ClC ₆ H ₄	C ₁₇ H ₁₄ Cl ₂ N ₂ O ₆ S ₂ (477.34)	42.78 42.72	2.96 2.95	5.87 5.92
4a	182–184	80	C ₆ H ₅	C ₁₇ H ₁₅ NO ₇ S ₂ (409.43)	49.87 49.93	3.69 3.72	3.42 3.48
4b	196–198	73	<i>P</i> -CH ₃ C ₆ H ₄	C ₁₉ H ₁₉ NO ₇ S ₂ (437.49)	52.16 52.20	4.38 4.42	3.20 3.25
4c	217–219	77	<i>P</i> -ClC ₆ H ₄	C ₁₇ H ₁₃ Cl ₂ NO ₇ S ₂ (478.32)	42.69 42.75	2.74 2.75	2.93 2.96
5a	210–212	73	C ₆ H ₅	C ₁₈ H ₁₆ N ₂ O ₇ S ₂ (436.46)	49.53 49.51	3.69 3.71	6.42 6.51
5b	221–223	70	<i>P</i> -CH ₃ C ₆ H ₄	C ₂₀ H ₂₀ N ₂ O ₇ S ₂ (464.51)	51.71 51.77	4.34 4.38	6.03 6.10
5c	230–232	67	<i>P</i> -ClC ₆ H ₄	C ₁₈ H ₁₄ Cl ₂ N ₂ O ₇ S ₂ (505.35)	42.78 42.86	2.79 2.73	5.54 5.58
6a	216–218	65	C ₆ H ₅	C ₂₀ H ₂₀ N ₂ O ₇ S ₂ (464.51)	51.71 51.76	4.34 4.39	6.03 6.09
6b	226–228	68	<i>P</i> -CH ₃ C ₆ H ₄	C ₂₂ H ₂₄ N ₂ O ₇ S ₂ (492.57)	53.64 53.71	4.91 4.96	5.69 5.66
6c	205–207	70	<i>P</i> -ClC ₆ H ₄	C ₂₀ H ₁₈ Cl ₂ N ₂ O ₇ S ₂ (533.40)	45.03 45.00	3.40 3.45	5.25 5.32
7a	128–130	73	C ₆ H ₅	C ₁₈ H ₁₆ N ₂ O ₆ S ₃ (452.52)	47.77 47.82	3.56 3.53	6.19 6.24
7b	142–144	71	<i>P</i> -CH ₃ C ₆ H ₄	C ₂₀ H ₂₀ N ₂ O ₆ S ₃ (480.58)	49.98 50.05	4.19 4.18	5.83 5.87
7c	150–152	75	<i>P</i> -ClC ₆ H ₄	C ₁₈ H ₁₄ Cl ₂ N ₂ O ₆ S ₃ (521.41)	41.46 41.49	2.71 2.73	5.37 5.43

Table 2
IR data of compounds 2–7.

Compound	IR (KBr) cm ^{−1}					
	SO ₂	C=S	N—C=O	CO ₂ Me	O—CO	NH
2a	1126	1341	—	—	1733	—
2b	1130	1338	—	—	1735	—
2c	1121	1339	—	—	1731	—
3a	1119	1340	—	1676	—	—
3b	1122	1330	—	1668	—	—
3c	1126	1332	—	1672	—	—
4a	1136	1336	—	1678	—	1736
4b	1129	1337	—	1664	—	1742
4c	1130	1331	—	1655	—	1748
5a	1134	1333	—	1661	—	—
5b	1131	1339	—	1667	—	—
5c	1128	1336	—	1671	—	—
6a	1125	1334	—	1674	—	—
6b	1126	1341	—	1668	—	—
6c	1131	1339	—	1662	—	—
7a	1135	1336	1488	1673	—	—
7b	1129	1332	1492	1679	—	—
7c	1127	1335	1497	1670	—	—

Table 3
¹H and ¹³C NMR data of compounds 2–7.

Compound	¹ H NMR (CDCl ₃ /DMSO-d ₆) δ, (ppm)	¹³ C NMR (CDCl ₃ /DMSO-d ₆) δ, (ppm)
2a	3.47 (dd, 1H, C ₂ '—H, <i>J</i> = 9.0, 14.6 Hz), 3.56 (dd, 1H, C ₂ '—H, <i>J</i> = 4.4, 14.8 Hz), 3.61 (s, 3H, OCH ₃), 3.69 (s, 3H, OCH ₃), 4.05 (d, 1H, C ₂ '—H, <i>J</i> = 8.4 Hz), 4.22–4.28 (m, 1H, C ₁ '—H), 7.18–7.59 (m, 10H, Ar-H)	39.7 (C-1'), 52.8 and 53.6 (OCH ₃), 54.9 (C-2'), 60.4 (C-2), 168.2 and 168.9 (CO ₂ Me), 128.2, 129.2, 130.4, 132.6, 133.2, 133.8, 135.4 (aromatic carbons)
2b	2.36 (s, 6H, Ar—CH ₃), 3.46 (dd, 1H, C ₂ '—H, <i>J</i> = 8.8, 14.2 Hz), 3.58 (dd, 1H, C ₂ '—H, <i>J</i> = 4.4, 14.4 Hz), 3.64 (s, 3H, OCH ₃), 3.66 (s, 3H, OCH ₃), 4.09 (d, 1H, C ₂ '—H, <i>J</i> = 8.6 Hz), 4.26–4.32 (m, 1H, C ₁ '—H), 7.23–7.76 (m, 8H, Ar-H)	22.5 (Ar—CH ₃), 39.2 (C-1'), 53.2 and 53.9 (OCH ₃), 54.8 (C-2'), 60.7 (C-2), 167.9 and 168.7 (CO ₂ Me), 127.8, 129.6, 130.1, 131.5, 133.7, 134.2, 135.9, 136.1 (aromatic carbons)
2c	3.49 (dd, 1H, C ₂ '—H, <i>J</i> = 9.1, 14.7 Hz), 3.58 (dd, 1H, C ₂ '—H, <i>J</i> = 4.6, 14.9 Hz), 3.65 (s, 3H, OCH ₃), 3.72 (s, 3H, OCH ₃), 4.08 (d, 1H, C ₂ '—H, <i>J</i> = 8.6 Hz), 4.30–4.38 (m, 1H, C ₁ '—H), 7.32–7.56 (m, 8H, Ar-H)	39.6 (C-1'), 53.0 and 53.1 (OCH ₃), 54.9 (C-2'), 59.7 (C-2), 166.5 and 167.1 (CO ₂ Me), 125.2, 128.7, 129.4, 131.6, 133.8, 134.9, 136.0, 138.5 (aromatic carbons)
3a	3.12 (dd, 1H, C ₂ '—H, <i>J</i> = 3.2, 15.0 Hz), 3.85 (dd, 1H, C ₂ '—H, <i>J</i> = 9.7, 14.9 Hz), 4.34–4.42 (m, 1H, C ₁ '—H), 4.49 (d, 1H, C ₄ '—H, <i>J</i> = 13.9 Hz), 7.12–7.75 (m, 10H, Ar-H), 9.11 (bs, 2H, NH)	51.6 (C-2'), 55.4 (C-1'), 63.1 (C-4), 171.6 (C-3 and C-5), 129.6, 130.5, 131.1, 132.0, 133.3, 133.7, 135.3, 136.8 (aromatic carbons)
3b	2.32 (s, 6H, Ar—CH ₃), 3.14 (dd, 1H, C ₂ '—H, <i>J</i> = 3.1, 14.8 Hz), 3.82 (dd, 1H, C ₂ '—H, <i>J</i> = 9.2, 14.6 Hz), 4.32–4.38 (m, 1H, C ₁ '—H), 4.46 (d, 1H, C ₄ '—H, <i>J</i> = 13.7 Hz), 7.21–7.81 (m, 8H, Ar-H), 9.08 (bs, 2H, NH)	21.9 (Ar—CH ₃), 51.2 (C-2'), 55.8 (C-1'), 62.7 (C-4), 172.3 (C-3 and C-5), 128.4, 129.9, 130.7, 131.6, 132.9, 133.2, 134.9, 135.7, 136.4 (aromatic carbons)
3c	3.16 (dd, 1H, C ₂ '—H, <i>J</i> = 6.3, 16.2 Hz), 3.80 (dd, 1H, C ₂ '—H, <i>J</i> = 8.4, 16.0 Hz), 4.35–4.41 (m, 1H, C ₁ '—H), 4.45 (d, 1H, C ₄ '—H, <i>J</i> = 13.9 Hz), 7.16–7.39 (m, 8H, Ar-H), 9.14 (bs, 2H, NH)	51.8 (C-2'), 56.2 (C-1'), 59.6 (C-4), 171.0 (C-3 and C-5) 125.7 128.9, 129.2, 131.9, 133.6, 134.3, 135.8, 137.5, (aromatic carbons)
4a	3.14 (dd, 1H, C ₂ '—H, <i>J</i> = 3.5, 15.1 Hz), 3.78 (dd, 1H, C ₂ '—H, <i>J</i> = 9.7, 14.9 Hz), 4.31–4.38 (m, 1H, C ₁ '—H), 4.48 (d, 1H, C ₄ '—H, <i>J</i> = 13.9 Hz), 7.07–7.84 (m, 10H, Ar-H), 10.10 (bs, 1H, NH)	50.7 (C-2'), 55.1 (C-1'), 62.9 (C-4), 172.6 (C-3), 178.7 (C-5), 129.6, 130.2, 131.5, 132.4, 132.9, 134.6, 136.1, 137.4 (aromatic carbons)
4b	2.38 (s, 6H, Ar—CH ₃), 3.16 (dd, 1H, C ₂ '—H, <i>J</i> = 3.4, 15.0 Hz), 3.87 (dd, 1H, C ₂ '—H, <i>J</i> = 9.5, 14.8 Hz), 4.36–4.42 (m, 1H, C ₁ '—H), 4.50 (d, 1H, C ₄ '—H, <i>J</i> = 13.7 Hz), 7.10–7.98 (m, 8H, Ar-H), 10.19 (bs, 1H, NH)	22.6 (Ar—CH ₃), 51.2 (C-2'), 55.9 (C-1'), 63.5 (C-4), 173.4 (C-3), 179.2 (C-5), 129.2, 130.7, 131.3, 132.9, 133.5, 134.9, 135.7, 136.8 (aromatic carbons)
4c	3.05 (dd, 1H, C ₂ '—H, <i>J</i> = 3.3, 14.9 Hz), 3.70 (dd, 1H, C ₂ '—H, <i>J</i> = 9.3, 14.7 Hz), 4.33–4.39 (m, 1H, C ₁ '—H), 4.46 (d, 1H, C ₄ '—H, <i>J</i> = 13.8 Hz), 7.16–7.36 (m, 8H, Ar-H), 9.08 (bs, 1H, NH)	50.9 (C-2'), 56.2 (C-1'), 63.1 (C-4), 172.9 (C-3), 178.6 (C-5), 129.7, 130.4, 131.9, 132.6, 133.7, 134.2, 136.7, 138.9 (aromatic carbons)
5a	3.09 (dd, 1H, C ₂ '—H, <i>J</i> = 3.6, 15.0 Hz), 3.75 (dd, 1H, C ₂ '—H, <i>J</i> = 9.7, 14.8 Hz), 4.34–4.41 (m, 1H, C ₁ '—H), 4.49 (d, 1H, C ₅ '—H, <i>J</i> = 13.6 Hz), 6.99–7.39 (m, 10H, Ar-H), 9.91 (bs, 2H, NH)	50.1 (C-2'), 57.3 (C-1'), 62.7 (C-5), 158.1 (C-2), 165.3 (C-4 and C-6), 127.3, 129.2, 130.7, 132.1, 132.8, 133.7, 134.6, 135.8 (aromatic carbons)
5b	2.34 (s, 6H, Ar—CH ₃), 3.04 (dd, 1H, C ₂ '—H, <i>J</i> = 3.5, 14.9 Hz), 3.71 (dd, 1H, C ₂ '—H, <i>J</i> = 9.5, 14.7 Hz), 4.37–4.44 (m, 1H, C ₁ '—H), 4.51 (d, 1H, C ₅ '—H, <i>J</i> = 13.5 Hz), 7.19–7.69 (m, 8H, Ar-H), 9.99 (bs, 2H, NH)	21.7 (Ar—CH ₃), 50.4 (C-2'), 57.6 (C-1'), 62.3 (C-5), 157.8 (C-2), 166.1 (C-4 and C-6), 128.5, 129.6, 130.3, 131.8, 132.3, 133.4, 135.7, 136.6 (aromatic carbons)
5c	3.07 (dd, 1H, C ₂ '—H, <i>J</i> = 3.7, 15.0 Hz), 3.74 (dd, 1H, C ₂ '—H, <i>J</i> = 9.7, 14.9 Hz), 4.25–4.36 (m, 1H, C ₁ '—H), 4.62 (d, 1H, C ₅ '—H, <i>J</i> = 13.6 Hz), 7.18–7.59 (m, 8H, Ar-H), 9.89 (bs, 2H, NH)	50.3 (C-2'), 60.1 (C-1'), 64.1 (C-5), 158.3 (C-2), 166.5 (C-4 and C-6), 125.8 128.4, 129.0, 129.8, 132.3, 133.3, 133.9, 135.1, (aromatic carbons)
6a	2.78 (s, 6H, N—CH ₃), 3.09 (dd, 1H, C ₂ '—H, <i>J</i> = 3.8, 14.9 Hz), 3.72 (dd, 1H, C ₂ '—H, <i>J</i> = 9.5, 14.7 Hz), 4.31–4.36 (m, 1H, C ₁ '—H), 4.47 (d, 1H, C ₅ '—H, <i>J</i> = 13.9 Hz), 7.09–7.81 (m, 10H, Ar-H)	27.5 (N—CH ₃), 52.7 (C-2'), 57.9 (C-1'), 64.4 (C-5), 157.1 (C-2), 167.9 (C-4 and C-6), 129.3, 130.6, 131.2, 132.4, 132.8, 133.6, 134.5, 135.2 (aromatic carbons)
6b	2.25 (s, 6H, Ar—CH ₃), 2.71 (s, 6H, N—CH ₃), 3.03 (dd, 1H, C ₂ '—H, <i>J</i> = 3.6, 14.8 Hz), 3.70 (dd, 1H, C ₂ '—H, <i>J</i> = 9.3, 14.6 Hz), 4.29–4.35 (m, 1H, C ₁ '—H), 4.43 (d, 1H, C ₅ '—H, <i>J</i> = 13.6 Hz), 7.11–7.74 (m, 8H, Ar-H)	22.3 (Ar—CH ₃), 27.9 (N—CH ₃), 51.9 (C-2'), 58.2 (C-1'), 63.9 (C-5), 156.7 (C-2), 168.7 (C-4 and C-6), 128.8, 130.3, 131.9, 132.6, 132.9, 133.0, 133.7, 134.9 (aromatic carbons)
6c	2.75 (s, 6H, N—CH ₃), 3.06 (dd, 1H, C ₂ '—H, <i>J</i> = 3.7, 14.9 Hz), 3.73 (dd, 1H, C ₂ '—H, <i>J</i> = 9.4, 14.7 Hz), 4.32–4.38 (m, 1H, C ₁ '—H), 4.46 (d, 1H, C ₅ '—H, <i>J</i> = 13.7 Hz), 7.12–7.40 (m, 8H, Ar-H)	27.4 (N—CH ₃), 51.2 (C-2'), 58.9 (C-1'), 63.3 (C-5), 157.3 (C-2), 167.7 (C-4 and C-6), 129.2, 130.9, 131.4, 132.2, 132.7, 133.4, 134.9, 136.2 (aromatic carbons)

(continued)

Table 3
(Continued)

Compound	¹ H NMR (CDCl ₃ /DMSO-d ₆) δ, (ppm)	¹³ C NMR (CDCl ₃ /DMSO-d ₆) δ, (ppm)
7a	3.10 (dd, 1H, C _{2'} —H, <i>J</i> = 3.9, 15.1 Hz), 3.78 (dd, 1H, C _{2'} —H, <i>J</i> = 9.6, 15.0 Hz), 4.33–4.37 (m, 1H, C _{1'} —H), 4.49 (d, 1H, C _{5'} —H, <i>J</i> = 13.9 Hz), 7.12–7.78 (m, 10H, Ar-H), 9.38 (bs, 2H, NH)	52.9 (C-2'), 59.4 (C-1'), 64.2 (C-5), 167.4 (C-4 and C-6), 171.3 (C-2), 128.2, 129.2, 130.6, 131.4, 132.0, 133.2, 134.5, 135.7 (aromatic carbons)
7b	2.27 (s, 6H, Ar-CH ₃), 3.08 (dd, 1H, C _{2'} —H, <i>J</i> = 3.8, 15.0 Hz), 3.75 (dd, 1H, C _{2'} —H, <i>J</i> = 9.5, 14.9 Hz), 4.31–4.35 (m, 1H, C _{1'} —H), 4.46 (d, 1H, C _{5'} —H, <i>J</i> = 13.6 Hz), 7.17–7.82 (m, 8H, Ar-H), 9.31 (bs, 2H, NH)	22.7 (Ar-CH ₃), 51.7 (C-2'), 58.8 (C-1'), 64.9 (C-5), 166.9 (C-4 and C-6), 172.5 (C-2), 128.9, 129.4, 131.3, 132.9, 133.4, 134.6, 135.0, 135.8 (aromatic carbons)
7c	3.11 (dd, 1H, C _{2'} —H, <i>J</i> = 3.9, 15.1 Hz), 3.79 (dd, 1H, C _{2'} —H, <i>J</i> = 9.7, 15.0 Hz), 4.29–4.36 (m, 1H, C _{1'} —H), 4.47 (d, 1H, C _{5'} —H, <i>J</i> = 13.8 Hz), 7.14–7.93 (m, 8H, Ar-H), 9.36 (bs, 2H, NH)	51.1 (C-2'), 58.2 (C-1'), 64.4 (C-5), 167.3 (C-4 and C-6), 171.9 (C-2), 129.6, 130.4, 131.7, 132.4, 133.7, 134.9, 135.6, 136.6 (aromatic carbons)

at δ 3.09, 3.75; 3.09, 3.72; 3.10, 3.78, a multiplet at 4.34–4.41, 4.31–4.36, 4.33–4.37, and a doublet at 4.49, 4.47, 4.49 which were accounted for C_{2'}—H, C_{1'}—H, C_{5'}—H. The compounds **5a** and **7a** displayed a broad singlet at δ 9.91, 9.38 ppm for NH which disappeared on deuteration. Besides, compound **6a** showed a singlet at 2.78 ppm for N-Me group. The structures of the compounds **5–7** were further confirmed by ¹³C NMR spectra (Table 3).

CONCLUSIONS

A new class of pyrazolidinedione, isoxazolidinedione, pyrimidinetrione, and thioxopyrimidinedione were developed from the synthetic intermediate *E*-1,2-diarylsulfonylethene adopting facile, simple, and well-versed synthetic methodologies.

EXPERIMENTAL

General. Melting points were determined in open capillaries on a Mel-Temp apparatus and are uncorrected. The purity of the compounds was checked by TLC (silica gel H, BDH, ethyl acetate/hexane, 1:3). The IR spectra were recorded on a Thermo Nicolet IR 200 FTIR spectrometer as KBr pellets and the wave numbers were given in cm^{−1}. The ¹H NMR spectra were recorded in CDCl₃/DMSO-d₆ on a Varian EM-360 spectrometer (300 MHz). The ¹³C NMR spectra were recorded in CDCl₃/DMSO-d₆ on a Varian VXR spectrometer operating at 75.5 MHz. All chemical shifts are reported in δ (ppm) using TMS as an internal standard. The microanalyses were performed on a Perkin-Elmer 240C elemental analyzer. The starting compound 1,2-diarylsulfonylethene (**1**) was prepared as per the literature procedure [20].

Dimethyl 2-(1',2'-diarylsulfonylethyl)malonate (2): General procedure. A mixture of dimethyl malonate (15 mmol), methyl ethyl ketone (5 mL), and anhydrous potassium carbonate (10 mmol) was cooled to 5–10°C. To this, compound **1** (10 mmol) was added and stirred for 2–4 h maintaining the same temperature. The contents of the flask were diluted with water and extracted with chloroform. The organic layer was

washed with water, brine and dried (anhyd. Na₂SO₄). The solvent was removed *in vacuo*. The resultant solid was recrystallized from 2-propanol.

4-(1',2'-Diarylsulfonylethyl)pyrazolidine-3,5-dione (3): General procedure. The compound **2** (1 mmol), hydrazine hydrate (1.5 mmol), MeOH (20 mL), and NaOMe (5 mL) were refluxed for 3–5 h. The solution was cooled and poured onto crushed ice containing conc. HCl. The solid obtained was filtered, dried, and recrystallized from methanol.

4-(1',2'-Diarylsulfonylethyl)isoxazolidine-3,5-dione (4): General procedure. To a solution of **2** (1 mmol) in MeOH (10 mL), hydroxylamine hydrochloride (1 mmol) and NaOMe (5 mL) were added and refluxed for 4–6 h. The reaction mixture was cooled and poured into ice-cold water containing conc. HCl. The solid separated was filtered, dried, and recrystallized from methanol.

5-(1',2'-Diarylsulfonylethyl)pyrimidine-2,4,6-trione (5)/5-(1',2'-diarylsulfonylethyl)-1,3-dimethylpyrimidine-2,4,6-trione (6): General procedure. A mixture of compound **2** (1 mmol), urea/N,N'-dimethylurea (1 mmol), MeOH (5 mL), and NaOMe (5 mL) was refluxed for 8–10 h. The contents were cooled and poured into ice-cold water containing conc. HCl. The separated solid was filtered, dried, and recrystallized from methanol.

5-(1',2'-Diarylsulfonylethyl)-2-thioxopyrimidine-4,6-dione (7): General procedure. To an equimolar mixture (1 mmol) of **2** and thiourea, MeOH (10 mL) and NaOMe (2 mL) were added and refluxed for 6–8 h. It was cooled and poured into ice-cold water containing conc. HCl. The solid separated was filtered, dried, and purified by recrystallization from methanol.

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REFERENCES AND NOTES

- [1] Trost, B. M. *Chem Rev* 1978, 78, 363.
- [2] Seebach, D.; Kolb, M. *Chem Ind (London)* 1974, 687.
- [3] (a) Johnson, C. R.; Lockard, J. P. *Tetrahedron Lett* 1971, 12, 4589; (b) Takaki, K.; Agawa, T. *J Org Chem* 1977, 42, 3303.
- [4] (a) Helder, R.; Doornbos, T.; Strating, J.; Zwanenburg, B. *Tetrahedron* 1973, 29, 1375; (b) Padmavathi, V.; Sumathi, R. P.; Padmaj, A.; Bhaskar Reddy, D. *Indian J Chem* 2001, 40B, 846; (c) Padmavathi, V.; Venugopal Reddy, K.; Balaiah, A.; Ramana Reddy,

- T. V.; Bhaskar Reddy, D. *Heteroatom Chem* 2002, 13, 677; (d) Padmavathi, V.; Jagan Mohan Reddy, B.; Rajagopala Sarma, M.; Thriveni, P. *J Chem Res (S)* 2003, 79; (e) Padmavathi, V.; Rajagopala Sarma, M.; Padmaja, A.; Bhaskar Reddy, D. *J Heterocycl Chem* 2003, 40, 933.
- [5] Bobranski, B.; Matczak, H. *Roczniki Chem* 1975, 49, 99.
- [6] Doran, W. J. In *Barbituric Acid Hypnotics*; Blicke, F. F., Cox, R. H., Eds.; John Wiley and Sons: New York, 1959; Vol. 4, p 5.
- [7] Zawisza, T.; Matczak, H.; Kowalczyk-Bronisz, S. H.; Jakobiec, T. *Pol Arch Immunol Ther Exp* 1981, 29, 235.
- [8] Buchi, J.; Ammann, J.; Lieberherr, R.; Eichenberger, E. *Helv Chim Acta* 1953, 36, 75.
- [9] Dante, N.; Elena, M.; Magistetti, M. J. *Arzneim Forsch* 1969, 19, 1721.
- [10] Mimi, L. Q.; Christopheer, D. E.; Ann, Y. L.; Richard, S. A.; Robert, M. K.; Lam, G.; Mathew, R. W.; Pancras, C. W.; Ruth, R. W. *J Med Chem* 1999, 42, 2760.
- [11] Dannahardt, G.; Kiefer, W.; Kramer, G.; Maehrlein, S.; Nowe, U.; Fiebich, B. *Eur J Med Chem* 2000, 35, 499.
- [12] Boarland, M. P. V.; McOmie, J. F. W.; Timms, R. N. *J Chem Soc* 1952, 4691.
- [13] Kornet, M. J.; Thorstenson, J. H.; Lubawy, W. C. *Pharm J Sci* 1974, 63, 1090.
- [14] Richon, A. B.; Maragoudakis, M. E.; Wasvary, J. S. *J Med Chem* 1982, 25, 745.
- [15] Hisashi, S.; Syoji, O.; Masahiro, T.; Tsutomu, S.; Megumi, I.; Korekiyo, W.; Itsuo, U. *J Med Chem* 1998, 41, 1927.
- [16] Nagai, A.; Matsushita, Y.; Ono, N.; Takechi, Y. *Jpn. Kokai Tokkyo JP. 04173780*, 1992; *Chem Abstr* 1992, 117, 212485.
- [17] Padmavathi, V.; Venkatasubbaiah, D. R. C.; Balaiah, A.; Chandra obulareddy, B.; Padmaja, A. *Indian J Chem* 2005, 44B, 2569.
- [18] Padmavathi, V.; Sudheer, K.; Padmaja, A. *Indian J Chem* 2008, 47B, 734.
- [19] Padmaja, A.; Payani, T.; Dinneswara reddy, G.; Padmavathi, V. *Eur J Med Chem* 2009, 44, 4557.
- [20] Bhaskar Reddy, D.; Chandrasekhar Babu, N.; Padmavathi, V.; Sumathi, R. P. *Synthesis* 1999, 491.