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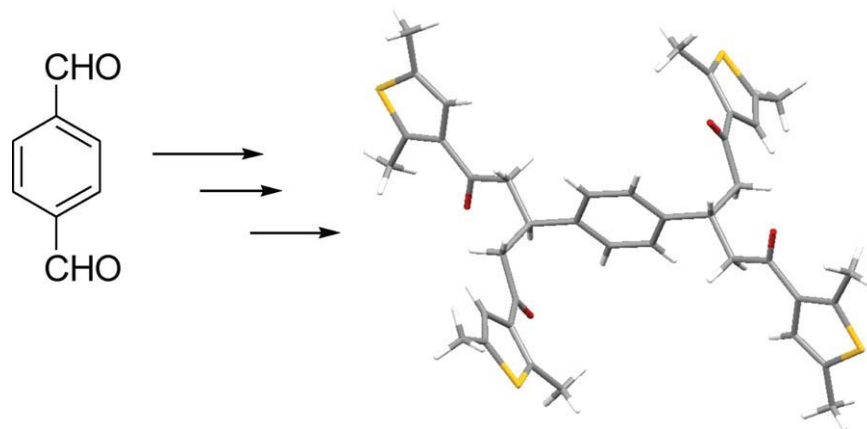
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Several 3,3'-(1,4-phenylene)bis(1,5-diones) and their chalcone precursors have been prepared in good to excellent yield via aldol addition and Michael addition starting from 3-acetyl-2,5-dimethylfuran or 3-acetyl-2,5-dimethyl-thiophene with terephthalaldehyde in the presence of appropriate base NaOH or lithium diisopropylamide. The kind and amount of alkali played a critical role in improving the reaction rates and yields of the products.

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INTRODUCTION

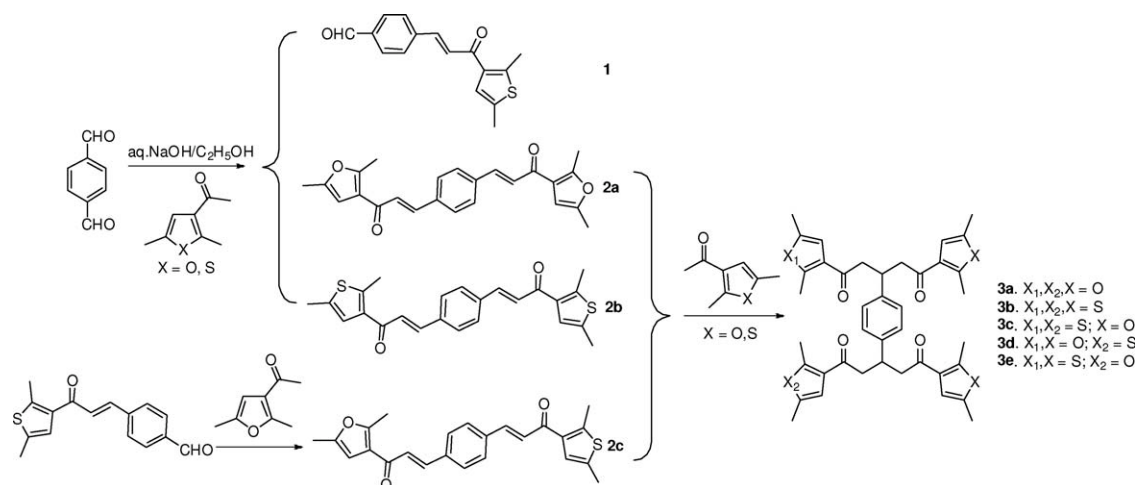
3,3'-(1,4-Phenylene)bis(1,5-diones) as versatile synthetic intermediates are important in the synthesis of photoluminescence [1–3], biosensors [4], and photochromism materials [5–8]. Li and Tian [9] synthesized hexatriene compounds provided by the 1,2-dithienylethene structure, which showed significant photochromic property. The target α , β -unsaturated compounds could also give rise to other heterocyclic compounds such as pyridines. Higuchi and coworkers [10] reported that Ru(II)-based metallo-supramolecular coordination polymers were potential photoluminescence device. The ligand terpyridines, which had rich coordination chemistry and generally displayed a high binding affinity to many metal ions, were obtained from the intermediate tetraketones. Thus, there is significant interest in the development of new compounds with tetraketones units. In the literature, we noticed that arylene diketones were prepared from terephthalaldehyde, isophthalaldehyde, 2,5-thiophenedicarboxaldehydes, and alkylphenyl ketones in the presence of alkali in ethanol [11–14]. Herein, we report the syntheses of a series of 3,3'-(1,4-phenylene)bis(1,5-diones) derivatives, which were well known to possess various biological activities [15,16] from 3-acetyl-2,5-dimethylfuran/3-

acetyl-2,5-dimethyl thiophene and terephthalaldehyde, as shown in Scheme 1. To improve the diversity of tetraketones, the tandem addition and humdrum reactants [11], which always lead to poor diversities, were avoided and a new chalcone **2c** was synthesized as precursor. Unsymmetrical intermediates can reduce photochromic molecular symmetry and broaden the absorption spectrum, which are very useful for multifrequency optical memories and displays [17,18].

RESULTS AND DISCUSSION

We envisioned the alkylation through the direct addition reactions of both 3-acetyl-2,5-dimethylfuran/3-acetyl-2,5-dimethylthiophene with terephthalaldehyde following ref. 14. However, the reaction occurred in aqueous sodium hydroxide (aq.NaOH) gave low yields. Maybe, it was attributed to poor activity and solubility of the reagents in alcohols. Both reactions carried in tetrahydrofuran (THF) and *N,N*-dimethylformamide (DMF) only gave byproducts. A flexible and diversified preparation of 3,3'-(1,4-phenylene)bis(1,5-diones) derivatives were carried out by convenient aldol addition and Michael addition.

Scheme 1

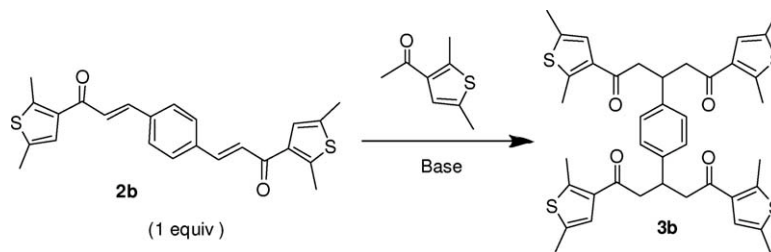


The requisite chalcones **2a**, **2b**, and **2c** were prepared from the corresponding commercially available 3-acetyl-2,5-dimethylfuran or 3-acetyl-2,5-dimethylthiophene

and terephthalaldehyde. Directly using a catalytic amount of 10% aq.NaOH, the desired chalcones **2a** and **2b** were obtained in 80 and 92% yields. Typically,

Table 1

Reaction conditions for the Michael addition of 3-acetyl-2,5-dimethylthiophene and **2b**^a.



Entries	Base	Solvent	T (°C)	Time (h)	Yield ^b
1	NaOH	— ^f	r.t.	2	0
2	aq.NaOH ^c	C ₂ H ₅ OH	r.t.	48	0
3	aq.NaOH ^a	C ₂ H ₅ OH	e	48	0
4	aq.NaOH ^a	C ₂ H ₅ OH/CH ₂ Cl ₂ ^d	r.t.	100	0
5	aq.NaOH ^a	C ₂ H ₅ OH/Toluene ^c	r.t.	100	0
6	aq.NaOH	C ₂ H ₅ OH/THF	r.t.	20	7
7	NaH(2.0 equiv)	DMF	r.t.	8	17
8	NaH(2.0 equiv)	DMF	r.t.	16	11
9	NaH(2.4 equiv)	DMF	r.t.	8	Trace
10	NaH(2.0 equiv)	DMF	0	24	Trace
11	LDA(2.4 equiv)	THF	−78 to −40	6	0
12	LDA(2.4 equiv)	THF	−78 to −40	20	0
13	LDA(4.8 equiv)	THF	−78 to −40	20	Trace
14	LDA(4.8 equiv)	THF	−25	2	68.5
15	LDA(4.8 equiv)	THF	−10	2	60
16	LDA(6.0 equiv)	THF	−25	2	60
17	LDA(4.0 equiv)	THF	−25	2	62

^a All products were characterized by ¹H, ¹³C NMR spectroscopy.

^b Isolated yields.

^c Reaction carried out under three different concentrations: 1, 10, and 50%.

^d Using TBAI as the phase transfer catalyst.

^e Reaction carried out under three different temperatures: r.t. (room temperature), 60°C, and 80°C.

^f Without using any solvent.

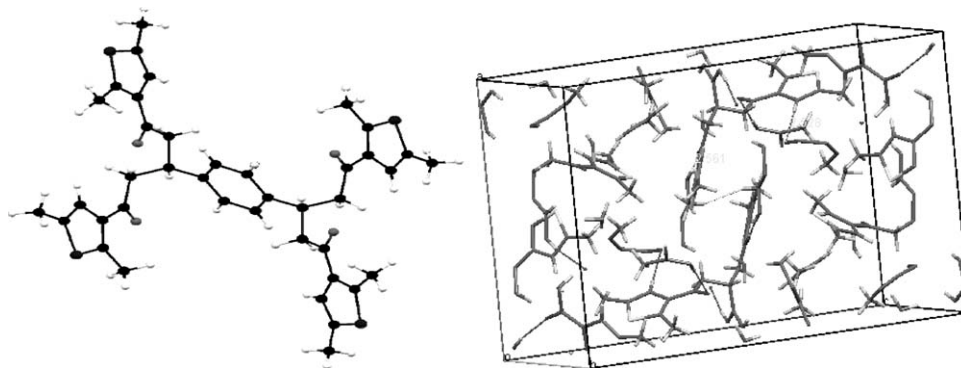


Figure 1. The molecular structure and crystal packing of compound **3b**.

terephthalaldehyde and 3-acetyl-2,5-dimethylthiophene were stirred in the presence of 0.1mol/L NaOH in ethanol to obtain mono-additive intermediate **1**, then treated with 3-acetyl-2,5-dimethylfuran and sodium hydroxide in ethanol to afford pale yellow powder **2c**. These conditions provided the expected products (**1**, **2c**) in well-pleasing NMR yields, 85 and 90%.

Table 1 showed the reaction conditions for the Michael addition of 3-acetyl-2,5-dimethylthiophene and **2b**, which was investigated by various parameters such as base, solvent, and time. The reaction was initially performed by dissolving and stirring the reactants in ethanol using aq.NaOH (1, 10, and 50%) without any catalyst. But even after stirring for 48 h, the reaction was incomplete and only byproducts were obtained (entries 2 and 3). Addition of the phase-transfer catalyst tetra-*n*-butylammonium iodide (TBAI) had no improvement in the occurrence of reaction (entries 4 and 5). Although the products could be obtained by adding THF, in which chalcones were well dissolved, the yield was only 7% because of the self-reaction of chalcone in the presence of NaOH (Table 1 entry 6). To improve the reaction efficiency, the effect of organic base was then investigated. It was found that lithium diisopropylamide (LDA) gave the best result, whereas potassium *tert*-butoxide and NaH were less active or ineffective (entries 7–17).

The reactions proceeded very smoothly and were completed within 2 h (entries 14–17), showing one major single spot on thin layer chromatography (TLC). The products were isolated through chromatography after quenched with water. The yields of the products were between 60 and 68%. The molecular structure of compound **3b** was also confirmed by X-ray diffraction analysis, as shown in Figure 1.

In the crystal structure of compound **3b**, $C_{40}H_{42}O_4S_4$, the dihedral angles between the tolyl ring and thiophene ring were 27.5(3) and 74.8(4)°, respectively. The two thiophene rings in a “diagonal line” were parallel. One

benzene C atom (C14) interacted with a carbonyl group O atom (O1) of an adjacent molecule through C—H...O hydrogen bond [2.561(2) Å], and one thiophene C atom (C8) interacted with a carbonyl group O atom (O2) of an adjacent molecule through C—H...O hydrogen bond [2.528 (3) Å] to form a three-dimensional supramolecular array.

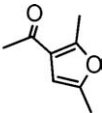
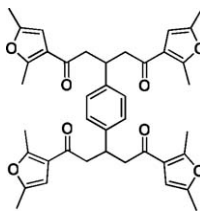
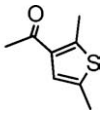
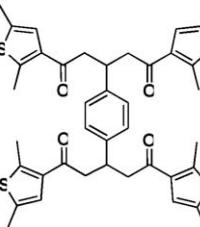
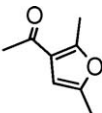
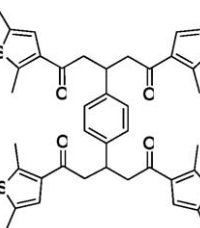
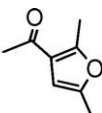
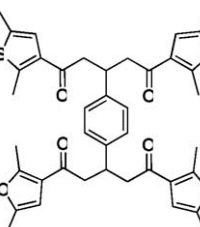
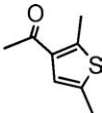
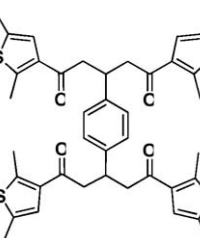
A variety of 3,3'-(1, 4-phenylene)bis(1,5-diones) have been prepared from the corresponding chalcones under optimized conditions described above. The results of these tetraketones are summarized in Table 2. The new chemistry provides the products in good to excellent yields.

In conclusion, a convenient and efficient method for preparing the important intermediate 3,3'-(1, 4-phenylene)bis(1,5-diones) derivatives and their chalcones precursors with the proper use of NaOH or LDA has been described. The kind and amount of alkali played a critical role in improving the reaction rates and yields of the products. The main features of the present method are: (1) the procedure is operationally simple using readily available chemicals and can provide various symmetrical 1,5-diketones in moderate to high yields of up to 70%; (2) employment of LDA as base is effective in improving the chemical yield in some cases; (3) unsymmetrical 1,5-diketones are also able to be prepared by the present method. Transformation of tetraketones into a new class of photoelectric compounds is in progress.

EXPERIMENTAL

Terephthalaldehyde and NaH were purchased from Aladdin Reagent Company. 3-Acetyl-2,5-dimethylfuran and 3-acetyl-2,5-dimethylthiophene were purchased from Alfa Aesar, LDA 2M sol. in THF/*n*-heptane/ethylbenzene from Acros Organics. Toluene and DMF were dried over 4 Å molecular sieves before use. THF was freshly distilled from sodium/benzophenone under a nitrogen atmosphere before use. All other solvents and reagents were purified by standard techniques or

Table 2
Synthesis of 3,3'-(1,4-phenylene)bis(1,5-diones) (**3a–3e**)^a.

Entries	Reagent	Product	Time (h)	Yield(%) ^b
1	2a + 		1	70
2	2b + 		2	68.5
3	2b + 		1.5	59
4	2c + 		2	65
5	2c + 		2	61

^a Reaction was conducted at -25°C .

^b Isolated yields.

used as supplied. TLC was carried out using commercially prepared 100–400 mesh silica gel plates (GF₂₅₄) that were visualized by irradiation (254 nm) and column chromatography was carried out using silica gel (200–300 mesh). Melting points (m.p.) were determined with an X4 melting point

inspection instrument and the thermometer was uncorrected. NMR spectra (¹H and ¹³C, respectively) were measured in deuteriochloroform (CDCl₃) on 400 MHz (operating frequencies: ¹H, 400.13 MHz; ¹³C, 100.61 MHz) FT spectrometers at ambient temperature with tetramethylsilane (TMS) as an

internal standard. Infrared (IR) spectra were obtained on a Bruke Tensor 27 spectrometer. Low-resolution mass spectra (MS) were recorded on an Esquire HCT plus. The mode of ionization used was atmospheric pressure chemical ionization (APCI) with methanol or THF as solvent. High resolution mass spectrometer [HRMS (FAB)] was recorded on MAT 95XP, Thermo.

X-ray diffraction analysis. All diffraction data were collected on a Bruker Smart 1000 CCD diffractometer with graphite-monochromated Mo K α radiation ($\lambda = 0.71073$ Å) at room temperature using the program SMART and processed by SAINT⁺. Space groups of these compounds were determined from systematic absences and further justified by the refinement results. In all cases, the structures were solved by direct methods and refined using full-matrix least-squares/difference Fourier techniques using SHELX. All nonhydrogen atoms were refined with anisotropic displacement parameters. All hydrogen atoms of the ligands were placed at idealized positions and refined as riding atoms with the relative isotropic parameters of the heavy atoms to which they are attached. The H atoms of water located from the difference Fourier map in the final stage of refinement.

4-(3-(2,5-Dimethylthiophen-3-yl)-3-oxoprop-1-enyl) benzaldehyde (1). A mixture of terephthalaldehyde (1.34 g, 10 mmol), 0.1 mol/L NaOH (20 mL), and 3-acetyl-2,5-dimethylthiophene (1.8 g, 12 mmol) was refluxed for 12 h. Water was added (10 mL) and pH was adjusted to 5–6 with 1M HCl. The yellow mixture was filtered, washed with water and cold ethanol and then crystallized from a mixture of CH₂Cl₂ and ethanol. A light yellow powder was obtained in 85% yield; m.p. 137–139°C; IR (KBr) ν : 2915, 1692, 1656, 1600, 1564, 1474, 1228, 1210, 1164, 1136, 988, 818, 732 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ (ppm): 10.04 (s, 1H), 7.92 (d, $J = 8.2$ Hz, 2H), 7.76 (d, $J = 8.3$ Hz, 2H), 7.72 (d, $J = 15.8$ Hz, 1H), 7.39 (d, $J = 15.7$ Hz, 1H), 7.10 (s, 1H), 2.73 (s, 3H), 2.46 (s, 3H); ¹³C NMR (101 MHz, CDCl₃) δ : 15.0, 16.0, 112.5, 125.7, 127.6, 128.7, 130.2, 135.6, 136.2, 137.0, 140.8, 141.5, 148.3, 185.5, 191.5; APCI-MS (m/z): 270.3.

General procedure for 2a–2c. Terephthalaldehyde or **1** (10 mmol) and 3-acetyl-2,5-dimethylfuran (22 mmol) were mixed together in a round-bottom flask with 30 mL ethanol, followed by the addition of 10% aq. NaOH (3 mL). The mixture was stirred for the appropriate time (0.5–1 h) at room temperature. After completion of the reaction indicated by TLC, water was added (10 mL), and the pH adjusted to 3–4 with 1M HCl; the yellow mixture was filtered, washed with water and cold ethanol, and crystallized from a mixture of CH₂Cl₂ and ethanol.

3,3'-(1,4-Phenylene)bis(1-(2,5-dimethylfuran-3-yl)prop-2-en-1-one) (2a). Yellow crystal, yield 3.0 g, 80%; m.p. 182–183°C; IR (KBr) ν : 2912, 1656, 1581, 1397, 1226, 1178, 1011, 973, 822, 711, 506 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ (ppm): 7.70 (d, $J = 15.8$ Hz, 2H), 7.62 (s, 4H), 7.21 (d, $J = 15.7$ Hz, 2H), 6.34 (s, 2H), 2.62 (s, 6H), 2.30 (s, 6H); ¹³C NMR (101 MHz, CDCl₃) δ : 13.3, 14.5, 105.6, 122.4, 125.0, 128.8, 136.7, 141.6, 150.1, 158.2, 185.6; APCI-MS (m/z): 374.0.

3,3'-(1,4-Phenylene)bis(1-(2,5-dimethylthiophen-3-yl)prop-2-en-1-one) (2b). Light yellow solid, yield 2.94 g, 92%; m.p. 185–186°C; IR (KBr) ν : 2915, 1653, 1592, 1479, 1358, 1331, 1224, 1196, 1138, 977, 815, 719, 679, 490 cm⁻¹; ¹H NMR

(400 MHz, CDCl₃) δ (ppm): 7.69 (d, $J = 15.7$ Hz, 2H), 7.63 (s, 4H), 7.29 (d, $J = 15.7$ Hz, 2H), 7.08 (s, 2H), 2.72 (s, 6H), 2.45 (s, 6H); ¹³C NMR (101 MHz, CDCl₃) δ : 15.0, 16.0, 125.8, 125.9, 128.8, 135.4, 136.5, 136.8, 142.3, 147.6, 186.0; APCI-MS (m/z): 406.5.

1-(2,5-Dimethylfuran-3-yl)-3-(4-((E)-3-(2,5-dimethylthiophen-3-yl)-3-oxoprop-1-enyl phenyl)prop-2-en-1-one) (2c). Light yellow solid, yield 3.4 g, 86%; m.p. 163–165°C; IR (KBr) ν : 2915, 1653, 1587, 1473, 1401, 1223, 1133, 972, 816, 709 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ (ppm): 7.70 (d, $J = 15.7$ Hz, 1H), 7.69 (d, $J = 15.7$ Hz, 1H), 7.63 (s, 4H), 7.31 (d, $J = 15.8$ Hz, 1H), 7.21 (d, $J = 15.8$ Hz, 1H), 7.09 (s, 1H), 6.34 (s, 1H), 2.72 (s, 3H), 2.62 (s, 3H), 2.45 (s, 3H), 2.30 (s, 3H); ¹³C NMR (400 MHz, CDCl₃) δ : 13.3, 14.5, 15.1, 16.0, 105.6, 125.0, 125.7, 125.8, 128.8, 135.4, 136.7, 141.6, 142.3, 150.1, 158.2, 185.6, 186.0; APCI-MS (m/z): 391.0.

General procedure for 3a–3e. After cooled to –25°C, to a round-bottom flask, 3-acetyl-2,5-dimethylfuran or 3-acetyl-2,5-dimethylthiophene (0.6 mmol) and 0.3 mL of LDA (2M) were added to 2.5 mL of dry THF. The yellow solution was stirred for 10 min and compounds **2a**, **2b**, and **2c** (0.12 mmol) were dissolved in dry THF (2 mL), and then added dropwise to the flask under Argon. After kept for 15 min at the temperature below –25°C, the reaction mixture was stirred at room temperature for 0.5–2 h. When the reaction was completed (monitored by TLC), the mixture was quenched with water, and pH was adjusted to 5–6 with 1M HCl. The solvent was removed by rotary evaporation. Dichloromethane (3 $\times$ 20 mL) was added to the residue to give a solid/liquid mixture. The mixture was filtered and the solid was thoroughly washed with dichloromethane (3 $\times$ 10 mL). The combined filtrates were concentrated in vacuum and the residue was purified by flash column chromatography and recrystallization. With this method, compounds **3a–3e** were prepared.

3,3'-(1,4-Phenylene)bis(1,5-bis(2,5-dimethylfuran-3-yl)pentane-1,5-dione) (3a). White crystal, yield 70%; m.p. 144–146°C; IR (KBr) ν : 2922, 1676, 1566, 1005, 948, 844 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ (ppm): 7.16 (s, 4H), 6.22 (s, 4H), 3.82–3.92 (m, 2H), 2.91–3.11 (m, 8H), 2.46 (s, 12H), 2.23 (s, 12H); ¹³C NMR (101 MHz, CDCl₃) δ : 13.2, 14.3, 36.0, 47.3, 105.6, 121.6, 127.5, 127.7, 149.8, 147.0, 157.0, 195.0; APCI-MS (m/z): 651.1. HRMS (FAB) calculated for C₄₀H₄₃O₈ ([M+H]⁺): 651.2952. Found: 651.2951.

3,3'-(1,4-Phenylene)bis(1,5-bis(2,5-dimethylthiophen-3-yl)pentane-1,5-dione) (3b). Colorless crystal, yield 68.5%; m.p. 180–182°C; IR (KBr) ν : 2916, 1660, 1546, 1476, 1365, 1216, 1130, 826 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ (ppm): 2.38 (s, 12H), 2.55 (s, 12H), 3.04–3.21 (m, 8H), 3.84–3.92 (m, 2H), 6.99 (s, 4H), 7.15 (s, 4H); ¹³C NMR (101 MHz, CDCl₃) δ : 15.0, 16.0, 36.3, 47.9, 126.0, 127.5, 135.0, 135.5, 142.2, 147.0, 194.7; APCI-MS (m/z): 714.7.

The single crystal of **3b** was obtained in dichloromethane and ethyl acetate solution (1:2). The crystal structure has been registered at the Cambridge Crystallographic Data Centre and allocated the deposition number CCDC 768841. C₄₀H₄₂O₄S₄, Mr = 715.02, space group Pbc_a, $a = 15.640(3)$ Å, $b = 10.139(2)$ Å, $c = 23.730(5)$ Å, $V = 3763.0(13)$ Å³, $Z = 4$, $D_x = 1.262$ g/cm⁻³, $F(000) = 1512$, $T = 293$ K, $R(\text{reflections}) = 0.0415(2493)$, $wR2(\text{reflections}) = 0.1157(3385)$, and $\theta_{\text{max}} = 25.200$.

3,3'-(1,4-Phenylene)bis(1-(2,5-dimethylfuran-3-yl)-5-(2,5-dimethylthiophen-3-yl)pentane-1,5-dione) (3c). White crystal, yield 59%; m.p. 138–140°C; IR (KBr) ν : 2919, 1661, 1566, 1481, 1364, 1226, 1136, 1003, 828, 665 cm^{-1} ; ^1H NMR (400 MHz, CDCl_3) δ (ppm): 7.15 (s, 4H), 6.99 (s, 2H), 6.21 (s, 2H), 3.82–3.92 (m, 2H), 2.92–3.23 (m, 8H), 2.54 (s, 6H), 2.46 (s, 6H), 2.39 (s, 6H), 2.23 (s, 6H); ^{13}C NMR (101 MHz, CDCl_3) δ : 13.2, 14.3, 15.0, 16.0, 36.1, 47.4, 47.9, 105.6, 121.5, 126.0, 135.0, 127.5, 135.0, 142.2, 147.3, 194.8, 157.0, 194.7; APCI-MS (m/z): 683.1. HRMS (FAB) calculated for $\text{C}_{40}\text{H}_{43}\text{O}_6\text{S}_2$ ($[\text{M}+\text{H}^+]$): 683.2496. Found: 683.2497.

3-(4-(1,5-Bis(2,5-dimethylfuran-3-yl)-1,5-dioxopentane-3-yl)phenyl)-1-(2,5-dimethylfuran-3-yl)-5-(2,5-dimethylthiophen-3-yl)pentane-1,5-dione (3d). White crystal, yield 65%; m.p. 136–137°C; IR (KBr) ν : 2920, 1675, 1566, 1481, 1392, 1229, 1004, 839 cm^{-1} ; ^1H NMR (400 MHz, CDCl_3) δ (ppm): 7.15 (s, 4H), 6.99 (s, 1H), 6.22 (s, 3H), 3.82–3.92 (m, 2H), 2.92–3.22 (m, 8H), 2.54 (s, 3H), 2.46 (s, 9H), 2.39 (s, 3H), 2.23 (s, 9H); ^{13}C NMR (101 MHz, CDCl_3) δ : 13.2, 14.3, 15.0, 15.9, 35.9, 36.1, 47.3, 47.4, 47.9, 105.6, 121.5, 126.0, 127.4, 127.5, 135.0, 135.5, 142.2, 147.3, 149.8, 157.0, 194.7, 195.0; APCI-MS (m/z): 667.0. HRMS (FAB) calculated for $\text{C}_{40}\text{H}_{43}\text{O}_7\text{S}$ ($[\text{M}+\text{H}^+]$): 667.2724. Found: 667.2722.

3-(4-(1,5-Bis(2,5-dimethylthiophen-3-yl)-1,5-dioxopentane-3-yl)phenyl)-1-(2,5-dimethylfuran-3-yl)-5-(2,5-dimethylthiophen-3-yl)pentane-1,5-dione (3e). White crystal, yield 61%; m.p. 149–150°C; IR (KBr) ν : 2918, 1668, 1571, 1481, 1365, 1256, 1136, 820 cm^{-1} ; ^1H NMR (400 MHz, CDCl_3) δ (ppm): 7.15 (s, 4H), 6.99 (s, 3H), 6.21 (s, 1H), 3.83–3.93 (m, 2H), 2.93–3.23 (m, 8H), 2.55 (s, 9H), 2.46 (s, 3H), 2.39 (s, 9H), 2.23 (s, 3H); ^{13}C NMR (101 MHz, CDCl_3) δ : 13.2, 14.3, 15.0, 15.9, 36.1, 36.3, 47.4, 47.8, 47.9, 105.6, 121.6, 126.0, 127.4, 127.5, 135.0, 135.5, 142.2, 147.3, 149.8, 157.0, 194.7, 195.0; APCI-MS (m/z): 699.0. HRMS (FAB) calculated for $\text{C}_{40}\text{H}_{43}\text{O}_5\text{S}_3$ ($[\text{M}+\text{H}^+]$): 699.2267. Found: 699.226.

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REFERENCES AND NOTES

- [1] Bhowmik, P. K.; Han, H.; Nedeltchev, A. K.; Mandal, H. D.; Jimenez-Hernandez, J. A.; McGannon, P. M. *J Appl Polym Sci* 2010, 116, 1197.
- [2] Bhowmik, P. K.; Han, H.; Nedeltchev, A. K.; Mandal, H. D.; Jimenez-Hernandez, J. A.; McGannon, P. M. *Polymer* 2009, 50, 3128.
- [3] Spiliopoulos, I. K.; Mikroyannidis, J. A. *J Polym Sci Part A: Polym Chem* 2001, 39, 2454.
- [4] Lu, Y.; Xiao, C. C.; Yu, Z. F.; Zeng, X. S.; Ren, Y.; Li, C. X. *J Mater Chem* 2009, 19, 8796.
- [5] Lucas, L. N.; de Jong, J. J. D.; van Esch, J. H.; Kellogg, R. M.; Feringa, B. L. *Eur J Org Chem* 2003, 155.
- [6] Dang, Y. Z.; Chen, Y. *Eur J Org Chem* 2007, 5661.
- [7] Liu, J. J.; Xu, Y.; Li, X. C.; Tian, H. *Dyes Pigm* 2008, 76, 294.
- [8] Chen, Y.; Zeng, D. X.; Xie, N.; Dang, Y. Z. *J Org Chem* 2005, 70, 5001.
- [9] Li, X. C.; Tian, H. *Macromol Chem Phys* 2005, 206, 1769.
- [10] Han, F. S.; Higuchi, M.; Kurth, D. G. *J Am Chem Soc* 2008, 130, 2073.
- [11] Krivun, S. V.; Dorofeenko, G. N. *Khimiya Geterotsiklichesikh Soedinenii* 1966, 5, 656.
- [12] Latva, M.; Takalo, H.; Simberg, K.; Kankare, J. *J Chem Soc Perkin Trans 2* 1995, 995.
- [13] Lin, F. Y.; Cheng, S. Z. D.; Harris, F. W. *Polymer* 2002, 43, 3421.
- [14] Huang, S. A. X.; Chuang, K. C.; Cheng, S. Z. D.; Harris, F. W. *Polymer* 2000, 41, 5001.
- [15] Cabrera, M.; Simoens, M.; Falchi, G.; Lavaggi, M. L.; Piro, O. E.; Castellano, E. E.; Vidal, A.; Azqueta, A.; Monge, A.; de Cerain, A. L.; Sagrera, G.; Scoane, G.; Cerecetto, H.; Gonzalez, M. *Bioorg Med Chem* 2007, 15, 3356.
- [16] Bhattacharya, A.; Mishra, L. C.; Sharma, M.; Awasthi, S. K.; Bhasin, V. K. *Eur J Med Chem* 2009, 44, 3388.
- [17] Irie, M. *Chem Rev* 2000, 100, 1685.
- [18] Higashiguchi, K.; Matsuda, K.; Tanifuji, N.; Irie, M. *J Am Chem Soc* 2005, 127, 8922.