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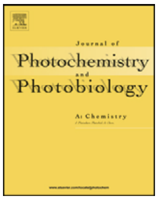

Morphology change of diarylethene derivatives having benzofuran derivatives—Photoinduced crystallization

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ABSTRACT

We have synthesized three diarylethene derivatives — 1,2-bis(2-(4-*n*-alkylphenyl)benzofuran-3-yl)ethenes (**1a**–**3a**). We have observed the morphology changes of the melted diarylethenes after heating. The crystal of **2a** melts at 102 °C. After heating at 130 °C, the liquid did not crystallize below 40 °C for more than 3 days because of the temperature above the eutectic point. When the liquid was irradiated with 365 nm light, the photoirradiated area crystallized, because of the generation of **2b**. After the sample was irradiated with visible light, the crystals changed to a colorless liquid. Photoinduced crystallization was also observed for the 4-*n*-dodecyl derivative (**3a**).

1. Introduction

Photochromism has attracted considerable attention because of its potential applications to molecular devices such as optical memories and switches [1,2]. Among various thermally irreversible photochromic compounds, diarylethene derivatives are the most promising compounds because of their fatigue resistance and thermal irreversibility [3–24]. Benzofuran derivatives have the nature of these properties.

The combination of self-assembly and photochromic behavior is very useful in opto electronic devices. It has been reported that diarylethene derivatives control liquid-crystalline phases by photochromism [20–24]. We report chiral photochromic dopants, (1*R*,2*R*)-*N*,*N*′-bis(4-{3-[1-(2-methylbenzo[*b*]thiophene-3-yl)hexafluorocyclopent-1-ene-2-yl]-2,4-dimethylthiophen-5-yl}benzilideneamino)cyclohexane [20] and (*R*)-1,1′-bis{3-[1-(2-methylbenzo[*b*]thiophene-3-yl)hexafluorocyclopent-1-ene-2-yl]-2,4-dimethylthiophen-5-yl}-bi-2-naphthol [21], which control the twisting power force in chiral nematic liquids by alternate irradiation with appropriate wavelengths of light. Polarization of a ferroelectric liquid crystal can be modulated by a diarylethene dopant, 1,2-bis(5-(4-*n*-(heptyloxy)phenyl)-2-methylthiophene-3-yl)hexafluorocyclopentene [22]. Photochromic diarylethenes connected to cyanobiphenyl groups, 1,2-bis(2-[11-{4-(4′-cyanophenyl-4-yloxy)undecyl}]benzo[*b*]thiophene-3-yl)perfluorocyclopentene [23,24], show liquid-crystalline properties. The diarylethene with a hexa(ethylene glycohol) side chains (*S*,*S*)-1,2-bis[2-methyl-5-(4-(*N*-(2-{2-[2-(2-{2-[2-(2-methoxy-ethoxy)-ethoxy]-ethoxy}-ethoxy)-ethoxy]-ethoxy}-1-methyl-ethyl))-carbamoyl-phenyl)-thiophene-3-yl]hexafluorocyclopentene [25], shows photoresponsive LCST (lower critical solution temperature) change in water, and formed a chiral self-assembled structure. Clouding is controlled by irradiation with the appropriate wavelengths of light. On solubility there is a large difference between benzo[*b*]thiophene and benzofuran derivatives. Although the solubility of 1,2-bis(2-methylbenzo[*b*]thiophene-3-yl)perfluorocyclopentene is 1×10^{-4} M in hexane, one of 1,2-bis(2-methyl-benzofuran-3-yl)perfluorocyclopentene is 1×10^{-2} M in hexane. The benzofuran derivatives undergo photochromism under the high concentration in solution. And there might be a difference between the open-ring and the closed-ring isomers on solubility. The solubility changes controlled by photoirradiation would make a novel category for functional materials. A partial crystallization would make the physical changes — such as transparency and polarization of the observed material. In this work, we synthesized benzofuran derivatives — the 1,2-bis(2-(4-*n*-alkylphenyl)benzofuran-3-yl)perfluorocyclopentene (**1a**–**3a**) and investigated their morphological behavior by photoirradiation. We attempted to construct a system in which crystallization can be induced from liquid phase using UV and visible lights.

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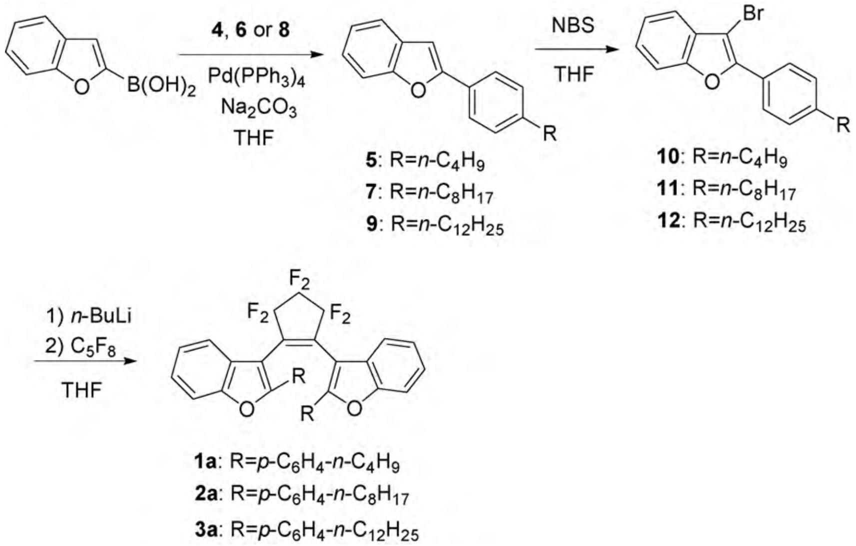
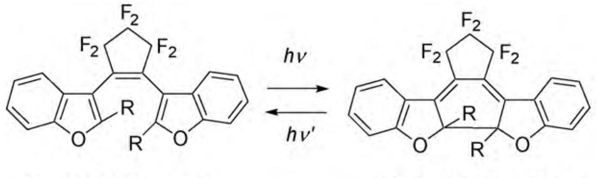


Scheme 1.



1a: R=p-C$_6$H$_4$-n-C$_4$H$_9$
2a: R=p-C$_6$H$_4$-n-C$_8$H$_{17}$
3a: R=p-C$_6$H$_4$-n-C$_{12}$H$_{25}$

1b: R=p-C$_6$H$_4$-n-C$_4$H$_9$
2b: R=p-C$_6$H$_4$-n-C$_8$H$_{17}$
3b: R=p-C$_6$H$_4$-n-C$_{12}$H$_{25}$

2. Experimental methods

2.1. General

The solvents used were spectrograde and were purified by distillation before use. The solubility of the diarylethene was measured in hexane (1 ml) at 25 °C. Absorption spectra were measured with a spectrophotometer (Hitachi, U-3310). For solution measurements, a mercury lamp (Ushio, 500 W) was used as the light source. Light of appropriate wavelength was isolated by passing light through a monochromator (RITSU MC-10N) or through Y-44 and UV-D33S filters. The quantum yields were determined by comparing the reaction rates of the diarylethene derivatives in hexane against that of furylfulgide in toluene. The samples were not degassed. Differential scanning calorimetry (DSC) measurements were carried out with a Pyris 1 apparatus (Perkin Elmer) with a liquid nitrogen cooling system (Perkin Elmer Cryofill). XRD analysis was carried out on a Mac Science MXP-18 (scan rate 4° min^{-1}, CuKα_1, 40 kV, 100 mA). Single crystals were observed using an OPTI-POL 2POL (Nikon) polarizing microscope. Photoirradiation for single-crystal measurements was carried out using 100 W mercury lamp (Nikon, C-SHG1 and LH-M100CB-1) as a light source. ^{1}H NMR were recorded on a Bruker AVANCE 400 spectrometer (400 MHz) with CDCl$_3$ as solvent and tetramethylsilane as an internal standard at room temperature. Good quality crystals (**1a**: 0.2 × 0.1 × 0.1 mm and **2a**: 0.25 × 0.15 × 0.1 mm) were selected for the X-ray diffraction study. These crystals were obtained from a solution of hexane and chloroform. Data collection was performed on a Bruker SMART 1000 CCD-based diffractometer (55 kV, 35 mA) with MoKα irradiation. We synthesized the 1,2-bis(2-(4-*n*-alkylphenyl)-1-benzofuran-3-yl)ethenes (**1a**–**3a**) (Scheme 1).

2.2. Synthesis of the diarylethene derivatives

2.2.1. 2-(4-n-Butylphenyl)-1-benzofuran (5)

1-Benzofuran-2-boronic acid (3.42 g, 21.2 mmol), tetrakistriphenylphosphine paradium (0) salt (763 mg, 0.96 mmol), sodium bicarbonate (2.5 g) and water (30 ml) were added to a solution of 1-bromo-4-*n*-butylbenzene (**4**) (4.05 g, 19.0 mmol) in THF (70 ml). The reaction mixture was heated for 3 h at 70 °C. The reaction mixture was poured into water (30 ml) and extracted with diethyl ether. The organic phase was dried over anhydrous magnesium sulfate and evaporated in vacuo. The crude product was purified by column chromatography on silica gel (hexane) to give 3.12 g of **5** in 66% yield.

5: colorless liquid; ^{1}H NMR (400 MHz, CDCl$_3$) $\delta = 0.94$ (t, $J = 7.6$ Hz, 3H), 1.34–1.44 (m, 2H), 1.60–1.68 (m, 2H), 2.65 (t, $J = 8.0$ Hz, 2H), 6.97 (s, 1H), 7.20–7.27 (m, 4H), 7.50–7.59 (m, 2H), 7.77–7.79 (m, 2H). Ms (EI) *m*/*z*: [M$^+$] 250. Anal Calcd. for $C_{18}H_{18}O$: C 86.36, H 7.25%. Found: C 86.37, H 7.10%.

2.2.2. 2-(4-n-Octylphenyl)-1-benzofuran (7)

Compound **7** was synthesized from 5.0 g (18.6 mmol) of 1-bromo-4-*n*-octylbenzene (**6**) by the same procedure as used for **5**. After purification, 3.88 g of **7** was obtained in 68% yield.

7: colorless liquid; ^{1}H NMR (400 MHz, CDCl$_3$) $\delta = 0.88$ (t, $J = 7.6$ Hz, 3H), 1.26–1.38 (m, 2H), 1.60–1.68 (m, 2H), 2.64 (t, $J = 8.0$ Hz, 2H), 6.97 (s, 1H), 7.20–7.27 (m, 4H), 7.50–7.58 (m, 2H), 7.77–7.79 (m, 2H). Ms (EI) *m*/*z*: [M$^+$] 306. Anal Calcd. for $C_{22}H_{26}O$: C 86.23, H 8.55%. Found: C 86.29, H 8.57%.

2.2.3. 2-(4-n-Dodecylphenyl)-1-benzofuran (9)

Compound **9** was synthesized from 6.5 g of 1-bromo-4-*n*-dodecylbenzene (**8**) (20 mmol) by a same procedure as used for **5**. After purification, 3.68 g of **9** was obtained in 51% yield.

9: colorless liquid; ^{1}H NMR (400 MHz, CDCl$_3$) $\delta = 0.94$ (t, $J = 7.6$ Hz, 3H), 1.34–1.44 (m, 18H), 1.60–1.68 (m, 2H), 2.65 (t, $J = 8.0$ Hz, 2H), 6.97 (s, 1H), 7.20–7.27 (m, 4H), 7.50–7.59 (m, 2H), 7.77–7.79 (m, 2H). Ms (EI) *m*/*z*: [M$^+$] 362. Anal Calcd. for $C_{26}H_{34}O$: C 86.13, H 9.45%. Found: C 86.02, H 9.58%.

2.2.4. 3-Bromo-2-(4-n-butylphenyl)-1-benzofuran (10)

To a stirred THF solution (30 ml) containing 2-(4-*n*-butylphenyl)-1-benzofuran (**5**) (3.12 g, 12.4 mmol), 2.66 g of *N*-bromosuccinimide (14.9 mmol) was slowly added at 5 °C, The

Table 1
Absorption characteristics and photoreactivity of diarylethene derivatives **1**–**3** in hexane.

Compound	ε (10^4 dm^3 mol^{-1} cm^{-1})		Quantum yield	
	a	*b*	Cyclization	Cycloreversion
1	3.81 (279 nm)	1.60 (482 nm)	0.43 (313 nm)	0.28 (500 nm)
2	4.02 (279 nm)	1.60 (482 nm)	0.46 (313 nm)	0.27 (500 nm)
3	4.25 (279 nm)	1.56 (482 nm)	0.47 (313 nm)	0.27 (500 nm)

reaction mixture was stirred for 15 h at room temperature. The reaction mixture was poured into sodium thiosulfate solution and extracted with diethyl ether. The organic layer was dried over anhydrous magnesium sulfate, and the solution was evaporated in vacuo. The crude product was purified by column chromatography on silica gel (hexane) to give 3.53 g of **10** in 87% yield.

10: colorless liquid; ^{1}H NMR (400 MHz, CDCl$_3$) δ = 0.95 (t, *J* = 7.8 Hz, 3H), 1.33–1.42 (m, 2H), 1.60–1.68 (m, 2H), 2.67 (t, *J* = 7.8 Hz, 2H), 7.32–7.36 (m, 2H), 7.48–7.56 (m, 1H), 8.07–8.09 (m, 1H). Ms (EI) *m*/*z*: [M$^+$] 328. Anal Calcd. for $C_{18}H_{17}BrO$: C 65.67, H 5.20%. Found: C 65.81, H 5.25%.

2.2.5. 3-Bromo-2-(4-n-octylphenyl)-1-benzofuran (**11**)

Bromination of **7** (6.88 g, 22.6 mmol) was carried out by a procedure similar to that used for **10**. The crude product was purified

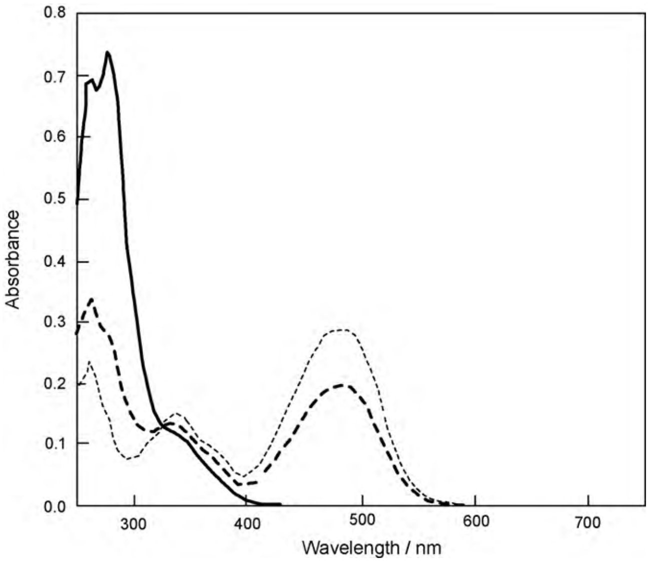


Fig. 1. Absorption spectra of **2a** (solid line), **2b** (dotted line) and at the photostationary state (dashed line) under irradiation with 313 nm light in hexane (1.8×10^{-5} mol/l).

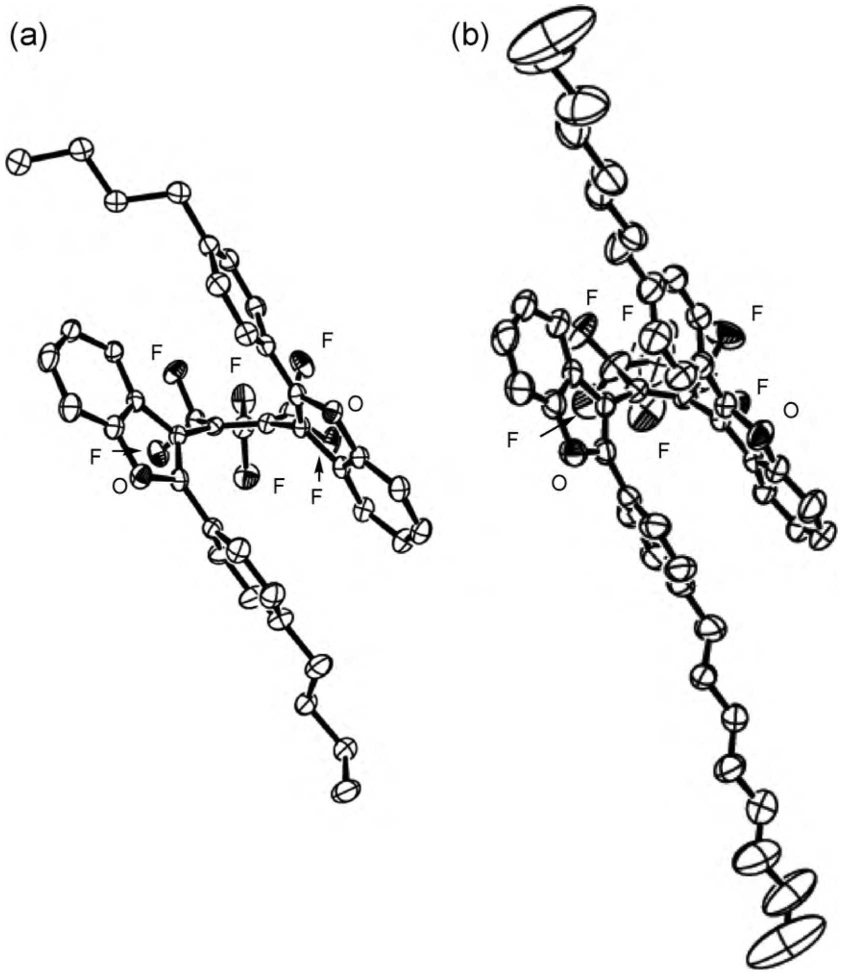


Fig. 2. ORTEP drawings of (a) **1a** and (b) **2a**. The ellipsoids represent 50% displacement of atoms. Hydrogen atoms are omitted for clarity.

by column chromatography on silica gel (hexane) to give 7.16 g of **11** in 83%.

11: colorless liquid; ^{1}H NMR (400 MHz, CDCl$_3$) $\delta = 0.88$ (t, $J = 7.2$ Hz, 3H), 1.20–1.40 (m, 18H), 1.62–1.68 (m, 2H), 2.67 (t, $J = 7.2$ Hz, 2H), 7.30–7.39 (m, 4H), 7.50–7.58 (m, 2H), 8.08–8.10 (m, 2H). Ms (EI) m/z: [M$^+$] 384. Anal Calcd. for $C_{22}H_{25}BrO$: C 68.57, H 6.54%. Found: C 68.39, H 6.48%.

2.2.6. 3-Bromo-2-(4-n-dodecylphenyl)-1-benzofuran (12)

Bromination of **9** (3.67 g, 10.1 mmol) was carried out by a procedure similar to that used for **10**. The crude product was purified by column chromatography on silica gel (hexane) to give 3.25 g of **12** in 73%.

12: colorless liquid; ^{1}H NMR (400 MHz, CDCl$_3$) $\delta = 0.88$ (t, $J = 7.2$ Hz, 3H), 1.20–1.40 (m, 18H), 1.62–1.68 (m, 2H), 2.67 (t, $J = 7.2$ Hz, 2H), 7.30–7.39 (m, 4H), 7.50–7.58 (m, 2H), 8.08–8.10 (m, 2H). Ms (EI) m/z: [M$^+$] 440. Anal Calcd. for $C_{26}H_{33}BrO$: C 70.74, H 7.53%. Found: C 70.54, H 7.52%.

2.2.7. 1,2-bis(2-(4-n-Butylphenyl)-1-benzofuran-3-yl)perfluorocyclopentene (1a)

To a stirred THF solution (30 ml) containing 3-bromo-2-(4-*n*-butylphenyl)-1-benzofuran (**10**) (2.78 g, 8.47 mmol), 8.47 ml of 1.6 M *n*-butyllithium hexane solution (9.31 mmol) was slowly added at −78 °C, and the solution was stirred for 15 min at −78 °C. Then the octafluorocycylpentene (0.561 ml, 0.424 mmol) was added slowly to the reactionmixture at −78 °C and left to stand with stirring at −78 °C to 30 °C for 12 h. The reaction mixture was poured into concentrated sodium chloride solution and extracted with diethyl ether. The organic phase was dried over anhydrous magnesium sulfate and evaporated in vacuo. The crude product was purified by column chromatography on silica gel (hexane) to give 0.569 g of **1a** in 40% yield.

1a: yellow crystals; ^{1}H NMR (400 MHz, CDCl$_3$) $\delta = 0.93$ (t, $J = 7.2$ Hz, 6H), 1.26–1.34 (m, 4H), 1.40–1.48 (m, 4H), 2.32 (t, $J = 7.2$ Hz, 4H), 6.77–6.79 (m, 4H), 7.00–7.07 (m, 6H), 7.14–7.16 (m, 4H), 7.25–7.27 (m, 2H). Ms (EI) m/z: [M$^+$] 672. Anal Calcd. for $C_{41}H_{34}F_6O_2$: C 73.20, H 5.09%. Found: C 73.31, H 5.21%. CCDC deposition number: 724436.

2.2.8. 1,2-bis(2-(4-n-Octylphenyl)-1-benzofuran-3-yl)perfluorocyclopentene (2a)

The reaction of 3-bromo-2-(4-*n*-octylphenyl)-1-benzothiophene (**11**) (2.00 g, 5.19 mmol), 1.6 M *n*-butyllithium in hexane (3.57 ml, 5.70 mmol), and octafluorocyclopentene (0.343 ml, 2.59 mmol) was carried out as described for compound **1a**. The crude product was purified by column chromatography on silica gel (hexane) to give 0.680 g of **2a** in 33% yield.

2a: yellow crystals; ^{1}H NMR (400 MHz, CDCl$_3$) $\delta = 0.90$ (t, $J = 7.2$ Hz, 6H), 1.27–1.35 (m, 20H), 1.40–1.48 (m, 4H), 2.30 (t, $J = 7.2$ Hz, 4H), 6.77–6.79 (m, 4H), 6.98–7.05 (m, 6H), 7.12–7.15 (m, 4H), 7.25–7.27 (m, 2H). Ms (EI) m/z: [M$^+$] 784. Anal Calcd. for $C_{49}H_{50}F_6O_2$: C 74.98, H 6.42%. Found: C 75.07, H 6.39%. CCDC deposition number: 724437.

2.2.9. 1,2-bis(2-(4-n-Dodecylphenyl)-1-benzofuran-3-yl)perfluorocyclopentene (3a)

The reaction of 3-bromo-2-(4-*n*-dodecylphenyl)-1-benzothiophene (**12**) (1.8 g, 4.07 mmol), 1.6 M *n*-butyllithium hexane solution (2.80 ml, 4.48 mmol), and octafluorocyclopentene (0.269 ml, 2.03 mmol) was carried out as described for compound **1a**. The crude product was purified by column chromatography on silica gel (hexane) to give 0.247 g of **3a** in 14% yield.

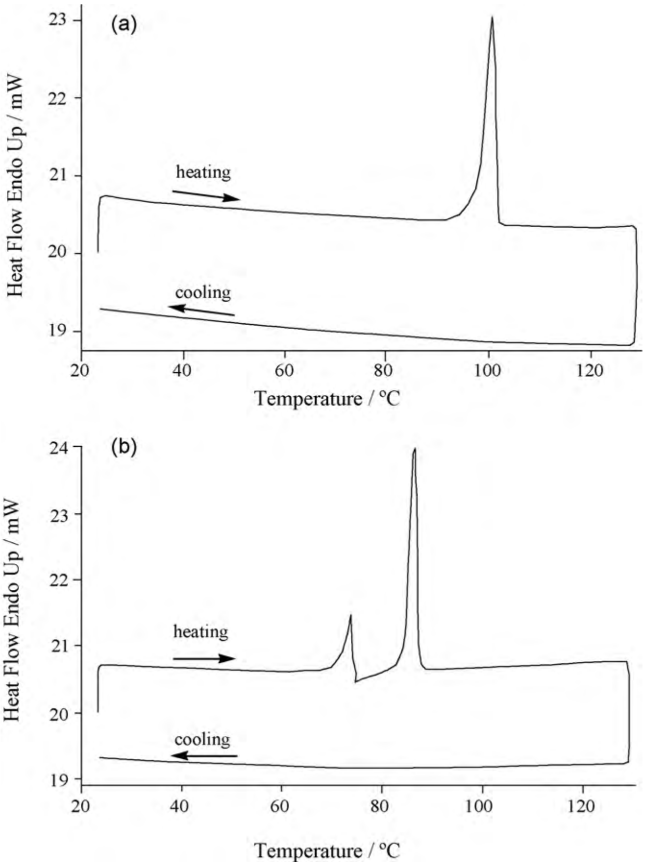


Fig. 3. DSC profiles of **2a**: (a) the first run and (b) the second run (heating rate: 4 °C/min, cooling rate: 4 °C/min).

3a: yellow crystals; ^{1}H NMR (400 MHz, CDCl$_3$) $\delta = 0.88$ (t, $J = 7.2$ Hz, 6H), 1.26–1.33 (m, 36H), 1.40–1.48 (m, 4H), 2.32 (t, $J = 7.2$ Hz, 4H), 6.76–6.78 (m, 4H), 6.98–7.05 (m, 6H), 7.12–7.16 (m, 4H), 7.24–7.26 (m, 2H). Ms (EI) m/z: [M$^+$] 897. Anal Calcd. for $C_{57}H_{66}F_6O_2$: C 76.31, H 7.42%. Found: C 76.32, H 7.47%.

3. Results and discussion

3.1. Synthesis of diarylethenes 1a–3a

Diarylethenes **1a**–**3a** were prepared from 2-(4-*n*-alkylphenyl)-3-bromo-1-benzofuran with octafluorocyclopentene. The detailed strategy was shown in Scheme 1. Their structures were confirmed by ^{1}H NMR, mass spectrometry, and elemental analysis. Diarylethenes **1a** and **2a** were confirmed by X-ray crystallography.

3.2. Photochromism of 1a–3a in hexane and in the crystalline phase

Diarylethenes **1a**–**3a** show photochromism in hexane solution. Fig. 1 shows absorption spectra of **2a**, **2b**, and photostationary state under irradiation with 313 nm light in hexane. Upon irradiation with 313 nm light, the open-ring isomers of **1a**, **2a**, and **3a** transform to the closed-ring isomers **1b**, **2b**, and **3b**, respectively. Upon irradiation with >440 nm light, **1b**, **2b**, and **3b** convert to **1a**, **2a**, and **3a**, respectively. Absorption characteristics and photoreactivity of **1**–**3** are summarized in Table 1. The cyclization quantum yields were increased with the increasing of chain length, but the cycloreversion quantum yield did not affect by the increasing of the chain length.

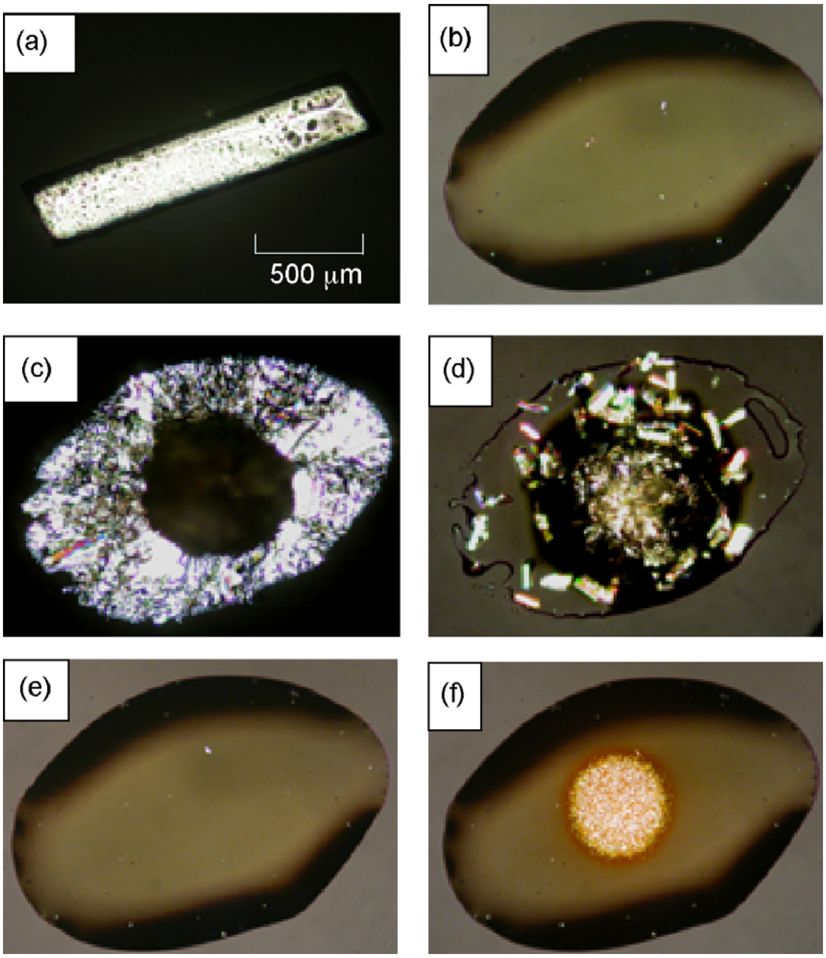


Fig. 4. Photographs of **2**: (a) a single crystal, (b) after being heated at 130 °C (observed at 40 °C), (c) after 3 h at 30 °C, (d) second heating at 80 °C, (e) after 3 days at 40 °C, and (f) upon irradiation with 365 nm light at 40 °C for 3 min.

Single crystals of **1a** and **2a** were obtained from hexane solution. The single crystals of **1a** and **2a** underwent photochromism in a single-crystalline phase. These crystals were analyzed by X-ray crystallography (Fig. 2) [26]. The aryl groups are in an anti-parallel conformation, and the distances between the reactive carbons are 0.382 and 0.399 nm, respectively. The distances are sufficient for the molecules to undergo photochromism in the single-crystalline phase [27]. The crystals **3a** did not analyze by X-ray crystallography because the suitable crystals did not obtained. The crystals showed photochromism in the crystalline phase upon alternate irradiation with 365 nm light and >440 nm light.

3.3. *DSC profiles and morphology changes of **1a**–**3a***

Fig. 3(a) shows DSC curve of **2a** on the first cycle. The cycle proceeds as follows: (1) heat from 30 to 130 °C at 4 °C/min, (2) hold for 2 min at 130 °C, and (3) cool from 130 to 30 °C at 4 °C/min. From the DSC measurement, sample **2a** melts at 102 °C When the sample was cooled to 30 °C, the crystallization of **2a** did not occur immediately, but occurred after 90 min. Fig. 3(b) shows DSC curves on the second cycle for recrystallized **2a** after standing for 1 day at 30 °C. In the cycle, two clear endotherms occur at 72 and 86 °C upon heating. A single crystal of **1a** melts at 164 °C. DSC measurements show that there is no change in the melting point between the first and second scans (see supplementary data).

In order to elucidate this phase change, we observed single crystals of **2a** under a polarized microscope under crossed nicol prisms. Fig. 4(a) shows a photograph of the single crystal of **2a**. The crystal of **2a** is square, and the molecules are packed in an anti-parallel conformation. After heating at 130 °C, the single crystals of **2a** melted (Fig. 4(b)). During the liquid was cooled to 30 °C after 3 h, the liquid changed into the crystals (Fig. 4(c)). During the second heating, part of the crystals melts at 80 °C, and only square crystals are observed (Fig. 4(d)). The square crystals melt at 86 °C, and the phase changes to the liquid, as indicated in Fig. 4(b). We confirmed that the sample did not decompose after heating at 130 °C by HPLC measurements. These results suggest that two (or more) conformations exist after heating at 130 °C. The existence of two kinds of crystals produces the liquid state of **2a** at 40 °C, because the temperature is above the eutectic point. The state is stable after more than 3 days at 40 °C (Fig. 4(e)).

The existence of two (or more) kinds of crystals was observed by XRD analysis. A series of phase transfer changes was also observed in the XRD pattern (Fig. 5). The sample in Fig. 5(a) is the same as that in Fig. 4(a), which is initially a square crystal. After the crystal of **2a** melts, the XRD spectrum is changed to the shallow pattern (Fig. 5(b)), which indicates that the state is liquid. The XRD pattern of **2a** after recrystallization sample is shown in Fig. 5(c). The spectrum indicates that two (or more) kinds of crystals are contained in the sample. The two states between Fig. 5(a) and (c) are not identical.

More interestingly, crystallization by UV light illumination was observed. Fig. 4(f) shows the photoinduced crystallization of **2** upon irradiation with 365 nm light for 3 min at 40 °C. The crystal color generated was red, and the crystal contained the closed-ring isomer. The crystal generated in the liquid dissolved upon irradi-

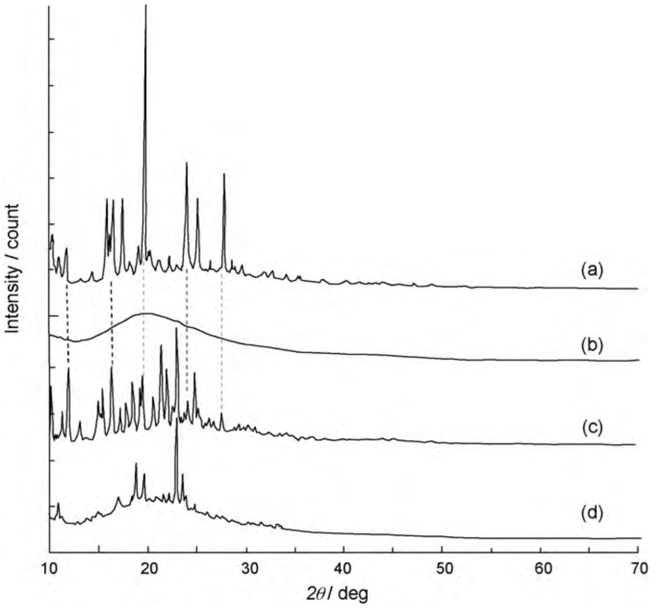


Fig. 5. XRD spectrum of **2**: (a) open form (square crystal), (b) melted sample, (c) recrystallized sample, and (d) crystallized sample under irradiation with 365 nm light.

ation with 440 nm light for 1 min at 40 °C. The crystallization by photoirradiation was reversed 10 times by alternating irradiation between 365 and 440 nm light. Crystallization by photoirradiation was observed at temperature between 40 and 70 °C. The photoinduced crystallization was also observed by XRD (Fig. 5(d)). The peaks of the XRD spectrum represent the crystals of **2b**, because the XRD pattern is consistent with one of the pure **2b** crystals (recrystallization solvent: diethyl ether). The solubility of **2a** is 0.1 M in hexane at 25 °C, but that of **2b** is 0.002 M in hexane at 25 °C. The poor solubility of **2b** induces crystallization in the bulk liquid of **2a**.

A similar photoinduced crystallization was observed for **3a**. The melting point of **3a** was 68 °C from DSC measurements. After heating at 80 °C, the single crystal **3a** melted, but the liquid did not change into the crystals at 30 °C. The crystallization by photoirradiation was reversed several times by alternating irradiation between 365 and 440 nm light at 30 or 40 °C.

4. Conclusions

We have synthesized 1,2-bis(2-(4-*n*-alkylphenyl)benzofuran-3-yl)ethene derivatives. A single crystal of **1a** melted at 164 °C. DSC measurements show that there is no change in the melting point between the first and second scans. The crystal of **2a** melted at 102 °C. After heating at 130 °C, the liquid did not crystallize below 40 °C for more than 3 days because of the temperature above the eutectic point. When the liquid was irradiated with 365 nm light, the photoirradiated area crystallized. From the XRD analysis, the photo-generated crystal was identical with **2b**. After the sample was irradiated with visible light, the crystals changed to a colorless liquid. Photoinduced crystallization was also observed for the 4-*n*-dodecyl derivative (**3a**) at 40 °C. We have constructed a photoinduced crystallization system based on this photochromic reaction.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jphotochem.2010.05.013.

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