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Photochromism of Heterocyclic Fulgides[1)]

Akihiko Tomoda*, Akira Kaneko, Hideki Tsuboi,
and Ryouka Matsushima†
Basic Research Development Laboratory, Toyooka Plant, Yamaha Co.,
Toyooka-mura, Iwata-gun, 438-01
† Department of Applied Chemistry, Faculty of Engineering,
Shizuoka University, Hamamatsu 432

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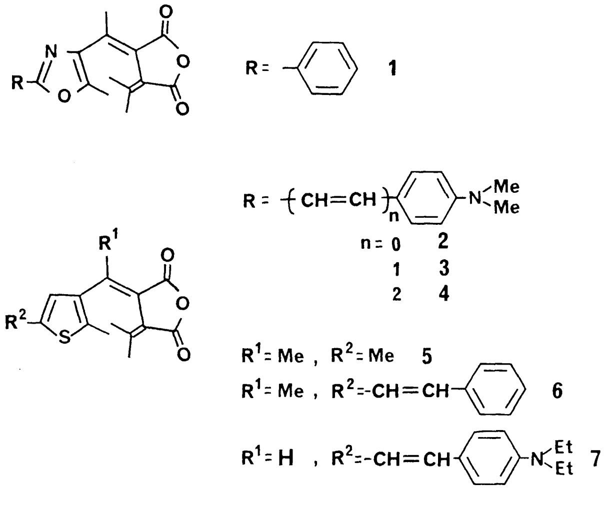
Aiming at practical uses of photochromic compounds as data-storage media, electron-donating (e.g. *p*-dimethylaminophenyl) and conjugate chain (e.g. *p*-dimethylaminostyryl) substituted heterocyclic fulgides, e.g. 2-[1-(2-substituted 5-methyl-4-oxazolyl)ethylidene]-3-isopropylidenesuccinic anhydride and 2-[1-(5-substituted 2-methyl-3-thienyl)ethylidene or methlyene]-3-isopropylidenesuccinic anhydride, were prepared. The effect on photochromic performance was also investigated; it was clarified that substituents had effects on the red-shift and a significant increase in the color intensity of a colored form. However, the rate of the discoloration reaction was also affected, depressed, by the substituent.

Recently, work on organic photochromic compounds as data-storage media has received much attention because of a potential to achieve higher signal densities in a re-writable recording system. There have been many attempts to improve the thermal stability of the two states of a photochromic reaction, e.g. uncolored and colored forms and, likewise, to improve the photochemical fatigue resistance in relation to the cycle-repeating durability.[2)] Irie and Mohri[3)] have discovered thermally stable photochromic 1,2-diarylethene derivatives, some of which have shown excellent photochemical fatigue resistance.

Another photochromic compound, fulgide, has also been studied by Heller[4)] since 1970. He showed that fulgides containing a heterocyclic structure, such as furyl, thienyl, and pyrrolyl, were excellent candidates for data-storage media because of their efficient thermal stability. Recent work on this type of compound has promoted improvements in the quantum yields for both coloration and discoloration reactions,[5)] and stimulated investigations on molecular structure using the MNDO-method[6)] and synthetic route.[7)]

Photochromic reactions involve molecular structural changes, and are significantly affected by the structure. Therefore, in order to practically utilize the photochromic compounds, clarifying the relationship between the chemical structure and photochromic properties is indespensable; it would hopefully lead to an approach to effectively design a new molecule. We[8)] have also studied various heterocyclic fulgides, showing that (4-oxazolyl)fulgide (**1**) possesses an efficient thermal stability as well as photochemical fatigue resistance, owing to the higher chemical stability of the aromatic structure. In this paper we report on the effect of an electron-donating or conjugate chain substituent on the heteroaromatic structure of a fulgide, such as (4-oxazolyl) or (3-thienyl)fulgide. (Scheme 1).



Scheme 1.

Results and Discussion

Photochromic and Spectroscopic Properties of Fulgides. Heterocyclic fulgides were prepared in 3—15% yields by Stobbe condensations of a 4-(3-)acetyl (or formyl)thiophene and 4-acetyl oxazole derivative with diethyl 2-isopropylidenesuccinate, followed by hydrolysis and dehydration, as described in the literature[4,5)]. Figure 1 shows the absorption spectrum change of [2-(*p*-dimethylaminophenyl)oxazol-4-yl]fulgide (**2**) in a dilute toluene solution (5×10^{-5} mol dm^{-3}). The photostationary state (pss) for coloration, due to a intramolecular cyclization, was attained by irradiation with UV light (366 nm) for 120 s, and showed an absorption maximum at 510 nm. Subsequent irradiation for 240 s with visible light (>500 nm) induced discoloration, e.g. a disclosure reaction leading to (**2**). HPLC measurements showed the formation of 87% of the colored form (**2C**) in pss under UV light irradiation, as well as a quantitative regeneration of the uncolored form (**2**) with visible light irradiation. Alternate irradiation with UV and visible light on a dilute toluene solution of (**2**) was repeated in the presence of air. After 500 times of cycle-repeating, the absorption intensity change of (**2C**) suggested that only 20% of the fulgides were photochemically lost.

Figures 2 and 3 illustrate the absorption spectra of colored forms of fulgide (**1**—**4**) and (**5**—**7**), respectively. The spectroscopic properties are summarized in Table 1. These spectra indicate that the introduction of electron-donating *p*-dimethylaminophenyl and other conjugate chains to a heteroaromatic structure of fulgide is remarkably effective to red-shift, while increasing the molecular absorption coefficient (ε) of the colored form. However, no significant difference in the absorption

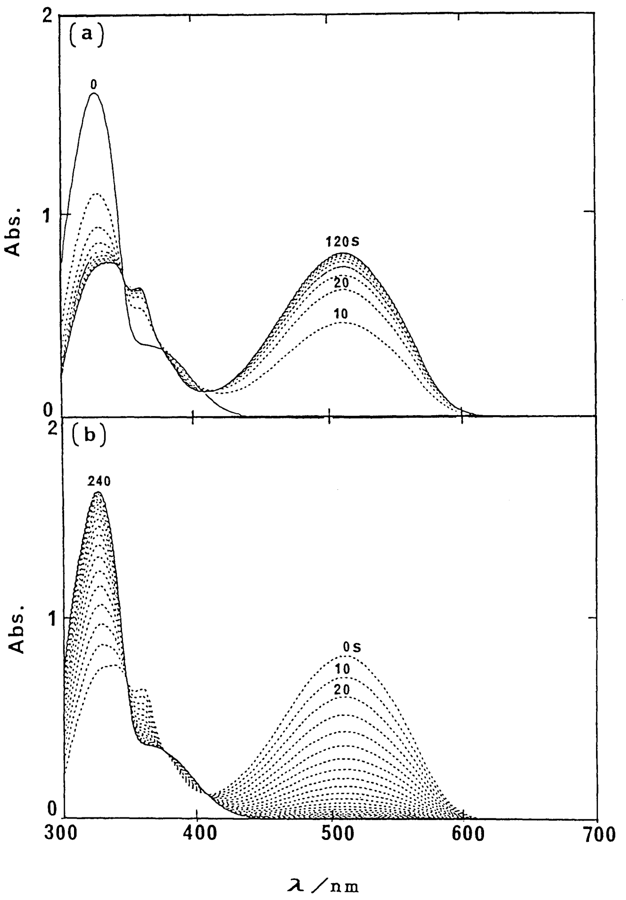


Fig. 1. Absorption spectrum change of (4-oxazolyl)-fulgide (**2**) in a dilute toluene solution (5×10^{-5} mol dm^{-3}) with UV (a) and visible light (b) irradiation. (a): Curve 0 is the spectrum of the uncolored form (**2**,*E*-isomer) absorption curves around 510 nm refer to the colored forms after 120 s UV light irradiation. The spectrum was recorded every ten seconds of irradiation time. (b): Curve 240 refers to uncolored form regenerated by visible light irradiation for 240 s.

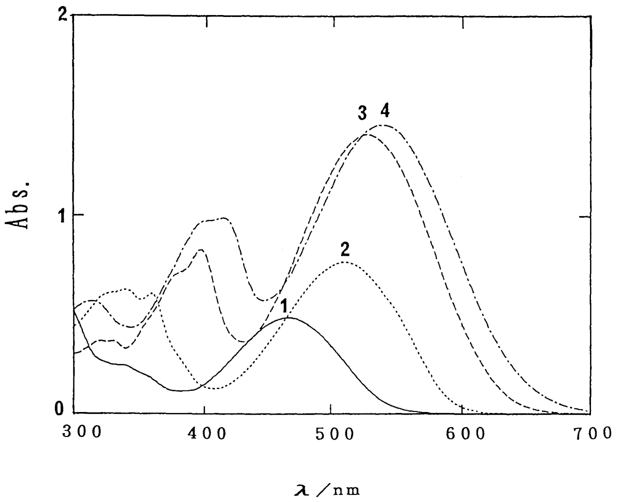


Fig. 2. Absorption spectrums of colored form of various (4-oxazolyl)fulgides (**1**—**4**) in a dilute toluene solution. The colored form was prepared by irradiation of UV light on the uncolored form solution (5×10^{-5} mol dm^{-3}).

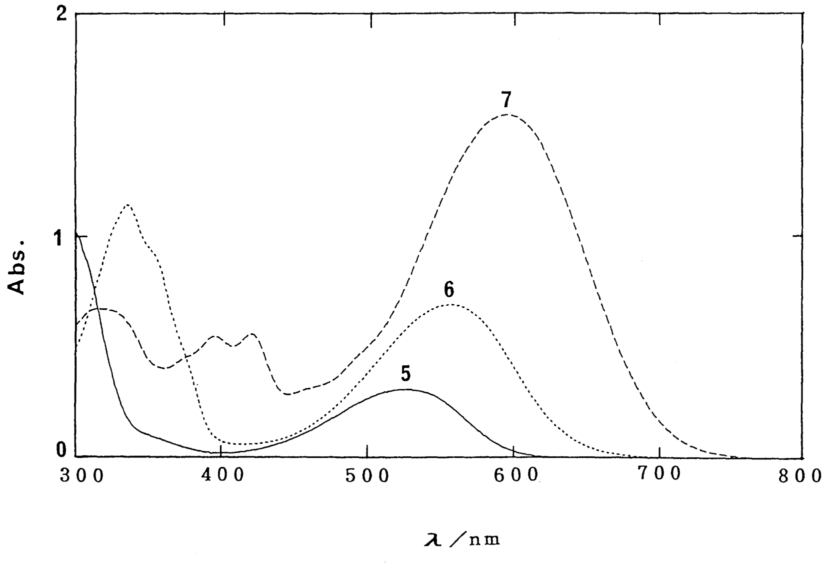


Fig. 3. Absorption spectrums of colored form of various (3-thienyl)fulgides (**5**—**7**) in a dilute toluene solution (5×10^{-5} mol dm^{-3}).

Table 1. Spectroscopic Properties of Fulgides in Toluene Solution[a)]

Fulgide	*E*	*Z*	*C*
	λ_{max}/nm (ε_{max}/mol^{-1} cm^{-1})	λ_{max}/nm (ε_{max}/mol^{-1} cm^{-1})	λ_{max}/nm (ε_{max}/mol^{-1} cm^{-1})
1	337 (10100)	347 (13800)	462 (11000)
2	325 (33300)	322 (31400)	510 (18300)
3	379 (36100)	379 (36800)	528 (26000)
4	—[b)]	403 (37800)	538 (27600)
5	334 (5670)	344 (7010)	526 (7180)
6	334 (31800)	—[b)]	557 (15200)
7	382 (41400)	—[b)]	595 (30500)

a) The absorption properties were determined by the HPLC measurement of the fulgide solution under UV or visible light irradiation. b) Not measured.

spectra between colored fulgide (**3**) and (**4**) was observed. In a previous paper[8)] we suggested that the electronic transition of the colored form involves an intramolecular electron transfer from the hetero-atom to the carbonyl group; the absorption maxima would tend to red shift with the electron-donating ability of the heterocyclic group. It is therefore understandable that an electron-donating group on a heteroaromatic structure affects the spectroscopic properties, as shown in Figs. 1 and 2. To the contrary, little changing the absorption spectra of colored fulgide (**4**), compared with (**3**), is attributable to a lack of the coplanarity, due to the presence of two flexible vinyl groups which inhibit an increase in the electron-donating ability of the heterocyclic group.

In Fig. 3, (3-thienyl)fulgides (**5**—**7**) also exhibit the tendency to red-shift along with an introduction of a substituent similar to (4-oxazolyl)fulgides.

Heller[9)] also had prepared (5-phenyl-3-thienyl)fulgide in place of the 5-methyl-3-thienyl derivative; he reported that the colored form showed a red-shifted λ_{max} (546 nm) and a higher ε (12000 mol^{-1} cm^{-1}). These results imply that the introduction of appropriate substituent on the heteroaromatic fulgides has practical significance in designing the color and color intensity.

It is also important to note that [5-(*p*-diethylaminostyryl)-2-methyl-3-thienyl]fulgide (**7**) converted to the colored form quantitatively (HPLC measurement showed us that 100% of the colored form was photo-generated). Again, Heller points out in his paper[9)] that to cause a photo-coloration reaction, R^1 of (3-thienyl) fulgide (scheme) should be a substituent (e.g. methyl), since the *E*–*Z* isomerization which occurred competitively with the coloration was inhibited by the bulkiness of the substituent (R^1). However, regardless of this suggestion, fulgide (**7**) (R^1; hydrogen) boasts rapid photo-coloration. This fact says that the electronic effect of the substituent contained in the heteroaromatic structure is favorable for a ring-closure reaction, rather than a steric effect.

The absorption spectroscopic data of the uncolored form is shown in Table 1. A significant increase of ε was observed. (4-Oxazolyl)fulgide (**3**) exhibited more than three times the quantity of ε compared to that of fulgide (**1**); the thienyl series also shows a greater tendency to increase ε. The wavelengths of the uncolored forms were hardly affected by these substituents.

Photo-Responsiveness of Fulgides. Figure 4 shows the absorption intensity change at λ_{max} of the colored form in a dilute toluene solution, according to the coloration and discoloration reaction upon UV and visible light irradiation. The discoloration rate was strongly dependant on the substituent.

The presence of *p*-dimethylaminophenyl and other conjugate chains depressed the discoloration rate remarkably in the following order: fulgide (**1**), (**2**), (**3**), and (**4**). The coloration, however, was not very much affected. Table 2 summarizes the quantum yields (Φ_{CE}) for the discoloration of fulgides (**1**—**7**). The 3-thienyl series also showed a similar depressed rate.

To the contrary, Yokoyama and Kurita[10)] reported (5-dimethylamino-3-indolyl)fulgide, which also showed a lowered discoloration rate (Φ_{CE}; 0.0001); a remarkable red-shift of the colored form was achieved.

It is known that the photo-discoloration rate is dependant upon the temperature, e.g the reaction is thermally activated. The activation energy for (3-thienyl)fulgide (**5**) had been estimated as being 50 cm^{-1} by Ulrichll). We also investigated the temperature-dependency of the reaction in PMMA thin film (ca. 1 μm thickness) for fulgide (**1**), (**2**), and (**3**). Arrhenius plots revealed that the discoloration of these fulgides was also thermally activated, and that the temperature-dependency are in the following order of (**3**), (**2**), and (**1**). The activation energies of fulgide (**1**), (**2**), and (**3**) were 2.2, 4.8, and 6.5 kcal mol^{-1}, repsectively. This indicates that the quantity of the activation energy barrier is clearly dependent upon the substituent. The large quantity of colored forms (**2C** and **3C**) is presumably due to a higher degree of delocalization over the fulgide molecule by the *p*-dimethylaminophenyl and *p*-dimethylaminostyryl groups, compared to phenyl.

On the other hand, as an explanation of the lowered discoloration rate we considered the possibility of a side-reaction which would included *E*–*Z* isomerization of the olefinic bond contained in colored fulgide (**3C** and **4C**). To clarify the relation between the *E*–*Z* isomerization

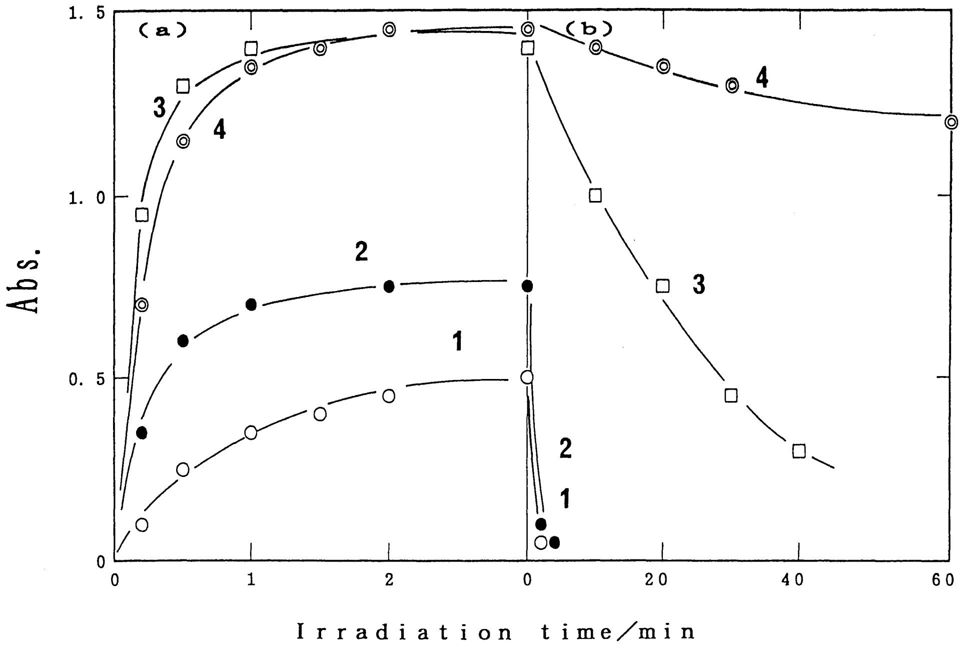


Fig. 4. Photo-responsiveness for coloration (a) and discoloration (b) of (4-oxazolyl)fulgides in dilute toluene solution (5×10^{-5} mol dm^{-3}) under UV and visible light irradiation.

Table 2. Quantum Yields for Discoloration of Fulgides in Dilute Toluene Solution

Fulgide	ϕ_{CE}[a]	
1	0.054	(477)[b]
2	0.0091	(511)
3	0.00019	(535)
4	0.000049	(535)
5	0.14	(535)
6	0.0092	(556)
7	0.0043	(591)

a) Visible light beams used were isolated by passing the light from xenon lamp through glass filters (Toshiba IRA-25S and KL-glass filters), and the photoreactions in a dilute toluene solution was monitored by HPLC or UV-vis spectrometer. The light intensities were determined with a power meter. b) Irradiation wavelength/nm.

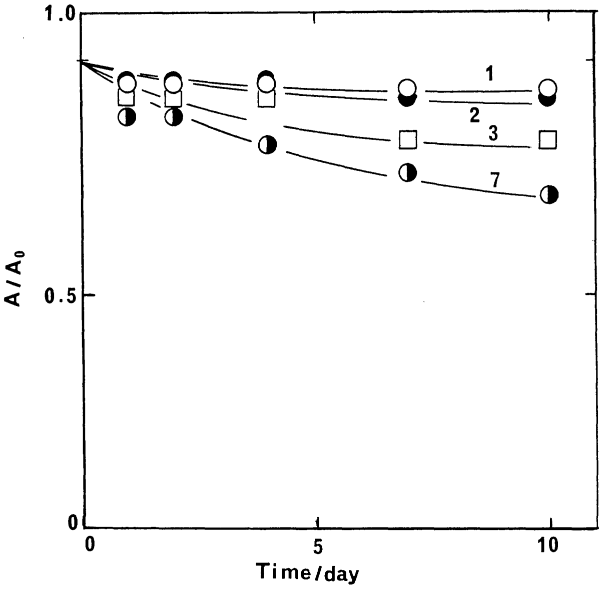


Fig. 5. Thermal degradation of the fulgides in PMMA film at 80 °C in the dark. PMMA films of 0.5 µm thickness containing fulgides (ca. 15 wt%) had been irradiated with UV light for coloration.

(triplet reaction) of the olefinic bond and the discoloration rate, we investigated the effect of triplet quenches, e.g. benzil, pyrene, anthracene, and benz[*b*]anthracene (Et value/kcal mol^{-1}; 54.3, 48.2, 42.7, and 29.3)[12] on the discoloration rate of fulgide (**3**) in a toluene solution. However, no quencher affected the rate. This means that no triplet reaction, e.g. *E*–*Z* isomerization of the styryl group, occurred.

From these results it is speculated that the substituents play a significant role in stabilizing the excited colored form, because of the higher delocalization of fulgide molecule, and effect the discoloration rate.

Thermal Stability of Colored Fulgides. In Fig. 5, thermal degradation of colored fulgides (**1**), (**2**), (**3**), and (**7**) in PMMA thin film (ca. 0.5 µm thickness) at 80 °C are illustrated. (4-Oxazolyl)fulgide (**1**), (**2**), and (**3**) exhibited good fatigue resistance. After 10 d, ca. 80—90% of the colored fulgide showed the same photochromic performance. (3-Thienyl) fulgide (**7**) showed little fatigue, and was slightly inferior to (4-oxazolyl) fulgides.

In conclusion, an attempt to red-shift the colored form of fulgide was made by substitution with electron-donating substituents; we clarified that the method is effective to red-shift and to increase the molar-absorption coefficient of the colored form. From a practical view point, e.g. the use of fulgide as a data-storage media, the effect seems to be of great advantage, owing to the greater sensitivity to read-out data. However, this attempt also caused a decrease in the discoloration rate.

Experimental

UV and ^{1}H NMR spectra were measured with a spectrophotometer (Hitachi U-3200) and 60- and 90-MHz spectrometers (JEOL JNM-PMX60 and EX-90 NMR). A mercury lamp (Ushio, 250 W) and a xenon lamp (Ushio, 300 W) were used as light sources for the coloration and discoloration reactions, respectively. A mercury line at 366 nm (ca. 50 mJ) and a visible light beam were isolated by passing the light through glass filters (UV light; Toshiba UV-D36C and UV-35, visible light; L-39, Y-40, O-50). The quantum yields for discoloration were determined by measuring the photoreaction rate, which was carried out in a toluene solution (5×10^{-5} mol dm^{-3}) by irradiation with visible light isolated through a glass filter (Toshiba KL-45, KL-48, KL-50, KL-53, KL-55, and KL-60). The light intensity was measured with a power meter AQ2710 (ANDO). The photochemical fatigue of fulgides was investigated by measurements of the absorption spectra of the colored forms. In the presence of air, toluene solutions of fulgide (1×10^{-4} mol dm^{-3}) were irradiated with UV and visible light, alternately, using a photo-irradiater cycle test system (OMROM). For measuring the thermal stability, a PMMA thin film (0.5 μm) was prepared on a glass plate by spin-coating a cyclohexanone solution containing a fulgide and PMMA (15 : 85 by weight); the surface was coated with acrylic film (SD-301 Dainihon-inki).

Materials. (4-Oxazolyl)fulgide (**1**) and (3-thienyl)fulgide (**5**) were prepared as described in the litarature.[4,5] Novel fulgides (**2**, **3**, **4**, **6**, and **7**) were also prepared by Stobbe condensation of 4-acetyl (or formyl)thiophene and 4-acetyloxazole derivative with diethyl 2-isopropylidenesuccinate using potassium *t*-butoxide[4] or lithium diisopropylamide[5,10] as a catalyst; this was followed by hydrolysis and dehydration reactions. The yields, physical constants and analytical data are summarized in Table 3.

(4-Oxazolyl)fulgide (2). The acetyl derivative, 4-acetyl-2-(*p*-dimethylaminophenyl)-5-methyloxazole was prepared in 48% yield from 3-hydroxyimino-2,4-pentanedione and *p*-dimethylaminobenzaldehyde, using the method described in the literature.[13] Mp 123-125 °C; ^{1}H NMR (CDCl$_3$) δ=2.55 (s, 3H), 2.63 (s, 3H), 3.00 (s, 6H), 6.63 (d, 2H), 7.80 (d, 2H).

A mixture of the acetyl derivative (9.3 g, 40 mmol) and diethyl 2-isopropylidenesuccinate (10 g, 48 mmol) was added dropwise to a refluxing solution of potassium *t*-butoxide (5.0 g) in *t*-butyl alcohol (60 ml); the solution was refluxed for 1 h. After removing the solvent, the residue was poured into ice-cooled water, and extracted with ether. The aqueous layer was acidified to liberate the half-ester which was extracted with ether and dried over anhydrous magnesium sulfate; the solvent was then removed. The ester was mixed with 10% ethanolic potassium hydroxide (100 ml) and boiled for 3 h. After cooling, the mixture was poured into water, acidified with 6 M (1 M=1 mol dm^{-3}) HCl, and extracted with ether. After drying, the solvent was removed, and the resulting diacid was boiled with acetic anhydride. After the solvent was removed, 0.74 g of fulgide (**2**) was purified by column-chromatography on silica gel and recrystalization from chloroform-hexane.

(4-Oxazolyl)fulgide (3). Starting material, 4-acetyl-2-(*p*-dimethylaminostyryl)-5-methyloxazole was prepared in 18% yield according to a method described in the literature[13] from 3-hydroxyimino-2,4-pentanedione and *p*-dimethylaminocinnamaldehyde. Mp 135—136 °C; ^{1}H NMR (CDCl$_3$) δ=2.52 (s, 3H), 2.62 (s, 3H), 2.97 (s, 6H), 6.50—7.46 (m, 6H).

To a THF solution of lithium diisopropylamide, which was freshly prepared from diisopropylamine (6 ml), 2,2′-bipyridil (50 mg) and 17.5 ml of an *n*-butyl lithium hexane solution in THF (200 ml), diethyl 2-isopropylidenesuccinate (4.5 g, 21 mmol) was added at −70 °C. The mixture was then stirred for 30 min, before the acetyl derivative (5.2 g, 19 mmol) was added. The solution was warmed to room temperature and further stirred for 1 h. The reaction mixture was poured into ice-cooled water, and acidified with 6 M (1 M=1 mol dm^{-3}) HCl. The solution was extracted with ether and dried over anhydrous magnesium sulfate before removing the solvent. An oily product was mixed with 10% ethanolic potassium hydroxide (100 ml) and boiled for 5 h. After cooling, the mixture was poured into water, acidified with 6 M HCl, and extracted with ether. After drying and removing the solvent, the diacid was

Table 3. Yields, Physical Constants and Analytical Data of Fulgide[a)]

Fulgide	Yield	Mp	^{1}H NMR, δ in CDCl$_3$		Found (Calcd)/%			m/z
	%	θ_m/°C	-CH$_3$ (3H, s)	-CH= and others	C	H	N	M$^+$
2Z	6	202—204	1.97, 2.32(6H), 2.44, 3.00(6H)	6.70(2H, d), 7.86(2H, d)	68.85 (68.83)	6.02 (6.05)	7.39 (7.65)	366
3Z	5	195—197	2.00, 2.28, 2.30, 2.43, 2.98(6H)	6.45—7.60(6H, m)	69.34 (70.40)	6.15 (6.17)	7.16 (7.14)	392
4Z	6	216—219	1.97, 2.25, 2.28, 2.45, 2.98(6H)	6.39—7.40(8H, m)	71.10 (71.74)	6.64 (6.26)	6.67 (6.69)	418
6Z	3	222—224	2.02, 2.14, 2.36, 2.44	6.85—7.30(3H, m) 7.52—7.64(5H, m)	71.31 (72.50)	5.46 (5.53)		364
7E	15	168—172	1.70, 2.40(6H)	1.15(6H, t), 3.32(4H, q) 6.62—7.47(8H, m)	70.68 (71.23)	6.37 (6.46)	3.00 (3.32)	421

a) Analytical data of fulgide (**1**) had been shown in a previous paper[8)] and the data of fulgide (**5**) was consistent with that reported in the literature.[4)] boiled with acetic anhydride. After the solvent was removed, 0.53 g (1.3 mmol) of fulgide (**3**) was purified by column chromatography on silica gel and recrystalization from chloroform–hexane. The Stobbe reaction, in which potassium *t*-butoxide or sodium hydride acts as a catalyst failed.

(4-Oxazolyl)fulgide (4). Starting material, 4-acetyl-2-[4-(*p*-dimethylaminophenyl)butadien-1-yl]-5-methyloxazole was prepared in 9% yield from 5-(*p*-dimethylaminophenyl)penta-2,4-dienal[14] and 3-hydroxyimino-2,4-pentanedione according to a method described in the literature[13]. Mp 153—156 °C; ^{1}H NMR (CDCl$_3$) δ=2.53 (s, 3H), 2.63 (s, 3H), 2.99 (s, 6H), 6.62—7.40 (m, 8H).

Fulgide (**4**) (0.11 g, 2.6 mmol) was obtained similarly to fulgide (**3**) from the (4-acetyl)oxazole derivative (1.3 g, 4.4 mmol) and the succinate (1.0 g, 4.6 mmol).

(3-Thienyl)fulgides (6) and (7). Fulgide (**6**) and (**7**) were prepared from the 3-acetyl and 3-formylthiophene derivative by a Stobbe condensation with potassium *t*-butoxide and lithium diisopropylamide used as a catalyst, respectively. The starting materials were obtained as described below.

3-Acetyl-2-methy-5-styrylthiophene. 2-Methyl-5-styrylthiophene was prepared in 53% yield by the Wittig reaction in the usual manner[15] from 5-methylthiophene-2-carbaldehyde and diethyl benzylphosphonate. A Friedel-Crafts reaction with acetic anhydride in the presence of anhydrous SnCl$_4$ as a catalyst[4] gave 3-acetyl-2-methyl-5-styrylthiophene in 5% yield. Mp 59—62 °C; ^{1}H NMR (CDCl$_3$) δ=2.42 (s, 3H), 2.65 (s, 3H), 6.80—7.37 (m, 8H).

5-(*p*-Diethylaminostyryl)-2-methylthiophene-3-carbaldehyde. 4-Bromo-5-methylthiophene-2-carbaldehyde; 5-methylthiophen-2-carbaldehyde was brominated with bromine according to a method described in the literature[16] to give 4-bromo-5-methylthiophene-2-carbaldehyde in 42% yield. Bp 105—110 °C/10 mmHg (1 mmHg=133.322 Pa) (CDCl$_3$) δ= 2.42 (s, 3H), 7.55 (s, 1H), 9.75 (s, 1H).

3-Bromo-5-chloromethyl-2-methylthiophene; 4-Bromo-5-methylthiophene-2-carbaldehyde was converted to the 5-hydroxymethylthiophene derivative with NaBH$_4$, and chlorinated with SOCl$_2$ according to the usual method[17] in 80 and 78% yields, respectively. Bp 89 °C/2 mmHg; ^{1}H NMR (CDCl$_3$) δ=2.33 (s, 3H), 4.63 (s, 2H), 6.85 (s, 2H).

5-(*p*-Diethylaminostyryl)-3-bromo-2-methylthiophene; The 5-chloromethylthiophene derivative was converted to thenylphosphonate in the usual manner.[15] To an ice-cooled solution of sodium ethoxide 2.3 g (35 mmol) and DMF 15 ml, a mixture of the thenylphosphonate (7.4 g, 23 mmol), *p*-diethylaminobanzaldehyde 4.1 g (23 mmol) and 10 ml of DMF was added. After stirring for 3 h at room temperature, the reaction mixture was poured onto crushed ice, and extracted with chloroform. After washing with water and dried, the solvent was removed. Recrystalization from 2-propanol gave 5-(*p*-diethylaminostyryl)-3-bromo-2-methylthiophene in 74% yield. Mp 115—116 °C; ^{1}H NMR (CDCl$_3$) δ=1.13 (t, 6H), 2.31 (s, 3H), 3.31 (q, 4H), 6.55—7.42 (m, 7H).

Butyllithium–hexane solution 9.3 ml (14 mmol) was added to a solution of the bromide 3.8 g (11 mmol) and THF 50 ml at −70 °C with stirring. Then 3 ml of DMF was added, and the mixture was stirred for 20 min. The reaction mixture was poured onto crushed ice, extracted with chloroform, and dried. After removing the solvent, the resulting solids were recrystalized from ethanol–chloroform, and giving 5-(*p*-diethylaminostyryl)-2-methylthiophene-3-carbaldehyde (1.6 g, 5.4 mmol) in 49% yield. Mp 131—132 °C; ^{1}H NMR (CDCl$_3$) δ=1.15 (t, 6H), 2.68 (s, 3H), 3.35 (q, 4H), 6.53—7.90 (m, 7H), 9.96 (s, 1H).

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