New π-Donor Precursor Molecule for the Preparation of Organic Metals, 4,5;4′,5′-Bis(1,4-dioxanediyl-2,3-dithio)-tetrathiafulvalene: Synthesis and Properties

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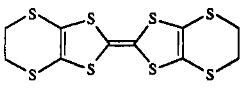
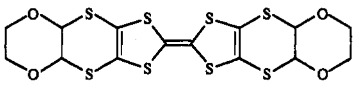
The synthesis of quasi two-dimensional systems based on radical cation salts of bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF or ET) permitted great progress to be made in the design of organic superconductors and in the raising of their critical temperatures.[1,2] The ET molecule contains eight sulfur atoms and noncoplanar CH_2–CH_2 groups.



BEDT-TTF (or ET)



BDDT-TTF

The sulfur atoms participate in multiple side-by-side S···S interactions in the cation radical layers, and the nonplanar region enhances the steric hindrance of intrastack S···S interactions, leading to the equalization of interactions inside the conductive layers. It should be noted that, for a long time, overall planarity of the donors and/or acceptors was considered to be one of the most important structural requirements for obtaining conducting ion radical salts.[3] However, it was predicted,[4] and later shown,[5] that one-dimensional interactions, resulting from the stacking of such planar molecules, lead to the dielectrization of the electronic system at low temperatures.

The synthesis of new π-donors from ET by chemical modification is an intensively studied approach to the design of organic metals and superconductors. Such chemical modifications of ET have primarily entailed the following: (i) variations on the terminal ethylene bridges, (ii) substitution of the S atoms by other heteroatoms (Se, O), and (iii) substitution of the central C=C bond by metal ions. This has led to numerous new organic metals and superconductors.[1,2,6-12]

Among the possible variations of the terminal ethylene bridges of ET is the extension of the molecule by the addition of heterocycles. In particular, the substitution of both terminal ethylene bridges of ET by 1,4-dioxane rings will lead to 4,5;4′,5′-bis(1,4-dioxanediyl-2,3-dithio)-tetrathiafulvalene, or BDDT-TTF. In the cation radical salts that derive from BDDT-TTF, additional side-by-side interactions occur due to the increased number of heteroatoms. Moreover, the noncoplanarity of the 1,4-dioxane rings with respect to the central ET molecular framework should hinder the formation of a dense face-to-face stacked quasi one-dimensional packing. Finally, and in contrast to the addition of benzene rings to the ET skeleton,[13] the addition of 1,4-dioxane rings should alter the electronic structure and solubility little. The latter property is of special significance to the synthesis of cation radical salts because the low solubility of a parent often causes difficulties in growing crystals and studying the physical properties of its salts.

In the present paper, we report the synthesis and characterization of BDDT-TTF, its cyclic voltammetry data, and its potential use in the formation of conducting cation radical salts.

Results and Discussion

BDDT-TTF (5) was prepared as shown in Scheme 1.

As a first step, the 4,5-(1,4-dioxanediyl-2,3-dithio)-1,3-dithiole-2-thione cycloadduct (3), was synthesized by [2 + 4] cycloaddition of compound 2 (4π) to 1,4-dioxene (2π) (1). The use of compound 2, which contains a dithiadiene moiety and can be described as an oligomer of 1,3-dithiole-2,4,5-trithione, was reported for the first time by Neiland *et al.*[14] to react with alkyl-substituted and electron-withdrawing alkenes, thus giving substituted 4,5-(ethylenedithio)-1,3-dithiole-2-thiones. This oligomer probably depolymerizes on heating to give the unstable 1,3-dithiole-2,4,5-trithione monomeric species, which forms Diels–Alder adducts with dienophiles.[14,15] This unstable 1,4-dioxane-2,3-dithione, possessing a dithiabutadiene fragment, is also known to undergo cycloaddition as a 4π-component to alkenes.[16]

Single crystals of adduct 3 were obtained, and the structure was determined by X-ray diffraction methods (monoclinic, space group $P2_1$; $a = 9.631(2)$ Å, $b = 5.491(1)$ Å, $c = 9.895(1)$ Å; $\beta = 92.04°$; Figure 1).[17] Only one stereoisomer was obtained that in which the methine hydrogen atoms are in the *cis*-position.

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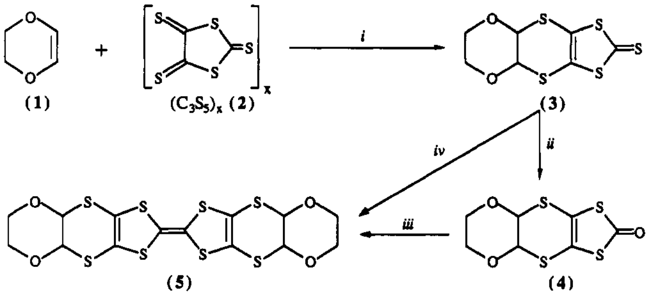
Scheme I[a]



[a] (*i*) 90 °C; (*ii*) $Hg(OAc)_2$ in $AcOH/CHCl_3$, 25 °C; (*iii*) and (*iv*) $P(OMe)_3$ in C_6H_6, 100 °C.

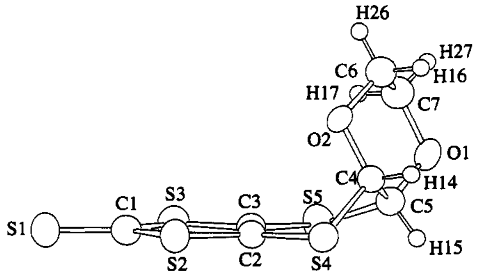


Figure 1. Structure of 4,5-(1,4-dioxanediyl-2,3-dithio)-1,3-dithiole-2-thione (**3**).

Oxidation of **3** with mercuric acetate in acetic acid affords the oxo derivative **4**, *i.e.* 4,5-(1,4-dioxanediyl-2,3-dithio)-1,3-dithiol-2-one. In the event of incomplete substitution, the reaction mixture may still contain some starting thione **3**, and the crystals obtained therefrom melt at a temperature higher by 1° than do those of pure ketone **4**. Such a variation in the melting temperature is characteristic of solid solutions of isostructural compounds.[18] In fact, the cell parameters of mixed crystals (monoclinic, space group $P2_1$; $a \approx 9.4$ Å, $b \approx 5.4$ Å, $c \approx 9.8$ Å; $\beta = 92°$) are similar to those of pure thione **3**. The thione concentration in a ketone–thione mixture can be determined from the extinction coefficient of the band at 390 nm in the UV spectra of the ketone **4**, thione **3**, and their solid solutions (supplementary material).

BDDT–TTF (**5**) is obtained by the self-coupling of thione **3**, or cross-coupling of **3** with ketone **4**, in the presence of $P(OEt)_3$ or $P(OMe)_3$. Various experimental conditions for these coupling reactions are summarized in Table 1. Higher yields are observed in the reactions with the ketone (or its equimolar mixture with the thione) and trimethyl phosphite. Lower yields are obtained when the pure thione is used. The yields are also dependent on the coupling reagent itself (trimethyl phosphite seems to be slightly more efficient than triethyl phosphite), on the coupling agent concentration, and on the temperature. The highest yield (42%) was obtained for a ≈12-fold molar excess of $P(OMe)_3$ and a temperature of 100 °C.

BDDT-TTF (**5**) and its precursor thione **3** and ketone **4** were characterized by elemental analysis and ^1H NMR and UV–vis–IR, and mass spectrometry. The molecular ion was observed for each compound. A fragment with $m/z = 86$ is the base peak. This indicates that the fragmentation of the molecular ion in these compounds involves the $C_4H_6O_2^+$ ion elimination ($M = 86$). The presence in the mass spectrum of **5** of peaks with $m/z = 414$ and 328 with an intensity ratio of 2:1, probably indicates the loss of two dioxene molecules from **5**.

The assignment of the frequencies of the vibration bands in the infrared spectra (supplementary material) was based on that proposed by Kozlov *et al.*[19,20] for ET and its corresponding precursor thione and ketone. In the spectrum of **3**, four intense bands are observed at 1092, 1060, 1047, and 1038 cm^{-1}. The latter three can probably be attributed to the stretching vibrations of the C=S bond and are not observed in the spectra of **4** and **5**. Three intense bands at 1671, 1637, and 1614 cm^{-1} are observed in the spectrum of **4** and are attributed to the stretching C=O vibrations. Some bands are slightly shifted compared to those of **3**; the largest shift is observed for the CH and C=C stretching vibrations at 2976 *vs* 2963 cm^{-1} and 1509 *vs* 1486 cm^{-1}, respectively. Large shifts of vibration frequencies and some rearrangement of the band intensities are observed when the spectrum of BDDT–TTF is compared to the spectra of its precursors; for example, the intensity of the C=C band of BDDT–TTF (1509 cm^{-1}) is considerably less than that of the same band for **3** (1486 cm^{-1}) and **4** (1509 cm^{-1}). The intense band at 1098 cm^{-1} for **5**, 1096 cm^{-1} for **4**, and 1092 cm^{-1} for **3** is likely due to stretching C–O vibrations.

The ^1H NMR spectra of thione **3** and BDDT–TTF are shown in Figure 2. A singlet due to the methine protons ($\delta = 5.86$ ppm) and two symmetric multiplets ($\delta = 4.17$ and 3.88 ppm) due to the methylene protons (Figure 2a) are observed in the spectrum of a solution of **3** in THF-d_8. The ratio of their integral intensities is 1:1:1. The multiplets are due to overlapping AB systems, in agreement with the nonsymmetrical environment of the methyl protons (see Figure 1). In the spectrum of a solution of **5** in CCl_4 (Figure 2b), the methine protons are seen as two closely located lines ($\delta = 5.27$ and 5.25 ppm). This is probably attributable to the presence of two stereoisomers of **5**. The ratio of the line intensities indicates that the stereoisomer with the methine proton signal at $\delta = 5.27$ ppm is the major product. A distortion of symmetry of the multiplets corresponding to the methylene protons is also observed, as expected for four nonequivalent AB systems in two stereoisomers. However, the ratio of integral intensities of the signals corresponding to the methine and methylene protons, respectively, is still 1:2, as was observed in **3**. It should be noted that a weak doublet, which we could not assign, is observed between the two multiplets of the methylene protons.

The redox potentials of BDDT–TTF were measured by cyclic voltammetry in CH_3CN/n-Bu_4NPF_6 (0.1 M) on a Pt electrode with a potential scan rate of 0.1 V/s. Two reversible redox waves are observed at $E_{1/2} = 0.58$ and 0.82 V *vs* SCE, and correspond to the BDDT–TTF/BDDT–TTF$^+$ and BDDT–TTF$^+$/BDDT–TTF^{++} couples, respectively. After electrolysis of a solution of BDDT–TTF at constant potential (0.7 V), the ESR frozen-glass spectrum of the postelectrolysis solution shows a signal at $g = 2.008$, corresponding to the BDDT–TTF$^+$ cation radical species. The redox potentials of BDDT–TTF are ≈40–50 mV higher than those of ET measured under the same conditions, but the ΔE potential separations between the oxidation waves are identical (0.24 V).

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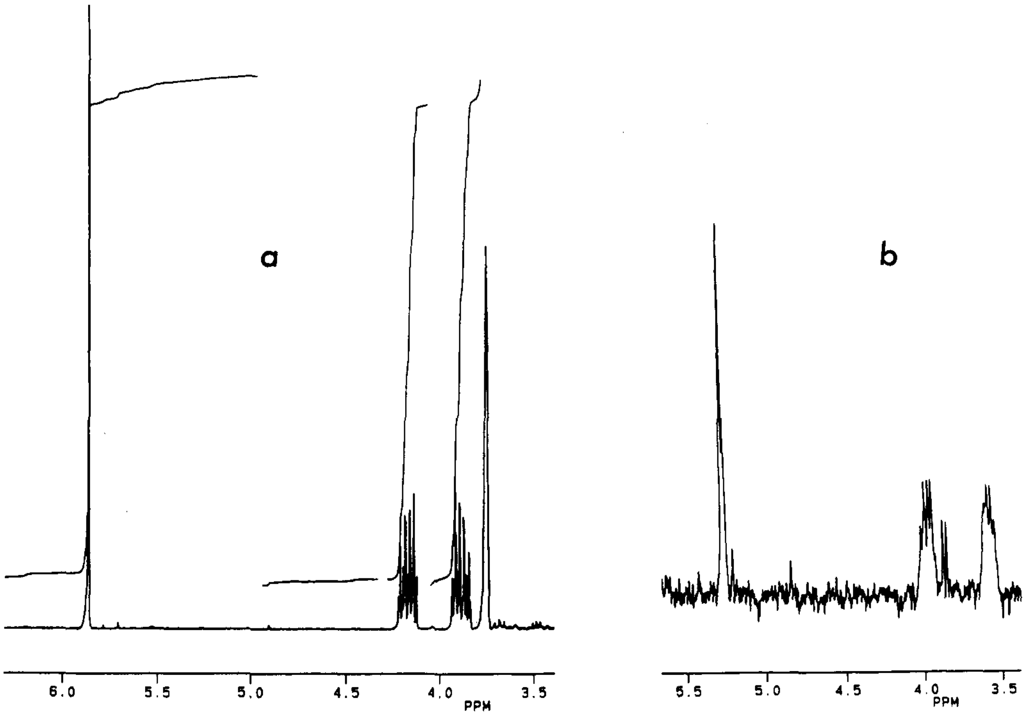

Figure 2. ^{1}H NMR spectra: (a) thione (3) in THF-d_8; (b) BDDT–TTF (5) in CCl_4; at 250 MHz.

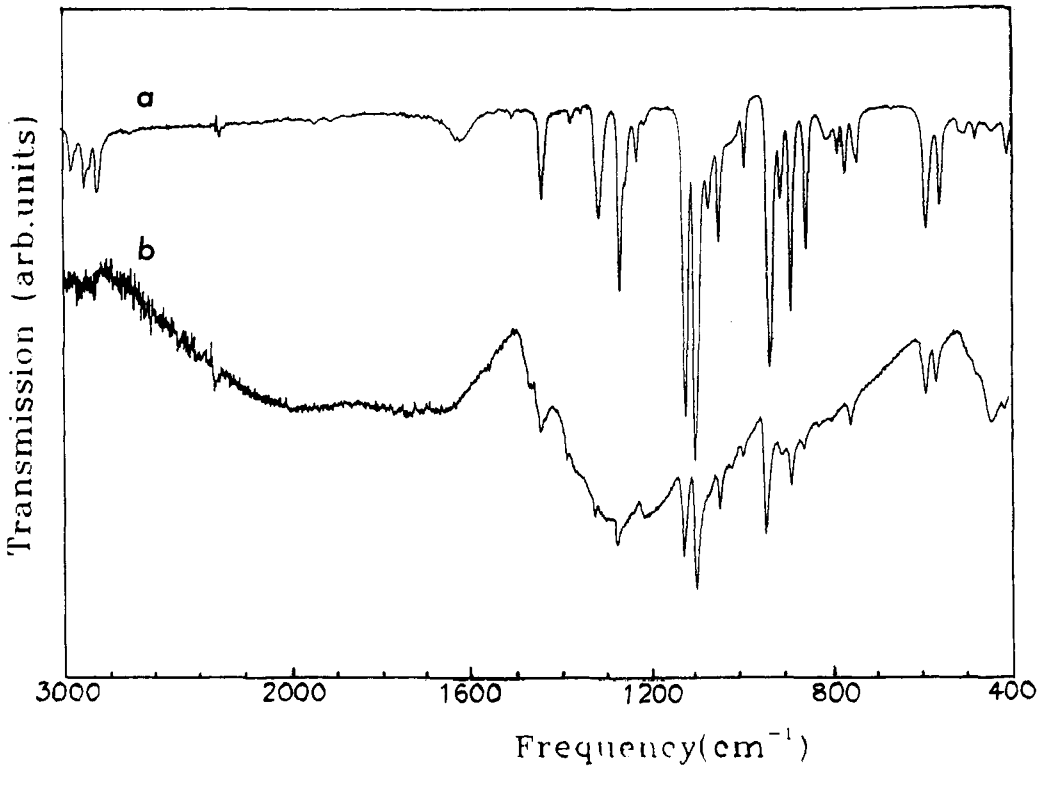

Figure 3. IR spectra in KBr: (a) BDDT–TTF (5); (b) BDDT–TTF cation radical salt with I_3^-.

BDDT–TTF does form crystalline salts upon oxidation of a benzonitrile solution by DDQ or upon electrochemical oxidation in the presence of inorganic anions (ClO_4^-, I_3^-). The IR spectra of BDDT–TTF and its radical cation salt with I_3^- are compared in Figure 3. The vibrational bands in the spectrum of the salt are noticeably widened in contrast to those of BDDT–TTF and are superimposed on a wide band in the region of 1200 cm^{-1}. Such an IR spectrum is characteristic of conducting low-dimensional organic compounds. The room-temperature compaction powder conductivity of the iodide salt, measured on a compressed pellet by a four-probe method, is equal to 0.3 Ω^{-1} cm^{-1}.

In conclusion, we have synthesized a new donor molecule BDDT–TTF (5), which is identical to ET in its central region but contains additional oxygen heteroatoms as a result of the substitution of the terminal ethylene bridges of ET by dioxane rings. The addition of dioxane rings to the ET skeleton does not cause significant alterations in the electronic structure, as shown by the similar redox

Table 1. Experimental Conditions for the Coupling Reactions Affording BDDT–TTF (5)

starting material[a] (g)	solvent[b] (mL)	coupling agent (mL)	T^c (°C)	reaction duration (h)	reaction yield (%)
3 (0.4)		$(EtO)_3P$ (15)	120	2	0
3 (0.1)	B (10)	$(EtO)_3P$ (2)	100	4.5	trace
3 (0.25)	D (3)	$(EtO)_3P$ (2)	110–120	3.5	0
4 (0.24)	D (3)	$(EtO)_3P$ (2)	110–120	2	11
3 (0.13) + **4** (0.1)	D (5)	$(EtO)_3P$ (2)	100–110	1	31
3 (0.25)		$(MeO)_3P$ (1)	110–120	0.75	14
3 (0.25)		$(MeO)_3P$ (2)	110–120	2	7
3 (0.2)	B (3)	$(MeO)_3P$ (1)	100	2	20
3 (0.2)	B (6)	$(MeO)_3P$ (1)	100	6	20
3 (0.2)	D (3)	$(MeO)_3P$ (1)	110–120	2	13
4 (0.2)	B (3)	$(MeO)_3P$ (1)	100	2	32
3 (0.1) + **4** (0.1)	B (6)	$(MeO)_3P$ (1)	100	3.5	42

[a] **3** = 4,5-(1,4-dioxanediyl-2,3-dithio)-1,3-dithiole-2-thione; **4** = 4,5-(1,4-dioxandiyl-2,3-dithio)-1,3-dithiol-2-one. [b] B = benzene, D = dioxane. *T* = temperature.

behavior of both donors. Preliminary studies indicate that BDDT–TTF can be used as a donor for the preparation of new conductive radical cation salts with inorganic anions such as I_3^-. Further work is underway to grow crystals of these salts of suitable size and quality with the hope of obtaining new organic metals and superconductors.

Experimental Section

General. All reactions were carried out under N_2. Melting points were determined using a Electrothermal melting point apparatus in sealed capillary tubes. The ^1H NMR spectra were recorded on a Bruker AM 250 spectrometer. The electronic impact mass spectra were obtained using a Nermag R 10-10H quadrupole mass spectrometer. IR (in KBr) and UV/vis spectra (in solution) were recorded using a Perkin-Elmer 283G and a Hewlett-Packard 8452A spectrometer, respectively. ESR spectra were recorded with a Bruker ER-200 TT spectrometer. The equipment used for cyclic voltammetry was identical to that described elsewhere.[21]

Reagents. 1,4-Dioxene (**1**) was obtained from Fluka Chemicals. The $(C_3S_5)_x$ oligomer (**2**) was synthesized by the methods described in ref 14. Triethyl phosphite and trimethyl phosphite (Aldrich Chemicals) were freshly distilled under N_2 prior to use. All solvents were dried by standard techniques, distilled, and degassed prior to use. All other reagents were used without purification.

4,5-(1,4-Dioxanediyl-2,3-dithio)-1,3-dithiole-2-thione (3). Dioxene (**1**) (2 g, 2.32 mmol) was added to a stirred suspension of oligomeric 1,3-dithiole-2,4,5-trithione (**2**) (7g, 3.87 mmol) in dioxane (150 mL). The mixture was heated up to 90 °C and stirred for 10 h at this temperature. The resulting mixture was filtered hot, and the remaining solid was further washed with hot ethanol (3 × 100 mL) for complete extraction of the product. The combined filtrates were treated with activated charcoal. The solvent was evaporated under vacuum using a rotary evaporator. The resulting solid was recrystallized from ethanol to give **3** as yellow needle-like crystals (2 g) in 25% yield, mp 188–189 °C dec R_f (silica gel, CH_2Cl_2) = 0.73. Anal. Calcd for $C_7H_6O_2S_5$: C, 29.77; H, 2.14. Found: C, 29.79; H, 2.27. MS: *m/z* (relative intensity) 282 (0.29, $M^{\bullet +}$), 86 (1.00, $C_4H_6O_2^+$), 76 (0.27, CS_2^+), 58 (0.23, $C_2H_2O_2^+$). ^1H NMR (250 MHz, THF-d_8): 5.86 (s, 2H), 4.17 (m, 2H), 3.88 (m, 2H). UV ($CHCl_3$): λ_{max} 266 nm (ϵ 7000), 308 (ϵ 2000), 336 (ϵ 1300), 388 (ϵ 5200).

4,5-(1,4-Dioxanediyl-2,3-thio)-1,3-dithiol-2-one (4). Method A. Thione **3** (0.518 g, 1.83 mmol) was dissolved in $CHCl_3$ (100 mL). Glacial acetic acid (25 mL), and $Hg(OAc)_2$ (1.17 g, 3.66 mmol) were added to this solution. The resulting solution was stirred at room temperature for 2 h. The precipitate was filtered off, and the filtrate washed with water (3 × 150 mL), aqueous $NaHCO_3$, and finally with water again. After drying (Na_2SO_4), the solvent was evaporated and the resulting precipitate was recrystallized from ethanol to give (**4**) as light-yellow needles (0.328 g; 67% yield), mp 150 °C dec.

Method B. Thione **3** (0.4 g, 1.42 mmol) was dissolved in CH_2Cl_2 (50 mL). Glacial acetic acid (25 mL) and $Hg(OAc)_2$ (0.722 g, 2.27 mmol) were added to this solution. The mixture was stirred at room temperature for 2 h. The solvent and acetic acid were removed under vacuum. The residue was suspended in CH_2Cl_2 (150 mL) and filtered off. The filtrate was kept over Na_2CO_2 for 12 h. After Na_2CO_3 was filtered off, the solvent was evaporated. The resulting precipitate was recrystallized from ethanol (0.25 g, 66% yield), mp 150–151 °C dec, R_f (silica gel, CH_2Cl_2) = 0.80. Anal. Calcd for $C_7H_6O_3S_4$: C, 31.56; H, 2.27. Found: C, 31.55; H, 2.27. MS: *m/z* (relative intensity) 266 (0.56, $M^{\bullet +}$), 86 (1.00, $C_4H_6O_2^+$), 76 (0.1, CS_2^+), 58 (0.1, $C_2H_2O_2^+$). UV ($CHCl_3$): λ_{max} 290 (ϵ 4000).

4,5;4′,5′-Bis-(1,4-Dioxanediyl-2,3-dithio)tetrathiafulvalene (BDDT–TTF) (5). $P(OMe)_3$ (1 mL, 8.5 mmol) was added to a stirred suspension of thione **3** (0.2 g, 0.7 mmol) in benzene (3 mL) under a dry nitrogen atmosphere. The resulting mixture was heated over an oil bath up to 100 °C and kept at this temperature for 2 h. The resulting mixture was allowed to cool to room temperature, and ethanol (5 mL) was added. The mixture was further cooled in a refrigerator. A bright orange solid precipitated and was filtered off, washed with ethanol, acetone, and ether, and dried under vacuum (0.041 g, 20% yield), R_f (silica gel, CH_2Cl_2) = 0.64. Anal. Calcd $C_{14}H_{12}O_4S_8$: C, 33.58; H, 2.42. Found: C, 33.46; H, 2.59. MS: *m/z* (relative intensity) 500 (0.2, $M^{\bullet +}$), 414 (0.41, $(M - C_4H_6O_2)^+$), 328 (0.2, $(M - 2C_4H_6O_2)^+$), 128 (0.61, S_4^+), 86 (1.00, $C_4H_6O_2^+$), 76 (0.2, CS_2^+), 58 (0.21, $C_2H_2O_2^+$). ^1H NMR: 250 MHz, CCl_4; 5.27, 5.25 (s, s, 4H), 3.98 (m, 4H), 3.58 (m, 4H). UV ($CHCl_3$): λ_{max} 316 (ϵ 13 000), 346 (ϵ 15 000).

BDDT–TTF Cation Radical Salt with I_3^-. Galvanostatic (1 μA) oxidation, using the apparatus and techniques described in ref 12, of a solution of BDDT–TTF (0.001 M) in 1,1,2-trichloroethane containing a large excess of n-Bu_4NI_3 yielded small crystals growing on the anode. The crystals were collected and washed with ethanol and ether. The very small size and the amount of crystals obtained did not allow characterization by elemental analysis of X-ray diffraction methods.

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Supplementary Material Available: Table 1, listing of the frequencies of the vibration bands in the infrared spectra of compounds **3–5** and Figure j, UV–vis spectra of compounds **3**, **4**, and mixed crystals (4 pages). This material is contained in libraries on microfiche, immediately follows this article in the microfilm version of the journal, and can be ordered from the ACS; see any current masthead page for ordering information.

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