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Bis(methylethylenedithio)tetrathiafulvalene

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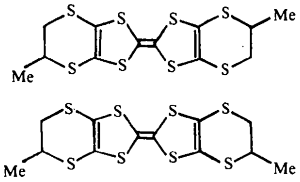
Abstract

The crystal of the title compound, $C_{12}H_{12}S_8$, is a mixture of the 7,7′-dimethylbis(ethylenedithio)tetrathiafulvalene and 7,8′-dimethylbis(ethylenedithio)tetrathiafulvalene isomers, in an almost 60:40 ratio. The crystal is disordered with respect to the position of one of the methyl substituents. The S atoms are involved in three-dimensional intermolecular S···S contacts with distances within 3.703 Å.

Comment

The organic π-donor bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF) has been the subject of numerous studies owing to the superconducting properties of several of its cation radical salts (Williams *et al.*, 1991; Bryce, 1991). The presence of two-dimensional S···S networks and C—H···(anion) interactions in the cation radical salts is important in determining the solid-state properties. Consequently, much synthetic effort has been devoted to systematically varying the structure of BEDT-TTF with consequential 'fine tuning' of the ionization potential, symmetry, size and S···S intermolecular contacts (Kumar, Singh, Das, Sinha & Mishnev, 1991; Moore & Bryce, 1992). The crystal of neutral BEDT-TTF is composed of pairs of BEDT-TTF molecules having a side-by-side arrangement, similar to that found in superconducting ion radical salts (Kobayashi, Kobayashi, Sasaki, Saito & Inokuchi, 1986).

The title compound, BMeEDT-TTF, a π-donor closely related to BEDT-TTF, has been reported previously (Papavassiliou, Zambounis & Yiannopoulos, 1987); however, no systematic investigations have been carried out. Recently, a modified synthesis and detailed characterization of the title compound have been reported (Singh & Singh, 1992) and ^{1}H NMR studies indicate it to be an isomeric mixture. In fact, in the 300 MHz spectrum of the compound, the proton resonances are split in a 60:40 ratio due to the presence of these isomers. The first ($^1E_{1/2}$) and second ($^2E_{1/2}$) ionization potentials of the present compound are slightly lower than those of BEDT-TTF. In this paper, we report the crystal structure and compare it with those of related compounds.

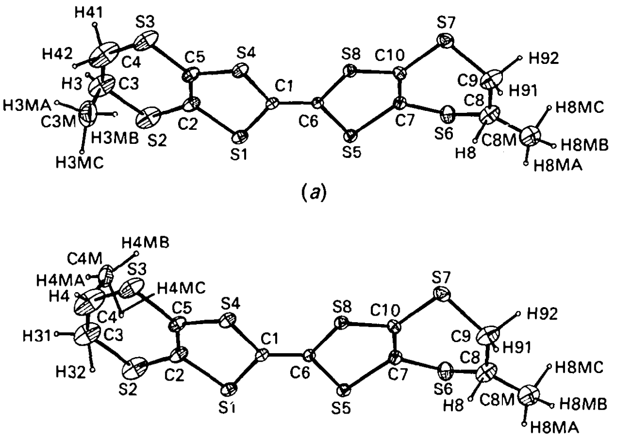


BMeEDT-TTF

The reaction of 4,5-dimercapto-1,3-dithiole-2-thione with (±)-1,2-dibromopropane at 198 K afforded 4,5-(methylethylenedithio)-1,3-dithiole-2-thione. The thione was coupled in the presence of triethylphosphite to give the title compound. The compound was purified by recrystallization from dichloromethane to give orange crystals.

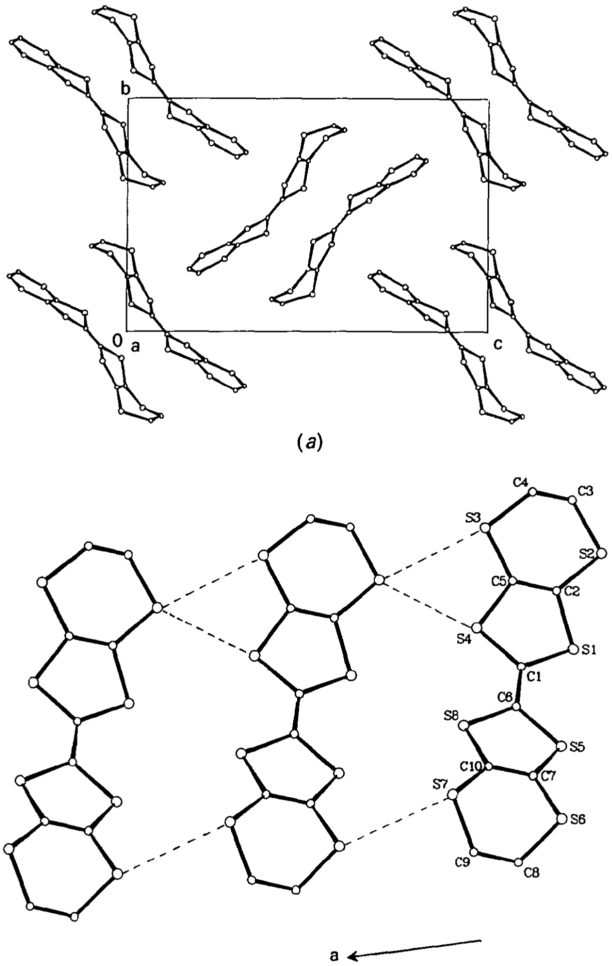
The molecule is disordered near the C(3)—C(4) region, as expected. The C(3)—C(3*M*) and C(4)—C(4*M*) bond lengths [1.720 (20) and 1.643 (22) Å, respectively] are longer than the normal C—C bond length. The disorder of the methyl groups located at the C(3) and C(4) atoms makes the C(3)—C(3*M*) and C(4)—C(4*M*) bond lengths, the C(3)···C(4*M*) and C(4)···C(3*M*) distances, and also distances involving some of the H atoms, deviate significantly from the corresponding ideal values. Also, the C(3), C(3*M*), C(4) and C(8*M*) atoms have higher temperature factors. Contrary to expectation, the molecule shown in Fig. 1(*b*) does not have any centre of symmetry as the ring formed by the atoms C(2), S(2), C(3), C(4), S(3) and C(5) is planar within 0.101 (11) Å, whereas the equivalent ring formed by the other ethylenedithio moiety has a half-chair conformation with a dihedral angle of 45.8 (4)° between the planes formed by C(7), C(10), S(7), S(6) and S(6), S(7), C(9), C(8). The S atoms are involved in intermolecular S···S contacts: S(3)···S(2$^{\text{i}}$) 3.533 (3), S(4)···S(2$^{\text{i}}$) 3.434 (3), S(7)···S(6$^{\text{i}}$) 3.703 (3), S(1)···S(5$^{\text{ii}}$) 3.572 (2),

$S(5)\cdots S(8^{iii})$ 3.585 (2) Å [symmetry codes: (i) $x + 1, y, z$; (ii) $-x, -y, -z$; (iii) $-x + 1, -y, -z$]. Details of the crystal packing of BEDT-TTF and related molecules have been reported (Kobayashi *et al.*, 1986; Porter, Kini & Williams, 1987; Kato, Kobayashi, Kobayashi & Sasaki, 1985; Das, Sinha, Kumar, Singh & Mishnev, 1992; Benming *et al.*, 1991). In the crystal, the title compound packs to form pairs of molecules and adjacent molecules are stacked along the *a* axis, as found in similar molecules.



(*b*)

Fig. 1. The two isomers of the title compound having (*a*) methyl groups at C(8) and C(3) and (*b*) methyl groups at C(8) and C(4).



(*b*)

Fig. 2. (*a*) The unit-cell packing projected onto the *bc* plane and (*b*) the molecular stacking along the *a* axis.

Experimental

Crystal data

$C_{12}H_{12}S_8$
$M_r = 412.71$
Monoclinic
$P2_1/n$
$a = 6.5687$ (7) Å
$b = 12.802$ (2) Å
$c = 20.219$ (2) Å
$\beta = 97.26$ (1)°
$V = 1686.65$ Å^3
$Z = 4$
$D_x = 1.625$ Mg m^{-3}

Cu $K\alpha$ radiation
$\lambda = 1.5418$ Å
Cell parameters from 25 reflections
$\theta = 15.47$–$36.20°$
$\mu = 9.269$ mm^{-1}
$T = 293$ K
Needle
0.3 × 0.2 × 0.2 mm
Orange

Data collection

Enraf–Nonius CAD-4 diffractometer
ω–2θ scans
Absorption correction: empirical (ψ scan)
$T_{\min} = 0.658$, $T_{\max} = 1.000$
3926 measured reflections
3782 independent reflections
3291 observed reflections [$F \geq 3.0\sigma(F)$]

$R_{\rm int} = 0.024$
$\theta_{\max} = 72.0°$
$h = 0 \rightarrow 8$
$k = 0 \rightarrow 15$
$l = -24 \rightarrow 24$
3 standard reflections
frequency: 60 min
intensity variation: none

Refinement

Refinement on F
$R = 0.0738$
$S = 1.5576$
2914 reflections
191 parameters
H-atom parameters not refined

Unit weights applied
$(\Delta/\sigma)_{\max} = 0.890$
$\Delta\rho_{\max} = 0.882$ e Å^{-3}
$\Delta\rho_{\min} = 0.784$ e Å^{-3}
Atomic scattering factors from *SHELX*76 (Sheldrick, 1976)

Table 1. *Fractional atomic coordinates and equivalent isotropic displacement parameters* (Å^2)

$$U_{\rm eq} = (1/3)\Sigma_i\Sigma_j U_{ij}a_i^*a_j^*\mathbf{a}_i.\mathbf{a}_j.$$

	x	y	z	$U_{\rm eq}$
C(1)	0.4142 (6)	−0.0128 (4)	0.1129 (2)	0.049 (3)
C(2)	0.2581 (8)	−0.1314 (6)	0.1949 (3)	0.074 (4)
C(3)	0.1910 (12)	−0.2521 (9)	0.3024 (5)	0.145 (5)
C(4)	0.3695 (13)	−0.2348 (11)	0.3266 (5)	0.187 (5)
C(5)	0.4556 (8)	−0.1260 (6)	0.2190 (3)	0.071 (4)
C(6)	0.4359 (6)	0.0664 (4)	0.0709 (2)	0.044 (3)
C(7)	0.3600 (9)	0.2327 (4)	0.0016 (3)	0.058 (4)
C(8)	0.4234 (13)	0.3627 (8)	−0.0979 (4)	0.113 (4)

C(9)	0.6244 (13)	0.3819 (8)	−0.0693 (4)	0.122 (5)
C(10)	0.5600 (8)	0.2385 (4)	0.0254 (3)	0.055 (4)
S(1)	0.1824 (2)	−0.0785 (1)	0.1156 (1)	0.063 (1)
S(2)	0.0579 (3)	−0.1912 (2)	0.2301 (1)	0.130 (3)
S(3)	0.5800 (3)	−0.1793 (2)	0.2927 (1)	0.126 (2)
S(4)	0.6164 (2)	−0.0615 (1)	0.1689 (1)	0.066 (1)
S(5)	0.2344 (2)	0.1150 (1)	0.0139 (1)	0.053 (1)
S(6)	0.2295 (3)	0.3292 (1)	−0.0470 (1)	0.080 (2)
S(7)	0.7215 (3)	0.3424 (1)	0.0138 (1)	0.080 (2)
S(8)	0.6724 (2)	0.1275 (1)	0.0657 (1)	0.052 (1)
C(3*M*)	0.3245 (23)	−0.3528 (13)	0.2715 (9)	0.177 (5)
C(4*M*)	0.2374 (23)	−0.1386 (15)	0.3540 (9)	0.119 (5)
C(8*M*)	0.3435 (15)	0.4196 (9)	−0.1553 (5)	0.144 (5)

Table 2. *Selected geometric parameters* (Å, °)

C(6)—C(1)	1.342 (7)	S(4)—C(5)	1.759 (6)
S(1)—C(1)	1.746 (5)	S(5)—C(6)	1.755 (5)
S(4)—C(1)	1.749 (5)	S(8)—C(6)	1.754 (5)
C(5)—C(2)	1.329 (8)	C(10)—C(7)	1.342 (8)
S(1)—C(2)	1.753 (6)	S(5)—C(7)	1.751 (6)
S(2)—C(2)	1.750 (7)	S(6)—C(7)	1.736 (6)
C(4)—C(3)	1.232 (11)	C(9)—C(8)	1.394 (11)
S(2)—C(3)	1.785 (10)	S(6)—C(8)	1.787 (9)
C(3*M*)—C(3)	1.720 (20)	C(8*M*)—C(8)	1.415 (13)
S(3)—C(4)	1.768 (11)	S(7)—C(9)	1.792 (9)
C(4*M*)—C(4)	1.643 (22)	S(7)—C(10)	1.737 (6)
S(3)—C(5)	1.746 (6)	S(8)—C(10)	1.754 (5)
S(1)—C(1)—C(6)	123.1 (4)	S(8)—C(6)—C(1)	122.7 (4)
S(4)—C(1)—C(6)	123.1 (4)	S(8)—C(6)—S(5)	113.8 (3)
S(4)—C(1)—S(1)	113.7 (3)	S(5)—C(7)—C(10)	117.0 (4)
S(1)—C(2)—C(5)	117.8 (5)	S(6)—C(7)—C(10)	123.7 (4)
S(2)—C(2)—C(5)	128.5 (5)	S(6)—C(7)—S(5)	119.0 (3)
S(2)—C(2)—S(1)	113.6 (3)	S(6)—C(8)—C(9)	120.7 (6)
S(2)—C(3)—C(4)	126.2 (9)	C(8*M*)—C(8)—C(9)	118.9 (9)
C(3*M*)—C(3)—C(4)	77.1 (9)	C(8*M*)—C(8)—S(6)	112.3 (7)
C(3*M*)—C(3)—S(2)	104.4 (8)	S(7)—C(9)—C(8)	123.0 (7)
C(4*M*)—C(3)—C(4)	62.6 (9)	S(7)—C(10)—C(7)	125.5 (4)
C(4*M*)—C(3)—S(2)	98.7 (8)	S(8)—C(10)—C(7)	117.2 (4)
C(4*M*)—C(3)—C(3*M*)	139.7 (8)	S(8)—C(10)—S(7)	117.1 (3)
S(3)—C(4)—C(3)	132.1 (9)	C(2)—S(1)—C(1)	93.9 (3)
C(3*M*)—C(4)—C(3)	63.2 (9)	C(3)—S(2)—C(2)	102.1 (3)
C(3*M*)—C(4)—S(3)	99.5 (7)	C(5)—S(3)—C(4)	100.6 (4)
C(4*M*)—C(4)—C(3)	75.7 (9)	C(5)—S(4)—C(1)	94.5 (3)
C(4*M*)—C(4)—S(3)	107.6 (10)	C(7)—S(5)—C(6)	93.9 (2)
C(4*M*)—C(4)—C(3*M*)	138.8 (9)	C(8)—S(6)—C(7)	99.2 (4)
S(3)—C(5)—C(2)	128.6 (5)	C(10)—S(7)—C(9)	101.0 (4)
S(4)—C(5)—C(2)	116.3 (5)	C(10)—S(8)—C(6)	93.7 (2)
S(4)—C(5)—S(3)	115.1 (3)	C(4)—C(3*M*)—C(3)	39.7 (5)
S(5)—C(6)—C(1)	123.5 (4)	C(4)—C(4*M*)—C(3)	41.7 (6)

The occupancies of the C(3*M*) and C(4*M*) methyl atoms were refined to 0.596 (3) and 0.404 (3), respectively. The H atoms were geometrically located and were not refined. However, the appropriate occupancy was assigned for structure-factor calculations.

Program(s) used to solve structure: *SHELX*86 (Sheldrick, 1985). Program(s) used to refine structure: *SHELX*76 (Sheldrick, 1976). Molecular graphics: *ORTEP*II (Johnson, 1976) and *PLUTO* (Motherwell & Clegg, 1978). Software used to prepare material for publication: *SWATAB* (Swaminathan, 1989).

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Lists of structure factors, anisotropic displacement parameters, H-atom coordinates, complete geometry, including H-atom geometry, least-squares-planes data and torsion angles have been deposited with the IUCr (Reference: CR1087). Copies may be obtained through The Managing Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

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N-(4,6-Diméthylpyrid-2-yl)-5-méthylpyrazine-2-carboxamide

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Abstract

The whole molecule of the title compound, $C_{13}H_{14}N_4O$, is approximately planar. There is a C—H···O [2.955 (6) Å, 107°] and an N—H···N

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