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Charge-Transfer Complexes of 1,4,5,8-Naphthalenetetrones and 1,4,9,10-Anthracenetetrones. Novel Acceptors for Electrically Conducting Materials

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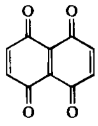
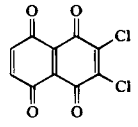
The cyclic voltammograms of 1,4,5,8-naphthalenetetrone (NTO), 2,3-dichloro-1,4,5,8-naphthalenetetrone (DClNTO), 1,4,9,10-anthracenetetrone (ATO), and 6,7-dichloro-1,4,9,-10-anthracenetetrone (DClATO) have indicated that they have suitable redox potentials as an acceptor component of electrically conducting charge-transfer complexes. They have actually been found to form stable anion radical salts with alkali metals and charge-transfer complexes with TTF-type donors. The NTO and DClATO complexes are found to be highly conductive, while the ATO and DClNTO complexes are not. The single-crystal X-ray structure analysis of TTF−ATO, $P2_1/c$, $a = 11.022(5)$, $b = 13.924(3)$, $c = 12.365(3)$ Å, $\beta = 110.99(2)°$, $V = 1768.5(1)$ Å^3, $Z = 4$, has confirmed that the oxidation state evaluated from the molecular geometries corresponds to that estimated from the infrared spectra. A partial charge-transfer state is found to be achieved in the conductive complexes.

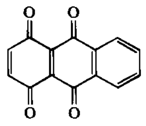
Introduction

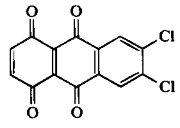
There has been marked progress in the study of organic conductors, and many metallic conductors and even superconductors have been produced. The key material for the progress is considered TTF−TCNQ[1,2] (TTF, tetrathiafulvalene, TCNQ, 11,11,12,12-tetracyanoquinodimethane). Most of donors or acceptors designed for molecular conductors are based on the molecular framework of TTF or TCNQ. For each of them, various chemical modifications have been reported.[3] On the other hand, so far, there has been only a little effort for finding other frameworks than TTF or TCNQ for molecular conductors.

Tetrahalo-*p*-benzoquinone is a rare example of other types of acceptors which provide conductive charge-transfer complexes.[4] The framework of it, *p*-benzoquinone, is known to act as an electron acceptor, e.g., in quinhydrone. Since the electron affinity of *p*-benzoquinone is not large enough to form conducting charge-transfer complexes, it is required to be modified with electron-withdrowing substituents on it for realizing electron-transfer between molecules. An extension of *p*-benzoquinone, 1,4,5,8-naphthalenetetrone (NTO), has attracted our attention, since it is expected to have larger electron affinity than *p*-benzoquinone. The reported reduction potential of NTO was indeed largely shifted to the positive side by more than 0.5 V compared with *p*-benzoquinone.[5,6] The molecule is also interesting from some other points of view; namely, its twin-type structure may reveal multiple reduction states, may reduce the on-site Coulomb repulsion, and may act as a strong proton acceptor for the formation of hydrogen bonds. NTO is not a new compound; the synthesis was reported in 1928.[7] However, the charge-transfer complex with NTO reported so far is only one, pyrene−NTO.[8]


NTO


DClNTO


ATO


DClATO

In this paper, we describe the properties of NTO as an acceptor. We also show those of three modifications of the framework, DClNTO, ATO, and DClATO. Electrical properties have been examined for charge-transfer complexes and alkali metal salts based on these acceptors. These results as well as the crystal structure of TTF−ATO are presented.

Experimental Section

Materials. NTO was synthesized by oxidation of naphthazarin with lead tetraacetate following the method reported[7] and was purified by recrystallization from chlorobenzene. Naphthazarin was commercially obtained. DClNTO, ATO, and DClATO were synthesized similarly from commercially obtained 2,3-dichloronaphthazarin, quinizarin, and 6,7-dichloroquinizarin, respectively.

⊗ Abstract published in *Advance ACS Abstracts,* August 15, 1994.

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Alkali metal salts of NTO, DClNTO, ATO, and DClATO were prepared by reaction with corresponding iodides in dried acetonitrile under nitrogen atmosphere. Anal. K−NTO: Calcd for 1:1 ($C_{10}H_4KO$): C, 52.85; H, 1.77. Found: C, 52.27; H, 2.23. K−DClNTO: Calcd for 1:1:1.5H_2O ($C_{10}H_5KO_{5.5}C_{l2}$): C, 37.17; H, 1.56; Cl, 21.94. Found: C, 37.47; H, 1.36; Cl, 21.26. K−ATO: Calcd for 1:1 ($C_{14}H_6KO_4$): C, 60.63; H, 2.18. Found: C, 60.07; H, 2.25. K−DClATO: Calcd for 1:1 ($C_{14}H_4KO_4Cl_2$): C, 48.57; H, 1.16. Found: C, 48.25; H, 1.15. Rb−ATO: Calcd for 1:1 ($C_{14}H_6O_4Rb$): C, 51.94; H, 1.87. Found: C, 52.00; H, 1.86. Cs−ATO: Calcd for 1:1 ($C_{14}H_6CsO_4$): C, 45.31; H, 1.63. Found: C, 45.22; H, 1.80. All the alkali metal salts obtained are relatively stable in the air; the electrical resistivities of the pressed pellets did not change with exposure to the air for a few days or after storing them in a refrigerator for 2 months, though some elemental analyses were indicative that the solids absorbed moisture.[9]

Charge-transfer complexes were prepared by mixing of the solutions of donor and acceptor. Solvents used for the preparation were benzene or chlorobenzene, since the acceptors are less stable in more polar solvents. It was also found that the acceptors were not stable when the temperature of the solution was near the boiling point. The temperature during the preparation was thus kept as low as possible. The donors examined are mainly TTF, TMTTF (tetramethyltetrathiafulvalene), and TMTSF (tetramethyltetraselenafluvalene) which are relatively soluble in benzene or chlorobenzene near room temperature. The highest temperature applied was 80 °C for the crystal growth of TTF−ATO. Anal. TTF−NTO: Calcd for 1:1 ($C_{16}H_8O_4S_4$): C, 48.95; H, 2.05; S, 32.68. Found: C, 48.87; H, 2.10; S, 32.53. TTF−DClNTO: Calcd for 1:1:0.33 chlorobenzene ($C_{18}H_{7.66}O_4S_4Cl_{2.33}$): C, 43.34; H, 1.55; S, 25.72; Cl, 16.56. Found: C, 43.16; H, 1.68; S, 25.87; Cl, 16.72. TMTTF−ATO: Calcd for 1:1 ($C_{24}H_{18}O_4S_4$): C, 57.87; H, 3.63; S, 25.72. Found: C, 57.65; H, 3.85; S, 25.88. TTF−DClATO: Calcd for 1:1 ($C_{20}H_8O_5S_4Cl_2$): C, 46.96; H, 1.57; S, 25.08; Cl, 13.86. Found: C, 46.60; H, 1.79; S, 24.66; Cl, 14.03. TMTTF−DClATO: Calcd for 4:5:4 $H_2O\cdot(C_{110}H_{76}O_{24}S_{16}Cl_{10})$: C, 49.87; H, 2.89; S, 19.37; Cl, 13.38. Found: C, 49.50; H, 2.56; S, 19.39; Cl, 13.53.

Measurements. Electrical conductivity measurements were performed by a conventional four-probe or two-probe method. It was found that the conducting paste, probably the thinner, used for making electrical contacts between electrodes (20 μm thickness gold wire) and the sample influenced the conductivity of charge-transfer complexes. Reliable data were obtained for most cases when a silver paste thinned with *n*-butyl acetate was used. The conductivity is much less when a silver or gold paste thinned with less volatile solvents was used.

Cyclic voltammograms were recorded on a YANACO polarographic analyzer P-1100 with a platinum working electrode and an Ag·AgCl reference electrode. The infrared absorption spectra of Nujol mull specimens were measured using a Perkin-Elmer 1650 FT IR spectrometer.

X-ray Structure Analysis. A single crystal of TTF−ATO with the dimensions of 0.52 × 0.22 × 0.12 mm^3 was grown from a chlorobenzene solution by slow cooling. An automated Rigaku AFC-5R diffractometer with graphite monochromatized Mo Kα radiation ($\lambda = 0.710\ 73$ Å) was used for data collection at room temperature. Twenty-five reflections with $20° < 2\theta < 30°$ were used to determine the lattice parameters. Crystal data are summarized in Table 1. The intensities of the three standards monitored every 150 data measurements showed no significant deviation; the deviations were within 1.3%. The structure was solved by a direct method (SHELX 86[10]). The positions of the hydrogen atoms were determined from difference synthesis maps. A block-diagonal least-squares technique (UNICS III[11]) with anisotropic thermal parameters for non-hydrogen atoms and isotropic for hydrogen atoms was employed for the structure refinement. The final *R* is 0.057, and R_w is 0.052.

Table 1. Crystallographic Data for TTF−ATO

molecular formula	$C_{20}H_{10}O_4S_4$
molecular weight	442.54
crystal system	monoclinic
space group	$P2_1/c$
a/Å	11.022(5)
b/Å	13.924(3)
c/Å	12.365(3)
β/deg	110.99(2)
V/Å^3	1768.5(1)
Z	4
D_{calcd}/g cm^{-3}	1.662
μ(Mo Kα)/cm^{-1}	5.41
2θ range	$5° < 2\theta < 60°$
range of h, k, and l	$0 \le h \le 15, 0 \le k \le 19, -17 \le l \le 16$
scan width/deg	$1.3 + 0.3 \tan\theta$
scan mode	$\omega-2\theta$
scan rate/$\theta°$ min^{-1}	6
no. of reflections measured	5609
no. of independent reflections observed ($F_o > 3\sigma(F_o)$)	3287
no. of parameters	294
R	0.057
R_w	0.052
weighting factor (g) $w^{-1} = \sigma^2 + (gF)^2$	0.015

Table 2. Electrochemical Data

acceptor	$E°$ (vs Ag/AgCl)^a/V	acceptor	$E°$ (vs Ag/AgCl)^a/V
NTO	+0.03	DClATO	+0.07
DClNTO	+0.23	*p*-chloranil	−0.02
ATO	−0.04	TCNQ	+0.15

[a] $E° = (E^a + E^c)/2$, where E^a and E^c are anodic and cathodic peak potentials, respectively (in acetonitrile with $Bu_4N\cdot BF_4$).

Results and Discussion

Cyclic Voltammograms. The potentials for the first redox process are summarized in Table 2. They are all reversible. On the other hand, the second redox processes are not completely reversible; the second reduction peaks were observed at 0.5−0.7 V more negative side from the first peak, but the peak is smeared for the reverse process. The value for NTO is in good agreement with that reported before.[5,6] The redox potential, +0.03 V, is slightly more positive than that of *p*-chloranil (−0.02 V) and is more negative than that of TCNQ (+0.15 V). The redox potentials of three other acceptors are in the range −0.04 V (ATO) to 0.23 V (DClNTO). ATO is therefore considered to be slightly weaker than *p*-chloranil and DClNTO is slightly stronger than TCNQ. Two others are between *p*-chloranil and TCNQ. As mentioned in the Introduction, both *p*-chloranil and TCNQ are typical electron acceptors which form electrically conducting charge-transfer complexes, especially with TTF-type donors. With simple application of the guidelines for the combination of donor and acceptor for electrically conducting charge-transfer complexes,[12,13] these acceptors can be considered to have potential to form conducting complexes with most of TTF-type donors.

Alkali Metal Salts. The alkali metal salts were all obtained as a powdered form. The composition, resis-

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[9] Some discrepancies between calcd and found values in elemental analysis for K-NTO and K-ATO are supposed to be due to this effect.

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Table 3. Properties of the Alkali Metal Salts

salts	composition	color	ϱ(RT)^a/Ω cm	$E_a{}^b$/eV
Li–NTO		dark purple	4×10^2	0.10
Li–DClNTO		dark purple	5×10^4	0.24
Li–ATO		dark purple	3×10^4	0.23
Li–DClATO		dark purple	2×10^5	0.44
K–NTO	1:1	bluish green	2×10^5	0.36
K–DClNTO	1:1^c	dark purple	1×10^5	0.49
K–ATO	1:1	dark green	8×10^6	0.61
K–DClATO	1:1	dark green	9×10^5	0.50
Na–ATO		purple	2×10^4	0.23
Rb–ATO	1:1	dark green	2×10^7	0.53
Cs–ATO	1:1	purple	1×10^7	0.48

^{a} Resistivity at room temperature for powder compaction. ^{b} Activation energy for conduction. ^{c} The salt contains crystal water.

tivity at room temperature, and activation energy for conduction are summarized in Table 3 for the lithium and potassium salts. The composition of the lithium salts is not clear, since the weight percent of lithium is too small to determine the composition from elemental analysis. For the potassium salts, 1:1 composition was confirmed by elemental analysis.

The resistivities of the potassium salts are nearly the same order as that of the TCNQ simple salt. On the other hand, the resistivities of the lithium salts, especially Li–NTO, are much lower. Since the measurements are for the compressed pellets, the resistivity along the molecular stacking direction is expected to be quite low. This fact also leads the question about the stoichiometry of these salts. The sodium, rubidium, and cesium salts were prepared for ATO. The resistivities at room temperature are 2×10^4, 2×10^7, and 1×10^7 Ω cm, respectively. The compositions are 1:1 except the sodium salt due to the same reason for the lithium salts. The salt with small cation tends to give low resistivity.

The preliminary results of the visible–near-infrared spectra for Nujol mull specimens of Li and K salts clearly indicate the difference between Li–NTO and the others; the electronic absorption well extended to the infrared region was observed for Li–NTO, while the charge-transfer bands for the others were observed between 0.7 and 1.2 eV.

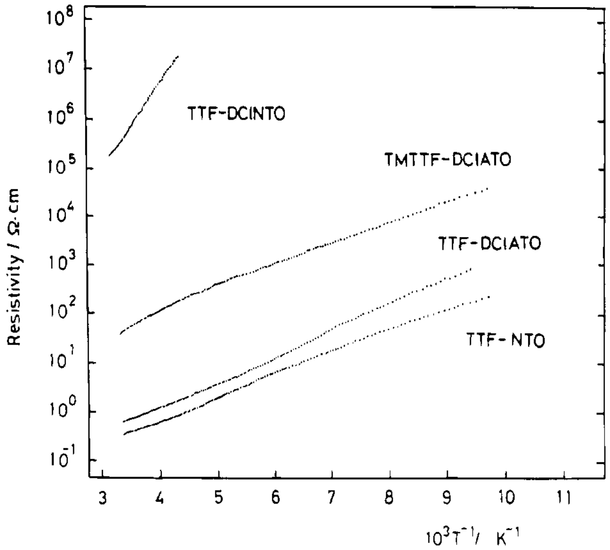

Figure 1. Temperature dependence of resistivity (powder compaction).

Table 4. Peak Position of the CO Stretching Band

compound	ν_{CO}/cm^{-1}	compound	ν_{CO}/cm^{-1}
NTO	1693	K–ATO	1625
K–NTO	1615	TTF–ATO	1642
TTF–NTO	1642	TMTTF–ATO	1668
DClNTO	1703	DClATO	1700
K–DClNTO	1631	K–DClATO	1625
TTF–DClNTO	1630	TTF–DClATO	1644
ATO	1702	TMTTF–DClATO	1659

Charge-Transfer Complexes of NTO. As mentioned before, the redox potentials of these acceptors are suitable for constructing conducting charge-transfer complexes with TTF-type donors. Among the donors examined, only TTF gave a complex as a powdered form by reaction with NTO . The temperature dependence of the resistivity is shown in Figure 1. The stoichiometry is 1:1 based on elemental analysis. Since the resistivity at room temperature is quite low, 2.0×10^{-1} Ω cm, it is naturally expected that partial charge-transfer from donor to acceptor is occurred in the complex. The degree of charge transfer is then estimated from the CO stretching band of NTO. Although it has not been established for NTO-type acceptors, it is well-known that the frequency of the CO stretching band of *p*-chloranil-type acceptors can be used for the estimation of charge on acceptor.[14] Actually, as shown in Table 4, the CO stretching band of NTO shows a large red-shift when it becomes anion radical. The frequency of the CO stretching band of TTF–NTO, 1642 cm^{-1}, is between those of the neutral and anion radical states. By assuming a linear correlation between the shift and the charge on NTO, the degree of charge transfer in TTF–NTO is roughly estimated to be 0.6–0.7. The value should be verified quantitatively by other methods such as X-ray structure analysis. This will be discussed in the structures of TTF–ATO (vide infra). The fact that the complex has quite a high conductivity despite the stoichiometry of 1:1 also supports that the degree of charge transfer is between zero and one.

Charge-Transfer Complexes of DClNTO. The TTF complex which has 1:1 stoichiometry was obtained as a powdered form. The electrical resistivity of this complex is relatively high ($\varrho(RT) = 2 \times 10^5$ Ωcm). The frequency of the CO stretching mode in the infrared spectrum is nearly the same as that of the potassium salt, which suggests that the complex is rather a simple salt. Since the redox potential of DClNTO is higher than that of TCNQ, the acceptor is probably too strong to form a partial charge-transfer complex with TTF. TMTTF and TMTSF did not give charge-transfer complexes.

Charge-Transfer Complexes of ATO. Reaction with TTF or TMTTF gave a 1:1 complex. As shown in Table 5, their resistivities are rather high. In contrast to the other complexes and the salts studied, TTF–ATO can be obtained as a crystalline form which has a suitable size for X-ray structure analysis. The molecular structures determined from the X-ray structure

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Table 5. Properties of the Charge-Transfer Complexes

complex	composition	$\varrho(\mathrm{RT})^a/\Omega$ cm	$E_a{}^b$/eV
TTF−NTO	1:1	2×10^{-1}	0.07
TTF−DClNTO	1:1^c	2×10^{5}	0.32
TTF−ATO	1:1	1×10^{6e}	0.40
TMTTF−ATO	1:1	2×10^{6}	0.43
TTF−DClATO	1:1	6×10^{-1}	0.10
TMTTF−DClATO	4:5^d	4×10^{1}	0.08

[a] Resistivity at room temperature for powder compaction. [b] Activation energy for conduction. [c] Crystal contains solvent molecules. [d] Only nominal value (it could be a mixture of two phases). [e] Value for single crystal.

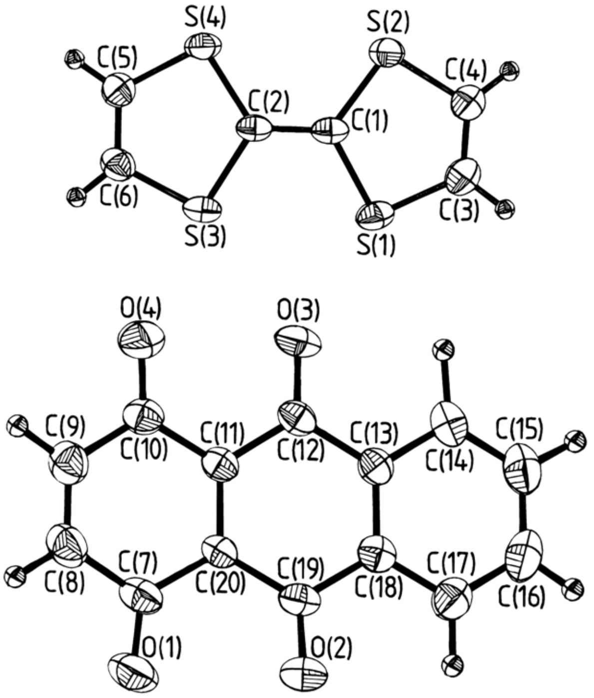


Figure 2. ORTEP drawing of TTF (top) and ATO (bottom) showing the atom numbering scheme.

analysis are shown in Figure 2 with the atom numbering scheme. The crystal structure is shown in Figure 3, and the atomic parameters are listed in Table 6. As expected from its high resistivity, donors and acceptors do not form segregated columns; the crystal completely loses the columnar structure and is constructed with the units which composed of a donor and an acceptor. In the crystal, the ATO plane of one unit is faced to another related by an inversion center (e.g., two ATO molecules centered at (0.5, 0.5, 0.5) in Figure 3). Interplanar distance between ATOs, namely, between the neighboring units, is however much longer (3.43 Å) compared with that between TTF and ATO within the unit (3.27 Å). Along the other directions, the units are arranged in a complicated way and are weakly bound to each other; the shortest interatomic contact between the units relative to the sum of the van der Waals radii is 3.020(3) Å for S(3)┅O(4). In the unit, there are short interatomic contacts as listed in Table 7. It is quite reasonable from this structure that this complex is not electrically conductive.

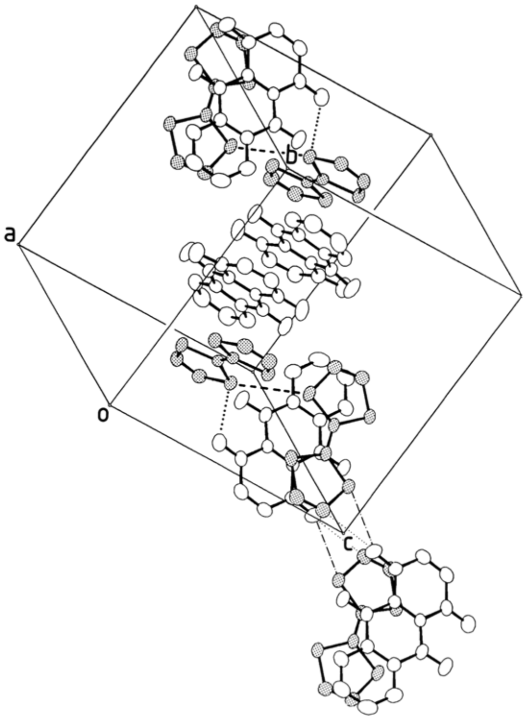


Figure 3. Crystal structure of TTF−ATO and short interatomic contacts between the units: thick dotted line, S(1)┅O(1); thick broken line, S(1)┅S(2); thin dotted line, C(6)┅O(4); thin dotted broken line, S(3)┅O(4) in Table 7.

Table 6. Atomic Parameters for TTF−ATO

	x	y	z	$B_{\mathrm{eq}}{}^a/\mathrm{Å}^2$
S(1)	0.2320(1)	0.1855(1)	0.4642(1)	3.24(2)
S(2)	0.2375(1)	0.2187(1)	0.2325(1)	3.38(2)
S(3)	−0.0002(1)	0.3516(1)	0.4088(1)	3.29(2)
S(4)	0.0040(1)	0.3639(1)	0.1729(1)	3.34(2)
C(1)	0.1729(3)	0.2509(2)	0.3367(2)	2.6(1)
C(2)	0.0745(3)	0.3170(2)	0.3128(3)	2.7(1)
C(3)	0.3407(3)	0.1190(3)	0.4231(3)	3.3(1)
C(4)	0.3431(3)	0.1333(3)	0.3174(3)	3.4(1)
C(5)	−0.1089(3)	0.4330(3)	0.2054(3)	3.6(1)
C(6)	−0.1112(3)	0.4275(3)	0.3119(3)	3.6(1)
O(1)	0.2468(3)	0.5160(2)	0.0742(2)	5.4(1)
O(2)	0.4765(3)	0.4280(2)	0.1936(2)	5.2(1)
O(3)	0.3293(3)	0.4132(2)	0.5639(2)	4.6(1)
O(4)	0.1555(3)	0.5472(2)	0.4670(2)	5.2(1)
C(7)	0.2366(4)	0.5253(3)	0.1701(3)	3.6(1)
C(8)	0.1389(3)	0.5915(3)	0.1807(3)	4.0(1)
C(9)	0.1154(4)	0.5991(3)	0.2794(3)	3.8(1)
C(10)	0.1839(3)	0.5408(2)	0.3796(3)	3.1(1)
C(11)	0.2864(3)	0.4745(2)	0.3738(3)	2.6(1)
C(12)	0.3592(3)	0.4161(2)	0.4766(3)	2.8(1)
C(13)	0.4697(3)	0.3576(2)	0.4726(3)	2.8(1)
C(14)	0.5381(3)	0.3005(3)	0.5697(3)	3.6(1)
C(15)	0.6403(4)	0.2441(3)	0.5652(3)	4.5(1)
C(16)	0.6777(4)	0.2466(3)	0.4675(4)	4.7(1)
C(17)	0.6123(3)	0.3040(3)	0.3723(3)	3.8(1)
C(18)	0.5065(3)	0.3598(2)	0.3747(3)	2.8(1)
C(19)	0.4351(3)	0.4198(3)	0.2730(3)	3.1(1)
C(20)	0.3173(3)	0.4711(2)	0.2735(3)	2.7(1)

[a] $B_{\mathrm{eq}} = {}^4/_3\Sigma_i\Sigma_j\beta a_i a_j$.

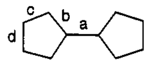
It is meaningful to examine whether the frequency of the CO stretching band is really correlated with the degree of charge transfer which can be estimated from the molecular geometry. The mean bond lengths of TTF are compiled in Table 8 together with those in different oxidation states. The central C=C bond is known to be sensitively changed by the charge on TTF. From comparison with other compounds, the charge on TTF is estimated to be nearly 0.6 in TTF−ATO. The degree of charge transfer estimated from the frequency of the CO stretching band shown in Table 4 is 0.7−0.8. The agreement is considered to be rather good.

The 1:1 TMTTF complex is also a poor conductor. Since the degree of charge transfer estimated from the frequency of the CO stretching band is 0.4−0.5, the

Table 7. Short Interatomic Distances (Å) between Molecules

Within a Unit			
S(1)–C(12)	3.484(3)	C(1)–C(11)	3.326(4)
S(2)–C(18)	3.462(3)	C(1)–C(12)	3.153(4)
S(2)–C(19)	3.472(4)	C(2)–C(10)	3.332(4)
S(3)–O(4)	3.162(3)	C(2)–C(11)	3.092(5)
S(3)–C(10)	3.416(4)	C(2)–C(12)	3.357(4)
S(4)–C(7)	3.419(4)	C(3)–C(14)	3.394(5)
S(4)–C(8)	3.479(4)	C(5)–C(9)	3.261(5)
Between Units			
S(1)–O(1)	3.094(3)	S(3)–O(4)	3.020(3)
S(1)–S(2)	3.556(2)	C(6)–O(4)	2.960(5)

Table 8. Mean Dimensions of TTF/Å



	TTF[a]	TTF−TCNQ[b]	TTF−Br[c]	TTF−ClO$_4$[d]	TTF−ATO
a	1.349	1.372	1.393	1.404	1.371
b	1.757	1.745	1.720	1.713	1.740
c	1.726	1.739	1.724	1.725	1.729
d	1.314	1.326	1.332	1.306	1.332

[a] Reference 15. [b] Reference 16. [c] Reference 17. [d] Reference 18.

crystal should be composed of mixed-stack type columns or isolated units like TTF−ATO.

Charge-Transfer Complexes of DClATO. Both TTF and TMTTF form charge-transfer complexes with DClATO. The composition of TTF−DClATO is determined as 1:1 from elemental analysis. On the other hand, the composition of TMTTF−DClATO could not be determined from elemental analysis. A nominal value has been obtained as 4:5, but it is not sure whether the powdery material obtained is homogeneous or a mixture of two complexes which have different compositions. The resistivities at room temperature are shown in Table 5. Compared with other complexes studied, these complexes show gradual change of resistivity by time. This effect is more pronounced in TMTTF−DClATO. Although the values listed in Table 5 are obtained immediately after preparation of the complex, it has been noticed that the resistivity is gradually increased during the attachment of probes. Therefore, the intrinsic resistivities of these complexes are expected to be lower than those listed in Table 5; probably the same order as that of TTF−NTO.

In conclusion, we have found that NTO-type acceptors give highly conducting charge transfer complexes with TTF-type donors. Since most of the conducting charge-transfer complexes found have 1:1 stoichiometry, the acceptors are also expected to form partially charge-transferred complexes with other donors than TTFs and partially reduced salts with some closed-shell cations, as found for TCNQ. Syntheses and properties of diquinone-type acceptors were extensively studied by Miller et al.[5,6,19] In their compounds, two *p*-benzoquinone groups are not directly bound but separated by aromatic rings. Some conducting solids have been obtained,[20] though the reduction potentials are slightly more negative than those of NTO-type acceptors. Our work as well as theirs indicates that a compound in which two or more *p*-benzoquinone groups are fused becomes a good acceptor for constructing electrically conducting salts or complexes.

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Supplementary Material Available: Tables of atomic parameters of hydrogen atoms, anisotropic thermal parameters of non-hydrogen atoms, bond lengths, bond angles, and least-squares planes and atomic deviations for TTF-ATO (7 pages); a list of *h*, *k*, *l*, F_o, and F_c for TTF-ATO (10 pages). Ordering information is given on any current masthead page.

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