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Requirements for an "Organic Metal"

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Current theories on the origin of high electrical conductivity in TCNQ-based materials emphasize the need for incomplete charge transfer as well as segregated stacks of donor and acceptor molecules and uniform spacing within the stacks. The requirements for an "organic metal" are discussed in terms of partial charge transfer. An examination of 61 structurally related TTF–TCNQ based complexes suggests that the combinations with the redox potential $-0.02 \leq E_1(\mathrm{D}) - E_1(\mathrm{A}) \leq 0.34$ V have a high possibility of being "organic metals." A method to arrange the complex by the difference between the electrochemical half-wave potentials of the donor and the acceptor molecule is proposed to predict electrical properties of the complex.

Since the discovery of highly electrically conducting charge transfer (CT) complexes[1,2] and anion-radical salts[3,4] of tetracyano-*p*-quinodimethane (TCNQ), several proposals have been forwarded to design new "organic metals." As early as five years after the first report of the electrical properties of TCNQ salts by the Dupont group,[5] LeBlanc proposed the importance of cation polarizability as a criterion for high conductivity.[6] Prior to this, Little[7] suggested a design for a high temperature organic superconductor by incorporation of high polarizability through suitable molecules pendant on a conjugated polymeric backbone. The efforts of organic chemists in the several years following to prepare "organic metals" solely on the basis of polarizability considerations met with little success.[8] Prior to 1973 only three anion radical salts of TCNQ displayed "metallic" behavior over a limited temperature range. The discovery of the TCNQ complex of tetrathiafulvalene (TTF) in 1973 and the demonstration of its metallic properties over a relatively wide temperature range reinstilled interest in the search for organic conductors. To date over 70 "organic metals" have been prepared and subjected to a battery of physical measurements. In general, these "organic metals" may be placed into one of four categories. An estimate of the number of known complexes for each classification is given in brackets.

1. Charge transfer complexes which are ionic at 300 K, with such notables as TTF–TCNQ, TTT (tetrathiatetracene)$_2$–TCNQ, Δ4,4′-bithiopyran–TCNQ and their derivatives and analogs. [>50]
2. Anion radical salts of TCNQ[3,4,9] with formally closed shell cations represented by quinolinium–(TCNQ)$_2$, acridinium–(TCNQ)$_2$ and *N*-methylphenazinium(NMP)–TCNQ. [≈7]
3. Cation radical salts[10] with formally closed shell anions typically halide (or related) salts of TTF, TTT, nickel phthalocyanine or perylene. [≈7]
4. Ternary salts[11] consisting of an amine complexed with TCNQ and iodide or water in varying proportions. [7]

In 1965, McConnel *et al.*[12] classified the alternating CT complexes in the solid state into two distinct categories —those with ionic ground states and those without on the basis of the magnitude of the molecular field energy, $E = \mathrm{N}[(I-A)\rho - M\rho^2/2]$ where I, A, M, and ρ are the ionization potential of the donor, electron affinity of the acceptor, electrostatic energy per DA pair, and the average charge density ($0 \leq \rho \leq 1$) per molecule, respectively, in N DA pairs in the crystal. Matsunaga[13] has pointed out the utility of IR spectra to distinguish the nature of the ground state as applied to a number of CT complexes including aromatic hydrocarbons, aromatic diamines and phenothiazines as donors with *p*-quinones, TCNQ, *etc.* as acceptors. He was able on the basis of IR and electronic spectra to place the alternating CT complexes into groupings similar to McConnell's classifications. In general, ionic complexes were more conducting than nonionic complexes.[14] It was also suggested that the *size* and *shape* of the component molecules were critical factors defining the ionic to nonionic boundary.[14a] It is in the borderline region between these phases that one may expect CT complexes or ion radical salts to display interesting properties like metallic conductivity due to incomplete charge transfer[15] or complex isomerism.[16–18]

Prerequisites to High Conductivity

The search for new materials and an understanding of those already in hand generated numerous criteria for achievement of the metallic state in these systems.[19–22] Some have yet to be fully tested. As a matter of convenience, we choose to divide these criteria into two categories but recognize that they are strongly correlated.

A. *Requirements for Molecular and Crystal Structure.*

1. The component molecule which is responsible for electron transport should be flat in the neutral and ionic state.[3]
2. The donor and/or acceptor should be highly symmetric to avoid strong random potentials which can drive the system insulating (Ref. 1 and references cited therein). The presence of weak electronic or structural disorder need not, in general, destroy the metallic state and may even be the source of stabilization for the uniform chain structure.[17,19,20]
3. The cation (or anion) should be of relatively small size to facilitate polarization effects.[1,2]
4. The geometry of the acceptor should be such that electron withdrawing functional groups are located at diametrically distant points on the molecule to enhance its electron affinity and minimize Coulombic repulsion of electrons in the doubly ionized state.[19]
5. Closely matched physical dimensions for the donor

and acceptor which facilitate packing in the crystal.[22] This militates against bulky substituents.

6. The donor and acceptor moieties must form segregated stacks.[3]

7. The spacing between component molecules in the donor and acceptor stacks must be uniform to delocalize electrons into a single metallic band[19] or into partially filled and partially empty bands which overlap (semimetal).

B. *Requirements for Electronic Structure of Component Molecules and Complexes.*

8. Unpaired electrons on the donor and/or acceptor molecules must be present in the complex.[3,19]

9. Incomplete charge transfer[15] (as will be seen later, this criterion is satisfied by moderately strong donors in combination with moderately strong acceptors such that their redox potential differences are closely matched.).

10. Minimal on site Coulomb repulsive energies to destabilize transitions to a Mott insulator.[19]

11. Enhanced polarizability derived from the introduction of heteroatoms or heavy atoms.[6,22]

12. Formation of a new aromatic sextet upon one electron oxidation or reduction with this aromaticity capable of migrating through the crystal.[21]

13. The dipole moment of the ions should be small.[9b,c]

The achievement of the metallic state in these systems is obviously a complicated interplay of many factors, some of which lend themselves to variability and control. Requirements 1 and 6—8 are fundamental. Unfortunately we are unable to predict *a priori* which systems will necessarily meet criteria 6 and 7. In addition, Torrance[15] has recently demonstrated the necessity of requirement 9. The degree of ionicity (or charge transfer) for several "organic metals" has been estimated by diffuse X-ray scattering measurements, thermopower studies or NMR experiments. Some examples are 0.91—0.94,[23a–c] *ca.* 0.5,[23d] or 0.66,[23e] 0.74,[24] 0.63,[24] and 0.48—0.67[25] for NMP–, HMTSF (hexamethylenetetraselenafulvalene)–, TSF(tetraselenafulvalene)–, and TTF–TCNQ respectively. Systems with complete charge transfer[12,22a] such as TTF–TCNQF$_4$ and TTT–TCNQF$_4$ are invariably insulators. The lower limit of the degree of ionicity for the appearance of metallic behavior has yet to be determined.

Discussion

In this paper we examine 61 of the complexes based on TTF (or TTT) and TCNQ together with their various derivatives and analogs in an attempt to predict other systems which are likely to yield highly conducting complexes. The choice of this series is two-fold. First, by far the largest number of "organic metals" are related to TTF–TCNQ and the physical properties of many of these have been extensively investigated. Second, the size and shape of the component molecules will affect their mode of packing the magnitude of the polarization energies and Madelung energies in the solid complexes and the repulsive Coulomb energies in the ionized molecules. Therefore, only

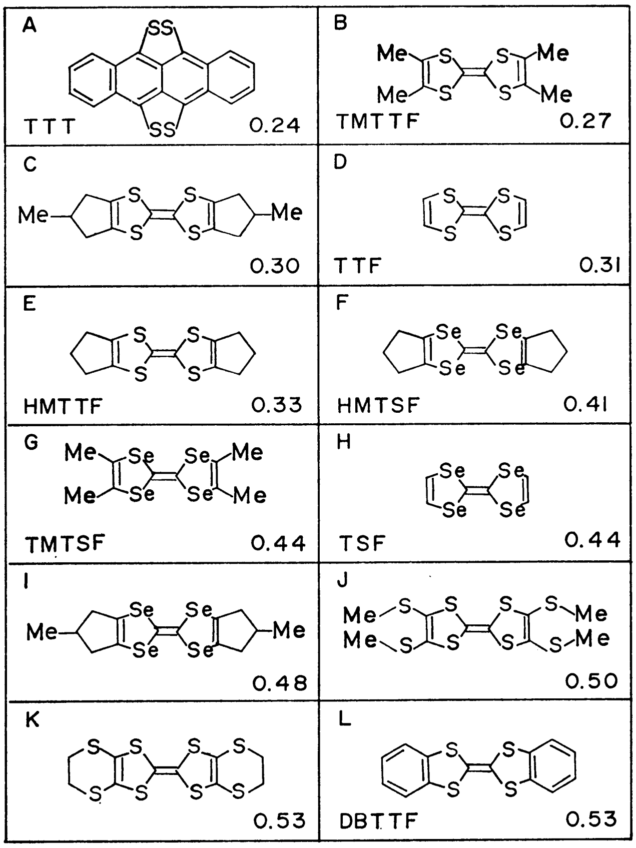
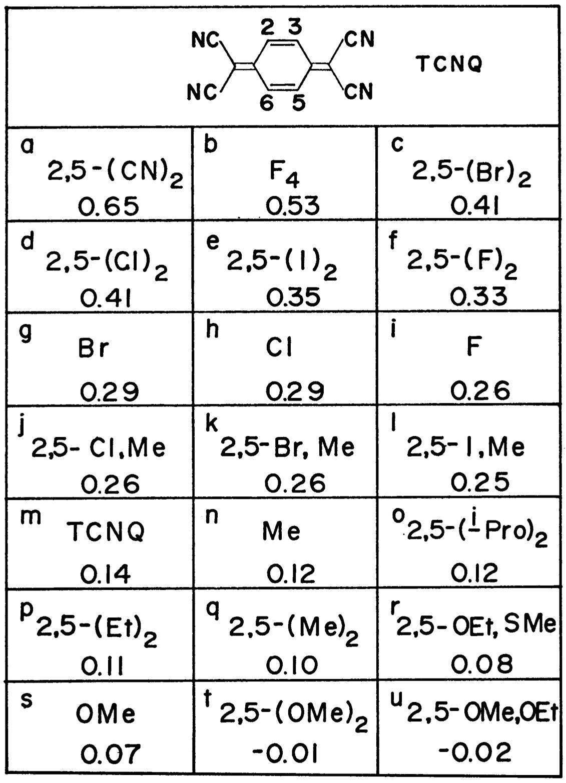


Fig. 1. Electron donors with corresponding electrochemical half-wave potentials.



a 2,5-(CN)$_2$ 0.65	b F$_4$ 0.53	c 2,5-(Br)$_2$ 0.41
d 2,5-(Cl)$_2$ 0.41	e 2,5-(I)$_2$ 0.35	f 2,5-(F)$_2$ 0.33
g Br 0.29	h Cl 0.29	i F 0.26
j 2,5-Cl,Me 0.26	k 2,5-Br, Me 0.26	l 2,5-I,Me 0.25
m TCNQ 0.14	n Me 0.12	o 2,5-(i-Pro)$_2$ 0.12
p 2,5-(Et)$_2$ 0.11	q 2,5-(Me)$_2$ 0.10	r 2,5-OEt,SMe 0.08
s OMe 0.07	t 2,5-(OMe)$_2$ -0.01	u 2,5-OMe,OEt -0.02

Fig. 2. Electron acceptors with corresponding electrochemical half-wave potentials.

an examination of a series with similar molecular structure can minimize the so called size and shape effect considerably. By this way one may discuss the certain parameter which controls the electric property of the complexes. In this meaning, Torrance's examination which includes a variety of structurally different donors such as Na, NMP, TTF, pyrene, *etc.* may have ambiguity.[15)]

In a broad sense these systems should meet all the requirements for high conductivity except number 9, which we may then abstract exclusively. In Figs. 1 and 2 respectively are tabulated the structures of the donor and acceptor molecules considered here along with their first electrochemical half-wave potentials ($D \rightarrow D^{+} + e$; $A + e \rightarrow A^{-}$). We have avoided asymmetric donors intentionally to minimize disorder effects. Since the TTF derivatives form an incomplete set we have also included several TTT complexes in this study. It is apparent that size and shape differences cannot be totally neglected but the differences in on-site Coulomb repulsive energies on the dications should be minimal across the series B-L. We have already shown there is virtually no enhanced on-site Coulomb repulsion present along the series, TCNQF$_n$ (n=0, 1, 2, 4).[26)] The lattice energies are expected to be substantially different for the TTT and TTF complexes, but Madelung energies are relatively constant for donors of *similar size* when complexed with TCNQ, *e.g.* −4.6 to −5.3 eV for Na, Rb, NH_4,[27)] and −2.3 to −2.4 eV for NMP- and TTF-TCNQ.[28)] Therefore as a first approximation we take the Madelung energies as almost constant across the series.

Conductivity results are plotted in Fig. 3.[29)] The complexes have been arranged here by placing the ionization potentials of the donors on the abscissa and electron affinities of the acceptors on the ordinate. We have approximated these two quantities by the solution electrochemical half-wave potentials E_1(D) and E_1(A) respectively.[30)] Similar treatments have been done by Wheland[22b)] and by Torrance,[15)] comparing the electric properties of the solid complexes with the solution electrochemical properties of the component molecules. Our treatment is, however, attempts to predict the properties of solid complexes from the solution electrochemical properties of the component molecules solely.

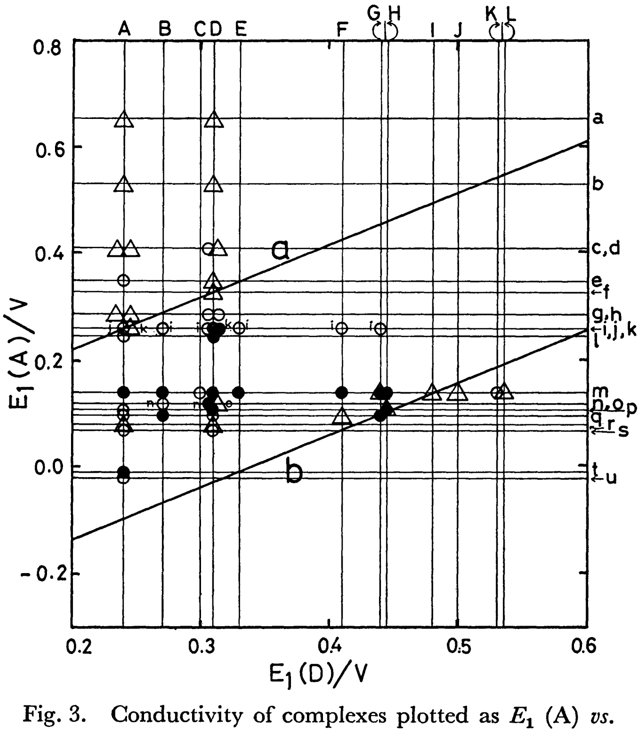


Fig. 3. Conductivity of complexes plotted as E_1 (A) *vs.* E_1 (D).
△=insulators or semiconductors; ○=highly conducting in compaction studies; ●="organic metals."

Those complexes in the upper left of the figure have small (I-A) values and favor the completely charge transferred ionic state. Those on the lower right favor the nonionic state. Those complexes which are insulators or semiconductors are represented by triangles whereas open circles designate highly conducting complexes based on compaction studies and closed circles are "organic metals." In general, complexes displaying substantial electrical conductivity fall between the two lines on the graph. The few exceptions are due to several factors. Complexes A–e and D–c are highly conducting due to heavy atom induced enhanced polarizability. Acceptors o and r are too bulky. It is uncertain why complex K–m is conducting. Some combinations yield complexes of variable stoichiometry: A–m (1: 2), B–m (1: 1), C–m (1: 1), and D–l (1: 1) are highly conductive but other compositions A–m (1: 1),[31)] B–m (1.3: 2),[32)] C–m (2: 1)[29g)] and D–l (2: 1)[33)] are semiconductors. There are two modifications of H–p[17)] and of G–m,[18)] a black "organic metal" and a red insulator. The structure of the latter for H–p has been assigned an alternating structure based on its optical spectrum.[17)] The proximity of these complexes to the lower boundary in the absence of heavy-atom effects may place them at the minimum for degree of charge transfer with retention of metallic behavior.

The two boundaries shown are defined by the upper line (a): $E_1(A) = E_1(D) + 0.02$ and the lower line (b): $E_1(A) = E_1(D) - 0.34$ V respectively. Therefore, to the degree our approximation is accurate, *related combinations with the redox potential* $-0.02 \leq E_1(D) - E_1(A) \leq 0.34$ V *have a high probability of being "organic metals," where* $0.24 \leq E_1(D) \leq 0.53$ V *and* $-0.02 \leq E_1(A) \leq 0.65$ V. This result is compared well with those of Wheland[22b)] ($|E_1(A) - E_1(D)| \leq 0.25$ V) and of Torrance[15)] ($-0.19 \leq E_1(D) - E_1(A) \leq 0.31$ V). Those above line (a) would be completely ionic, those below line (b), nonionic. The idea that CT complexes display a whole spectrum of ionicity is not novel. As early as 1961 Kommandeur classified crystalline organic compounds into five areas based on the magnitude of ($I - A - P_{+} - P_{-} - Q = E^{*}$) where P_{+} and P_{-} are the polarization energies of the cation and anion and Q the electrostatic Coulomb energy.[34)] Recently, Torrance *et al.*[28)] have emphasized that in cases where the net electrostatic binding energy $E_B = (I - A) - M/2$ is small ($\approx$ 2 eV), partial charge transfer may be energetically favored. Those complexes falling between lines (a) and (b) satisfy Torrance's criterion with $1.5 \leq (I - A) - M/2 \leq 1.9$ eV (using $I =$

6.85 eV for TTF,35) $A=2.84$ eV for TCNQ36) and $M/2=\approx 2.3$ eV for these complexes.28) The question still remains what degree of ionicity is necessary for metallic behavior? Investigation of complexes near the boundaries such as D–f, t, u, and D with Me_3TCNQ and Me_4TCNQ should provide the answer.37) The latter two acceptors are still under construction but their E_1(A) values are estimated to be 0.02—0.03 and −0.01 to 0.00 V respectively. Figure 3 is far from complete and further studies of complexes of B, E—H and L with other known acceptors and weaker TCNQ's are required.

Some of the usages of the diagram are as follows. 1: The electrical property of an unknown TTF–TCNQ based complex can be easily predicted with the aid of the diagram. If the electrochemical potential were obtained for a TTF based donor or a TCNQ based acceptor, the other component molecules can be selected to give metallic complexes baring steric and/or some unforeseen effects. 2: A new TCNQ derivative which gives complexes with metallic behavior can be designed. Metallic complexes with *weak* partial charge transfer, *i.e.* in the proximate region of the lower line, may well be worth synthetic efforts. 3: In the course of the synthesis of TTF derivatives and analogs, the electrical properties of their TCNQ complexes have been compared with that of TTF–TCNQ. Considering the importance of partial charge transfer, this comparison should be made between the complexes belonging to the metallic region in the diagram.

One may extrapolate (with some danger) this treatment to the *p*-quinone acceptors, assuming values for Madelung energies, polarization energies and intermolecular spacings in segregated stacks are similar to their TCNQ counterparts though the *p*-quinones have larger disproportionation energies (*ca.* 0.3 eV)38) when compared to the TCNQ series with the same E_1(A). Then *p*-chloranil ($E_1(A)=0.01$ V), *p*-bromanil (0.00 V) and *p*-fluoranil (−0.04 V) complexes with TTF and TMTTF39) locate just above line(b) while 2,3-dichloro-5,6-dicyano-*p*-benzoquinone (0.51 V) complex with DBTTF40) locates just below line (a).

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Note added in proof: A recent paper by J-M Fabre, M. Vigroux, E. Torreilles, and L. Giral (*Tetrahedron Lett.* **1980**, 607) examined TMTTF-Et_2TCNQ (B-p) and found unusual stoichiometry (3 : 2) and structure. The electrical conductivity is metallic: $\sigma_{300K}\simeq 130\Omega^{-1}\ cm^{-1}$.

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