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The crystal structure of the electron acceptor 2,5-difluoro-7,7,8,8-tetracyano-p-quinodimethane, 2,5-TCNQF2, has been determined by single-crystal X-ray diffraction methods. The layered crystal structure is dominated by the antiparallel coupling of cyano bond moments of symmetry-related molecules. The molecular and electronic structure (INDO approximation) of 2,5-TCNQF2 are extensively compared to that of its parent TCNQ and the perfluoro analog TCNQF4. Crystal data for 2,5-TCNQF2 are as follow: monoclinic, space group C2/m, $a = 10.208(4)$ Å, $b = 6.026(2)$ Å, $c = 8.836(3)$ Å, $\beta = 106.64(3)°$, $V = 520.8$ Å^3. The site symmetry (2/m; C_{2h}) in the crystal is identical to that for the free 2,5-TCNQF2 molecule.

INTRODUCTION

Recently, we and others have been devoting considerable effort to the understanding and extension of the solid-state chemistry and physics of organic charge-transfer salts derived from fluorinated analogs of the electron acceptor tetracyano-p-quinodimethane, TCNQ.[2–9] Synthetic progress has been such that routes to three fluorinated TCNQ derivatives are available: the asymmetric monofluoro derivative,[6] TCNQF; the symmetric difluoro analog,[7] 2,5-TCNQF2; and the perfluorinated acceptor TCNQF4.[10]

Important to the charge-transfer chemistry of this family of TCNQ derivatives has been the recognition[6–7,11] that there is a monotonic increase in acceptor electron affinity (EA) as the number of fluoro substituents is increased. Thus, these fluorinated acceptors will be expected (when the acceptor EA is the dominant effect) to yield salts with higher degrees of electron transfer (z)

than the analogous salts of TCNQ. Consistent with this reasoning, the charge-transfer salts of TTF (tetrathiafulvalene),[2,12] HMTSF (hexamethylenetetraselenafulvalene),[3] HMTTF (hexamethylenetetrathiafulvalene),[4] and DBTTF (dibenzotetrathiafulvalene)[5] with the strong electron acceptor TCNQF4 (EA = 3.20 eV; EA(TCNQ) = 2.84 eV[11-13]) all have $z = 1.0$ whereas their TCNQ salts are fractionally charged ($z = 0.59$ for TTF-TCNQ:[14] $z = 0.74$ for HMTSF-TCNQ:[15] $z = 0.72$ for HMTTF-TCNQ;[16] and $z = 0.47$ for DBTTF-TCNQ[17]).

In this context, the electron acceptor 2,5-TCNQF2, with its intermediate EA (3.02 eV[11]), appears on the basis of limited studies to be more versatile. For example, the salt TTF-2,5-TCNQF2,[7-8] with $z = 1.0$, is noteworthy in that its room-temperature crystal structure[8] consists of *dimerized* segregated stacks of donors and acceptors in a motif similar to that exhibited by DBTTF-TCNQF4.[5] Of additional interest is the salt of 2,5-TCNQF2 with the more difficult to oxidize[5,18] donor DBTTF. In this salt, z is ca. 0.65e[19] (near that of TTF-TCNQ, *vide supra*) but its crystal structure[20] is composed of mixed donor-acceptor stacks in a very similar fashion to DBTTF-TCNQ.[17]

In addition to the charge-transfer chemistry of these fluorinated acceptors, we have been investigating their crystal structures,[11,21] and, in particular, the variation in their principal modes of intermolecular interaction.[22] It is well-known that one of the dominant interactions in the structure of TCNQ is intermolecular stacking in which strong quinone ring-over-cyano group overlap is found.[23] In neutral TCNQF,[21] antiparallel cyano-cyano coupling is observed and proposed to be a conduit for the coupling of the permanent molecular electric dipole moments. Finally, in the crystal structure of TCNQF4,[11] the negative inductive effect of the fluoro substituents is sufficient to promote donor/acceptor (acid-base) type interactions to a key intermolecular role.

Herein, we report on the crystal structure of the neutral acceptor 2,5-TCNQF2, Figure 1, and emphasize two derived features: (1) the zero charge-

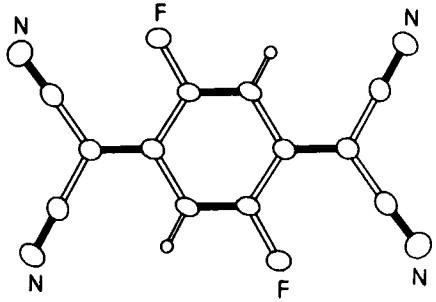


FIGURE 1 Molecular structure of 2,5-TCNQF2. Shaded bonds indicate bond orders greater than unity in the quinone resonance form.

transfer limit geometry of the acceptor; and, (2) the mode of intermolecular self-association in the solid state. Detailed comparisons of these derived properties with those of other members of this series of fluorinated TCNQ derivatives are given.

EXPERIMENTAL

The 2,5-TCNQF2 utilized in this study was synthesized according to the published procedure.[7] Crystals were grown by total evaporation of an acetonitrile (distillation purified) solution. A single crystal was found with some difficulty as many specimens were poorly formed or twinned (with (010) as the twin plane). Preliminary work indicated the crystal system to be monoclinic and the space group to be C2, Cm or C2/m (systematic absences: hkl, $h + k = 2n + 1$). The crystal density, 1.53(1) g cm^{-3}, was measured by the neutral buoyancy method in a mixture of cyclohexane and carbon tetrachloride. The calculated crystal density for $Z = 2$ (MW for $C_{12}N_4F_2H_2 = 240.17$) is 1.531 g cm^{-3}. We have assumed space group C2/m throughout our analysis; the presence of two formula units per cell requires that the molecules lie at position of site symmetry 2/m(C_{2h}) − the full symmetry of the 2,5-TCNQF2 molecule.

The yellowish-orange crystal chosen for intensity measurements was polyhedral in habit with the following face assignments and mean interfacial distances: (010)–($0\bar{1}0$), 0.12 mm; (001)–($00\bar{1}$), 0.15 mm; (110)–($\bar{1}\bar{1}0$), 0.35 mm. They crystal was mounted with the [110] direction approximately along the ϕ axis of a Syntex $P\bar{1}$ automated diffractometer. Precise unit-cell dimensions and their associated standard deviations were obtained from a least-squares fit to the setting angles of 15 carefully-centered reflections: $a = 10.208(4)$ Å, $b = 6.026(2)$ Å, $c = 8.836(3)$ Å, $\beta = 106.64(3)°$, $V = 520.8(3)$ Å^3.

The intensities of a total of 1399 reflections in the +h hemisphere to $2\theta = 55°$ were measured using graphite monochromatized Mo Kα radiation ($\bar{\lambda} = 0.71069$ Å) and the θ–2θ scan technique [scanning rate = 1.5°/min in 2θ]. Three standards were checked after every 100 reflections; the intensities of these standards showed no systematic variation over the course of the experiment. The measured intensities were assigned observational variances based on counting statistics plus a term $(pI)^2$, where p was taken to be 0.03 and represents an estimate of the error proportional to the diffracted beam intensity.[24] Lorentz and polarization corrections were applied, but no correction for absorption effects was deemed necessary (maximum and minimum transmission factors were estimated to be 0.98 and 0.96, respectively). Symmetry averaging of the data yielded an R value on averaging[25] of 0.014 and a set of 631 non-zero intensities (out of a possible 663 reflections). The data were subsequently placed on an absolute scale by the method of Wilson.[26]

In the assumed space group C2/m, the 2,5-TCNQF2 molecules are rig-

orously planar and lie perpendicular to the *b* axis. The only molecular degree of freedom is the orientation of the molecule within the (010) plane. Analysis of a Patterson synthesis by Vector Superposition methods readily yielded the molecular orientation and the positions of all but the F and H atoms of the molecule. A subsequent structure factor-Fourier synthesis allowed the positioning of the unique F atom; later in the analysis, the hydrogen atom was located by a difference-Fourier synthesis. This model was refined by full-matrix least squares methods, minimizing the quantity $\Sigma w(|F_o| - |F_c|)^2$ where $w = 4F_o^2/\sigma^2(F_o^2)$ and employing anisotropic thermal parameters for the nonhydrogen atoms and isotropic thermal parameters for the H atom. The final R value was 0.054.[27] The final weighted R value and goodness-of-fit parameter were 0.040 and 1.9, respectively.[27] The largest residuals on a final difference-Fourier map were ± 0.25 e/Å^3.

Neutral scattering factors for the nonhydrogen atoms[28] and the hydrogen atom[29] were taken from common sources. The scattering curves for the nonhydrogen atoms were corrected for anomalous dispersion effects.[30] A rigid-body analysis of the parameters of the 2,5-TCNQF2 molecule was performed employing the method of Schomaker and Trueblood.[31] The RMS difference between the least squares and rigid-body thermal parameters was 0.0028, indicating that the 2,5-TCNQF2 molecule can effectively be treated as a rigid body. We have employed the librational tensor from the rigid-body analysis to obtain corrected positional coordinates and derived bond lengths and angles (*vide infra*). Uncorrected positional parameters and average thermal parameters are given in Table I. Tables of nonhydrogen thermal parameters and observed and calculated structure factor amplitudes have been deposited. The X-ray analysis was performed with a standard set of computer programs.[33]

Table I

Final nonhydrogen atom ($\times 10^4$) and hydrogen atom ($\times 10^3$) positional parameters† and equivalent isotropic thermal parameters for neutral 2,5-TCNQF2.

Atom	x	y	z	$B_{eq.}$
F	2586(1)	0	9762(1)	4.5
N(1)	4744(2)	0	2984(2)	5.2
N(2)	1696(2)	0	5494(2)	4.6
C(1)	1299(2)	0	9854(2)	3.1
C(2)	9726(2)	0	1471(2)	3.1
C(3)	1122(2)	0	1417(2)	2.9
C(4)	2182(2)	0	2791(2)	3.2
C(5)	3607(2)	0	2868(2)	3.7
C(6)	1912(2)	0	4300(2)	3.4
H	952(2)	0	246(2)	2.8

† Estimated standard deviations of the least-significant figure are enclosed in parentheses.

RESULTS AND DISCUSSION

Crystal structure. The crystal structure of 2,5-TCNQF2 is depicted in the (010) projection of Figure 2. The restriction of the 2,5-TCNQF2 molecules to lie at sites of $2/m(C_{2h})$ symmetry in space group C2/m demands exact molecular planarity and leads to a structure consisting of rigorously planar sheets parallel to (010) and separated by 3.013 Å ($= |b|/2$). Two aspects of the intermolecular interactions in the structure of 2,5-TCNQF2 are of particular interest. Firstly, there is an intralayer interaction of the type C≡N---H—C which connects molecules separated by one translation along *c*. The H---N distance in this interaction is 2.47 Å and falls short of the sum of the van der Waals radii (Σ vdW)[34] of H and N by 0.23 Å. As the hydrogen atom involved in this contact probably bears a small positive charge (owing to the inductive influence of the fluoro substituent, *vide infra*) and the cyano N atom surely bears a significant negative charge, we classify this interaction as hydrogen bond-like in character.

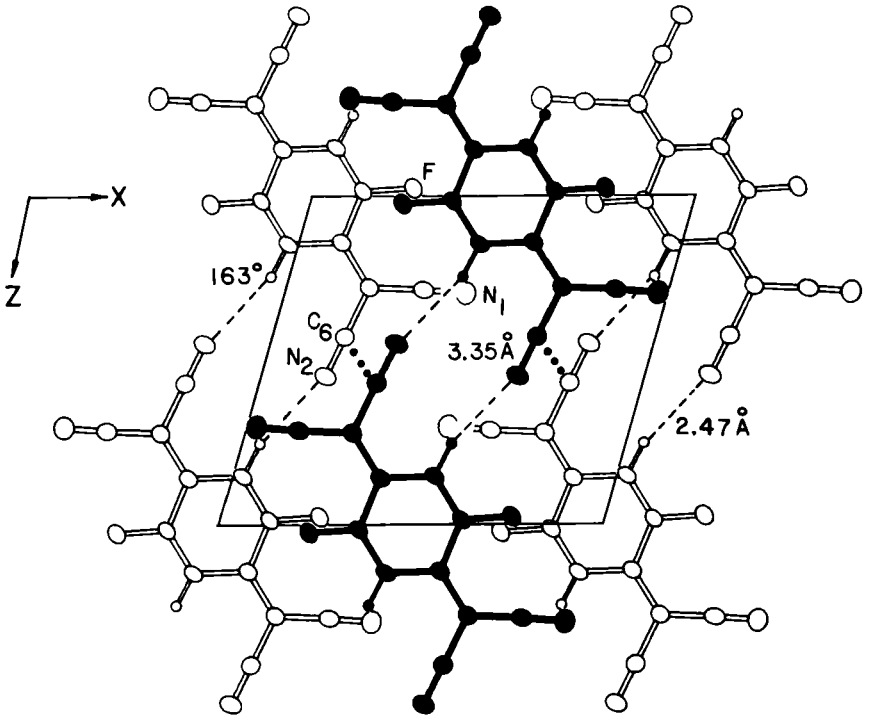


FIGURE 2 The (010) projection of the crystal structure of 2,5-TCNQF2. The shaded molecules lie in the plane at $y = \frac{1}{2}$; the unshaded molecules lie in the plane at $y = 0$. Dotted lines indicate the intermolecular antiparallel coupling of cyano bond moments, while the dashed lines denote intermolecular interactions of the type CH---N, see the text.

Secondly, interlayer extension is dominated by the antiparallel coupling of cyano bond moments, Figure 2, in a pattern similar to that shown by the crystal structure of TCNQF.[21] Contact distances for the coupling of cyano moments in the structure of 2,5-TCNQF2 are C···C = 3.35 Å (Σ vdW = 3.50 Å) and C···N = 3.31 Å (Σ vdW = 3.25 Å); we emphasize that these contact distances are *identical* to those shown by the antiparallel cyano coupling in the crystal structure of TCNQF.[21] As clearly shown in Figure 3, there is, however, a notable difference in the geometrical arrangement of alternate molecules in the crystal structures of TCNQF and 2,5-TCNQF2. The coupling of local moments in the crystal structure of TCNQF involves an interleaving of molecules, Figure 3, while that in the crystal structure of 2,5-TCNQF2 does not. Each of these modes apparently offers the same degree of local moment interaction as indicated by the equivalence of the contact distances. The mode present in the structure of TCNQF allows, however, a closer distance between

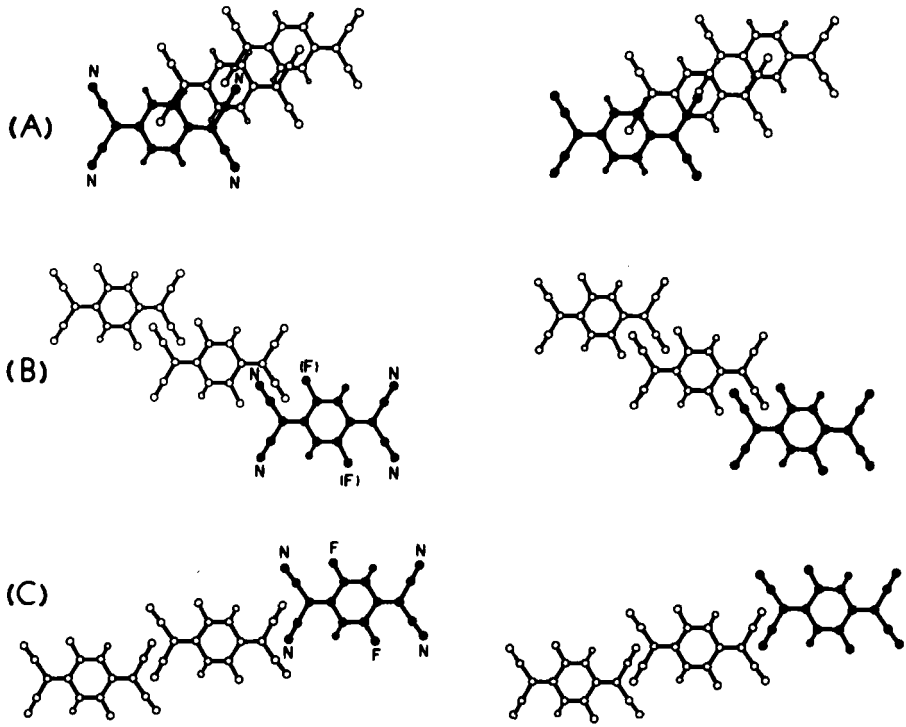


FIGURE 3 *Stereoviews of (A) the intermolecular overlap in the columns found in the crystal structure of TCNQ (intermolecular spacing, D = 3.45 Å), (B) the mode of intermolecular coupling of cyano bond moments in the twinned crystal structure of TCNQF (D = 3.07 Å), and (C) the mode of intermolecular coupling of cyano bond moments in the crystal structure of 2,5-TCNQF2 (D = 3.01 Å).*

molecular centers (8.20 Å vs. 9.35 Å for 2,5-TCNQF2). The coupling scheme chosen in the crystal structure of TCNQF presumably arises out of a desire to minimize the intermolecular separation and therefore maximize the intermolecular coupling of permanent electric dipole moments of the TCNQF molecules.

Finally, we note that the crystal structures of TCNQ,[23] TCNQF,[21] and 2,5-TCNQF2 all contain arrays of parallel molecules, Figure 3, and all rely to varying degrees on the presence of local cyano bond moments for crystal stability. The antiparallel cyano-cyano coupling in the crystal structures of TCNQF and 2,5-TCNQF2 have been described above. In the crystal structure of TCNQ, one finds (Figure 3) columns of molecules interacting *via* the cyano bond moment and the polarizable quinone system. Only in the crystal structure of TCNQF4[11] does one find the absence of parallel, interacting molecules. In that system, the principal intermolecular forces are acid-base (donor/acceptor) in character and appear to maximize at an interplanar angle near 90°.[11]

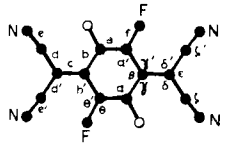
Molecular geometry. The molecular topology and derived bond lengths and bond angles for the 2,5-TCNQF2 molecule are presented in Figure 4. In Table II, we report the averaged, libration-corrected[35] molecular geometry for 2,5-TCNQF2 and compare it to the corrected molecular geometries of TCNQ and TCNQF4.[36]

The bond lengths in the 2,5-TCNQF2 molecule are, in general, very similar to those found in TCNQ[23] and TCNQF4.[11] The lone exception is the interior C=C bond (labelled a in Table II) which is noticeably shorter (0.017 Å) in 2,5-TCNQF2 than in TCNQ and slightly shorter (0.007 Å) than in TCNQF4. We expect that this shortening is primarily due to the influence of the fluoro substituents on the sigma orbitals of the quinone ring system.

It is obvious, however, from Table II that the effect of the fluoro substituents is more prominently displayed in the intramolecular bond angles. Quantitatively, the intramolecular angles can be divided into two categories based on the reduction in symmetry (D_{2h} to C_{2h}) on going from TCNQ or TCNQF4 to 2,5-TCNQF2: a) those angles for which two chemically independent values are anticipated (α and α', for example, of Table II); and, b) those for which only one independent value is expected (β, for example, of Table II). For bond angles of the first type, one uniformly finds that the primed angle is ca. 4° larger than the unprimed angle. For α and α', we believe this results primarily from a rehybridization of the C atom atomic orbitals owing to the electronic effect of the bonded F atom. For γ and γ' and δ and δ', the rehybridization of the atomic orbitals of the respective C atoms is more likely related to the steric influence of the fluoro substituents. Interestingly, for bond angles of the second type (β and ϵ), one finds a clearly discernible trend on going from TCNQ

Table II

Comparison of libration corrected molecular geometries for TCNQ (D_{2h} symmetry), 2,5-TCNQF2 (C_{2h}) and TCNQF4 (D_{2h})



Bond Length[a]	TCNQ	2,5-TCNQF2	TCNQF4
a	$1.346_{(1)}$Å	$1.329_{(1)}$Å	$1.336_{(1)}$Å
b	$1.448_{(2)}$	$1.445_{(1)}$	$1.442_{(2)}$
b′		$1.448_{(1)}$	
c	$1.374_{(1)}$	$1.377_{(1)}$	$1.374_{(1)}$
d	$1.441_{(2)}$	$1.442_{(1)}$	$1.443_{(2)}$
d′		$1.441_{(1)}$	
e	$1.140_{(2)}$	$1.140_{(1)}$	$1.143_{(2)}$
e′		$1.138_{(1)}$	
f		$1.342_{(1)}$	$1.338_{(2)}$
Bond Angle[a]	**TCNQ**	**2,5-TCNQF2**	**TCNQF4**
α	$120.8_{(2)}°$	$120.5_{(1)}°$	$123.1_{(2)}°$
α'		$124.0_{(1)}$	
β	$118.3_{(1)}$	$115.5_{(1)}$	$113.7_{(1)}$
γ	$120.8_{(2)}$	$120.4_{(1)}$	$123.2_{(2)}$
γ'		$124.1_{(1)}$	
δ	$121.9_{(2)}$	$120.4_{(1)}$	$123.7_{(2)}$
δ'		$124.8_{(1)}$	
ϵ	$116.1_{(1)}$	$114.8_{(1)}$	$112.6_{(1)}$
ζ	$179.5_{(2)}$	$179.9_{(1)}$	$175.2_{(2)}$
ζ'		$177.6_{(1)}$	
θ		$119.0_{(1)}$	$118.6_{(2)}$
θ'		$117.0_{(1)}$	$118.2_{(2)}$

[a] Subscripts indicate the number of chemically-independent bond lengths or bond angles contained in the average value quoted.

to 2,5-TCNQF2 to TCNQF4: as the number of fluoro substituents increases, β and ϵ monotonically decrease.

Less certainty can be attached to the variation in the angles ζ and ζ' as they are very susceptible to environmental effects. Although, we do note that these C—C≡N bond angles are farthest from linearity in TCNQF4, which may again reflect the steric influence of the fluoro substituents.

Finally, we comment on the relatively high symmetry shown by 2,5-TCNQF2 in its crystal structure. In most cases,[37] TCNQ-related molecules occupy sites having symmetry no higher than $\bar{1}(C_i)$. It is even rarer that these molecules take on their full gas phase symmetry, particularly the rigorously planar structure demanded for 2,5-TCNQF2. We expect that the site symme-

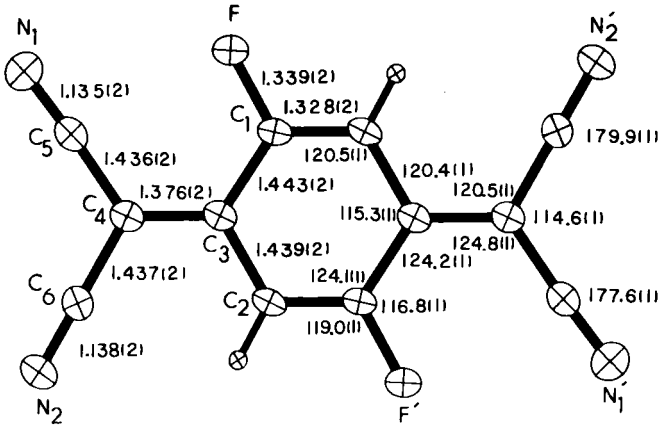


FIGURE 4 Intramolecular bond lengths and bond angles for the 2,5-TCNQF2 molecule as observed in its crystal structure.

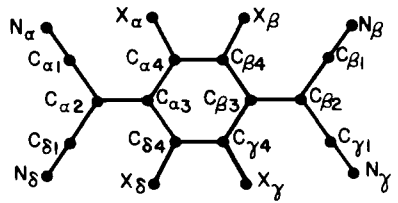
try displayed by the 2,5-TCNQF2 molecule is a direct result of the intermolecular interactions dictated by the layered nature of its crystal structure.

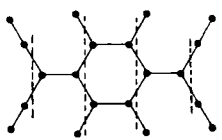
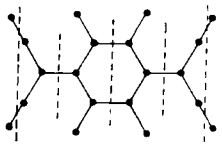
Electronic structure. Knowledge of the electronic structure of TCNQ and its derivatives is central to an understanding of the charge-transfer chemistry of such electron acceptors. In an attempt to give a qualitative picture for 2,5-TCNQF2, we have examined its electronic structure utilizing the INDO molecular orbital approximation[38] and the observed crystalline molecular geometry. In Figure 5, we illustrate three results of the INDO computation for 2,5-TCNQF2: the molecular orbital coefficients for the π-HOMO [symmetry a_u in point group C_{2h}] and the π-LUMO [symmetry b_g] and the partial atomic charges derived from a Mullikan population analysis.

These results are best appreciated in a comparative sense and, thus, we present in Table III a comparison of the π-HOMO and π-LUMO coefficients for TCNQ,[39] 2,5-TCNQF2, and TCNQF4.[40] As expected[41] the progressive substitution of the quinone ring H atoms by F atoms does not have a large effect on the π-HOMO and π-LUMO coefficients. There are, however, small, but systematic, changes in the energies of the π-HOMO and π-LUMO on going from TCNQ through 2,5-TCNQF2 to TCNQF4. In particular, the steady increase in the binding character of the π-LUMO along the series expectedly parallels the progression in electron affinities, Table IV. The most noticeable effect of the fluoro substituents is in the charge distribution; for 2,5-TCNQF2, the F atoms have negative charges rivaling those of the cyano N atoms. Similarly, the fluorine substituted quinone ring C atoms bear reasonably high positive charges, Figure 5.

Table III

Comparison of the π-frontier molecular orbitals for TCNQ, 2,5-TCNQF2 and TCNQF4



Highest Occupied Molecular Orbital		Lowest Unoccupied Molecular Orbital
	Nodal Planes	
Coefficient Comparison		Coefficient Comparison

Atom	TCNQ[a] ($X_\alpha - X_\delta = H$)	2,5-TCNQF2 (X_α, $X_\gamma = F$; X_β, $X_\delta = H$)	TCNQF4 ($X_\alpha - X_\delta = F$)	Atom	TCNQ[a] ($X_\alpha - X_\delta = H$)	2,5-TCNQF2 (X_α, $X_\gamma = F$; X_β, $X_\delta = H$)	TCNQF4 ($X_\alpha - X_\delta = F$)
N_α	0.22	0.25	0.24	N_α	0.19	0.23	0.23
N_β	0.22	0.24	0.24	N_β	−0.19	−0.23	−0.23
N_γ	0.22	0.25	0.24	N_γ	−0.19	−0.23	−0.23
N_δ	0.22	0.24	0.24	N_δ	0.19	0.23	0.23
$C_{\alpha 1}$	0.04	0.07	0.06	$C_{\alpha 1}$	−0.07	−0.06	−0.06
$C_{\beta 1}$	0.04	0.06	0.06	$C_{\beta 1}$	0.07	0.07	0.06
$C_{\gamma 1}$	0.04	0.07	0.06	$C_{\gamma 1}$	0.07	0.06	0.06
$C_{\delta 1}$	0.04	0.06	0.06	$C_{\delta 1}$	−0.07	−0.07	−0.06
$C_{\alpha 2}$	−0.47	−0.43	−0.42	$C_{\alpha 2}$	−0.44	−0.44	−0.44
$C_{\beta 2}$	−0.47	−0.43	−0.42	$C_{\beta 2}$	0.44	0.44	0.44
$C_{\alpha 3}$	−0.21	−0.23	−0.23	$C_{\alpha 3}$	0.30	0.27	0.26
$C_{\beta 3}$	−0.21	−0.23	−0.23	$C_{\beta 3}$	−0.30	−0.27	−0.26
$C_{\alpha 4}$	0.25	0.22	0.25	$C_{\alpha 4}$	0.26	0.26	0.23
$C_{\beta 4}$	0.25	0.27	0.25	$C_{\beta 4}$	−0.26	−0.22	−0.23
$C_{\gamma 4}$	0.25	0.22	0.25	$C_{\gamma 4}$	−0.26	−0.26	−0.23
$C_{\delta 4}$	0.25	0.27	0.25	$C_{\delta 4}$	0.26	0.22	0.23
X_α	0	−0.10	−0.11	X_α	0	−0.08	−0.07
X_β	0	0	−0.11	X_β	0	0	0.07
X_γ	0	−0.10	−0.11	X_γ	0	0.08	0.07
X_δ	0	0	−0.11	X_δ	0	0	−0.07

[a] Taken from ref. 39.

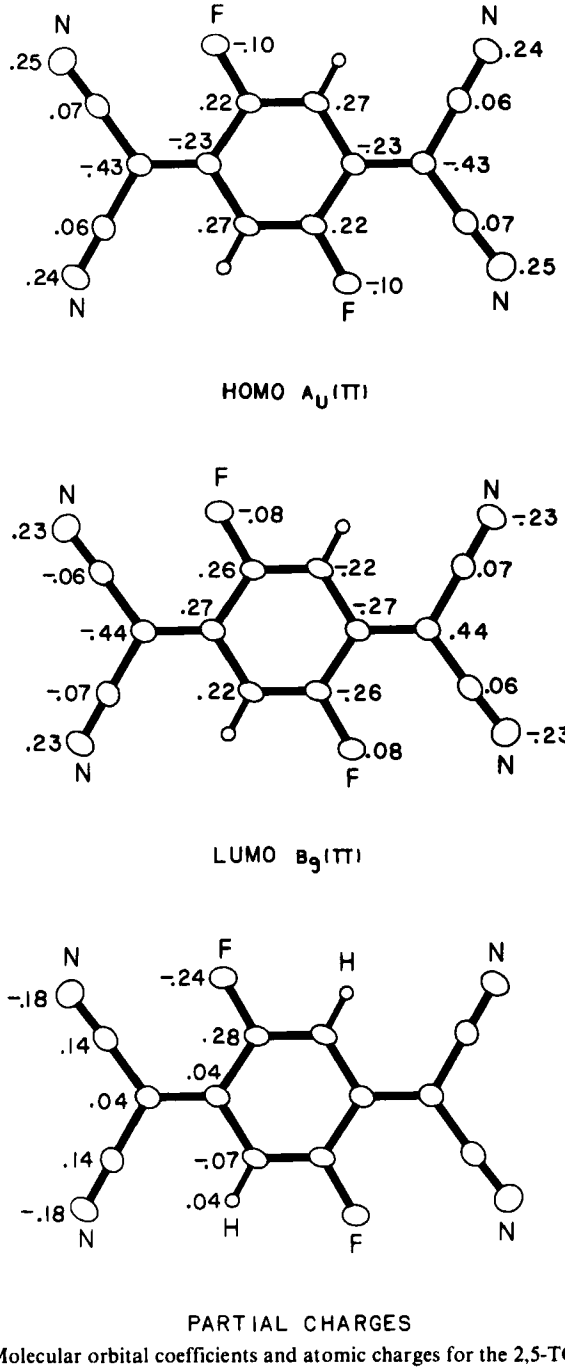


FIGURE 5 Molecular orbital coefficients and atomic charges for the 2,5-TCNQF2 molecule obtained from an INDO LCAO-MO analysis.

Table IV

Comparison of frontier molecular orbital energies (eV) and electron affinities (eV) for TCNQ, 2,5-TCNQF2 and TCNQF4

Acceptor	E (HOMO)	E (LUMO)	Electron Affinity[a]
TCNQ	−9.54	−0.74	2.85
2,5-TCNQF2	−9.87	−1.09	3.02
TCNQF4	−10.03	−1.48	3.20

[a]Taken from ref. 11.

Some of the consequences of the variation in the electronic structures of TCNQ and its fluorinated derivatives are expected to be the following:

(1) Charge-transfer salts of 2,5-TCNQF2 and TCNQF4 will usually have higher degrees of charge transfer than analogous TCNQ salts owing to their higher electron affinity (more favorable π-LUMO energies). As noted in the Introduction, experimental results bear out this contention.

(2) Changes in molecular geometry owing to the transfer of electron density into the π-LUMO of the acceptor will be similar. Empirical data thus far support comparable bond length variations for unit and fractionally-charged salts of TCNQ, 2,5-TCNQF2 and TCNQF4.[3-5,8,12,20]

(3) Arguments based on π-banding effects (e.g., electrical, magnetic and spectral anisotropies) in low-dimensional salts of TCNQ should be essentially applicable to 2,5-TCNQF2 and TCNQF4 salts with similar structural motifs and charge-transfer integrals.

(4) Charge redistribution almost certainly places the N---H—C intralayer interaction in the crystal structure of 2,5-TCNQF2 and the ubiquitous N···C—F interactions in the crystal structure of TCNQF4 among the key intermolecular interactions in those systems. Similarly, the presence of the C—F bond moment and its contribution to the permanent molecular moment of TCNQF is instrumental in determining its crystalline motif.[21-22]

SUMMARY

We have described in detail above the crystal, molecular and electronic structure of the acceptor 2,5-TCNQF2 and made comparisons with TCNQ and its extant fluorinated analogs. Noteworthy for 2,5-TCNQF2 are its high molecular symmetry in the solid, its intermediate electronic properties, and the inclusion in its crystal structure of elements found in the crystal structures of other fluorinated derivatives.

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Supplementary material

Tables of atomic thermal parameters and observed and calculated structure factor amplitudes have been deposited.

This material may be obtained by contacting Gordon & Breach, One Park Avenue, New York, NY 10016, attn. P. Bardi, Ref. No. MCLC. . .

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$$R_{AVE} = \sum_{i=1}^{N} \sum_{j=1}^{n} |F_{ij}^2 - \bar{F}_i^2| / \sum_{i=1}^{N} \bar{F}_i^2,$$

where N = the total number of unique observations and n = number of times each observation was measured.
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b) $R_w = \{\Sigma w(|F_o| - |F_c|)^2/\Sigma w|F_o|^2\}^{1/2}$
c) $GOF = \{\Sigma w(|F_o| - |F_c|)^2/(NO - NV)\}^{1/2}$, where NO = 631 observations and NV = 58 parameters varied.
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