

Supporting Information

Electric and Thermosolient Properties of a Charge Transfer Complex Exhibiting a Minor Valence Instability Transition

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1. DSC with using polycrystalline sample

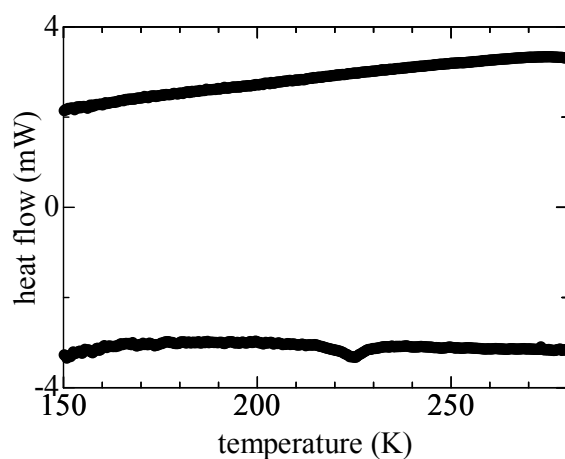
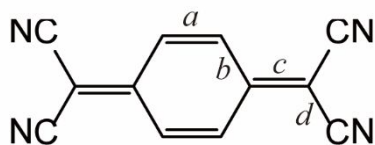


Figure S1. Heat flow of TMB-TCNQ crystals. The peak of the cooling process is obscured in comparison with the DSC using the well-ground sample adopted in the main text. This is caused by the crystals jumping in the sample holder.

2. CT degree in TMB-TCNQ



$$\rho = 22.43 - 23.81 (a + c)/(b + d)$$

Figure S2. The relation between CT degree (ρ) and molecular geometry of TCNQ. CT degree of CT complexes containing TCNQ molecule are able to be estimated from the bond lengths of the C=C (a and c) and C-C (b and d) in the TCNQ skeleton [S1]. The ρ in HT and LT phase were 0.24 and 0.36 respectively, according to the bond length summarized in Table S1.

Table S1. The bond length in TCNQ skeleton

	HT	LT
a	1.3436(15)	1.351(3), 1.348(3) average: 1.3495
b	1.4364(14), 1.4329(14) average: 1.43465	1.436(3), 1.435(3), 1.437(3), 1.433(3) average: 1.43475
c	1.3804(15)	1.390(3), 1.389(3) average: 1.3895
d	1.4232(15), 1.4258(15) average: 1.4245	1.427(3), 1.425(3), 1.431(3), 1.421(3) average: 1.426

[S1] Kistenmacher, T. J.; Phillips, T. E.; Cowan, D. O., The crystal structure of the 1:1 radical cation–radical anion salt of 2,2'-bis-1,3-dithiole (TTF) and 7,7,8,8-tetracyanoquinodimethane (TCNQ). *Acta Crystallogr., Sect. B: Struct. Crystallogr. Cryst. Chem.* **1974**, *30*, 763-768.