# Magnetic and/or Conducting Complexes with π-Acceptor Ligands DCNQIs and ortho-Analogues DCNAs

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*N*,*N*'-dicyano-p-quinonediimines (DCNQIs) exhibit unique features such as strong π–d interactions in their Cu salts through tetrahedral coordination of the terminal cyanoimino (=N–C≡N) groups [1]. Aiming for new structures and properties different from those of DCNQIs, we have developed ortho-analogues, *N*,*N*'-dicyano-acenaphthoquinonediimines (DCNAs), in which the two cyanoimino groups occupy positions next to each other [2]. We also prepared AMNN, which is the $Me_4N^+$ ion bearing nitronyl nitroxide (NN) radical in a methyl group, as a counter cation of acceptor-based conductors in order to construct novel molecular systems with cooperation between conducting and magnetic electrons. In this paper, we deal with (1) the anion radical salts of DCNQI or DCNA with the organic magnetic cation AMNN, (2) the salts with the transition metal cations, and (3) complexes of a neutral DCNA with the magnetic metal ions.

Black needles were obtained by electrochemical reductions of DCNQIs in the presence of the $PF_6^-$ salt of AMNN. They were low conductivity ($\sigma_{RT}$ = $10^{-4}$ S/cm) and almost non-magnetic, suggesting undesired reduction of AMNN. Chemical reductions of DCNQI or DCNA using the halide ion as reducing reagent are now examined to obtain the magnetic salts. Black tiny needles or powders were obtained by reduction of DCNA or DBr-DCNA with the metal salts of Li, Cu or Co. Among them, the Li-DCNA salts show semiconductive behavior with $\sigma_{RT}$ = $10^{-5}$ S/cm. Metal complexes were obtained by reaction between DCNA or DF-DCNA and $M(hfac)_2$ (M = Mn, Co, Ni or Cu) as green or yellow powder. IR, UV-Vis and CHN combustion analysis of the Cu complex suggest one-sided coordination of the bidentate DCNA to form $[Cu(DCNA)_2(hfac)_2]$, and preliminary magnetic measurement shows weak antiferromagnetic interactions ($\theta = -1.5$ K).

We found that DCNAs and AMNN are new building blocks for magnetic and/or conducting complexes, and continue to search for novel materials using them.

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