Spectroscopic data and complex-formation equilibrium constants are reported for some two dozen charge-transfer complexes of tetracyanoethylene (TCNE) with a variety of electron donors—mostly aromatic hydrocarbons. TCNE is found to be the strongest pi-electron acceptor yet reported. [The SCF indicates that this paper has been cited in over 405 publications since 1958.]

This paper was the second of a group of 12 papers with a total of 21 different authors that were published in the same issue of the Journal of the American Chemical Society. These papers announced the synthesis of tetracyanoethylene (TCNE), the first example of a percyanoolefin, and reported results on the complexes, the principal conclusion of this work was that TCNE was by far the strongest pi-electron acceptor examined up to that time.

Although research on cyanocarbon chemistry continued at DuPont for several more years, Phillips and I both moved on to other areas after the work reported in this paper—he to nuclear magnetic resonance and I to the physics of molecular crystals. We were both to return to the subject of cyanocarbons some time later with the discovery of tetracyanoquinodimethane (TCNQ) and its highly conducting ion-radical salt. I am not sure why this paper has been highly cited. Neither the experimental methods employed nor the theoretical interpretations invoked were particularly novel. Most likely it is because of the unique nature of TCNE and the unusual properties of cyanocarbons in general.