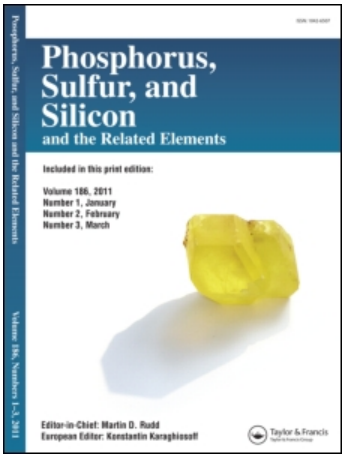

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Ohki Sato[a]; Makoto Sato[a]; Takaaki Kuramochi[a]; Takashi Shirahata; Kazuko Takahashi
[a] Department of Chemistry, Faculty of Science,Saitama University, Japan

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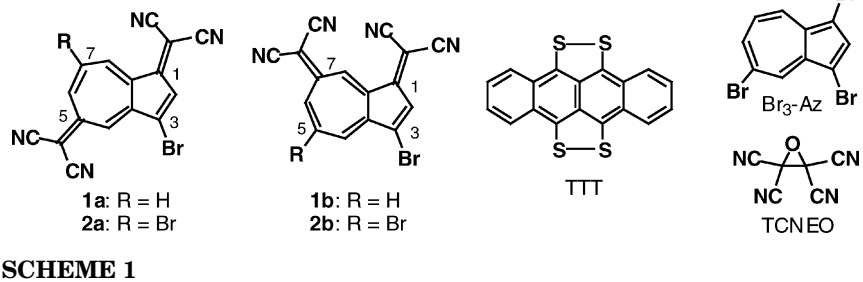
Synthesis and Conducting Properties of Tetracyanoazulenequinodimethane-Tetrathiotetracene Complexes

Ohki Sato
Makoto Sato
Takaaki Kuramochi
Department of Chemistry, Faculty of Science, Saitama University, Saitama, Japan

Takashi Shirahata
RIKEN

Kazuko Takahashi
Tohoku University

Tetracyanoquinodimethane (TCNQ) and their derivatives have proved to be the most impotant π-acceptor molecules for organic conductors. We are interested in new nonbenzenoid TCNQ-type acceptors, and now synthesized tetracyanoazulenequinodimethanes (TCNAzQDMs: **1a,b** and **2a,b**), and have clarified conducting properties of their tetrathiotetracene (TTT) complexes, which are reported herein. Synthesis of the new acceptors was carried out by the reaction of 1,3,5-tribromoazulene (Br_3-Az) with tetracyanoethylene oxide (TCNEO), affording



SCHEME 1

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Address correspondence to Ohki Sato, Department of Chemistry, Faculty of Science, Saitama University, Shimo-okubo 255, Saitama 338-8570, Japan. E-mail: ohkisato@chem.saitama-u.ac.jp

TABLE I Appearances, Conductivities, Activation Energy, and IR Data of TCNAzQDM-TTT Complexes

Complex	Appearance	$\sigma_{r.t.}$ S · cm^{-1}	E_a/eV	ν_{CN}/cm^{-1}
[**1a**][TTT]$_2$	Black powder	3.0×10^{-6}		2205
[**1b**][TTT]$_2$ [TCB]$_{0.5}$	Black powder	5.3×10^{-2}	9.9×10^{-2}	2208
[**2b**] [TTT]	Black violet fine needles	3.8×10^{-3}		2196

3-bromo-TCN-1,5-AzQDM **1a**, -1,7-AzQDM **1b** and their dibromo derivatives **2a,b** in 14, 14, 4, and 4% yields, respectively.

Preparation of charge-transfer complexes was performed by the direct reactions of **1a** and **1b** with TTT in 1,2,4-trichlorobenzene (TCB). The obtained [**1b**][TTT]$_2$[TCB]$_{0.5}$ showed a room-temperature conductivity of 5.3×10^{-2} S · cm^{-1}, which is 10^4 times higher than that of [**1a**][TTT]$_2$.

The reaction of **2b** with TTT in 1,2,4-TCB afforded [**2b**][TTT] complex as black violet fine needles. From the X-ray analysis, mixed-stacks of **2b** and TTT molecules were revealed.