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Synthesis and Conducting Properties of Tetracyanoazulenequinodimethane- Tetrathiotetracene Complexes

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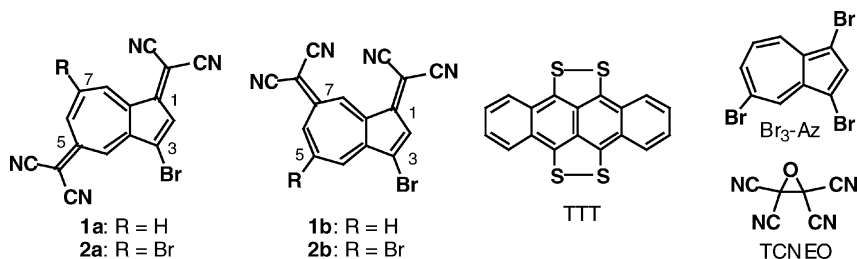
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Tetracyanoquinodimethane (TCNQ) and their derivatives have proved to be the most important π -acceptor molecules for organic conductors. We are interested in new nonbenzenoid TCNQ-type acceptors, and now synthesized tetracyanoazulenequinodimethanes (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 ($\text{Br}_3\text{-Az}$) with tetracyanoethylene oxide (TCNEO), affording



SCHEME 1

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

Complex	Appearance	$\sigma_{\text{r.t.}} \cdot \text{S} \cdot \text{cm}^{-1}$	E_{a}/eV	$\nu_{\text{CN}}/\text{cm}^{-1}$
[1a][TTT] ₂	Black powder	3.0×10^{-6}		2205
[1b][TTT] ₂	Black powder	5.3×10^{-2}	9.9×10^{-2}	2208
[TCB] _{0.5}				
[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 $5.3 \times 10^{-2} \text{ S} \cdot \text{cm}^{-1}$, 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.