



Multiple-Bridged Bis-Tetrathiafulvalenes: New Synthetic Protocols and Spectroelectrochemical Investigations

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Résumé en anglais

Synthetic strategies for preparing dimeric tetrathiafulvalenes (TTFs) linked by either one, two, or four bridges have been developed. In particular, we report efficient few-step protocols for the preparation of face-to-face overlapped quadruple-bridged bis-TTFs. The ready interconversion of *cis* and *trans* TTFs in the presence of catalytic amounts of acid was implemented in one synthetic protocol as a way to control the isomeric outcome. The compounds were characterized by NMR spectroscopy, mass spectrometry, and elemental analysis. Moreover, the X-ray crystal structure of the macrocycle **4b** is presented and compared to semiempirical (PM3) geometry optimizations. Cyclic voltammetry and spectroelectrochemistry were used to describe the interactions established between two TTF units upon oxidation, that is, their ability to form mixed-valence complexes and π -dimers either intra- or intermolecularly. The length, flexibility, and number of bridging units in a bis-TTF, as well as the specific TTF positions being connected, determine the extent of these interactions. Thus, rigid linkers enhance the formation of intermolecular mixed-valence complexes. For **4b**, the absorption spectrum of this mixed-valence state of TTF in solution has been recorded for the first time. Finally, preliminary complexation experiments with different electron-deficient molecules are described.

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