

Bandgap engineering in π -extended pyrroles : a modular approach to electron-deficient chromophores with multi-redox activity.

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Rok wydania

2016

Czasopismo

Journal of the American
Chemical Society

Numer woluminu

138

Strony

11390-11398

DOI

10.1021/jacs.6b07826

Kolekcja

Naukowa

Język

Angielski

Typ publikacji

Artykuł

Streszczenie

A family of bandgap-tunable pyrroles structurally related to rylene dyes was computationally designed and prepared using robust, easily scalable chemistry. These pyrroles show highly variable fluorescence properties and can be used as building blocks for the synthesis of electron-deficient oligopyrroles. The latter application is demonstrated through the development of π -extended porphyrins containing naphthalenediamide or naphthalenediimide units. These new macrocycles exhibit simultaneously tunable visible and near-IR absorptions, an ability to accept up to 8 electrons via electrochemical reduction, and high internal molecular free volumes. When chemically reduced under inert conditions, the most electron-deficient of these macrocycles revealed reversible formation of eight charged states, characterized by remarkably red-shifted optical absorptions, extending beyond 2200 nm. Such features make these oligopyrroles of interest as functional chromophores, charge-storage materials, and tectons for crystal engineering.

Adres publiczny

<http://dx.doi.org/10.1021/jacs.6b07826>

Strona internetowa wydawcy

<https://www.acs.org/content/acs/en.html>