Perylene Diimide-Derived Bis(Macrocyclic) Ligand Affords Chiral Multinuclear Pd(II) Complexes via Direct Bay-Region Palladation

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Streszczenie

Macrocyclic ligands provide powerful access to structurally defined multi-metallic architectures, yet direct π-core metalation of imide-functionalized polycyclic aromatic dyes remains difficult to control. Herein, we present the design and synthesis of a new nitrogen-rich bis(macrocyclic) ligand (**1**) based on a perylene diimide (PDI) scaffold. Coordination of Pd(II) to this ligand triggers direct C—H palladation at the bay region of the PDI, yielding two distinct complexes—a bis-Pd and a tetra-Pd species (**1-Pd$_2$** and **1-Pd$_4$**)—each featuring asymmetrically coordinated, bridged Pd centers within the macrocyclic cavities. Furthermore, palladation distorts the PDI core and induces conformational chirality in the macrocyclic framework. Ultrafast transient absorption measurements further reveal systematic changes in the excited-state response across the series. In addition, OTTLE spectroelectrochemical studies show that direct palladation creates strongly redox-responsive low-energy optical states, with markedly enhanced near-IR absorption in the reduced Pd complexes. This work provides a feasible synthetic route for the direct metalation of perylene diimide dyes, enabling strong dye-metal electronic coupling and establishing such chiral multinuclear complexes as functionally tunable multi-metallic π-systems.

Słowa kluczowe

chirality, macrocyclic ligands, multi-metallic complexes, palladium, perylene diimide

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