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（博士課程）

論文審査の要旨及び審査員

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論文審査の要旨（2000 字程度）

This thesis describes the preparation of a novel photosensitizer system based on a tris(4-pyridyl) hexaazaphenalene (TPHAP) molecule and its derivatives for photocatalytic reactions. This thesis was deemed appropriate as a doctoral dissertation based on the impact and novelty of these research results. Likewise, the presented experimental methods and discussions were considered a suitable criterion. First, the thesis described the photoactivity of TPHAP molecule which showed two photochemical processes, namely energy transfer and charge transfer, upon light excitation in the presence of oxygen, as determined by spectroscopic analysis and selective photooxidation reactions to generate reactive oxygen species. Furthermore, the photocatalytic transformation of cyclohexane was carried out using the TPHAP molecule as a homogeneous photocatalyst and TPHAP-based porous coordination network as a heterogenous catalyst, which exhibited moderate catalytic performance and photostability. A reaction mechanism was proposed based on control experiments and trapping agents. TPHAP-based systems are expected to be promising alternatives for photocatalytic applications in several important transformations of hydrocarbon compounds. In the next part, the modification of the TPHAP molecule via the N-alkylation reaction of pyridine into pyridinium groups was performed in order to decrease the excitation energy. The crystal structures of these compounds revealed a plethora of additional secondary interactions. The electronic and electrochemical properties were interrogated by spectroscopy, voltammetry, and theoretical calculations. The change of excitation energy of new pyridinium-TPHAP analogues relative to the parent compound was explored in the last part. One of the resultant pyridinium TPHAP molecules and its coordination network were tested as a photocatalyst towards a photooxidation reaction selective for the singlet oxygen, displaying an improved catalytic activity due to better matching of the excitation energy. In addition to the three main research sections, comprehensive introduction and thesis summary were described, showing the background, state of the art research, and future directions for the field. Overall, this thesis qualifies to be a doctoral dissertation in terms of novelty to the photocatalysis field and also methodology guidelines aimed at improving the photocatalytic activity of the pyridine-based molecules.