

論文 / 著書情報
Article / Book Information

題目(和文)	多環芳香族骨格を有するポリカチオン性三次元ナノ構造体の構築
Title(English)	Construction of Polycationic Nanostructures with Well-Defined Polyaromatic Cavities
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Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

論文要旨

THESIS SUMMARY

専攻： 化学環境学 専攻
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申請学位 (専攻分野)： 博士 (工学)
Academic Degree Requested Doctor of
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Academic Advisor(main)
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要旨 (英文 800 語程度)

Thesis Summary (approx.800 English Words)

In a history of supramolecular chemistry, a variety of three-dimensional nanostructure with well-defined cavities have been designed and synthesized by using neutral organic frameworks. However, the incorporation of cationic moieties into the neutral frameworks of hollow structures has been seldom explored so far.

In this thesis, I developed new methods to construct polycationic nanostructures with polyaromatic panels. I utilized bent bispyridine ligands containing polyaromatic rings such as an anthracene and an acridinium panel. By using bent bispyridine ligands, I designed and prepared novel polycationic nanostructures with well-defined cavities. In addition, I revealed the function of the polyaromatic nanospace.

A synthesis of polyaromatic molecular bowl was accomplished by the use of two bent anthracene-embedded bispyridine ligands linked by two methylene spacers. The bowl-shaped structure was confirmed by NMR, ESI-TOF MS, and X-ray crystallographic analyses. The bowl provides a ~1 nm-sized hemi-spherical cavity surrounded by four anthracene panels and includes two dicationic (Lewis acidic) bis-pyridinium moieties. The molecular bowl has high water solubility because of the cationic units, and they bound carbonyl compounds through hydrophobic and hydrogen bonding interactions.

A polyaromatic molecular tube was prepared by using Zincke reaction of a diamino anthracene dimer and a Zincke salt of the bis-pyridine ligand. The X-ray single crystal analysis revealed that the tube has a 1 nm-sized cylindrical cavity encircled by four anthracene panels. Interestingly, the tube bound a variety of long hydrocarbons in aqueous solutions, and selectively bound branched and/or unsaturated hydrocarbons even without coordinative groups.

The quantitative formation of polycationic capsular and tubular structures was realized by using a new bent bispyridine ligand containing two acridinium rings as a

cationic moiety. The cationic ligand was prepared in three-step reactions. The 2:1 combination of the ligand and Pd(II) ions led to the quantitative formation of a dodeca-cationic M_2L_4 molecular capsule. On the other hand, mixing a 1:1 ratio of the ligand and Pd(II) ions resulted in the selective formation of deca-cationic M_2L_2 molecular tube. The two structures were interconvertible by addition of the ligands or metal ions.

An M_3L_4 double capsule, two capsular molecules linked in close proximity, was prepared by self-assembly of metal ions and new W-shaped tripyridine ligands. The W-shaped ligand is containing four anthracene panels linked by two *meta*-phenylene spacers. Simple combination of Pd(II) ions and the W-shaped ligand led to the quantitative formation of the M_3L_4 double capsule. The product provides two 1 nm-sized cavities fully covered by eight anthracene panels, and these cavities are located at a close distance. This double capsule encapsulated one or two neutral molecules in each cavity.

Through above works, I developed new synthetic methods for polycationic nanostructures with well-defined cavities encircled by multiple polyaromatic panels. The resultant structures showed distinctive properties due to their frameworks and shapes. These results would contribute to the development of a new methodology to design and construct functional three-dimensional nanostructures.

備考：論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。

Note: Thesis Summary should be submitted in either a copy of 2000 Japanese Characters and 300 Words (English) or 1 copy of 800 Words (English).

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