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学 位 論 文 題 名
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Synthesis of Transition Metal Substituted Pentacene Derivatives and Their Application

(遷移金属を置換基にもつペンタセン誘導体の合成及び応用研究)

博士学位論文審査等の結果について (報告)
Results of Evaluation of the Doctoral Dissertation (Report)

Recently, graphene ribbons have attracted much attention as next generation semiconductors. Several papers were reported for their synthesis. However, there is no report for pentacene based graphene ribbon. The pentacene oligomers were important precursors of pentacene based graphene ribbon.

In chapter 2, the author focused on the synthesis and characterization of palladated pentacene firstly. Oxidative addition of 6,13-dibromo-5,14-dihydropentacene to $\text{Pd}(\text{PPh}_3)_4$ was followed by aromatization. The corresponding central ring palladated pentacene was obtained as an unexpectedly stable complex. By the ligands exchanged reaction, the PPh_3 ligands could be changed to PMe_3 or PEt_3 . After aromatization, the corresponding palladated pentacene with PMe_3 or PEt_3 was prepared successfully. Moreover, a series of dipalladated pentacene complexes and mixed transition metals substituted pentacene complexes were prepared successfully.

In chapter 3, the author focused on development of a new method for introduction of substituents into pentacene from electrophiles and nucleophiles selectively. For the formation of substituted pentacene derivatives, many methods have been reported. In Takahashi's homologation method and coupling method, the substituents come from starting alkynes. As for pentacenequinone method and cross-coupling method, the substituents come from nucleophiles. In this part, introduction of substituents using Pd-substituted pentacenes will be reported. The advantage of this method is the substituents of these pentacene derivatives come from not only nucleophiles but also electrophiles.

In chapter 4, the author focused on investigation of the dimerization of pentacene. With the central ring palladated dihydropentacene in hand, the dimerization was studied. However, central ring side dimer of pentacene was not obtained by the cross-coupling reaction. Probably the steric hindrance of the central ring was too high. Second ring and terminal ring palladated pentacene derivatives were then prepared and used for dimerization reaction. However, the second ring dimer of pentacene was not obtained due to the bulkiness of two pentacene derivatives. Compared with central ring and second ring palladated pentacenes, the terminal ring palladated pentacene is less bulky. Interestingly, first ring side dimer of pentacene was obtained in high yield under the same reaction conditions.

In chapter 5, selective oligomerization of pentacene derivatives using platinum was developed. For formation of pentacene based graphene ribbons, the pentacene oligomers were important precursors. In chapter 4, the dimer of pentacene using first ring palladated pentacene derivative was prepared. However, the substituents of this dimer are opposite. This is because the same side of two pentacene derivatives coupled in the cross-coupling. In this chapter, by use of platinated pentacene, a new dimer was obtained. The substituents of this dimer are in the same

direction. Furthermore, this method could be applied for selective oligomerization of pentacene. For example, a pentacene trimer was prepared from the dimer successfully. The substituents of three pentacene moieties were aligned in the same direction.

In conclusion, the author has new findings on the synthesis of transition metals substituted pentacene derivatives and the selective oligomerization of pentacene derivative. These results contribute to prepare pentacene based graphene ribbon. Therefore, the author is qualified to be granted the Ph.D degree (Pharmaceutical Sciences) from Hokkaido University.