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Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

（博士課程）
Doctoral Program

論 文 要 旨

THESIS SUMMARY

専攻： Department of	Electronic Chemistry 専攻	申請学位（専攻分野）： Academic Degree Requested	博士 Doctor of (Engineering)
学生氏名： Student's Name	Iryane Binti Ismail	指導教員（主）： Academic Advisor(main)	Assoc. Prof. Fusao Kitamura
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要旨（英文 800 語程度）
Thesis Summary (approx.800 English Words)

Electrochemical behaviour of DA (dopamine) depends on solution pH, its concentration, potential scan rate etc. which are closely associated with the intracyclization reaction (ICR) and polymerization. In this thesis, electrochemical behaviour of DA has been studied experimentally and theoretically. The obtained results were used to support the proposed model of electrooxidation pathway of DA. Also DA-modified electrodes, in which DA is covalently bound to chemically modified electrodes, were successfully fabricated by amide bond formation through simple stepwise technique of diazonium reduction on electrode surface. This doctoral thesis is composed of 7 chapters.

In Chapter 1, the importance, outline and motivation of the present research are demonstrated.

In Chapter 2, electrochemical reaction of DA was investigated. At acidic pH, ICR did not occur because of the protonated amino group. ICR leads to formation of leucodopaminechrome/dopaminechrome couple, and the redox peak of this couple was easily observed from the first potential cycle in solutions of pH 6 and above. The electrochemical reaction of DA proceeds in an ECC mechanism at pH 9 and below. On the other hand, at pH above 10, ICR and the subsequent chemical reactions occur rapidly. Higher concentration of DA (i.e., 1 mM or more) led to faster polymerization reaction resulting in the polymerized DA adsorbed on electrode surface. A CEC mechanism proceeds at pH 10.

In Chapter 3, it was found that redox reaction of 5-MDA is simpler than that of DA at pH 9 and below and that ICR does not occur. This is because the methyl group at C5 position of benzene ring prevents the amino group from nucleophilic attack at C5 position. Electrochemistry of 5-MDA is quite similar. On the other hand, at pH above 10, 5-MDA was largely electroinactivated due to the attack by hydroxide ions in the alkaline solution. 5-MDA is more easily oxidized due to the methyl group compared to DA. Comparison of HOMO/LUMO energies between DA and 5-MDA provided the explanation on the question of why 5-MDA was more easily oxidized. 5-MDA has higher HOMO energy compared to DA, meaning that 5-MDA more easily donates electron than DA.

In Chapter 4, the incorporation of DA and 5-MDA at Nafion-modified electrode is better in acidic solutions compared to that in neutral solution. This is because the positively charged amino groups of DA and 5-MDA are electrostatically attracted to the negatively charged sulfonate group of Nafion. Comparing DA and 5-MDA at acidic pH, 5-MDA was more easily incorporated into the Nafion film probably due to the hydrophobic attraction of the methyl group. At acidic pH, the redox reaction of DA (and 5-MDA) at Nafion-modified electrode gives formal potential shifted negatively compared to that on bare electrode due to hydrophobic interaction between the oxidized form of DA (and 5-MDA) and the hydrophobic moieties of Nafion.

In Chapter 5, covalently bound DA to the p-aminobenzoic acid (PABA)-modified electrode was fabricated in two step-technique. The first step is the reduction of PABA diazonium on the electrode surface. The PABA-modified electrode bears the carboxyl terminal group. In the second step, the carboxyl group was bound to the amino group of DA (and 5-MDA) through amide bond coupling. DA was found to be covalently bound to PABA since no redox peak of the leucodopamichrome/dopaminechrome couple could be observed. This indicated that ICR could not occur due to the amide bond formation between amino group of DA and carboxyl group of PABA.

In Chapter 6, two redox molecules (pyrolloquinoline quinone (PQQ) and DA) were covalently bound to the chemically modified electrode. Similar technique of electrode modification as shown in Chapter 5 was employed with a little modification where (in the new technique) three steps were involved. The first step is reduction of p-phenylenediamine (PDA) diazonium on the electrode surface. The PDA-modified electrode bears the amino terminal group. The second step is the amide bonding between the amino group of PDA and carboxyl group of PQQ. The final step is the binding of amino group of DA and another carboxyl group of PQQ. Both redox molecules were stably confined on the electrode surface.

In Chapter 7, concluding remarks on the present doctoral thesis are addressed.

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 1copy of 800 Words (English).

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