

論文 / 著書情報
Article / Book Information

題目(和文)	
Title(English)	Hydration states of ionic liquids stabilizing protein structures and evaluation of proteins ' activity on model organic surfaces
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出典(和文)	学位:博士(理学), 学位授与機関:東京工業大学, 報告番号:甲第12342号, 授与年月日:2023年3月26日, 学位の種別:課程博士, 審査員:林 智広,北本 仁孝,曾根 正人,柘植 丈治,田中 祐圭
Citation(English)	Degree:Doctor (Science), Conferring organization: Tokyo Institute of Technology, Report number:甲第12342号, Conferred date:2023/3/26, Degree Type:Course doctor, Examiner:,,,,
学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

論文要旨

THESIS SUMMARY

系・コース：	ライフエンジニアリング	系
Department of, Graduate major in		コース
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申請学位 (専攻分野)：	博士	(Science)
Academic Degree Requested	Doctor of	
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要旨 (英文 800 語程度)

Thesis Summary (approx.800 English Words)

Protein-material interaction in the two-dimensional (2D) environment and three-dimensional (3D) environment is completely different. A 3D environment refers to the protein material interaction in a solution, and 2D environment refers to the protein material interaction at the interface between solid and liquid phase. In a 3D environment, protein molecules simultaneously interact with water molecules and excipient molecules, such as salt, sugar, and polymers in a highly competitive manner. However, in the case of a 2D environment, the protein-material interaction is limited to the boundary between the solid surface and the bulk liquid. Understanding the protein-material interaction in 2D and 3D environment is essential for the development of protein based biomedical applications, for instance, drug delivery systems biomedical implants, biosensors, etc.

This thesis discusses the strategies to investigate the protein material interaction in 2D and 3D environment using cytochrome c (cyt c) as a model protein. 1) Investigation of the hydration state of protein-stabilizing ionic liquids (ILs) (3D environment) using an IR-NMR combinational approach. 2) Investigation of the activity of protein immobilized on model organic surfaces (2D environment) using a slab optical waveguide (SOWG) UV/Vis spectroscopy.

Certain Hy ILs have shown the excellent capability to stabilize protein molecules. Questions such as, “why do only certain Hy ILs stabilize protein molecules?” and “what is the hydrogen bonding state of the water in such Hy ILs?” are yet to be answered. To address these questions, we probed the IL-water interaction of protein-stabilizing and protein-denaturing Hy ILs using attenuated total internal reflectance infrared (ATR-IR) spectroscopy and proton nuclear magnetic resonance (¹H NMR) spectroscopy. From the OH stretching band of water molecules from ATR-IR spectroscopy, we observed that the ratio of the weakly hydrogen-bonded water molecules to the strongly hydrogen-bonded water molecules was close to that of pure water, which indicates that the hydrogen bonding network of water molecules may be relatively less perturbed in protein-stabilizing Hy ILs. From the HOH bending spectra of water molecules from ATR-IR spectroscopy, I observed that the bending vibration of the water molecules was higher in c-stabilizing Hy ILs. This higher bending frequency of water molecules indicates that protein-stabilizing Hy ILs have a strong electric field effect on the water molecules. I believe that the strong electric field effect and pure water-like hydrogen bonding network of water molecules may play an essential role in the stabilization capability of certain Hy ILs.

Investigation of the activity of proteins molecules immobilized at the solid-liquid interface is complicated due to the lack of surface sensitive and selective techniques. Using SOWG UV/Vis spectroscopy can circumvent the complications in real-time observation of proteins immobilized at the solid-liquid interface. In this chapter, I investigated the activity of the cyt c protein molecules immobilized on hydrophobic self-assembled monolayers (SAMs) such as octadecyl silane (ODS) SAM. Trichloromethyl silane (TCMS) SAM and ODS-TCMS mixed SAM. Furthermore, the activity of cyt c was observed on hydrophilic bare glass as a control. I observed that on bare glass, the cyt c immobilized without any change in the activity. However, on hydrophobic SAMs, the immobilized cyt c underwent a change in the redox activity; the immobilized cyt c were oxidized at the solid-liquid interface. One possible explanation for this oxidation could be due to the oxygen molecules trapped in the nanobubbles formed on the surface of the hydrophobic SAMs, which when interacting with immobilized cyt c molecules results in oxidation.

This thesis answers the following questions: “Why certain Hy ILs stabilize protein structures?” (3D

environment) and “what is the activity of cyt c protein molecules immobilized on hydrophilic and hydrophobic surfaces?” (2D environment). In a 3D environment, the pure water-like hydrogen bonding network of water molecules in the ion hydration shell is essential for mediating the interaction with the protein molecules. The hydration shell around the ions creates a microenvironment that protects the structure and the function of protein molecules. However, in 2D environment, the formation of nanobubbles on the hydrophobic surfaces could be a possible reason for achieving the change in the redox activity of the protein immobilized at the solid-liquid interface. In this thesis, the observations made from protein-material interaction in 2D and 3D environment can contribute to the development of IL-based drug delivery systems and hydrophobic coatings for biosensors and medical implants.

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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