論文 / 著書情報
Article / Book Information

題目(和文)	二次元ナノ材料上における自己組織化ペプチドによるバイ オエレクトリック界面の動的形成の理解
Title(English)	Dynamic formation of bioelectronic interfaces by self-organized peptides on 2D materials
著者(和文)	関貴一
Author(English)	Seki Takakazu
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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	有機高分子物質　専攻	申請学位（専攻分野）： Academic Degree Requested	博士　工学 Doctor of Engineering
学生氏名： Student's Name	関　貴一	指導教員（主）： Academic Supervisor(main)	早水　裕平
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要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

This doctoral thesis entitled "Dynamic formation of bioelectronic interfaces by self-organized peptides and 2D materials" describes the effect of electrochemically biased environment on the behavior of self-organized peptides on two-dimensional (2D) nano-materials and development of a new bio-sensing platform to investigate the local molecular events at the interface between pseudo-biological environment and electronic semiconductor.

Boundaries between living systems and artificial systems, especially electronic devices are regarded as bioelectronic interfaces. One of long-standing scientists' dreams in this research field is realization of bidirectional flow of biological signals by bridging the interface between electronic device and bio-modules such as functional proteins and living cells, where not only small molecules but also ions play significant roles. To date, primitive but successful demonstrations have been reported as exemplified by neuronal field-effect transistor devices and a variety of bio-sensing devices which have functional biomolecules responding to bio-related substances. In reality, surfaces of living cells are decorated with various type of functional molecules such as membrane proteins, and the functionalities and surrounding ionic environments do not have uniform geometry, which may be attributed to efficient cell-cell communications. Besides, such ionically uniform space may offer undulated two-dimensional ionic distribution which is difficult to be understood from the viewpoint of the traditional electrochemical double layer. However, despite the growing significances, there is still lack of the methodology to investigate the biological surface phenomena from the electronic point of view. To shed light on this topic, a bioelectronic model for pseudo-biological environment has developed, and using optical poperies of the 2D nano-material, ionic behavior and interactions between ions and the self-organized peptides at the interface have been investigated.

Firstly, the surface charge effect on surface behaviors of the self-organized peptide is discussed in chapter 2. The self-organized peptides have ability to form coherently ordered structure on various electronic materials surface and the coherent characteristics have been expected to act as molecular scaffolds for functional molecules on the device surface and also to offer a platform to study the effect of

surface charges (potential) to the surface biomolecules. The first question toward understanding the bioelectronic interface is how the surface biomolecules behaves under the electrochemical bias and how much their organized structure is perturbed. To answer these questions here, an electrochemical cell composed of highly-oriented pyrolytic graphite (HOPG) and graphite-binding peptides (GrBPs) was made. Using this electrochemical cell, the self-organized structure under various electrochemical bias in water were investigated by atomic force microscope (AFM). The correlation between the amino-acids sequence of peptides and structural stability of organized mono-layer against applied electrochemical bias and also the effect of local pH on the peptide-organized structure are described.

Secondly, photoluminescence behavior of molybdenum disulfide (MoS_2) under pH-biased aqueous solution is described in chapter 3. MoS_2 is one of attractive layered materials, which belongs to a class of transition metal chalcogenide (TMD). Single layer MoS_2 has prominent properties arising from the semi-conductive nature, e.g., strong photoluminescence (PL) and tunable electronic conductivity with a field effect transistor (FET) configuration. Large area crystal of MoS_2 and the PL behavior can be used for the spatial visualization of the complex behavior of the bioelectronic interfaces. However, compared to the FET-type MoS_2 application, optical sensing under aqueous condition application has not been demonstrated although MoS_2 has several advantages such as high sensitivity of optical behavior of MoS_2 to surrounding solvent and molecular adsorptions. The second questions here are how PL of MoS_2 is modulated and how MoS_2 PL response can be affected by surrounding ions. To address these questions, MoS_2 PL behavior was investigated by *in situ* PL measurement. The results revealed a large PL modulation by surrounding pH condition. The origin of this pH response was systematically studied with various types of MoS_2 using X-ray photoelectron spectroscopy and electron density analysis by PL measurements and a FET device.

Finally, using an electrochemical cell consisting of MoS_2 optical sensor and a gate electrode, time-dependent PL behaviors under various electrolyte solutions are investigated. PL was modulated with electrochemical pulse wave modulation. The modulated PL showed anomalously long decay behaviors, which is two or three orders of magnitude larger than the typical duration of the formation of electrical double layer, and the decay time constants were evaluated under aqueous solutions. Functionalization of MoS_2 surface with self-organized peptides largely affected the decay response to ions. These PL responses and the effects of biomolecular functionalization are described in chapter 4.