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学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称　　博士（理学）　　氏名　ムラリダハラン　ムルガン

学 位 論 文 題 名

Enhancement Mechanism of Microbial Current Production by Conductive Iron Sulphides Biosynthesized by Sulphate Reducing Bacteria
（硫酸還元細菌により生合成された導電性硫化鉄による微生物生成電流増大機構に関する研究）

Microbial Fuel Cell (MFC) is one of the novel energy harvesting technology which are designed to provide renewable form of energy dependent on microbial metabolic activities. In these bio-electrochemical systems, the microbes oxidize organic substrates found in the natural environments and converts the energy stored in the chemical bonds into electricity. This system mimics the interactions of bacteria with insoluble electron donors and acceptors. Since there are different bacterial communities can be involved in the MFC current generation, they can use a wide range of substrates for their metabolism. This has given the use of various biodegradable substrates found in the waste water for the microbial metabolism and showing simultaneous electricity generation and municipal waste water treatment. Here the electrons that are produced by oxidation of substrates at the anode surface are transferred through the external circuit to reach cathode where reduction occurs. A proton exchange membrane allows the flow of protons across the anodic and cathodic chambers for charge balance.

Sediment Microbial Fuel Cell (SMFC) is a type of MFC which usually gets operated by microbial metabolisms in the sediments. Since the MFCs can utilize different kind of substrates, the solid phase substrates found in sediments, contaminated soil and sludge can also be utilized for energy harvesting. Especially, the aquatic sediments can provide sufficient organic substrates for bacterial metabolism for a long time. SMFC make use of the sedimental substrates to power electroactive microbes for electricity generation. This recently developing technology has been proven to be cost effective and can provide uninterrupted power supply for many low power marine devices like oceanographic sensors and deep-sea wireless devices. Here the anode part is usually submerged inside the anaerobic sediments where oxidation of organic substrates occurs and at the cathode which is suspended in the overlying water, which has relatively oxic environments.

Although the electricity production from the SMFCs was observed, efforts are being made to scale up the production and increase the electricity generation for longer durations. The interactions of microbial communities among themselves and their impact on anode current generation is now well understood. Even though many studies have been made on electrode materials, SMFC designs and operational conditions, a closer look at the microbial communities on the electrode surface seems to be important.

In addition to the interactions among bacterial communities, the minerals found in the sediments can also impact the electricity generation. Previous studies showed that the presence of conductive iron sulphide (FeS) nanoparticles actively increased the current production from the Iron Reducing Bacteria (IRB) *Shewanella oneidensis* MR-1. The marine sediments are rich in various metal sulphides having better electrical conductive properties. The anodic current production in the SMFC can be influenced by conductive minerals observed in the marine sediments. The sulphate reducing bacteria which usually dominate the marine sediments are capable of biomineralize various forms of iron sulphide minerals. Thus, in this work, the involvement of biomineralized conductive iron sulphides on the microbial current generation was studied.

The structure of the present thesis is as follows.

In Chapter 1, the general introduction for MFC, SMFC and their electricity generation mechanisms and designs are introduced. The mechanisms for direct and indirect extracellular electron mechanisms are briefly discussed followed by applications and challenges of SMFC systems. Biomineralization of conductive iron sulphides by sulphate reducing bacteria and past works based on microbial current enhancement by conductive nanoparticles are discussed.

In Chapter 2, the description of the experimental details such as the three-electrode electrochemical system design, microbial cultivation, conditions for electrochemical experiments, electrochemical measurements, characterization techniques like Scanning Electron Microscopy, Fluorescent In-Situ Hybridization and X-ray Photoelectron Spectroscopy Analysis are given.

In Chapter 3, the impact of FeS biosynthesis on the current production of SRB is explained. *Desulfovibrio vulgaris* Hildenborough has been used as a model organism to investigate the mechanism behind anodic current generation in SRB at sulfidic environments. The anodic current generation by SRB pure culture and the role of FeS on the current density is explained. The mechanism of FeS and hydrogen sulphide mediated anodic current generation is shown after analyzing the characterization results.

In Chapter 4, the current generation mechanism in SRB and IRB cocultures and its relation with the biomineralized FeS is discussed. *Shewanella oneidensis* MR-1 and *Desulfovibrio vulgaris* Hildenborough were used as the model organism for IRB and SRB respectively. Here, the dominance of FeS biomineralized by SRB on the coculture current is shown. The thickness of coculture bioagglomerates formed on the anode surface by making cross sectional thin sections and its electrical conductive properties by performing electrochemical gate experiments by interdigitated electrode array (IDA) are explained. Oxidation of FeS after lactate depletion and reduction in the conductive property of the coculture bioagglomerate is discussed.

In Chapter 5, the experimental data showing the symbiotic interaction of SRB and IRB are shown. Increase in the production of FeS bioprecipitates by SRB in the presence of IRB is discussed with experimental data measuring the increase in the dry weight of the system. The total protein concentration as a measurement of increase in the microbial growth is shown. The microscopic analysis showing increase in the IRB growth in the coculture system is explained. This chapter deals with the symbiotic relationship between SRB and IRB for both growth and FeS generation.

In Chapter 6, the dependence of microbial electricity production by other metal sulphides are shown. Metal sulphides which have better electrical properties than FeS are made to bioprecipitate inside the coculture system. This chapter deals with methods to improve the MFC current density by using various metal sulphides with better electrical conductive properties and subsequently their potential for alternatively use the heavy metal sulphides for MFC performance improvement.

In Chapter 7, the general conclusion of the work and the future prospects are given.