BIO-ELECTROCHEMICAL POWER GENERATION FROM PETROCHEMICAL WASTEWATER USING AS SUBSTRATES IN MICROBIAL FUEL CELL

SUMAYA SARMIN

MASTER OF SCIENCE

UNIVERSITI MALAYSIA PAHANG

SUPERVISOR'S DECLARATION

We hereby declare that we have checked this thesis and in our opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Master of Science.

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(Supervisor's Signature)

Full Name : DR. MD. MAKSUDUR RAHMAN KHAN

Position : PROFESSOR

Date :

____

(Co-supervisor's Signature)

Full Name : DR. ASMIDA BINTI IDERIS

Position : SENIOR LECTURER

Date :

STUDENT'S DECLARATION

I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

____

(Student's Signature)

Full Name : SUMAYA SARMIN

ID Number : MKC18005

Date :

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SUMAYA SARMIN

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ABSTRAK

Air sisa petrokimia (PCW) dari kilang asid akrilik mempunyai permintaan oksigen kimia (COD) yang sangat tinggi kerana adanya asid akrilik (AA) bersama dengan asid organik lain. Rawatan PCW dengan kaedah aerobik dan anaerob konvensional memerlukan tenaga. Namun, perlakuan PCW dengan penjanaan tenaga serentak dengan menggunakan sel bahan bakar mikroba (MFC) dapat menjadi alternatif yang berpotensi untuk menyelesaikan masalah tenaga dan alam sekitar. Rintangan utama untuk rawatan PCW di MFC adalah mencari inokulum yang sesuai berdasarkan interaksi substrat-inokulum, untuk menguraikan mekanisme pemindahan elektron yang membawa kepada penjanaan kuasa tinggi serta kecekapan penyingkiran COD yang tinggi. Tujuan kerja ini adalah untuk mengetahui inokulum yang sesuai yang mempunyai sifat elektrogenik dan fermentasi, untuk menjelaskan mekanisme pemindahan elektron dan akhirnya untuk menyiasat kinetik pemindahan caj anod. MFC dikendalikan menggunakan PCW dari loji AA tempatan dan enapcemar anaerob (AS) sebagai biokatalis di mana AS disesuaikan untuk menyiapkan inokulum yang berkesan. Mikrob yang didominasi dikenal pasti yang merangkumi genera elektrogenik iaitu Pseudomonas aeruginosa (PA) dan Bacillus cereus (BC) bersama dengan metanogenik archea Methanobacterium spp. Komponen utama PCW, seperti asid akrilik, asid asetik (ACA) dan dimetil phthalate (DMP) digunakan sebagai makanan bagi MFC untuk menilai interaksi substrat-inokulum. Prestasi MFC dinilai dari segi penjanaan voltan / arus serta penjanaan kuasa maksimum menggunakan polarisasi dan kurva kuasa. Vertammetri siklik (CV) dan spektroskopi impedans elektrokimia (EIS) digunakan untuk menjelaskan kinetik pemindahan caj anod dan model Nernst-Monod-Butler-Volmer digunakan untuk mengesahkan dan meramalkan prestasi MFC. Hasil kajian menunjukkan bahawa substrat campuran dengan AS yang disesuaikan dapat menghasilkan daya tinggi (0.78 W/m^3) dibandingkan dengan AA dengan PA (0.24 W/m^3), AA dengan BC (0.22 W/m^3), ACA dengan PA (0.39 W/m^3), ACA dengan BC (0.32 W/m^3), DMP dengan PA (0.24 W/m^3) dan DMP dengan BC (0.21 W/m^3) masing-masing. Data penjanaan tenaga berkorelasi dengan pola pertumbuhan mikroba yang menunjukkan pembentukan sinergi berdasarkan substrat-inokulum dalam sistem AS yang disesuaikan dengan substrat. Kajian ini selanjutnya diperluas ke PCW sebenar yang menunjukkan bahawa PCW dengan COD awal 45,000 mg/L dapat menghasilkan ketumpatan daya 850 mW/m^2 (pada kepadatan arus 1500 mA/m^2) menggunakan AS yang diaklimatisasi sebagai biokatalis. Kecekapan penghapusan COD dan kecekapan coulombic (CE) didapati 40% dan 21%, masing-masing setelah 11 hari beroperasi menggunakan COD awal 45000 mg/L. Penyiasatan CV mengesahkan peranan pyocynin dan hydroquinone sebagai ulang-alik elektron. Semasa membandingkan data CV biofilm dan anolit bebas inokulum setelah 11 hari beroperasi, arus puncak redoks tinggi diperhatikan untuk kes terakhir yang menunjukkan dengan jelas peranan utama mekanisme pemindahan caj tidak langsung untuk penjanaan kuasa menggunakan PCW dan AS yang disesuaikan. Kinetik pemindahan caj dijelaskan dengan menggunakan Tafel slop. Parameter kinetik dinilai dengan pas data kinetik dalam model Nernst-Monod-Butler-Volmar di mana COD eksperimen dan pengeluaran ketumpatan semasa didapati sesuai dengan model yang dicadangkan. Model tersebut dapat digunakan untuk mengoptimumkan prestasi MFC yang diberi makan PCW.

ABSTRACT

The petrochemical wastewater (PCW) from the acrylic acid plant possesses a very high chemical oxygen demand (COD) due to the presence of acrylic acid (AA) along with other organic acids. The treatment of PCW by conventional aerobic and anaerobic methods is energy-intensive. However, the treatment of PCW with concurrent power generation by employing microbial fuel cell (MFC) could be a potential alternative to solve the energy and environmental issues. The main hurdle for the treatment of PCW in MFC is to find out the suitable inoculum based on the substrate-inoculum interaction, to unravel the mechanism of electron transfer leading to the high power generation as well as high COD removal efficiency. The goal of the present work is to find out the suitable inoculum possessing electrogenic and fermentative properties, to elucidate the electron transfer mechanism and finally to investigate the anode charge transfer kinetics. MFCs were operated using PCW from local AA plant and anaerobic sludge (AS) as biocatalyst where AS was acclimatized to prepare effective inoculum. The predominated microbes were identified which include the electrogenic genera namely *Pseudomonas aeruginosa* (PA) and *Bacillus cereus* (BC) along with methanogenic archea *Methanobacterium spp*. The major constituents of the PCW, such as acrylic acid, acetic acid (ACA) and dimethyl phthalate (DMP) were used as feed for MFC to evaluate the substrate-inoculum interaction. The performance of the MFC was evaluated in terms of voltage/current generation as well as maximum power generation using polarization and power curve. Cyclic votammetry (CV) and electrochemical impedance spectroscopy (EIS) were employed to elucidate the kinetics of anode charge transfer and Nernst-Monod-Butler-Volmer model was used to validate and predict the performance of MFC. The results revealed that the mixed substrates with acclimatized AS could produce high power (0.78 W/m^3) compared to AA with PA (0.24 W/m^3), AA with BC (0.22 W/m^3), ACA with PA (0.39 W/m^3), ACA with BC (0.32 W/m^3), DMP with PA (0.24 W/m^3) and DMP with BC (0.21 W/m^3) respectively. The power generation data was correlated with the microbial growth pattern which indicated the formation of substrates-inoculum based synergy in the mixed substrate-acclimatized AS system. The study was further extended to the real PCW which demonstrated that the PCW with an initial COD of 45,000 mg/L could generate power density of 850 mW/m^2 (at a current density of 1500 mA/m^2) using acclimatized AS as biocatalyst. The COD removal efficiency and the coulombic efficiency (CE) were found to be 40% and 21%, respectively after 11 days of operation using initial COD of 45000 mg/L. CV investigations confirmed the role of pyocynin and hydroquinone as electron shuttles. While comparing the CV data of the biofilm and the inoculum free anolyte after 11 days of operation, the high redox peak current was observed for the latter case which clearly demonstrated the predominant role of indirect charge transfer mechanism for power generation using PCW and acclimatized AS. The charge transfer kinetics was elucidated using the Tafel slop. The kinetic parameters were evaluated by fitting the kinetic data in Nernst-Monod- Butler-Volmar model where the experimental COD and current density production was found to be in good agreement with the proposed model. The model can be used for optimization of the performance of the PCW-fed MFC. The results of the present study showed that the electrocatalytic activity of anaerobic sludge can be improved by acclimatization which can be effectively used for simultaneous power generation and treatment of PCW.

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