PREPARATION AND PROPERTIES OF REDUCED GRAPHENE OXIDE FOR DIRECT METHANOL FUEL CELL APPLICATION

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I dedicate my thesis for my beloved

♥ Father, Abdul Halim Mohd Saad
♥ Mother, Hamimah Md Dali
♥ Siblings
♥ Friends

Thank you for your love and continuous supports.
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Reduced graphene oxide has recently attracted great attention due to its unique chemical and physical properties. In this study, reduced graphene oxide is introduced in direct methanol fuel cell application as this system had been delayed by several reasons which are related to the lack of materials with good methanol permeability and conductivity properties. Therefore, the objective of this research is to develop and characterize a high conductive reduced graphene oxide as additive in membrane electrode assembly (MEA). Reduced graphene oxide was prepared using reduction of the oxide with different reducing agents and the degree of reducibility of the reducing agents used was compared in order to determine the alternative reducing agent to replace hydrazine. The reduced graphene oxide was successfully prepared from the pristine graphite by reductive precipitation of graphene oxide aqueous solution using sodium borohydride which acted as the reducing agent for 24 hours reaction time at room temperature. Subsequently, MEA from sulfonated poly (ether ether ketone) (SPEEK) consists of reduced graphene oxide as additive was fabricated for direct methanol fuel cell (DMFC) performance testing. The physicochemical properties of the reduced graphene oxide were characterized by Fourier transform infrared (FTIR) spectroscopy, X-ray diffraction (XRD), transmission electron microscopy (TEM) and conductivity measurement. The FTIR spectrum of the reduced graphene oxide shows that the carbonyl group was successfully removed and the reduced graphene oxide contains some water. The XRD spectrum shows reduced graphene oxide has hexagonal structured crystal with a sharp peak at $2\theta = 26.4^\circ$ which can be assigned as 002 peak associated with reduced graphene oxide. The exhibited conductivity was in the range of $1.80 \times 10^3 - 1.30 \times 10^4$ S m$^{-1}$. TEM micrograph showed that reduced graphene oxide has multilayer structures. The DMFC performance of the blended reduced graphene oxide with SPEEEK55 exhibited substantial improvement by 5 – 15% of open circuit voltage (OCV), initial voltage, power density, and stabilization period as compared to the commercial nafion membrane. In conclusion, reduced graphene oxide is a potential material as catalyst support in MEA and additive in SPEEK55 membrane to improve the DMFC performance.
ABSTRAK

Sejak kebelakangan ini, grafena oksida terturun telah menarik banyak perhatian kerana sifat kimia dan fiziknya yang unik. Dalam kajian ini, grafena oksida terturun diperkenalkan dalam aplikasi sel bahan api metanol terus kerana sistem ini telah tertangguh disebabkan oleh beberapa sebab yang berkaitan dengan kekurangan bahan dengan kebolehtelapan metanol dan sifat kekonduksian yang baik. Kajian ini bertujuan untuk membangunkan dan mencirikan grafena oksida terturun sebagai tambahan dalam himpunan elektrod membran (MEA). Grafena oksida terturun disediakan dengan menggunakan agen penurunan yang berbeza untuk mengkaji kuasa penurunan dan sebagai satu pendekatan dalam mencari agen penurunan alternatif untuk menggantikan hidrazin. Penghasilan grafena oksida terturun berjaya dilakukan bermula dari grafit tulen dengan pemendakan penurunan grafena oksida oleh natrium borohidrat sebagai agen penurun dengan tindak balas selama 24 jam pada suhu bilik. Selepas itu, MEA oleh sulfonat poli (eter eter keton) (SPEEK) mengandungi grafena oksida terturun telah direka untuk ujian prestasi sel bahan api metanol terus (DMFC). Sifat fizik dan kimia grafena oksida terturun dicirikan menggunakan spektroskopi infra-merah transformasi Fourier (FTIR), pembelauan sinar-X (XRD), mikroskopi penghantaran elektron (TEM) dan pengukuran kekonduksian. Spektrum FTIR yang diperoleh menunjukkan bahawa kumpulan karbonil telah berjaya disingkirkan dan grafena oksida terturun mengandungi air. Spektrum XRD menunjukkan bahawa grafena oksida terturun mempunyai kristal berstruktur heksagon dengan pencak tajam pada $2\theta = 26.4^\circ$ yang boleh ditetapkan sebagai puncak 002 yang dikaitkan dengan grafena oksida terturun. Kekonduksian dipamerkan dalam julat $1.80 \times 10^{-3}$ – $1.30 \times 10^{-4}$ S m$^{-1}$. Mikrograf TEM menunjukkan bahawa grafena oksida terturun mempunyai struktur berlapis-lapis. Prestasi DMFC campuran grafena oksida terturun dengan SPEEK55 mempamerkan peningkatan yang ketara sebanyak 5 – 15% daripada voltan litar terbuka (OCV), voltan awal, ketumpatan kuasa dan tempoh penstabilan berbanding dengan membran nafion komersial Kesimplelannya, grafena oksida terturun adalah bahan yang berpotensi sebagai tambahan dalam membran SPEEK55 untuk meningkatkan prestasi DMFC.