THE EFFECT OF AMMONIUM NITRATE AND ETHYLENE GLYCOL IN CARBOXYMETHYL CELLULOSE SOLID POLYMER ELECTROLYTES FOR RECHARGEABLE PROTON BATTERY

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ABSTRACT

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School	**: School of Fundamental Science**

Solid polymer electrolytes (SPEs) have been recognised as promising electrolytes to overcome the limitation in gel/liquid counterparts. In this research, the potential of carboxymethyl cellulose (CMC)-based SPEs was studied. Two systems, namely System I which comprised of CMC doped ammonium nitrate (NH_4NO_3) salt and System II which comprised of CMC-NH_4NO_3 plasticized ethylene glycol (EG) are prepared via solution casting technique. The interactions between polymer, salt and plasticizer in both systems are observed by FTIR analysis. The amorphousness and thermal behaviour of both systems are obtained by XRD and TGA-DTG analyses, respectively. The incorporation of 45 wt.% NH_4NO_3 salt in System I has optimized the ambient temperature ionic conductivity to $(7.71 \pm 0.04) \times 10^{-3}$ Scm^{-1}. Ionic conductivity is enhanced to $(3.80 \pm 0.06) \times 10^{-2}$ Scm^{-1} on the addition of 20 wt.% EG plasticizer in System II. The temperature-dependent ionic conductivity for both electrolyte systems is Arrhenian and thermally activated. The activation energy did not show the significant effect to the enhancement of ionic conductivity in both systems. However, the activation energy in System I is higher than in System II. Ionic conductivity is observed to be influenced by the number density (n), mobility (μ) and diffusion coefficient (D) of mobile charge carriers in System I. Whereas, ionic conductivity in System II is affected only by n and inversely proportional to μ and D. These transport parameters are affected by the fraction of ionic species in both systems. The majority of mobile charge carriers is recognized to be ions as evidenced by transference number measurement via DC polarization technique. This analysis also supported that the mobile charge carriers in both systems are H^+ cations or protons. Conduction mechanism in both systems is described by the relation of Jonscher's universal power law and hopping mechanism models. Quantum mechanical tunneling model was identified to represent the ionic conduction mechanism in both systems. Electrical studies show that the SPEs are ionic conductors. The electrical behaviour of SPEs is attributed to the conductivity relaxation process. The highest conducting SPEs in both systems were applied in the fabrication of rechargeable proton batteries. Based on the batteries performance, it can be concluded that the rechargeable proton batteries in the present work are suitable to be applied in low current based electrochemical device applications.

ABSTRAK

Abstrak tesis yang dikemukakan kepada Senat Universiti Malaysia Terengganu sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

KESAN AMONIUM NITRAT DAN ETILENA GLIKOL DALAM ELEKTROLIT PEPEJAL POLIMER KARBOKSILMETIL SELULOSA UNTUK BATERI PROTON CAS SEMULA

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Februari 2016

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Pusat Pengajian : Pusat Pengajian Sains Asas

Elektrolit pepejal polimer (SPEs) telah dikenalpasti sebagai elektrolit yang berupaya untuk mengatasi kekurangan yang dialami oleh elektrolit gel/cecair. Dalam kajian ini, potensi karboksilmetil selulosa (CMC) berasaskan SPEs telah dikaji. Dua sistem iaitu Sistem I terdiri daripada CMC berdop garam ammonium nitrat (NH_4NO_3) dan Sistem II terdiri daripada CMC-NH_4NO_3 berpemplastik etilena glikol (EG) telah disediakan melalui teknik penuangan larutan. Interaksi-interaksi diantara polimer, garam dan pemplastik dalam kedua-dua sistem telah dicerap melalui analisis IR. Keamorfusan dan perlakuan haba kedua-dua sistem diperolehi daripada analisis XRD dan TGA-DTG. Dengan penambahan 45 wt.% garam NH_4NO_3 dalam Sistem I telah mengoptimumkan kekonduksian ion suhu ambien/sekeliling pada $(7.71 \pm 0.04) \times 10^{-3}$ Scm^{-1}. Kekonduksian ion dipertingkatkan kepada $(3.80 \pm 0.06) \times 10^{-2}$ Scm^{-1} dengan penambahan 20 wt.% pemplastik EG dalam Sistem II. Analisis kebergantungan kekonduksian ion terhadap suhu menunjukkan kedua-dua sistem elektrolit adalah Arrhenian dan diaktifkan secara terma. Tenaga pengaktifan tidak menunjukkan kesan yang ketara terhadap penambahan kekonduksian ion dalam kedua-dua sistem. Walau bagaimanapun, tenaga pengaktifan bagi Sistem I didapati lebih tinggi berbanding Sistem II. Kekonduksian ion dipengaruhi oleh kepadatan nombor (n), mobiliti (μ) dan pekali peresapan (D) pembawa cas bergerak dalam Sistem I. Manakala, kekonduksian ion dalam Sistem II hanya dipengaruhi oleh n dan berkadar songsang dengan μ dan D. Parameter-parameter angkutan ini dipengaruhi oleh pecahan spesies-spesies ion. Majoriti pembawa cas bergerak dikenalpasti sebagai ion seperti yang telah dibuktikan oleh pengukuran jumlah pemindahan melalui teknik pengkutuban DC. Analisis ini juga menyokong bahawa pembawa cas bergerak dalam kedua-dua sistem adalah H^+ kation atau proton. Mekanisma kekonduksian untuk kedua-dua sistem diterangkan berdasarkan hukum kuasa sejagat Jonscher dan model-model mekanisme lompatan. Model penerowongan mekanik kuantum telah dikenalpasti untuk mewakili mekanisma kekonduksian ion dalam kedua-dua sistem. Kajian elektrik menunjukkan bahawa SPEs adalah pengalir/konduktor ion. Perlakuan elektrik SPEs adalah disebabkan oleh proses kekonduksian relaksasi/kekenduran. SPEs dengan kekonduksian tertinggi dalam kedua-dua sistem telah digunakan dalam fabrikasi bateri proton cas semula. Berdasarkan prestasi daripada bateri-bateri tersebut, ia boleh disimpulkan bahawa bateri proton cas semula dalam kajian ini sesuai untuk diaplikasikan dalam penggunaan peranti elektrokimia berarus rendah.