EXPERIMENTAL CHARACTERISATION AND MODELING OF A VANADIUM REDOX FLOW BATTERY

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EXPERIMENTAL CHARACTERISATION AND MODELING OF A VANADIUM REDOX FLOW BATTERY

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Thesis submitted in fulfilment of the requirements
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LIST OF SYMBOLS

Symbol	Meaning	Units
$C_{V(II)}$	Concentration of V(II) species	Mol dm^{-3}
$C_{V(III)}$	Concentration of V(III) species	Mol dm^{-3}
$C_{V(IV)}$	Concentration of V(IV) species	Mol dm^{-3}
$C_{V(V)}$	Concentration of V(V) species	Mol dm^{-3}
$\varnothing$	Current efficiency	
C_{oxd}	Concentration of oxidised species	Mol dm^{-3}
C_{red}	Concentration of reduced species	Mol dm^{-3}
C_{ap}	Activation polarisation capacitance (transient response)	F
C_{cp}	Concentration polarisation capacitance (transient response)	F
E_{cell}	Cell potential; measured across unit cell stack	V
$E_{cell(ORP)}$	Open-circuit cell potential across two electrode or Oxidation-reduction redox potential; measured across two working electrode at reference cell (otherwise $E_{V(IV)/V(V)} - E_{V(III)/V(II)}$)	V
E_{drop}	Cell potential after IR_o effect	V
$E_{cell,oc}$	Open-circuit cell potential	V
E^o_{cell}	Standard cell potential; potential different across the two half-cells with respect to the standard hydrogen electrode (SHE) (otherwise $E^o_{cell,+} - E^o_{cell,-}$)	V

$E^{o}_{cell,+}$	Standard positive half-cell potential; standard electrode potentials of positive half-reactions potential with respect to the standard hydrogen electrode (SHE)	V
$E^{o}_{cell,-}$	Standard negative half-cell potential; standard electrode potentials of negative half-reactions potential with respect to the standard hydrogen electrode (SHE)	V
$E_{V(IV)/V(V)}$	Positive half-cell potential for V-RFB; measured at positive half-cell reference cell across working electrode and Hg/Hg_2SO_4 reference electrode	V
$E_{V(III)/V(II)}$	Negative half-cell potential for V-RFB; ; measured at negative half-cell reference cell across working electrode and Hg/Hg_2SO_4 reference electrode	V
E	Energy	Wh
E_d	Energy density	Wh L^{-1}
F	Faraday’s constant	Ah mol^{-1}
I	Current	A
IR_o	Drop of cell potential due to internal resistance of V-RFB cell	V
I_{shunt}	Shunt current; measured at positive terminal of dc supply and load during charge-discharge cycle at range 100 mV/A	A
I-V	Electrical power	P
j	Current density	mA cm^{-2}
K, L, M, N	Concentrations of oxidation and reduction species in Nernst equation	Mol dm^{-3}
k,l,m,n	Stoichiometric factors in Nernst equation	-

M_m	Molar mass of the reactant	g mol^{-1}
P	Power	W
P_d	Power density	W L^{-1}
P_p	Percentage purity	
p	Pressure	Pa
q	Charge	C
q/V	Volumetric charge density	Ah L^{-1}
Q	Equilibrium constant	-
R_o	Ohmic polarisation	Ω
R_{ap}	Activation polarisation resistance (Reaction rate loss)	Ω
R_{cp}	Concentration polarisation resistance (Mass transport loss)	Ω
S_g	Specific gravity of H_2SO_4	-
t	Time	second
T	Temperature	^oC
$\%\eta_V$	Voltage efficiency	%
$\%\eta_C$	Charge efficiency	%
$\%\eta_E$	Energy efficiency	%
Δc	Change in the reactant concentration	mol L^{-1}
V	Volume of electrolyte	L
W_{cell}	Amount of work in electrochemical cell	Joules, J
z	No. of electrons transferred during the redox reaction	-

LIST OF ABBREVIATIONS

AER	–	All electric range
BEV	–	Battery electric vehicle
EGM	–	Electric generator/motor
EKF	–	Extended Kalman filter
EV	–	Electric vehicle
FC	–	Fuel cell
FCEV	–	Fuel cell electric vehicle
HEV	–	Hybrid electric vehicle
HR	–	Hybridization ratio
ICE	–	Internal combustion engine
PHEV	–	Plug-in HEV
OCV	–	Open-circuit voltage
ORP	–	Oxidation-reduction redox potential
PVC	–	Polyvinyl chloride polymer
RESS	–	Rechargeable energy storage system
RFB	–	Redox flow battery
SHE	–	Standard hydrogen electrode
SOC	–	State-of-charge
UNSW	–	The University of New South Wales, Australia
V-RFB	–	Vanadium RFB

ABSTRACT

This thesis presents the summarization of work on experimental characterisation and modeling of a vanadium redox flow battery (V-RFB). This thesis presents background material and motivation factors of the studies; research goals, a review of previous work and discussion on related issues with respect to energy storage technologies, emphasising on V-RFB system. The aim of the study is to investigate the performance of V-RFB through experimental characterisation of V-RFB at different operating parameters and develop electrical circuit modeling of V-RFB. Preliminary experiment on 100 cm^2 unit cell laboratory unit V-RFB has helped in familiarising with V-RFB setup and its design weaknesses and factors leading the cell into failure mode are highlighted. Based on observation on 100 cm^2 unit cells, new design of 25 cm^2 unit cell laboratory unit V-RFB has been proposed with an improvement of efficiency and reduction of contact resistance are observed. Theoretically studies by using Faraday's law of electrolysis and Nernst equation are used to relate the equilibrium cell's potential with the concentration changes in vanadium species, back-up with experimental data from a divided, open-circuit potentiometric cell approach. Two different approaches has been presented, with newly proposed approach of a divided, open-circuit potentiometric cell, via Hg/Hg_2SO_4 reference electrodes and graphite rod working electrodes present superiorities in estimating the state-of-charge (SOC) of V-RFB. System characterisation has been carried-out for the new 25 cm^2 unit cell laboratory unit V-RFB under different of operating parameters such as current densities, temperatures, flow rates, concentrations and material properties. The cell exhibits highest energy efficiency at 82.1 %, operating at 308 K, 60 mA cm^{-2} current density and 3 cm^3 s^{-1} volumetric flow rate for 250 cm^3 (each reservoir) of 1.6 mol dm^{-3} *V(III)/V(IV)* in 4 mol dm^{-3} H_2SO_4. Formation charge of mixture of vanadium species into single electro-active species at positive and negative electrodes are highlighted. A method for estimating the V-RFB to complete its formation charge using electrochemical calculation of Faraday's constant are also presented. New equivalent electric circuit model for V-RFB has been proposed which consists of an open-circuit cell potential in series of ohmic internal resistance and the parallel n-RC network. Extended Kalman filter is used for parameter identification of dynamic characterisation of V-RFB. Numerical simulations are compared to experimental data at different pulse voltages at few SOCs and experimental charge-discharge characterisation of V-RFB system, demonstrating good agreement.

ABSTRAK

Tesis ini membentangkan ringkasan hasil kerja lapangan mencirikan dan memodelkan vanadium redox bateri teralir (V-RFB). Tesis ini membentangkan latar belakang dan faktor yang memotivasikan untuk pengajian, matlamat kajian, penilaian hasil kerja yang lepas dan perbincangan berkaitan dengan teknologi penyimpanan tenaga, dengan penekanan kepada sistem V-RFB. Tujuan pengajian ini adalah untuk mengkaji prestasi V-RFB melalui pencirian eksperimen V-RFB pada parameter operasi yang berbeza dan membangunkan model litar elektrik V-RFB. Eksperimen awal pada 100 cm^2 sel unit makmal yang membantu membiasakan dengan pembinaan V-RFB dan juga kelemahan rekaannya dan faktor yang menyebabkan bateri gagal berfungsi ada dinyatakan. Berdasarkan pemerhatian pada 100 cm^2 sel unit makmal, satu rekaan baru iaitu 25 cm^2 sel unit makmal dicadangkan dengan peningkatan tahap kecekapan dan penurunan rintangan sentuhan dapat diperhatikan. Kajian secara teori hukum Faraday elektrolisis dan persamaan Nernst digunakan untuk mengaitkan potensi sel keseimbangan dengan perubahan kepekatan spesies vanadium, dibantu dengan data eksperimen dari litar terbuka-terbahagi-sel pendekatan potentiometrik. Dua pendekatan yang berbeza telah dikemukakan, dengan pendekatan baru dari litar terbuka-terbahagi-sel pendekatan potentiometrik, melalui Hg/Hg_2SO_4 elektrod rujukan dan rod grafit bekerja-elektrod memberikan pendekatan terbaik dalam menganggarkan tahap caj (SOC) V-RFB. Pencirian sistem telah diberikan untuk sistem baru 25 cm^2 sel unit makmal V-RFB di bawah variasi parameter operasi seperti kepadatan arus, suhu, kadar aliran, kepekatan dan sifat bahan. Sel telah mencatatkan kecekapan tenaga tertinggi pada 82.1%, beroperasi pada 308 K, 60 mA cm^{-2} ketumpatan arus dan 3 cm^3 s^{-1} kadar aliran isipadu 250 cm^3 (setiap satu takungan) 1.6 mol dm^{-3} *V (III) / V (IV)* dalam 4 mol dm^{-3} H_2SO_4. Cas pembentukan campuran spesies vanadium ke spesies elektro-aktif tunggal pada elektrod positif dan negatif telah diketengahkan. Satu kaedah untuk menganggarkan V-RFB untuk melengkapkan cas pembentukan campuran spesies vanadium ke spesies elektro-aktif tunggal menggunakan pengiraan elektrokimia pemalar Faraday juga dibentangkan. Litar setara model elektrik yang baru untuk V-RFB telah dicadangkan yang terdiri daripada voltan terbuka litar sel sesiri dengan rintangan dalaman dan selari dengan rangkaian n-RC. Penapis Lanjutan Kalman digunakan untuk mengenalpasti parameter pencirian dinamik V-RFB. Simulasi berangka berbanding dengan data eksperimen pada voltan nadi berbeza pada beberapa SOCs dan eksperimen cas-discas pencirian sistem V-RFB, menunjukkan penemuan yang baik.

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APPENDIX - LIST OF PUBLICATIONS

Table A presents the entire articles that have been submitted or published in peer-reviewed Journals and Conferences as progressive contributions of this research work.

Table A: List of submitted or published in peer-reviewed Journals and Conferences for Experimental Characterisation and Modeling of a Vanadium Redox Flow Battery

No	Journals Publication Details	Impact Factor	Status	Thesis Chapter
1	**Mohamed, M.R.**, Ahmad, H. and Abu Seman, M.N. 2012. State-of-the-art of vanadium redox flow battery: A review on research prospects. *International Review of Electrical Engineering (IREE)* **7** (5): 5610-5622.	1.364	Published	Chapter 2
2	**Mohamed, M.R.**, Sharkh, S.M., Ahmad, H., Abu Seman, M.N. and Walsh, F.C. 2012. Design and development of unit cell and system for vanadium redox flow batteries (V-RFB). *International Journal of the Physical Sciences* **7** (7): 1010 - 1024.	0.540	Published	Chapter 3
3	**Mohamed, M.R.**, Ahmad, H. and Abu Seman, M.N. In Press. Estimating State-of-Charge of Vanadium Redox Flow Battery using a Divided, Open-circuit Potentiometric Cell. *Elektronika Ir Elektrotechnika* **19** (3): 37-42	0.913	Published	Chapter 4
3	**Mohamed**, **M.R.**, Leung, P.K., Ahmad, H., Abu Seman, M.N., *Renewable & Sustainable Energy Reviews*, Submitted on 29 Oct 2012 (RSER-D-12-01127).	6.619	Under Review	Chapter 5
4	**Mohamed**, **M.R.**, Ahmad, H., Abu Seman, M.N., Razali, S., Najib, M.S. *Journal of Power Sources*, In Press, Accepted Manuscript, Available online 3 April 2013	4.951	Published	Chapter 6

No	Conferences Publication Details	Thesis Chapter
1	**Mohamed, M.R.**, Sharkh, S.M., Ahmad, H., Abu Seman, M.N. and Walsh, F.C., In : Malaysian Postgraduate Conference on Electrical, Electronic and Control Technology (MCEECT 2012) 16-17 February 2012, Universiti Malaysia Pahang.	Chapter 2
2	**Mohamed, M.R.**, Ahmad, H., Abu Seman, M.N. (2012) In: International Conference on Green Technology & Ecosystems for Global Sustainable Development 2012 (ICGTEC2012), 28-30	Chapter 2

	May 2012, University of Tuzla, Bosnia.	
3	**Mohamed, M.R.**, Sharkh, S.M., Walsh, F.C., in: The 5th International IEEE Vehicle Power and Propulsion Conference (VPPC'09), IEEE, Dearborn, Michigan, 2009, pp. 551-557.	-