

ELECTROCHEMICAL IMPEDANCE SPECTROSCOPY STUDY OF AQUEOUS NA-ION BATTERY DEGRADATION

Aivaras Labžentis¹, Laurynas Staišiūnas², Linas Vilčiauskas²

¹Faculty of Physics, Vilnius University, Lithuania

²Center for Physical Sciences and Technology, Saulėtekio al. 3, LT-10257 Vilnius, Lithuania
aivaras.labzentis@ff.stud.vu.lt

Increasing energy consumption and changing people attitude towards more sustainable energy calls for new ways how to produce and store electricity [1]. One of the alternatives is the use of aqueous Na-ion batteries due to their low cost, environmental friendliness and durability. However, water based electrolytes suffer due to the limited electrochemical window of water and limited electrode material stability [1, 2].

The main goals of this study are to choose and investigate the effects of cell type and geometry to the Electrochemical Impedance Spectroscopy (EIS) measurements, record the EIS spectra of a negative Na-ion battery electrode during the galvanostatic charge/discharge cycling, fit the data using the equivalent circuit model and analyze the results in terms of electrode degradation mechanisms [3]. All the studied electrodes were developed in house using the NASICON-structured $\text{Na}_3\text{Ti}_2(\text{PO}_4)_3$ as active material and 1M $\text{Na}_2\text{SO}_4(\text{aq.})$ as electrolyte. All measurements were conducted at room temperature.

In the simulation an equivalent electrical circuit is used in order to fit the EIS results (Fig. 1). The data presented in Table 1 show the increase in one of the resistances (R_1) upon cycling, which correlates directly with the shrinking electrode charge capacity. This indicates the electrode degradation process caused by such processes as electrode material dissolution and subsequent deposition of certain blocking layer or corrosion of electrode component due to water decomposition reactions taking place. Nevertheless, the Coulombic efficiency does not deteriorate but actually increases with cycle number indicating a certain stabilization and corrosion inhibition processes.

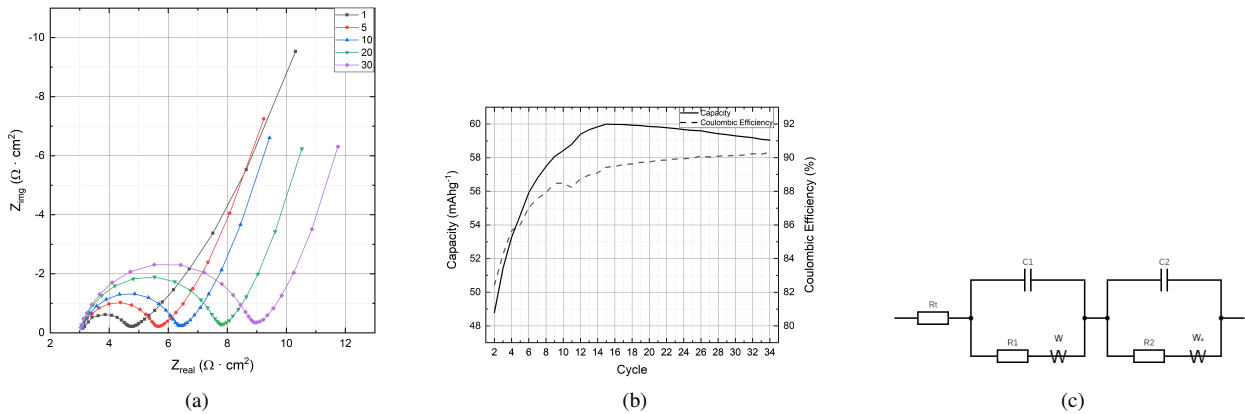


Fig. 1. Electrode degradation. (a) EIS change with cycle number, (b) charge capacity and Coulombic efficiency change with cycle number, (c) fitted electrical circuit.

Table 1. Fitted measurement data

Cycle No.	R_t	C_1	R_1	W	C_2	R_2	W_{or}	W_{oc}
	$\Omega \cdot \text{cm}^2$	F/cm^2	$\Omega \cdot \text{cm}^2$	$\Omega \cdot \text{cm}^2/\text{s}^{1/2}$	F/cm^2	$\Omega \cdot \text{cm}^2$	$\Omega \cdot \text{cm}^2/\text{s}^{1/2}$	$\text{s}^{1/2}$
1	3,15	$1,29 \cdot 10^{-4}$	0,43	3,69	$6,83 \cdot 10^{-6}$	1,32	0,90	0,52
5	3,12	$7,64 \cdot 10^{-5}$	0,77	2,74	$8,53 \cdot 10^{-6}$	1,65	0,79	0,48
10	3,11	$4,56 \cdot 10^{-5}$	1,37	2,34	$9,34 \cdot 10^{-6}$	1,77	0,78	0,49
20	3,08	$2,81 \cdot 10^{-5}$	2,74	2,13	$1,16 \cdot 10^{-5}$	1,74	0,89	0,57
30	3,07	$2,57 \cdot 10^{-5}$	3,74	2,20	$1,26 \cdot 10^{-5}$	1,82	1,04	0,68

Acknowledgements:

This project has received funding from the European Regional Development Fund (Project No. 01.2.2-LMT-K-718-02-0005) under grant agreement with the Research Council of Lithuania (LMTLT).

[1] J. Liu, C. Xu, Z. Chen, S. Ni, Z. X. Shen. Progress in aqueous rechargeable batteries, Green Energy & Environment 3 (1), 20-41 (2018).

[2] R. Demir-Cakan, M. R. Palacin, L. Croguennec, Rechargeable aqueous electrolyte batteries: from univalent to multivalent cation chemistry, Journal of Materials Chemistry A 7, 20519–20539 (2019).

[3] M. E. Orazem, B. Tribollet, Electrochemical Impedance Spectroscopy (Wiley, 2008).