

EQCM-D-Based Surface-Acoustic-Wave Spectroscopy: Emerging Innovations in Material Characterizations for Energy Storage and Conversion

Mikhael D. Levi

Department of Chemistry, Bar-Ilan University, Ramat-Gan 52900, Israel



Depending on stiffness/softness of the electrode coatings (attached with no-slip to quartz crystal surface), their thickness and morphology, the quartz crystal resonator can operate in either gravimetric or beyond-gravimetric modes providing important information about the mechanical properties of the electrodes.^{1,2} Starting from a simple gravimetric monitoring of dynamics of ions adsorption into nanoporous carbon supercapacitors,³ we have demonstrated a couple of years ago that the use of multiharmonic Electrochemical Quartz Crystal Microbalance with Dissipation Monitoring (EQCM-D) constitutes the basis of a new powerful *in situ* method of highly sensitive tracking intercalation-induced dimensional and porous structure changes in operating battery⁴ and supercapacitor⁵ electrodes. The resonance frequency and dissipation changes caused by contact of a porous solid electrode with electrolyte solutions are recorded on multiple harmonics and fitted to a suitable hydrodynamic impedance model returning the structural parameters of the electrode. Later on this new methodology was extended to a continuous monitoring of viscoelastic changes in binder-free 2D electrodes such as Mxene ($\text{Ti}_3\text{C}_2(\text{OH})_x$) caused by insertion of water molecules modulated by intercalation-deintercalation of Li-ions in aqueous solutions.⁶ Monitoring viscoelastic properties of solid-electrolyte interface (SEI) on a high-voltage anode such as LTO was shown to be an extremely effective means for fast optimization of cycling behavior of this electrode.⁷ Our recent paper makes focus on *in situ* acoustic diagnostics of particle-binder interactions in battery electrodes: accommodation of intercalation-induced volume changes significantly depend on the stiffness/softness of the binder used,⁸ on one hand, and on the extent to which the size of the guest cation matches the size of the host sites in which the cations are accommodated.⁹ The different worked examples of the successful use of EQCM-D-based surface-acoustic-wave spectroscopy for material characterization of energy storage electrodes have been summarized in a recent review.¹⁰

References

1. A.R. Hillman, J. Sol. State Electrochem. 2011, **15**, 1647-1660.
2. D. Johannsmann, The Quartz Crystal Microbalance in Soft Matter Research, 2014, Springer.
3. M.D Levi, G. Salitra, N. Levy, D. Aurbach, J. Maier, Nature Materials, 2009, **8**, 872-875.
4. N. Shpigel, M.D. Levi, S. Sigalov, O. Girshevitz, L. Daikhin, D. Aurbach, P. Pikma, M. Marandi, A. Jänes, E. Lust, N. Jäckel, and V. Presser, Nature Materials, 2016, **15**, 570-575.
5. N. Shpigel, M.D. Levi, S. Sigalov, L. Daikhin, D. Aurbach, and V. Presser, J. Phys.: Condens. Matter (2016) **28**, 114001.
6. N. Shpigel, M. R. Lukatskaya, S. Sigalov, Chang E. Ren, P. Nayak, M. D. Levi, L. Daikhin, D. Aurbach, Y. Gogotsi, ACS Energy Lett. 2017, 1407–1415.
7. V. Dargel, N. Shpigel, S. Sigalov, P. Nayak, M.D. Levi, L. Daikhin, D. Aurbach, Nature Communications, 2017, 1389.
8. V. Dargel, N. Jäckel, N. Shpigel, S. Sigalov, M.D. Levi, L. Daikhin, V. Presser, D. Aurbach, In Situ Multilength-Scale Tracking of Dimensional and Viscoelastic Changes in Composite Battery Electrodes, ACS Applied Materials & Interfaces 2017, 27664–27675.
9. N. Shpigel, S. Sigalov, Mikhael D. Levi, T. Mathis, L. Daikhin, A. Janes, E. Lust, Y. Gogotsi, D. Aurbach, In Situ Acoustic Diagnostics of Particle-Binder Interactions in Battery Electrodes, Joule 2018, doi.org/10.1016/j.joule.2018.02.014
10. N. Shpigel, M.D. Levi, S. Sigalov, O. Girshevitz, L. Daikhin, D. Aurbach. Accounts Chem. Research, 2017, **51**, 69-79.