Interfacial Studies of Sn thin Film Electrode in Electrolytes with additives using In Situ Electrochemical Quartz Crystal Microbalance-Dissipation (EQCM-D)

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Since its inception, the Li-ion battery has evolved from a mere high energy density device into intensified configurations that meet high energy, high power density, long lifetime, and safety demands.<sup>1</sup> These demands have driven battery research to worldwide attention. However, much of chemistry and processes underlying the battery system still remain to be understood, among which are the interphases between electrodes and electrolyte, known as solid electrolyte interphases (SEIs). Of special interest are the characteristics of SEI layer formed at the negative electrode electrolyte interface because this passivation layer is one of the dominant parameters that can influence the charge transfer kinetics and the overall capacity upon aging and cycling.<sup>2</sup> Therefore, the in-depth understanding about SEI chemistry and formation is essential for optimizing the LIB systems through tuning the composition and quality of the SEI layer on the surface of the negative electrodes.

We have recently utilized Electrochemical Quartz Crystal Microbalance with Dissipation (EQCM-D) to characterize SEI formation on tin thin film electrodes and probe the mass gain/loss during lithium intercalation-deintercalation processes upon electrochemical cycling. As shown in Figure 1, the mass change at the Sn/electrolyte interface was monitored in real time by measuring the decrease/increase of the resonance frequency in the cell. The results revealed that Li insertion is accompanied by apparent accumulation of surface species that partially dissolve during the anodic steps. Also the effects of the electrolyte additive fluoroethylene carbonate (FEC) on SEI formation are carefully studied and presented (Figure 2).

The key points of employing EQCM-D in this work are the extension of measurements to higher harmonics and incorporation of dissipation. The capabilities enable the detection of viscoelastic variation which contributes to the measured frequency shifts particularly in the presence of liquid medium. Therefore, given the unique advantage of EQCM-D, it is expected that the full picture of SEI formation, including mechanic properties and compositional change, can be reflected.



**Figure 1.** Typical EQCM-D experiment for the realtime acquisition of frequency shifts and corresponding mass change on Sn surface during 5 consecutive electrochemical cycles in Gen 2 electrolyte.



**Figure 2.** Frequency comparison between Gen 2 with/without FEC additives.

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## References

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