

INTERFACIAL PROCESSES IN MODEL LITHIUM ION SYSTEMS PROBED WITH VIBRATIONAL SUM FREQUENCY GENERATION SPECTROSCOPY

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Vibrational sum frequency generation (SFG) spectroscopy was used to probe electrochemical processes taking place at the interface between metal anodes and the liquid phase in model lithium ion systems.

Lithium ion batteries have been extensively studied and characterized by numerous techniques. However, the mechanisms behind many properties are still unclear due to the lack of techniques that can directly probe them in situ. The formation of the electrode passivating layer known as solid-electrolyte interphase (SEI) is one such example. During the first charging cycle of a battery, some of the electrolyte undergoes reduction at the electrode surface forming an electrically isolating barrier that prevents the subsequent reduction of more electrolyte molecules.

The SFG selection rules suppress signals from molecules in centrosymmetric environments such as electrolyte layers, so SFG is a selective probe of interfacial environments such as the SEI.

In this study, ethylene carbonate's (EC) response to potential cycling was observed. EC is commonly used as a high permittivity solvent in batteries and is widely believed to be the main component of the SEI in its reduced form, lithium ethylene dicarbonyl. EC's carbonyl stretch (1850 cm^{-1}) was measured in conjunction with cyclic voltammetry experiments. The SFG intensity showed remarkable agreement with the changing potential, as seen in the figure below. The shoulders on each side of the peaks in (a) are especially interesting, as they correspond to the potentials where lithium metal is oxidized and reduced. Vibrational modes found at $1300\text{-}1400\text{ cm}^{-1}$, usually assigned to the reduced form of EC, are also being studied in order to provide more information on the nature of the SEI.

