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Heterogeneity and Fluctuations in Electrochemical Sensors JEAN-LUC FRAIKIN, MICHAEL REQUA, MICHAEL STANTON, ANDREW CLELAND, UC Santa Barbara — Metal electrodes submerged in aqueous electrolytes biased with very small voltages frequently display a capacitive low-frequency electrical impedance, which is primarily imaginary but typically displays a $1/f^\alpha$ frequency dependence, with $0.7 \leq \alpha \leq 1$. This electrode-electrolyte interface is phenomenologically modeled as a constant phase element (CPE). There are a number of explanations for the observed frequency dependence, including geometric arguments based on the assumption of fractal surface geometries, but it is difficult to quantitatively match such models to experiment. We propose a new model to explain this phenomenon, as well as other low frequency electrical characteristics of the electrode-electrolyte interface, using a model that relies on microscopic heterogeneity, allowing for local variations in capacitance and diffusion coefficients. We will present the basic aspects of our model, and describe measurements under way to validate this model, using a combination of impedance measurements and electrochemical noise spectroscopy.

Jean-Luc Fraikin
UC Santa Barbara

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