

The faculty of Engineering of the Vrije Universiteit Brussel invites you to attend the public defense leading to the degree of

DOCTOR OF ENGINEERING SCIENCES

of **Freja Vandeputte**

The public defense will take place on **Friday 6th November 2026 at 5pm** in room **D.0.07** (Building D, VUB Main Campus)

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**FREQUENCY-DOMAIN ESTIMATION OF FRACTIONAL-ORDER MODELS
FOR ELECTROCHEMICAL IMPEDANCES**

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Abstract of the PhD research

Electrochemical impedance spectroscopy (EIS) is a widely-used non-invasive technique for characterising electrochemical systems. By measuring the voltage response to a current excitation, or vice versa, the impedance of the system can be estimated. Commonly, EIS is used as a nonparametric, purely data-driven estimation method, meaning that the impedance is only estimated at the excited frequencies. In this thesis, however, we propose parametric, physics-informed alternatives, based on equivalent circuit models (ECMs) for the impedance. Specifically, we use ECMs containing fractional-order elements, like Warburg diffusion elements and constant phase elements, in order to model low-frequency diffusion and non-ideal capacitors.

The thesis considers two electrochemical applications: batteries and the photoelectrochemical (PEC) cells. In classical EIS, an impedance is assumed to be a linear time-invariant (LTI) system. When a battery is under operation however, so when it is charging or discharging, its impedance not only behaves in a nonlinear way, but it also varies over time. Hence, operando EIS was introduced for estimating a time-varying impedance. This work thus develops parametric estimation methods for both classical and operando EIS data from batteries. The resulting fractional-order, optionally timevarying, impedance model has the advantage that it can be evaluated in every frequency of interest. To estimate the arbitrary fractional orders, we present a separable total least squares estimator, which first eliminates the linear parameters using their total least squares solution, and then minimises the resulting nonlinear least squares problem over the fractional orders. For PEC cells, classical EIS is used to estimate the reaction rate constants of the oxygen evolution reaction at the photoanode. Therefore, this work details a sample maximum likelihood estimator that integrates not only multiple frequencies, but also multiple potentials simultaneously.