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Variance-based Global Sensitivity Analysis: A Methodological Framework and Case Study for Microkinetic Modelling

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Abstract

Uncertainty in the physical modelling parameters is inherent to all chemical models. Sensitivity analysis has proven to be a useful tool to quantify and trace this uncertainty to specific physical input parameters and also to gain insight into the model input-output relationships. In this work, we present the methodological procedure of a prominent global sensitivity analysis method, Sobol's variance-based method, for a chemical modelling audience, and demonstrate the advantages of the application of this method specifically in the context of microkinetic modelling.

In Sobol's method, the fractional contribution of each of the physical parameters to the model output variance is represented in first-order and total-effect sensitivity indices. These describe a parameters' contribution excluding and including its interaction with other parameters, respectively. The analysis constitutes of Monte-Carlo-style model simulations in which the physical parameters are sampled following the quasi-random Sobol's sampling sequence. As the computational cost of Monte-Carlo simulations is the primary limitation of variance-based analyses, we take advantage of three common sensitivity index convergence criteria, that vary in required computational effort: (1) parameter screening, (2) parameter ranking and (3) full convergence of the analysis results. Each is able to provide distinct information regarding the influence of the physical input parameters on the output uncertainty.

We aim for Sobol's method to be easily applicable to other models within the field of chemical modelling and beyond. Therefore, we developed the method in this study as an analysis framework. In this framework, the procedure to determine the sensitivity remains largely separated from the analysed model, which can therefore be altered or exchanged independently.

We successfully demonstrate the application of this method by means of two example cases from the field of microkinetic modelling that differ in complexity and number of input parameters: a CO oxidation model, and a model describing the oxygen evolution reaction occurring at the photo-anode within a photo-electrochemical cell. We compare the analysis results obtained with the different convergence criteria and present several commonly used visualisation methods. The results obtained give insights into which parameters contribute most and least to the output uncertainty. Based on this, we propose a workflow incorporating the sensitivity analysis into the process of system modelling, which is aimed at reducing the output uncertainty and improving the modelled system.