Global Sensitivity Analysis of the Single Particle Lithium-Ion Battery Model with Electrolyte

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***Abstract*— The importance of global sensitivity analysis (GSA) has been well established in many scientific areas. However, despite its critical role in evaluating a model's plausibility and relevance, most lithium ion battery models are published without any sensitivity analysis. In order to improve the lifetime performance of battery packs, researchers are investigating the application of physics based electrochemical models, such as the single particle model with electrolyte (SPMe). This is a challenging research area from both the parameter estimation and modelling perspective. One key challenge is the number of unknown parameters: the SPMe contains 31 parameters, many of which are themselves non-linear functions of other parameters. As such, relatively few authors have tackled this parameter estimation problem. This is exacerbated because there are no GSAs of the SPMe which have been published previously. This article addresses this gap in the literature and identifies the most sensitive parameter, preventing time being wasted on refining parameters which the output is insensitive to.**

Keywords*— *Lithium ion battery, Modelling, Sensitivity Analysis, Error Propagation, Single Particle Model with Electrolyte

I. Introduction

Sensitivity analysis (SA) aims to quantify how the uncertainty in the output of a mathematical model (numerical or otherwise) can be attributed to the model input factors' (MIF) uncertainty [1, 2], as illustrated in Fig. 1. In the context of SA, MIFs include every type of model inputs, e.g. assumptions, errors in the data, resolution, and parameters.

In practice, the analysis normally involves computing the model output with alternative assumptions or different input values with the aim of determining the influence of an assumption or variable on the model output. SA is commonly performed on the model parameters because this analysis establishes confidence in the model simulations by evaluating model robustness, i.e. how sensitive the model is to changes in parameters[3]. Furthermore, this process enables each parameter to be ranked with respect to its contribution to the uncertainty in the model output. Identifying the sensitive model parameters prevents time being wasted on refining parameters which the output is insensitive to [4]. Ultimately, SA underpins the better model development and can identify important connections between model input and outputs [5, 6].

The critical role of SA in the process of building models has has been well established in many scientific areas [7]. It is also

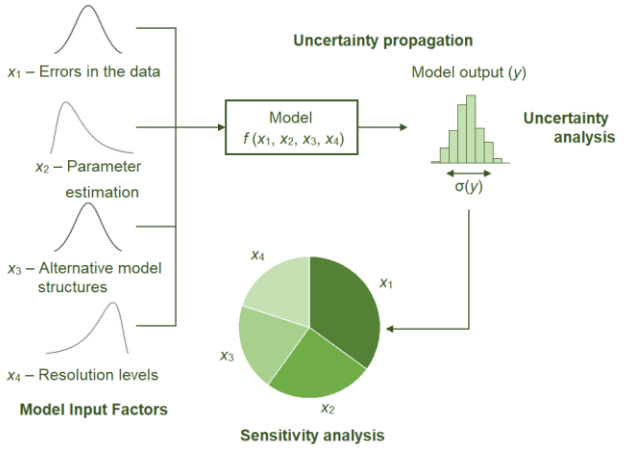

Fig. 1. Sensitivity analysis diagram

included in international institutions official guidelines, such as the United States Environmental Protection Agency [8], the White House Office of Management and Budget [9], and the European Commission [10].

Despite its importance in evaluating a model's plausibility and relevance, most lithium ion battery (LIB) models are published without SA, for example see [11–18]. This is a common issue across numerous scientific disciplines and affects even high impact journals; a recent review in Science and Nature exposed that only 4% of modelling and SA papers contained a global SA [19]. Global methods consider the sensitivity across the whole input space as opposed to local methods, which only investigate perturbations around a single 'nominal value' point [20]. Local methods are unable to detect the presence of interactions between MIFs [21] and it is universally accepted in the statistical literature that they are inadequate; e.g. Saltelli & Annoni 2010 have geometrcially demonstrated local methods are perfunctory due to the so called *curse of dimensionality* [22].

LIBs are monitored and maintained within their region of safe operations by battery management systems (BMS), which also use mathematical models to estimate important metrics that cannot be measured directly: e.g. the battery state of charge (SOC: remaining energy compared to a fully charged battery)

and its state of health (remaining capacity when compared to a new battery). Lumped parameter equivalent circuit models (ECM) are commonly used in conventional BMS since they can be implemented in real time. However, these models rely on experimental data and therefore the battery behaviour predicted cannot be extrapolated beyond the experimental range. The ECMs also struggle with low temperatures, low SOC and large depth of discharge (DOD). Under these conditions, the model accuracy is greatly reduced. Furthermore, accounting for battery degradation (capacity and power fade) is challenging due to the lack of physical significance of the model parameters. In order to improve the lifetime performance of battery packs, researchers are investigating the application of physics based electrochemical models instead of ECMs in the BMS and the use of higher fidelity models as part of an integrated BMS telemetry system. Due to their physical basis, these models are likely to maintain good accuracy at low temperatures, low SOCs and large DOD. Furthermore, such first-principle models can provide insights into battery ageing because they can more easily be coupled to degradation models. Physics based models could be solved online in next-generation BMSs to enable health-aware management and control algorithms that use internal electrochemical safety limits (e.g. local overpotential or lithium concentration) instead of the commonly employed terminal voltage and ad-hoc safety limits (e.g. temperature current). However, this is a challenging research area from both the parameter estimation and modelling perspective. One key challenge is the number of unknown parameters: the single particle model with electrolyte (SPMe), which is a simplified version of the "Doyle-Fuller-Newman" pseudo-2-D lithium-ion battery model, contains 31 parameters, many of which are themselves non-linear functions of other parameters. In general, parameters in this model are obtained from the literature, and whilst this is useful for initial design studies of cell performance, it has been asserted that the model must be parametrised specifically for the cells used in the pack if it is for use in a BMS. In order to support researches tackling this parameter estimation problem, this article presents a global sensitivity analysis on the SPMe.

A. Aims

The objective is to identify the most sensitive parameters in the SPMe. This may offer insights into the underlying mechanisms that govern the system and improve model understanding, as well as highlight expected future areas of research.

II. Literature Review

A full literature review of LIB SA is available in [23], which is briefly outlined in this section.

Schmidt *et al.*, 2010 [24], Rahimi-Eichi *et al.* 2013 [25], and Pózna *et al.* 2017 [26] have used local methods to perform SA on LIB ECM. Whilst ECM have a linear model structure, they are non-linear in the parameters (functions of SOC and temperature). Consequently, local SA is not suitable to evaluate the robustness of ECM based inference as the model should be linear (or at least additive) [22]. Similarly, local sensitivity studies of Doyle-Fuller-Newman model [25, 26, 24, 27] are not robust due to the so called *curse of dimensionality* [22].

Other SA of the ECM and physicochemical model [28–32] are based on scenario analyses, that is to say the methods only consider certain parameter combinations (scenarios). Factorial designs are superior since they study all the possible combinations. In addition, the scenario analyses have only considered minor parameter variations, i.e. small perturbations as oppose to exploring the full multidimensional input space. Limiting the analyses to narrow subsections within the range of possible parameter values is likely to bias results, as information is lost. Lastly, these methods are incapable of detecting, let alone quantify, the presence of interactions between MIFs.

Zhao & Howey 2016 [33] have implemented the Morris screening method [34] and the so called enhanced Morris screening method [35] to perform an appropriate global SA on a linear second order ECM. One limitation of this approach is that it does not discern between the main effect and interactions between parameters. In practice, we are usually interested in the main effects [36] therefore it is important that this effect is not biased by interactions.

Lastly, Lin *et al.* 2018 [37] used Sobol indices to perform a GSA of LIB 3D multiphysics model with 46 parameters. However, instead of using Monte Carlo methods, the indices are computed with polynomial chaos expansion, which suffers from the *curse of dimensionality* [22]. We have recently compared GSA methods for LIB coupled electro-thermal models [23], however LIB physics based electrochemical models are yet to be analysed. This article addressed this gap in the literature.

III. Methodology

A. Single Particle Model

In this paper, a single particle model with electrolyte (SPMe) dynamic is used as a physics based electrochemical battery model. The SPMe model proposed by [38] is a simplification of the Newman model in [39]. The SPMe is derived under several assumptions regarding [38]. The governing equations of the SPMe model are

$$\frac{\partial c_s^{\pm}}{\partial}(r,t) = \frac{1}{r^2}\frac{\partial}{\partial r}\left[D_s^{\pm} r^2 \frac{\partial c_s^{\pm}}{\partial r}(r,t) \right] \tag{1}$$

$$c_{ss}^{\pm}(t) = c_s^{\pm}(R_s^{\pm}, t) \tag{2}$$

$$\frac{\partial c_e^j}{\partial}(x,t) = \frac{\partial}{\partial x}\left[\frac{D_e(\varepsilon_e^j)^b}{\varepsilon_e^j}\frac{\partial c_e^j}{\partial x}(x,t) \right] \mp \frac{(1-t_c^0)}{\varepsilon_e^j F L^j} I(t) \tag{3}$$

$$\text{for } \; j \in \{-, s, +\}$$

$$\Phi_e^+(t) - \Phi_e^-(t) = \frac{L^+ + 2L^{sep} + L^-}{2\kappa} I(t) + \frac{2RT}{F}(1-t_c^0)\ln\frac{c_e^+}{c_e^-} \tag{4}$$

$$j_n^{\pm}(t) = \mp \frac{I(t)}{F a^{\pm} L^{\pm}} \tag{5}$$

$$\eta^{\pm}(t) = \frac{RT}{\alpha F}\sinh^{-1}\left(\frac{\mp I(t)}{2a^{\pm}L^{\pm}i_0^{\pm}(t)}\right) \quad (6)$$

$$i_0^{\pm}(t) = k^{\pm}\left[c_{ss}^{\pm}(t)\right]^{\alpha}\cdot\left[c_e^{\pm}(t)\left(c_{s,\max}^{\pm} - c_{s,sur}^{\pm}(t)\right)\right]^{\alpha} \quad (7)$$

$$\Phi_s^{\pm}(t) = \eta^{\pm}(t) + \Phi_e^{\pm}(t) + U^{\pm}\left(c_{ss}^{\pm}(t)\right) + FR_f^{\pm}j_n^{\pm}(t) \quad (8)$$

where the nomenclature, 9 fixed parameters and 22 model input factors (MIF) are defined in Tables I, II, and III respectively, and the specific interfacial surface area is given by

$$a^{\pm} = \frac{3\varepsilon_s^{\pm}}{R_s^{\pm}}. \quad (9)$$

Full details of the derivation of the SPMe equations are published in [38]. Briefly, (1-7) comprise the solid and electrolyte phase lithium concentrations, and the overpotential is found by solving the Butler-Volmer kinetics equations. In the SPMe, each electrode is idealised as a single spherical porous particle, and the molar ion flux $j_n^{\pm}$ is as proportional to current $I(t)$ as in (5). The lithium concentration in the solid phase for both positive c_s^+ and negative electrodes c_s^- in SPMe are expressed as (1). The electrolyte phase lithium diffusion follows (2). The block diagram of the SPMe is shown in Fig. 2. The terminal voltage output is a nonlinear function of solid state concentrations $c_s^{\pm}$, electrolyte state concentrations $c_e^{\pm}$, and current $I(t)$.

The SPMe model still contains partial-differential equations (PDE) to express the concentrations. The PDEs can be solved by finite difference or finite element method. However, these methods result in a very high order model (circa 350th order). Model order reduction techniques can be used to develop a lower order model that is suitable for control-oriented modelling. Residue grouping method is applied to the SPMe model to reduce the model order number to 16. The solid-state diffusion equations of the two electrodes employ residue grouping analytically; conversely, the liquid-state diffusion applied residue grouping numerically. The process to solve the PDE in SPMe model is identical to [40].

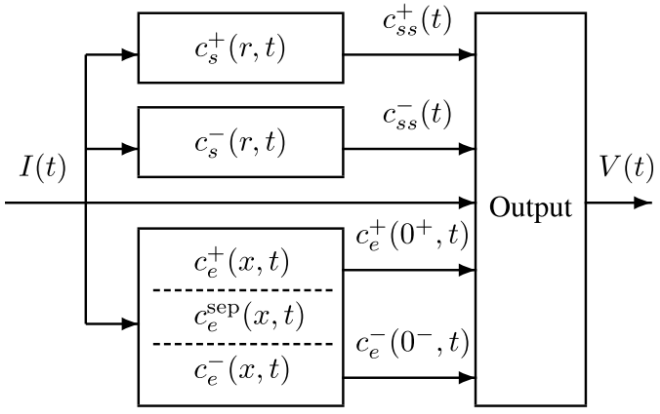

Fig. 2. Block diagram of SPMe. The subsystems in each block are independent of one another [38]

TABLE I. NOMENCLATURE

States and variables	Symbol	Unit
Lithium concentration in solid phase	$c_s^{\pm}$	molm^{-3}
Lithium concentration in anode	c_e^-	molm^{-3}
Lithium concentration in electrolyte	c_e^s	molm^{-3}
Lithium concentration in cathode	c_e^+	molm^{-3}
Solid electrode potential	$\Phi_s^{\pm}$	V
Electrolyte electric potential	$\Phi_e^{\pm}$	V
Molar ion flux	$j_n^{\pm}$	molm^{-3}s^{-1}
Exchange current density	$i_0^{\pm}$	Am^{-2}
Over-potential	$\eta^{\pm}$	V
Li conc. at solid particle surface	$c_{ss}^{\pm}$	molm^{-3}
Open circuit voltage (anode)	U^-	V
Open circuit voltage (cathode)	U^+	V
Distance from particle centre	r	μm
Distance from anode collector	x	μm
Time	t	s
Applied current	I	A
Terminal voltage	V	V

TABLE II. FIXED PARAMETERS

Transport parameters	Symbol	Nominal value
Charge transfer coefficients	α	0.5 Ωm^{-2}
Anode-electrolyte resistivity	R_f^-	0 Ωm^{-2}
Cathode-electrolyte resistivity	R_f^+	0 Ωm^{-2}
Conductivity of electrolyte	κ	0.95 Sm^{-1}
Faraday's constant	F	96485.33289 Cmol^{-1}
Universal gas constant	R	8.314472 Jmol^{-1}K^{-1}
Fixed electrolyte concentration	c_e	1000 molm^{-3}
Cell maximum voltage	V_{max}	4.115 V
Cell minimum voltage	V_{min}	2.5 V

TABLE III. MODEL INPUT FACTORS

Geometric	Symbol	Nominal value	Range
Thickness of (anode)	L^-	22 μm	± 1 μm
Thickness of separator	L^s	20 μm	± 1 μm
Thickness of (cathode)	L^+	35 μm	± 1 μm
Particle Radii			
Anode	R_s^-	11.5 μm	± 2 μm
Cathode	R_s^+	14.5 μm	± 2 μm
Volume fractions			
Anode	ε_s^-	0.635	± 0.085
Cathode	ε_s^+	0.476	± 0.054
Electrolyte (anode)	ε_e^-	0.295	± 0.160
Electrolyte for separator	ε_e^s	0.44	± 0.10
Electrolyte (cathode)	ε_e^+	0.443	± 0.112
Diffusion coefficient			
Solid for anode	D_s^-	0.1 μm^{2}s^{-1}	0.1 – 1 nm^{2}s^{-1}
Solid for cathode	D_s^+	0.8 μm^{2}s^{-1}	0.1 – 1 nm^{2}s^{-1}
Electrolyte	D_e	278.8 μm^{2}s^{-1}	± 10%
Miscellaneous			
Bruggeman porosity	b	1.5	1 – 3
Ambient temperature	T	298.15 K	± 1 K
Transference number	t_c^0	0.363	0.350 – 0.400
Nominal Reaction rates			
Anode	k^-	5 x 10^{-6}	5 x 10$^{-6\,-\,-12}$
Cathode	k^+	7 x 10^{-6}	5 x 10$^{-6\,-\,-12}$
Maximum Concentrations			
Anode	c_m^-	31,389 molm^{-3}	± 10%
Cathode	c_m^+	36,292 molm^{-3}	± 10%
Initial conditions			
Anode 100%	c_{100}^-	28,320	± 1,000
Cathode 100%	c_{100}^+	3,153	± 1,000

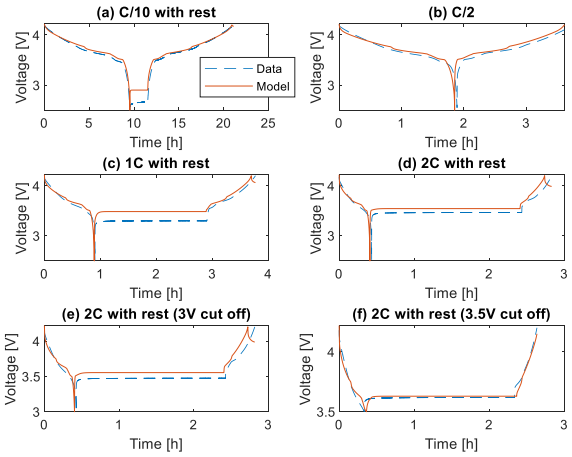

Fig. 3. Coin cell single particle model with electrolyte (SPMe) predictions vs real data for different scenarios

Six scenarios, as shown in Fig. 3, are used as inputs into the coin cell model:

(a) C/10 discharge followed by a 2h rest and C/10 charge,
(b) C/2 discharge, immediately followed by a C/2 charge,
(c) 1C discharge followed by a 2h rest and 1C charge,
(d) 2C discharge followed by a 2h rest and 2C charge,
(e) 2C discharge to 3V followed by a 2h rest and 2C charge,
(f) 2C discharge to 3.5V followed by a 2h rest and 2C charge.

B. *Sensitivity Analysis Method*

The most commonly used screening methods are based on elementary effects and have relatively low computational cost [34, 35]. The Morris screening method is used in this article as it is the most complete and most costly screening technique [42].

IV. Results and Discussion

A. *Morris Method*

The results of the Morris screening method for the C/10 discharge followed by a 2h rest and C/10 charge are shown in Fig. 1(a). The anode volume fraction, ε_s^-, anode thickness, L^-, anode diffusion coefficient, D_s^-, cathode diffusion coefficient, D_s^+, anode initial condition, c_{100}^-, and cathode volume fraction, ε_s^+, have strong non-linear effects and/or interactions effect (large μ and large σ). The anode particle radius, R_s^-, maximum cathode concentration, c_m^+, and cathode initial condition, c_{100}^+, have average non-linear effects and/or interactions effect (average μ = 18.1mV and average σ =24.3mV). The remaining MIFs are deemed to have no effect since they are one order of magnitude smaller than ε_s^- (μ < 4.6mV).

The results of the Morris screening method for the C/2 discharge, immediately followed by a C/2 charge are shown in Fig. 1(b). The cathode diffusion coefficient, D_s^+, anode volume fraction, ε_s^-, anode thickness, L^-, anode diffusion coefficient, D_s^-, and anode particle radius, R_s^-, have strong non-linear effects and/or interactions effect (large μ and large σ). The cathode volume fraction, ε_s^+, cathode initial condition, c_{100}^+, maximum cathode concentration, c_m^+, anode initial condition, c_{100}^-, separator thickness, L_s, cathode thickness, L^+, maximum anode concentration, c_m^-, and electrolyte separator volume fraction, ε_e^s, have average non-linear effects and/or interactions effect (average μ = 10.7mV and average σ = 26.6mV). The remaining MIFs are deemed to have no effect since they are one order of magnitude smaller than D_s^+ (μ < 4.0mV).

The results of the Morris screening method for the 1C discharge followed by a 2h rest and 1C charge are shown in Fig. 1(c). The anode diffusion coefficient, D_s^-, and cathode diffusion coefficient, D_s^+, have strong non-linear effects and/or interactions effect (large μ and large σ). The anode particle radius, R_s^-, anode volume fraction, ε_s^-, cathode volume fraction, ε_s^+, maximum cathode concentration, c_m^+, and anode initial condition, c_{100}^-, have average non-linear effects and/or interactions effect (average μ = 13.9mV and average σ =14.8mV). The remaining MIFs are deemed to have no effect since they are one order of magnitude smaller than D_s^+ (μ < 6.2mV).

The results of the Morris screening method for the 2C discharge followed by a 2h rest and 2C charge are shown in Fig. 1(d). The anode diffusion coefficient, D_s^-, has strong non-linear effects and/or interactions effect (large μ = 154mV and large σ = 145mV). The cathode diffusion coefficient, D_s^+, anode volume fraction, ε_s^-, and anode particle radius, R_s^-, have average non-linear effects and/or interactions effect (average μ = 15.4mV and average σ =19.7mV). The remaining MIFs are deemed to have no effect since they are one order of magnitude smaller than D_s^+ (μ < 12.3mV).

The results of the Morris screening method for the 2C discharge to 3V followed by a 2h rest and 2C charge are shown in Fig. 1(e). The anode diffusion coefficient, D_s^-, has strong non-linear effects and/or interactions effect (large μ = 97.6mV and large σ = 151mV). The anode volume fraction, ε_s^-, cathode diffusion coefficient, D_s^+, anode particle radius, R_s^-, anode thickness, L^-, cathode volume fraction, ε_s^+, have average non-linear effects and/or interactions effect (average μ = 10.8mV and average σ =16.1mV). The remaining MIFs are deemed to have no effect since they are one order of magnitude smaller than D_s^+ (μ < 8.5mV).

The results of the Morris screening method for the 2C discharge to 3.5V followed by a 2h rest and 2C charge are shown in Fig. 1(f). The anode diffusion coefficient, D_s^-, and cathode diffusion coefficient, D_s^+, have strong non-linear effects and/or interactions effect (large μ and large σ). The anode volume fraction, ε_s^-, cathode volume fraction, ε_s^+, cathode electrolyte volume fraction, ε_e^+, maximum cathode concentration, c_m^+, anode initial condition, c_{100}^-, anode particle radius, R_s^-, and anode volume fraction, ε_s^-, have average non-linear effects and/or interactions effect (average μ = 9.5mV and average σ =17.7mV). The remaining MIFs are deemed to have no effect since they are one order of magnitude smaller than D_s^+ (μ < 5.6mV).

The results shown in Fig. 1 reveal that the SPMe sensitivity varies significantly for different input scenarios. Overall, the anode diffusion coefficient, D_s^-, and cathode diffusion coefficient, D_s^+, are the most sensitive model input factors (MIFs).

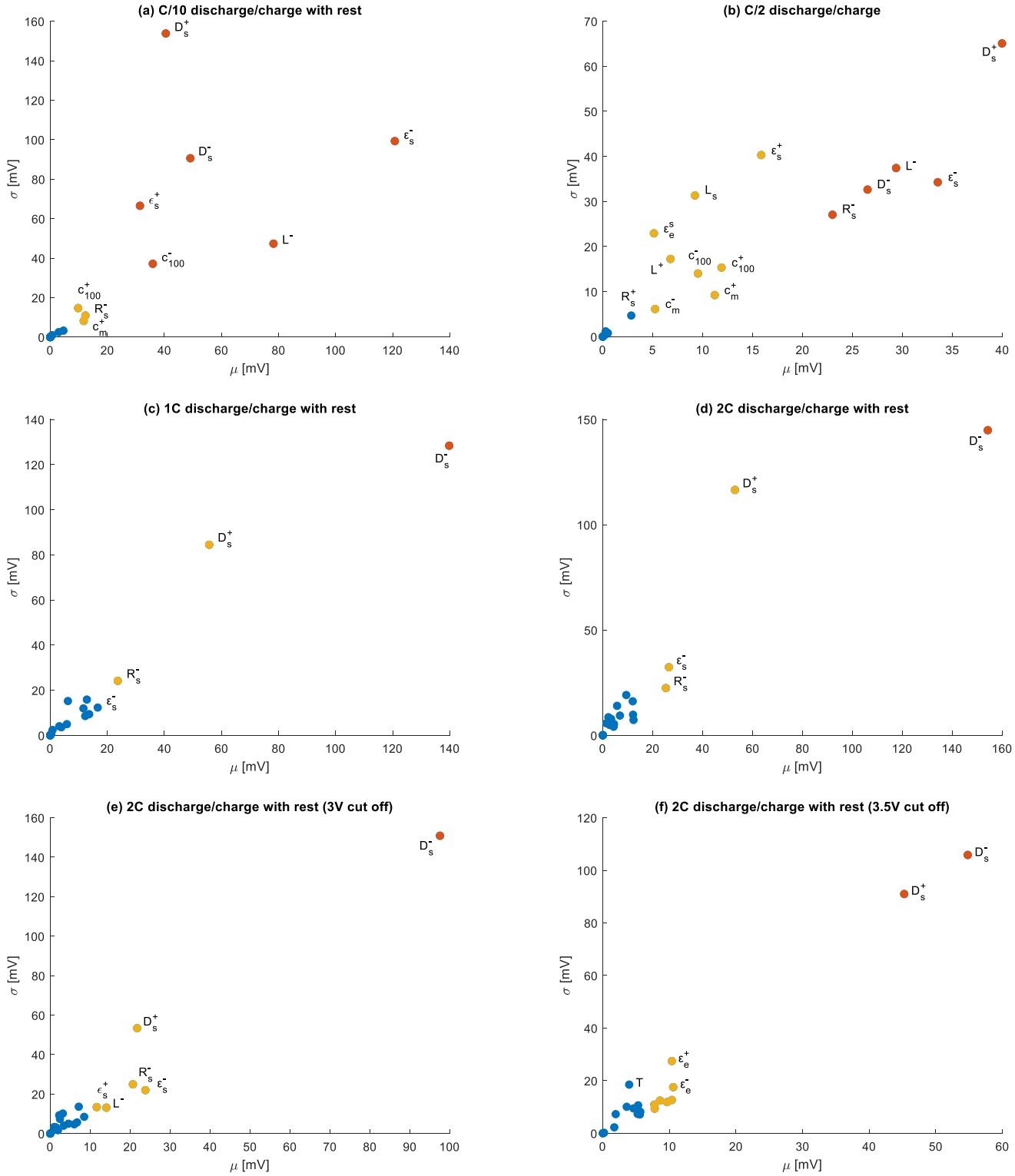

Fig. 4. Morris Screening results (strong non-linear effects and/or interactions effect – red, average strong non-linear effects and/or interactions effect – yellow, negligible effect – blue) for different scenarios

B. *Further work*

Now the most influential MIFs for the SPMe have been identified, a further sensitivity analysis is to be performed on the parameter estimation experiments. It is crucial that

parameters which are estimated from experimental data have a large first-order effect on the measured data. A parameter can be observable but have mainly second-order and higher effects on the experimental data. These are caused by interactions with other parameters and if these effects are larger than the effect of the parameter on its own (first-order effect) then the confidence in the numerical estimates for this parameter will be low. Since the MIFs identified are the most influential, it is crucial that the numerical estimates are accurate.

V. Conclusions

Battery engineers and researchers require an understanding of the sources and relative contribution of errors and inaccuracies in the lithium ion battery (LIB) models used. Both to improve the model development process but also to underpin more efficient experimentation and parameter estimation. Global sensitivity analyses (GSA) are crucial for evaluating model robustness and establishing confidence in the model simulations. It prevents time being wasted on improving the accuracy of model input factors (MIF) the output is insensitive to, which improves elucidation of the dominant mechanisms inherent in the model. These analyses are crucial for understanding the variance in model predictions, improving model accuracy, and validating the model.

This study uses the Morris Screening method to classify which parameters have a large effect. The anode diffusion coefficient, D_s^-, and cathode diffusion coefficient, D_s^+, are found to be the most sensitive model input factors across six different input scenarios.

Acknowledgment

This research is supported by WMG centre High Value Manufacturing (HVM) Catapult and Jaguar Land Rover. The EPSRC Faraday Institution Multi-Scale Modelling project (EP/S003053/1, grant number FIRG003)

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