Modelling inhomogeneous lithium plating with a distributed equivalent circuit network

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Model

The model used in this work is a thermally coupled distributed equivalent circuit model reflecting the true inner structure of the battery cell [1], upgraded to segregate the positive and negative half cell.

NE Potential = $V_{-} - V_{RE} < 0$ indicates plating risk

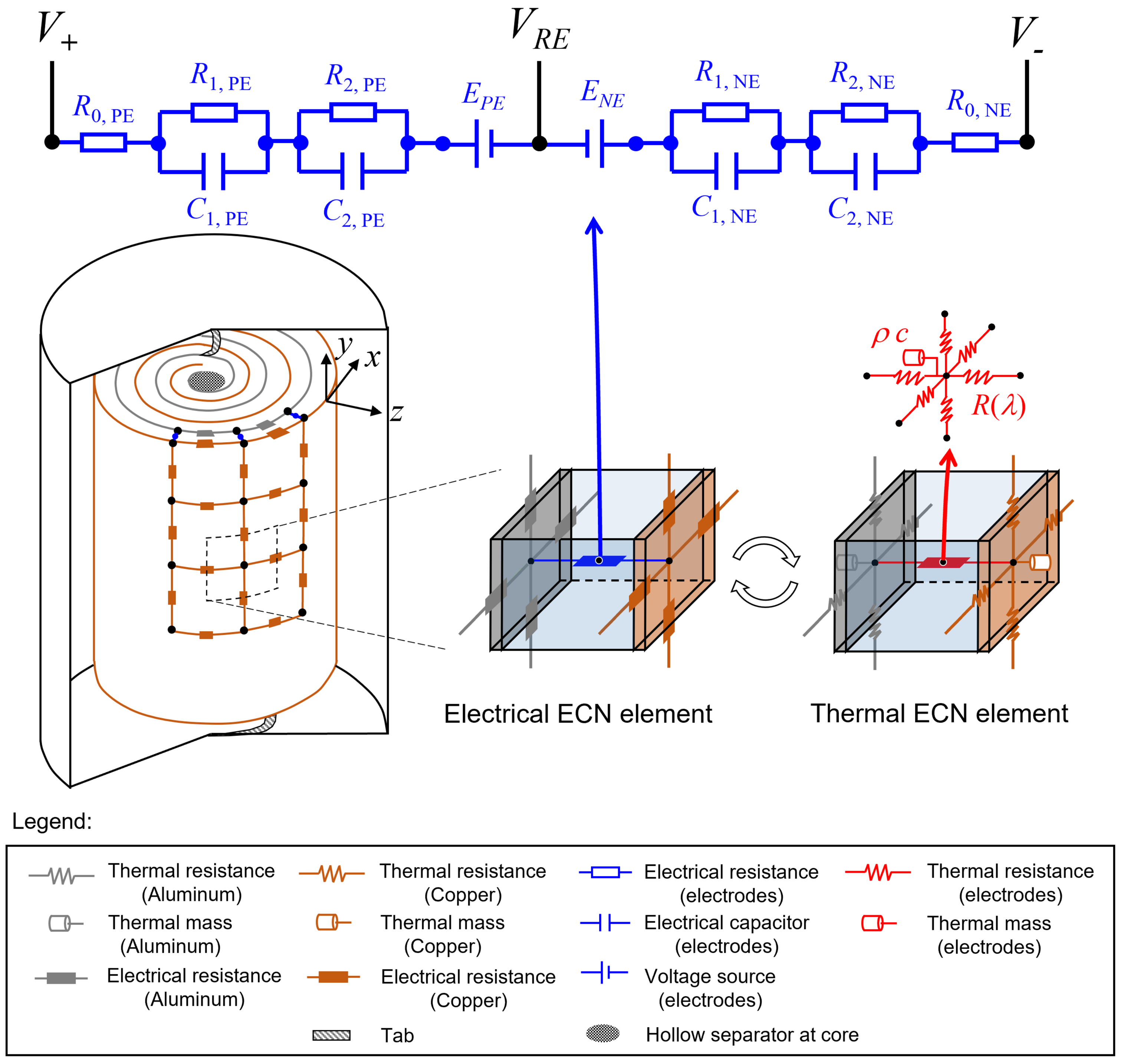

Fig 1: Structure of the cell in the distributed model, adapted from [1].

Parameterisation

- Predictions from a pseudo-2D isothermal model in PyBaMM [2] with parameters for the LGM50 (NMC/Graphite+Si) [3] used for parameterisation.
- Reference electrode placed on negative side of separator to enable extraction of half-cell potentials during GITT.
- Second-order (2RC-pair) equivalent circuit models parameterised from each half-cell GITT profile.

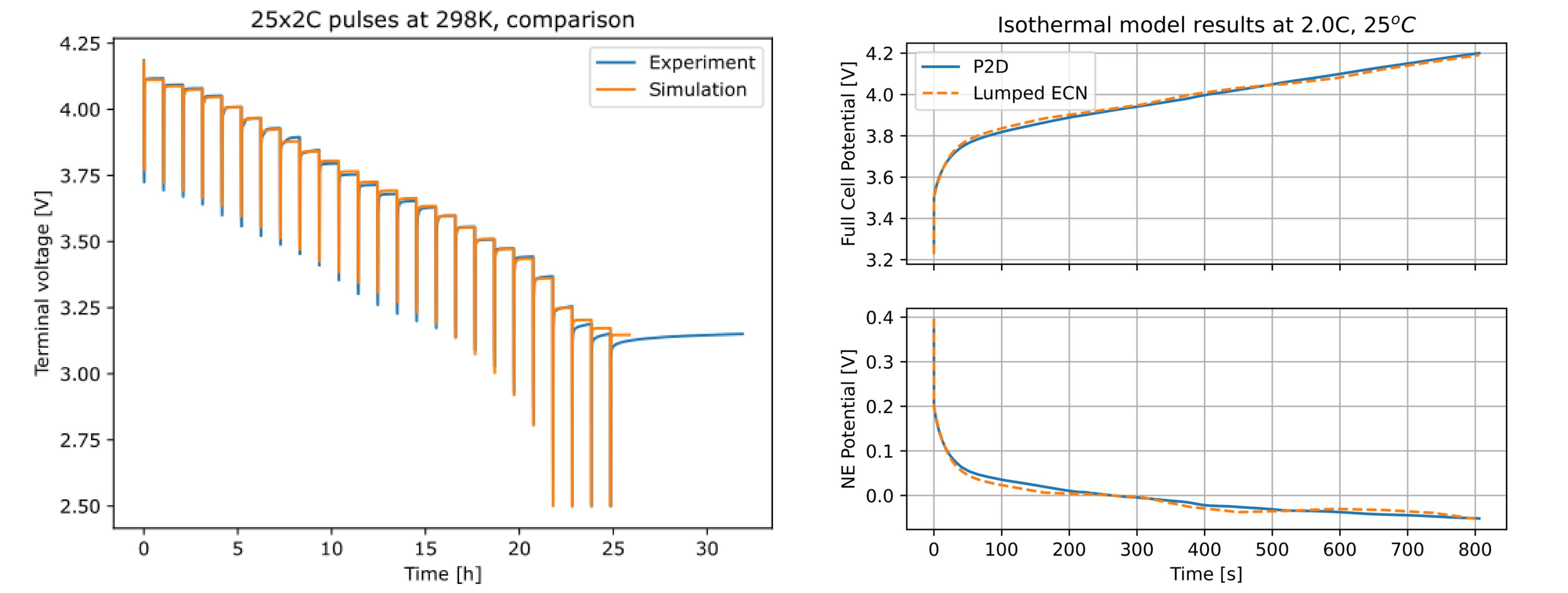

Fig 2: Validation of the pseudo-2D model against discharge GITT alongside outputs from the pseudo-2D model and the fitted lumped ECN model for a constant-current charge in isothermal conditions.

Results

For 2C charge from 10% SOC at 10°C ambient

(a) The lumped ECN model does not predict the negative electrode (NE) potential dropping below 0 V (Fig 3) because the tab placement causes significant current density gradients across the electrode.

(b) As the cell charges, a temperature gradient forms across the jelly roll which reduces the resistance in the core of the cell. Therefore, the NE potential is lowest on the colder outer surface of the cell.

(c) At the end of charge, the lowest NE potentials are found close to the tabs, where the electrode is in a higher state of charge leading to a larger diffusion resistance.

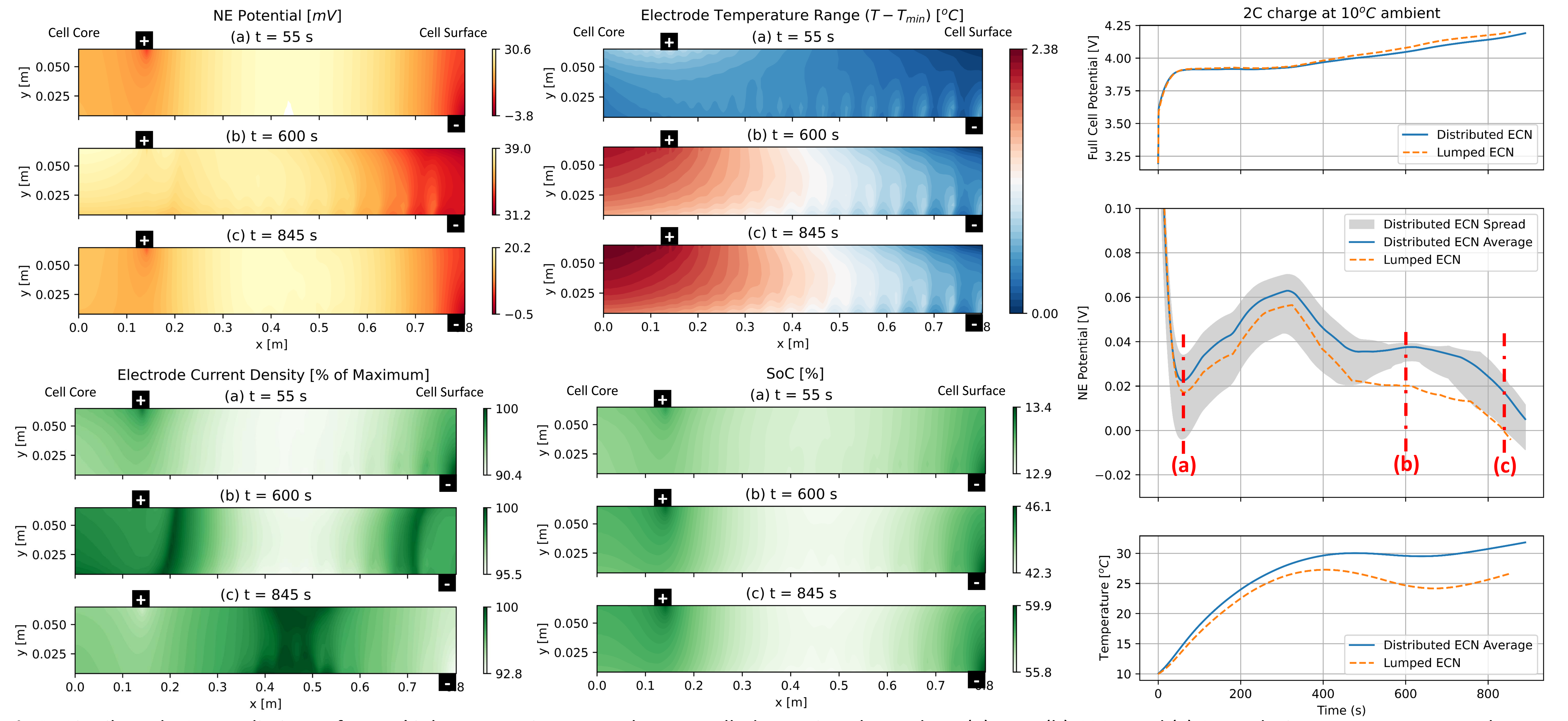

Fig 3: Distributed ECN predictions of states' inhomogeneity across the unravelled negative electrode at (a) 55 s, (b) 600 s and (c) 845 s during constant current charge. Cell potential and temperature plotted for distributed and lumped ECN models.

Conclusions

- The distributed model predicts favourable conditions to plating considerably earlier during charge than the lumped model.
- Local minima of the negative electrode potential result from the coupled effects of inhomogeneities in temperature, current density and state of charge.
- Such inhomogeneities are likely to arise during operating conditions associated with lithium plating, so understanding their effects is crucial for accurately predicting lithium plating in batteries.

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References

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[3] Kieran O'Regan *et al.* (2022), *Electrochimica Acta*, vol. 425, pp. 140600