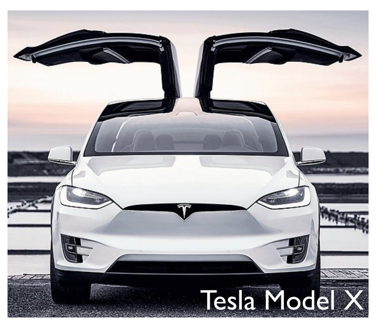
Numerical Investigation of Influence of Transport Properties of Highly Concentrated Electrolytes on Li-Dendrite Growth

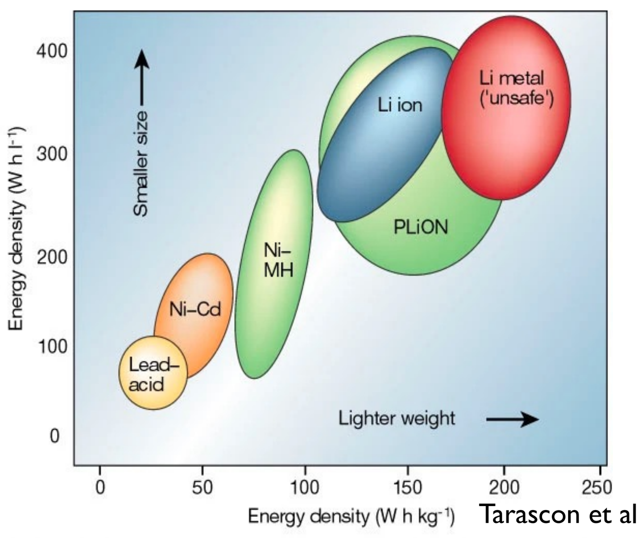
戴承寧 Chen-Ning Tai
Advisors: Chih-Hung Chen, Kuo-Ching Chen
Institute of Applied Mechanics, National Taiwan University, Taipei, Taiwan
Email: r10522834@ntu.edu.tw

國立臺灣大學 應用力學研究所

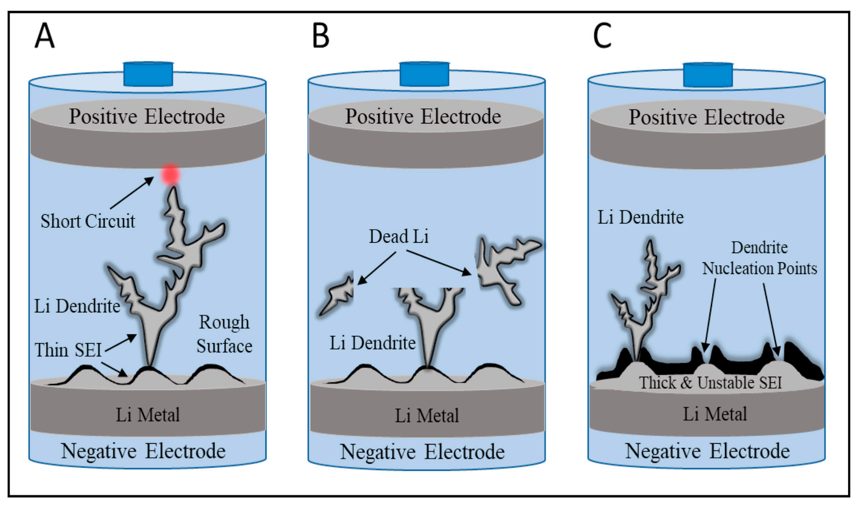
Motivation

- Recent battery technology can hardly meet the growing demands of high-energy storage in the battery market, such as electric vehicles.
- High-energy-density batteries made from the most promising anode material, Li, however, suffers from the formation of dendrites during charging/discharging, leading to its low battery performance and short cycling life.





Tarascon et al. (2001)

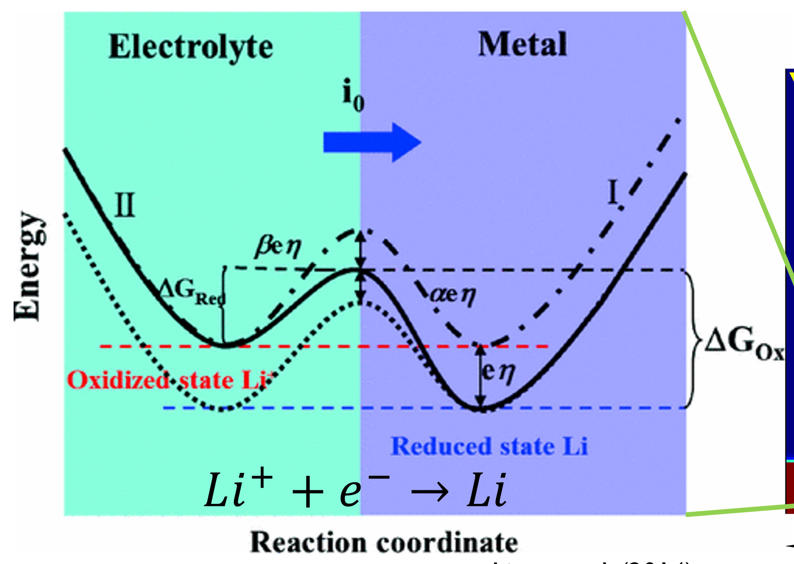


Foroozan et al. (2020)

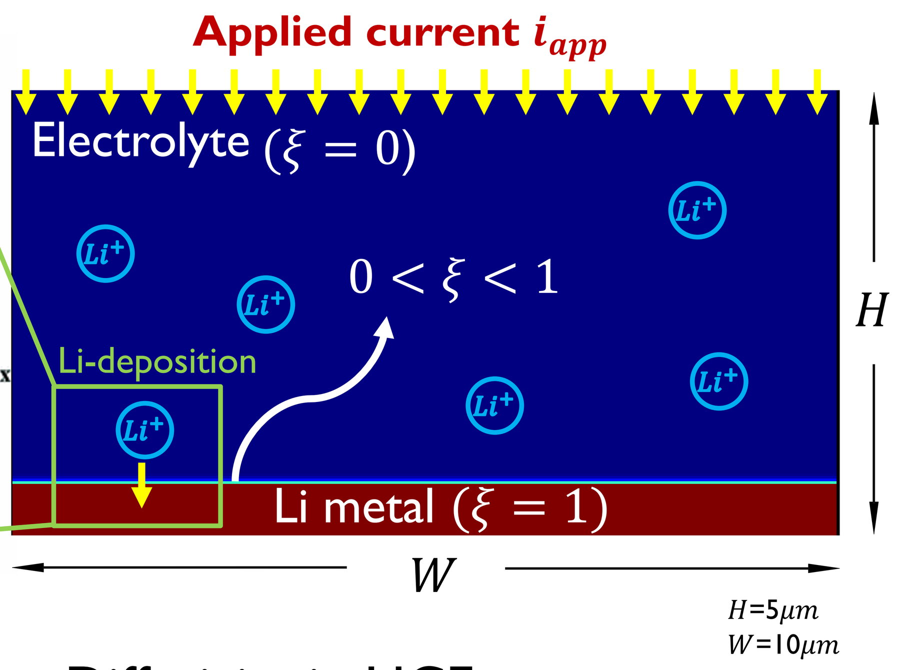
- Highly concentrated electrolytes (HCEs) are promising solutions to stabilize dendrite formation, but their transport properties are strongly influenced by salt concentration.
- In this study, the influence of transport properties of HCEs are systematically discussed by numerical method.

Methodology

Li-deposition process:



Liang et al. (2014)

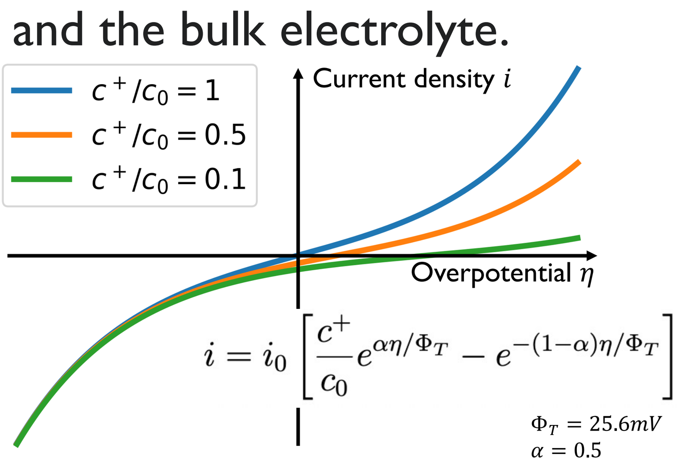


Ionic diffusion & migration:

$$\begin{cases} \frac{\partial c^+}{\partial t} = \nabla \cdot (\overbrace{D^+ \nabla c^+}^{diffusion} + \overbrace{\frac{D^+ c^+}{\Phi_T} \nabla \phi}^{Ele-migr.}) - \overbrace{\dot{R}_{Li^+}}^{reduction} \\ \frac{\partial c^-}{\partial t} = \nabla \cdot (D^- \nabla c^- - \frac{D^- c^-}{\Phi_T} \nabla \phi) \end{cases}$$

Butler-Volmer equation:

BV-equation describes how the electrical current i through an electrode depends on the voltage difference η between the electrode and the bulk electrolyte.

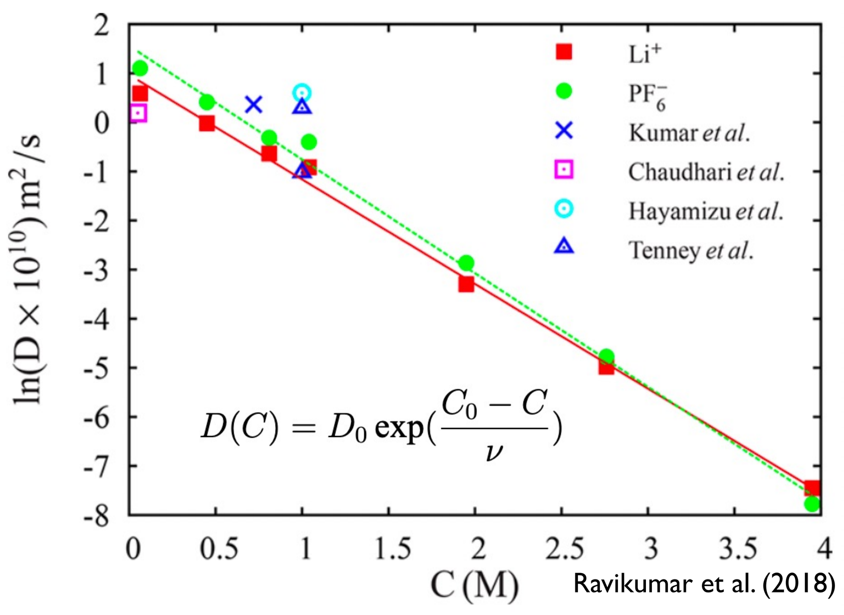


$$i = i_0 \left[\frac{c^+}{c_0} e^{\alpha\eta/\Phi_T} - e^{-(1-\alpha)\eta/\Phi_T} \right]$$

$\Phi_T = 25.6mV$
$\alpha = 0.5$

Diffusivity in HCEs:

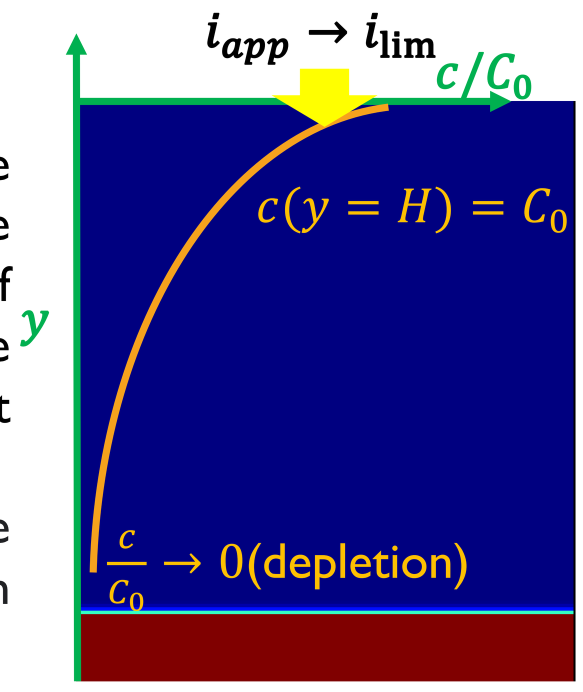
Diffusivity dominates the velocity of ionic transport in electrolytes and is highly dependent on salt concentration in HCEs.



Ravikumar et al. (2018)

Ionic depletion:

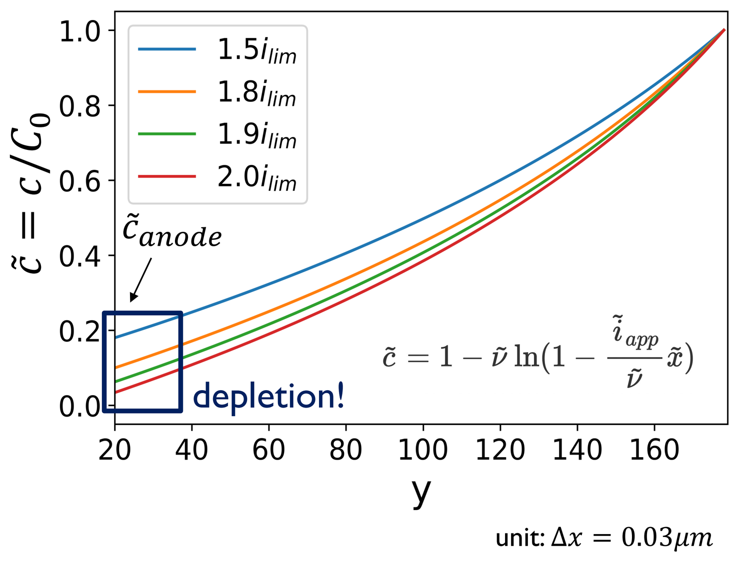
- Limiting current density i_{lim} refers to the largest sustainable current density that can be passed through the electrolyte, at this value of current density the ionic concentration at the anode is zero, and operating at current densities above this value results in cell failure.
- Rapid depletion of Li-ions on electrode surface is the underlying driving force for non-uniform electrodeposition of Li.



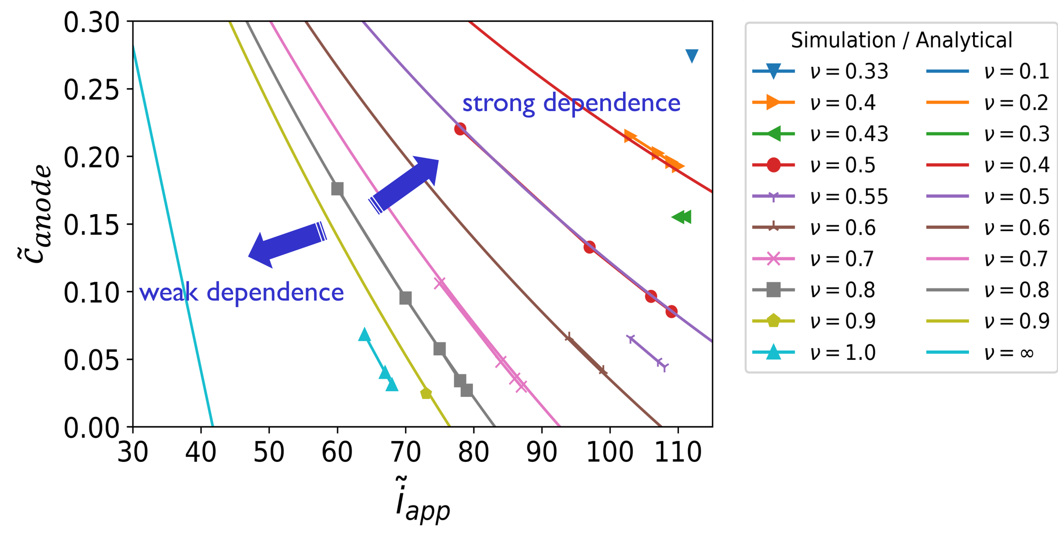
Results: One-Dim. Ionic Depletion

- Analytical solution of ionic depletion:

Concentration map ($\nu = 0.8$):



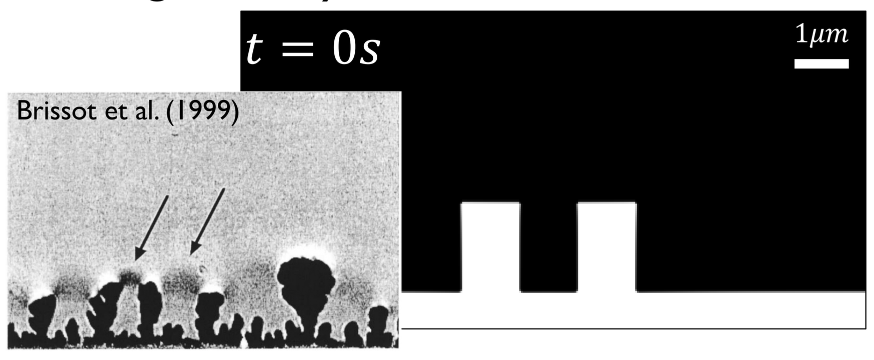
Concentration near interface – applied current



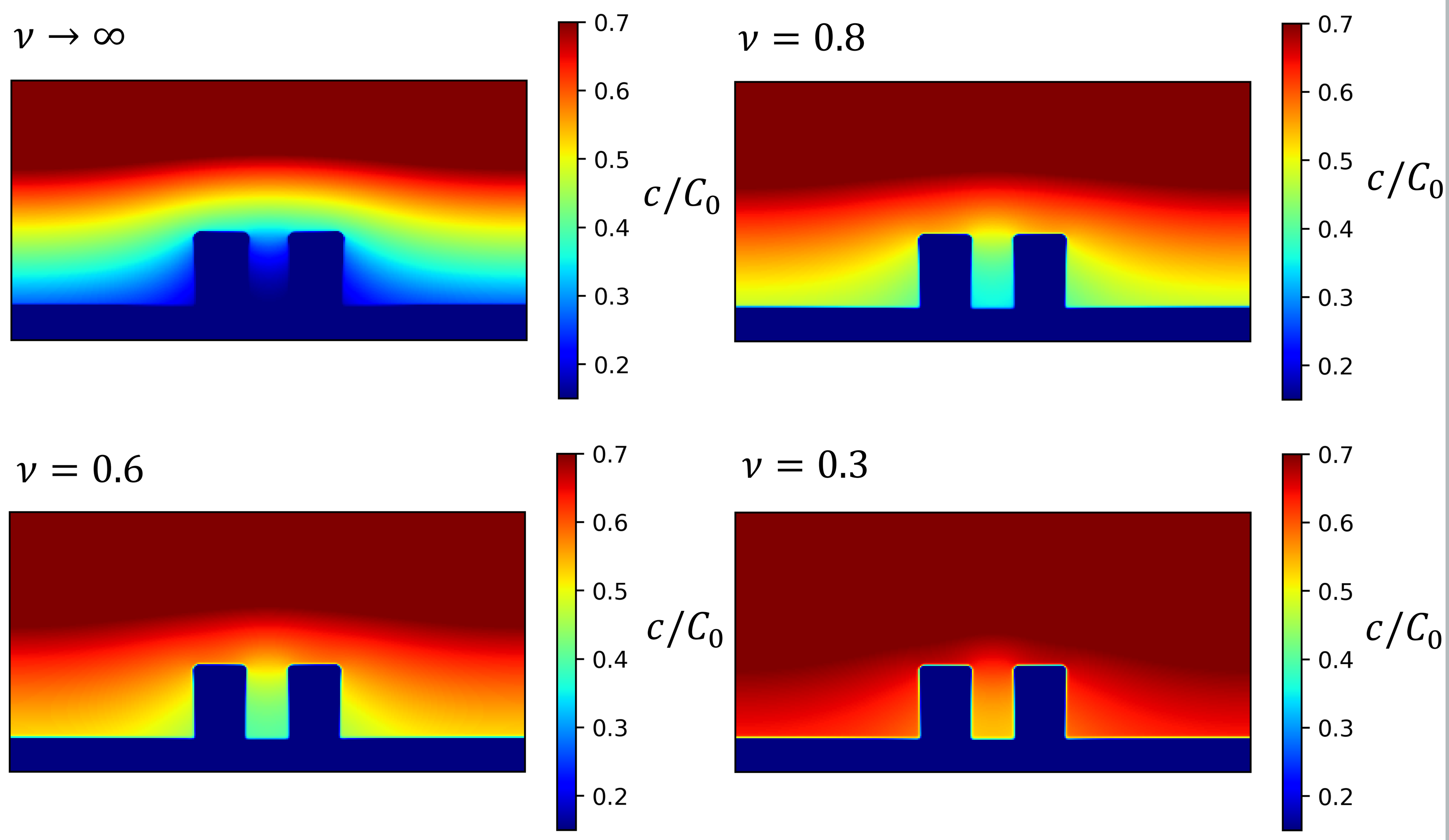
Results: Uneven Geometries

To precisely simulate the Li-deposition process under various conditions, we built a substrate with two rectangular pillars to mimic a real substrate and the initial geometry of a Li electrode.

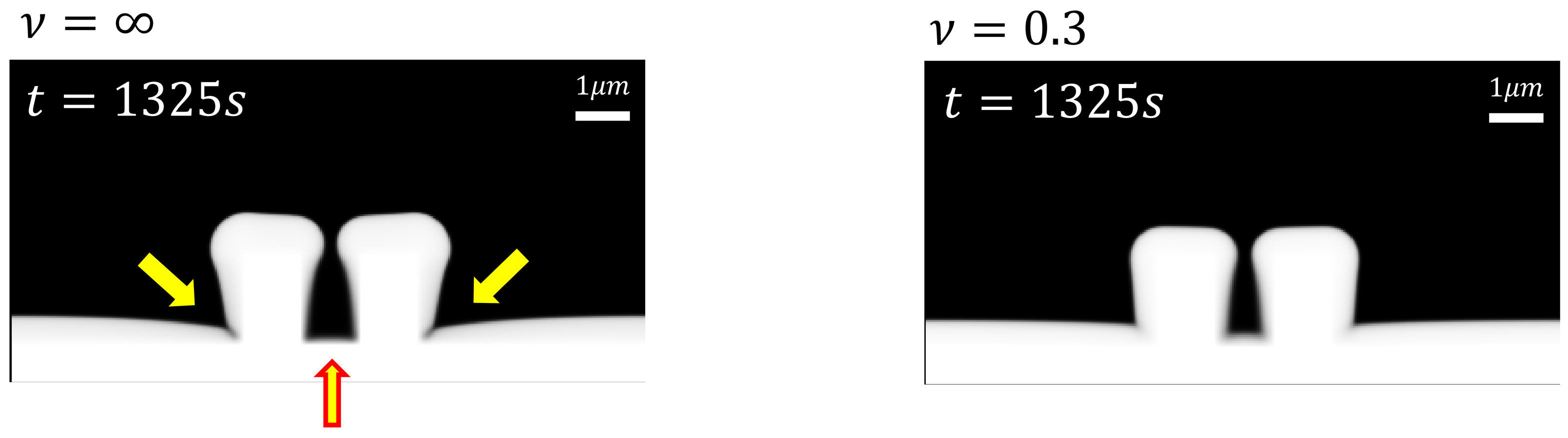
Initial geometry:



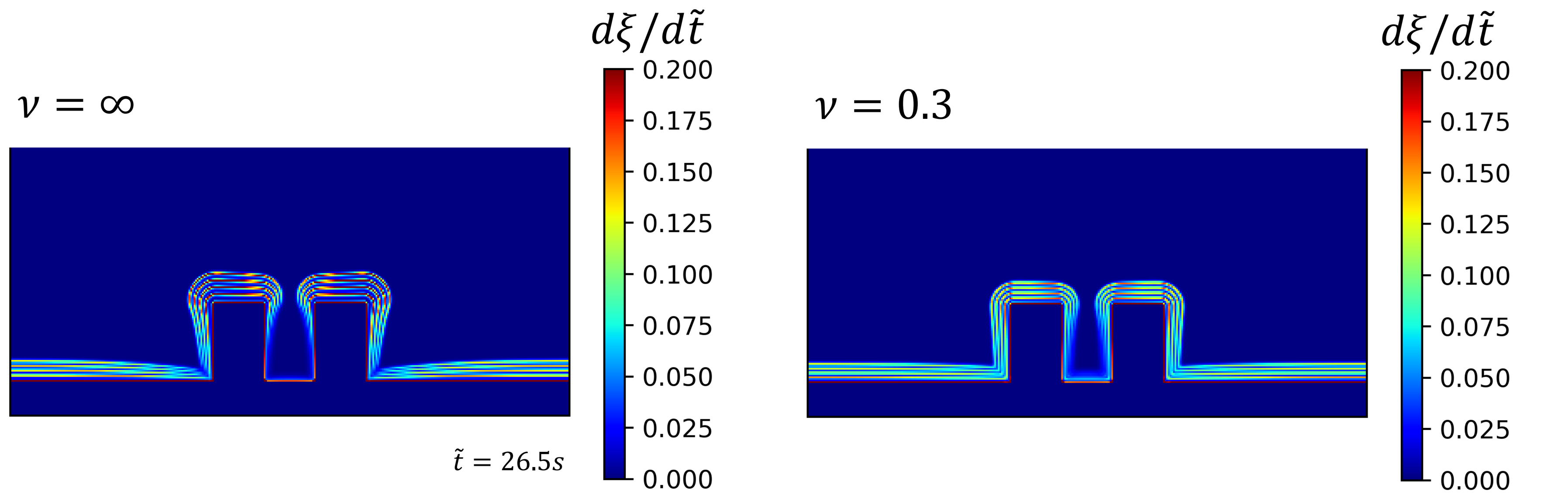
- Steady state concentration profile under a specific anode geometry:



- Anode geometry after deposition:



- Time evolution of anode geometry:



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