

Phase-field Theory of Dendritic Electrodeposition

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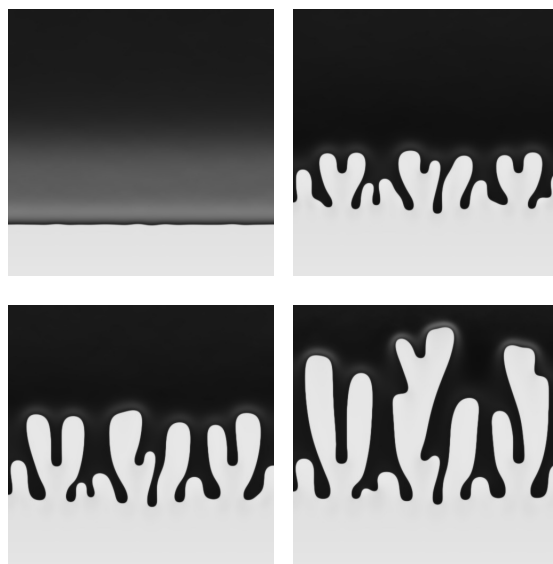
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Dendrite growth during electrodeposition is a challenging theoretical and numerical problem with important technological relevance for advanced battery technologies. Modeling efforts have largely focused on diffusion limited aggregation (DLA), which has been studied in detail and is relatively simple to simulate. However, DLA is applicable only at the limit of very small currents, and is therefore unable to produce many of the morphologies that have been observed experimentally during electrodeposition.

A complete theoretical understanding of the formation of different electrochemical dendrite morphology in binary electrolytes has not been developed. Chazalviel analyzed the space charge layer in front of an interface and related it to a potential drop across the interface that results in instability [1-2]. He found that the speed of the moving interface was determined by the speed of the anions and predicted that it lead to unstable interfaces, but was unable to simulate morphology.

The phase-field method has been very successful at quantitatively modeling morphology dendritic solidification, but has had only limited application in electrochemical systems. The advantage of the phase-field method is that it allows for a rigorous thermodynamic treatment of material properties while implicitly track boundaries and incorporating curvature-driven phase boundary motion. Phase-field methods have been extended to include electrochemistry [3-4], and have been applied to dendritic electrodeposition with a supporting electrolyte [5], but have not been applied to electrodeposition with binary electrolytes.

In this work I develop an electrochemical phase-field model for electrodeposition in a binary electrolyte, and use it to interpret the various morphologies that have been observed experimentally. The phase-field model is based on the model of Guyer *et al.* [3-4], which provides a diffuse-interface description of charge separation at an electrochemical interface, and considers electrochemical double layers and Butler-Volmer kinetics. The interface is modeled with a phase order parameter, and anion, cation, and electron diffusion are considered as well. The model provides insight into the parameters and mechanisms that lead to interface instability in electrochemical systems.



Simulation of dendritic growth during electrodeposition.

References

- [1] J. N. Chazalviel, *Phys. Rev. A* 42, 7355 (1990).
- [2] M. Rosso, J. N. Chazalviel, and E. Chassaing, *J. Electroanal. Chem.* 587, 323 (2006).
- [3] J. E. Guyer, W. J. Boettinger, J. A. Warren, and G. B. McFadden, *Phys. Rev. E* 69, 021603 (2004).
- [4] J. E. Guyer, W. J. Boettinger, J. A. Warren, and G. B. McFadden, *Phys. Rev. E* 69, 021604 (2004).
- [5] Y. Shibuta, Y. Okajima, and T. Suzuki, *Science and Technology of Advanced Materials* 8, 511 (2007).