

Balance between precision and scalability: Kinetic Monte Carlo Simulation of Electrodeposition Processes

Efe Mehmet Peker¹, Ozlem Ozcan¹, Franziska Heß²

¹ Bundesanstalt für Materialforschung und-prüfung, Berlin, Germany

² Technische Universität Berlin, Berlin, Germany

efe-mehmet.peker@bam.de

Abstract

The atomistic structure of electrodeposited metal films determines their properties such as activity and stability. The nucleation behaviour, frequency and arrangement of grain boundaries within electrodeposited layers as well as their atomic surface morphology have been of great interest due to their strong influence on their functional properties. Research on this topic has been largely dominated by empirical experimental studies, mainly because of the vast number of variables affecting the electrodeposition process and its outcomes. Moreover, methods capable of simulation of film growth kinetics were not applicable to complex systems. With the increasing performance of computational power over the last couple of decades, there has been a shift of attention toward numerical simulations to better understand and model atomistic-scale electrodeposition phenomena. The present study focusses on the atomistic simulation of the film growth processes in metal thin film electrodeposition. We have developed the tools to dimensionally augment the ZACROS Kinetic Monte Carlo simulation (KMCs) Software ^[1] for application to three dimensional processes and defined the necessary elementary steps for SC, FCC and BCC lattice structures. The events have been simplified down to two main categories, deposition of a single atom from the electrolyte surface layer onto the metal surface and diffusion of an atom from a discrete place on the crystal structure surface to another. MACE ^[2] energy model

was centered around an energy cluster expansion framework used to calculate the cluster energies necessary for the tuning of the discrete event rates. The accuracy of the MACE model approximations of surface structures and the cluster energies obtained from the developed framework were compared with respective DFT calculations. The simulation results were benchmarked against studies performed using the embedded atom model (EAM) ^[3], and energy values were compared with DFT calculations.

Our augmented ZACROS approach in combination with ever improving MACE energy models opens up new possibilities in scaling the kinetic modelling of surfaces and interfaces. The presentation will summarize the technical details of the 3D augmentation scheme, utilization of the AI energies and offer deep dives into the case studies performed with Cu electrodeposition.

References

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- [2] Batatia I, Kovács DP, Simm GNC, Ortner C, Csányi G. MACE: Higher Order Equivariant Message Passing Neural Networks for Fast and Accurate Force Fields. *Adv Neur In* 2022.
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Figures

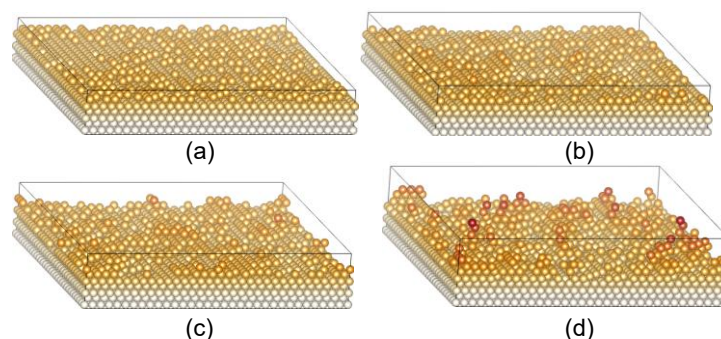


Figure 1. Surface snapshots of simulations operating under 0.1 (a), 1 (b), 10 (c) and 100 (d) mA/cm² current densities consisting of comparable number of copper atoms demonstrating surface roughening effect of increasing current density