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Machine Learning-Enhanced Modelling of Calcium Carbonate Nucleation in Porous Media under Counter-Diffusion Conditions

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This study introduces a novel numerical framework for simulating the nucleation of calcium carbonate ($CaCO_3$) polymorphs (calcite,vaterite, and amorphous calcium carbonate, shortly ACC) in porous media. Classical Nucleation Theory (CNT), and Non-Classical Nucleation Theory (n-CNT) were implemented into a high-resolution 3D Lattice Boltzmann Model (LBM) to reproduce the results of a dedicated counter-diffusion experiment characterized at the pore scale with advanced synchrotron-based chemical imaging techniques. By incorporating experimentally derived pH-dependent saturation index (SI) [1,2] thresholds for nucleation and accounting for stochastic induction times, the model attempts to capture the timing and distribution of nucleation of $CaCO_3$ polymorphs and to provide insights on the influence of supersaturation gradients, spatial constraints, and nucleation probabilities on the onset of precipitation.

Further sensitivity analyses reveal that nucleation thresholds critically govern both the spatial and temporal sequence of polymorph formation. While the simulations successfully capture the preferential nucleation of vaterite under high-pH conditions, the modeled calcite nucleation—occurring under narrowly defined SI and pH regimes—deviates from experimental observations. These discrepancies demonstrate the limitations of the typical nucleation threshold model. Nevertheless, our refined modelling framework is highly flexible and can readily incorporate alternative nucleation threshold models tailored to different experimental or theoretical conditions. This adaptability is a key strength and underscores the fundamental contribution of this work.

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