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Three-Dimensional nucleation with diffusion controlled growth: A comparative study of electrochemical phase formation from aqueous and deep eutectic solvents

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Abstract

The nucleation of Ag onto vitreous carbon from aqueous 3 M NaCl or 0.6 M NaClO₄ and deep eutectic solvent (DES) 1:2 molar mixture of choline chloride:urea solutions containing Ag⁺, has been studied analyzing the chronoamperometric response to single potential steps. From the coordinates of the maxima observed in the current responses, the nucleation frequencies A (s⁻¹) and number densities of nucleation sites N_0 (cm⁻²) were obtained from the standard model of nucleation with diffusion-controlled three-dimensional growth. Analysis of the overpotential dependence of nucleation frequencies using the classical electrochemical nucleation theory allowed to calculate the Gibbs free energy of nucleation $\Delta\tilde{G}(n_c)$ and critical nucleus size n_c as well as the exchange current density j_0 , transfer coefficient α and surface tension σ of silver nuclei. The kinetics of Ag⁺ reduction is two orders of magnitude slower in DES compared to both aqueous systems studied, and values of $\alpha \ll 0.5$ were found in both aqueous and DES media, indicating either that the intermediate state for metal ion reduction is located close to the initial state, i.e., the solvated or complexed metal ion in solution, or that the metal ion is specifically adsorbed on the surface and the symmetry factor involved requires an alternative electron transfer formalism. The low $\Delta\tilde{G}(n_c)$ and n_c values observed indicate that the discharge of a

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