

Formation of gold nanoparticle monolayers of controlled structure and electrokinetic properties

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Deposition mechanism of gold nanoparticles on poly(allylamine hydrochloride) (PAH) modified gold substrate was studied under *in situ* conditions by using quartz crystal microbalance (QCM), streaming potential measurements and scanning electron microscopy (SEM). The studies were carried out for various bulk concentrations of gold suspensions, flow rates, ionic strengths and pHs. It was shown that the particle deposition for the low coverage regime was governed by the bulk mass transfer step that results in a linear increase of the coverage with the time indicating that the hydration of the monolayers was negligible. This allowed one to derive a universal kinetic equation that describes the mass transfer rates in the QCM cell for various flow rates and diffusion coefficients. Kinetic measurements were also performed for longer times and for various ionic strengths and pHs. Negligible desorption of particles was confirmed that allowed one to precisely determine the maximum coverage of monolayers. A significant increase in the maximum coverage with ionic strength was interpreted as due to the decreasing range of the lateral electrostatic interactions among deposited particles.

The experimental results were quantitatively interpreted in terms of the extended random sequential adsorption (eRSA) model where the bulk and surface transfer steps were simultaneously considered. It was also shown that the QCM data matched the *ex situ* SEM results indicating that the monolayer hydration was also negligible for higher coverage range. The structure of monolayers obtained for various ionic strength was analyzed in terms of the radial distribution function by using the SEM micrographs of deposited particles. In this way, the significance and the range of electrostatic interactions was directly determined. Besides significance to basic sciences, the obtained results can be exploited for developing an efficient procedure of preparing gold-nanoparticle monolayers of desired coverage and structure having potential applicability as biosensors.

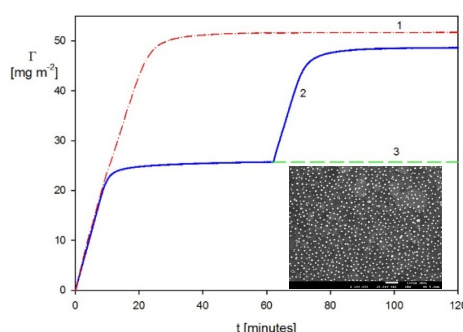


Figure 1. The kinetics of gold nanoparticle deposition on PAH modified Au sensor determined by QCM, pH 7.4 (PBS). Lines 1 and 3 show the results obtained for single step gold nanoparticle deposition for ionic strength 10^{-2} M (red) and 10^{-3} M (green) whereas line 2 presents a two-stage deposition kinetics obtained for ionic strength change from 10^{-3} M to 10^{-2} M. The inset shows the SEM image of nanoparticle monolayer at the gold sensor of the coverage of 0.15.

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