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The electrochemical desorption of n-alkanethiol monolayers from polycrystalline Au and Ag electrodes

Cindra A. Widrig ... Marc D. Porter [1]

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Abstract

Monolayers formed at Au and Ag surfaces by the spontaneous adsorption of n-alkanethiols were characterized voltammetrically to examine the chemistry of the bound thiol head group. Electrode reactions that correspond to the oxidative- and reductive-desorption of the adsorbed n-alkanethiol monolayer are reported for the first time. Analysis of the data indicates that upon adsorption at both Au and Ag surfaces the hydrogen of the thiol group is lost and the sulfur atom is oxidized by one electron. Based on the charge required for the reductive-desorption of the monolayer, the surface coverage of the oxidized n-alkanethiol species is 9.3Ã—$10^{â^'10}$ mol/cm^2 and 7.0 Ã—$10^{â^'10}$ mol/cm^2 on Au and Ag, respectively. The value of the surface coverage at Au is slightly greater than that expected for a closest-packed overlayer commensurate with a Au(111) substrate, whereas the value at Ag is somewhat less than that expected for

Au(111) substrate, whereas the value at Ag is somewhat less than that expected for layers commensurate at any of the low index Ag surface planes. The low apparent coverage observed for Ag electrodes is attributed to a portion of the monolayer being electroinactive at accessible applied voltages. As a probe of the mechanism for monolayer formation, films that were deposited on Ag and Au surfaces from solutions containing sodium n-octadecanethiolate were also characterized. Examination of the resulting layers with infrared reflection spectroscopy and optical ellipsometry indicates that only at Ag substrates were the films deposited from the thiolate solution complete and structurally similar to monolayers formed from n-octadecanethiol solutions. Based on this observation, it is postulated that the thiol hydrogen of the n-alkanethiol molecule participates in the reduction reaction that is concomitant to the thiol oxidation during adsorption on Au, whereas adsorption on Ag proceeds through the reduction of the Ag(I) surface species of the native oxide. A model for the electric double layer at the monolayer-coated-electrode/solution interface is also suggested based on the observed chain-length dependence of both the capacitance and the potential for the reductive desorption.

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1 To whom correspondence should be addressed.

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