Coverage dependent energetics for sulfur poisoning of Ni based anodes

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Recently, we reported an *ab-initio* thermodynamics model for the Ni(111)-H_2-H_2S system where the energetics for the competitive adsorption of H_2 and H_2S on a Ni surface was derived from density functional theory (DFT) calculations.[1] In that study, we did not explore the influence of surface coverage on the energetics and the resulting effect on the predicted sulfur and hydrogen coverages.

In this work, we extend our *ab-initio* thermodynamics model by explicitly including adsorbate coverage effects. Our results show that the adsorption energy for both H_2S and H_2 on Ni is a strong function of surface species coverage (Figure 1) while the entropy of adsorption is weakly dependent on coverage (Figure 2). We use the coverage-dependent Gibbs energy values for the adsorption reactions to predict the equilibrium coverages of S and H on Ni across a wide range of industrially relevant reaction conditions and compare our results with data in the literature.

The above reaction energetics can also be combined with a surface transport-reaction model for patterned anodes. This enables the direct simulation of H_2S poisoning on a Ni based anode while accounting for the near-triple-phase-boundary processes.

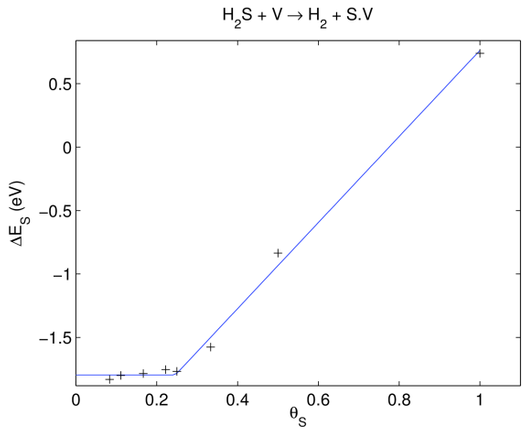

Figure 1: Adsorption energy of S on Ni as a function of S coverage

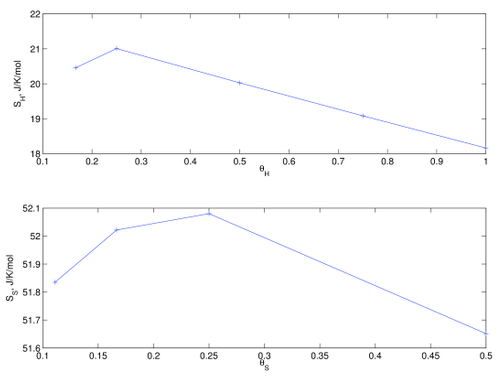

Figure 2: Entropy of adsorption (1073 K) as a function of species coverage

References:
1. D. S. Monder and K. Karan, *Journal of Physical Chemistry C,* **114**, 22597 (2010).