

Electrochemical Reactions using Heterogeneous Molecular Catalysis Plots of volcanoes and tafel plots of catalysis

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Abstract

We investigate the reasons for the occurrence or non-occurrence of volcanoes in the context of heterogeneous molecular catalysis by graphing the kinetics of the catalytic reaction against the stabilisation free energy of the major intermediate of the catalytic process. Modern energy difficulties, particularly those involving tiny molecules such as water, hydrogen, oxygen, proton, and carbon dioxide, are a powerful motivator for such investigations, just as they are for homogenous molecular catalysis or catalysis by surface active metallic sites. This is especially true in the case of heterogeneous molecular catalysis, which is frequently chosen over homogeneous molecular catalysis by the same molecules, if only for chemical separation and electrolytic cell construction.

Introduction

The basic assumption that the kinetic responses depend on a single descriptor, namely the stabilisation free energy of the primary intermediate, is the key shortcoming of the volcano plot technique, as it is with the other two catalysis types. Clearly, more thorough methodologies that investigate reactions to the greatest number of experimental conditions and can be expressed as catalytic Tafel graphs should be favoured. This is especially true in the case of heterogeneous molecular catalysis, where additional transport variables in the supporting film might influence current-potential responses. The occurrence of maxima in catalytic Tafel plots, as well as their reliance on the cyclic voltammetric scan rate, attest to this. In the context of today's energy challenges, an ever-increasing number of molecular systems, mostly transition metal complexes, are proposed for catalysing electrochemical reactions, particularly the oxidation or reduction of small molecules such as water, hydrogen, oxygen, proton, and carbon dioxide. Although homogeneous molecular catalysis is a promising technique, films containing the same molecules put on the electrode surface are likely to be preferable, if only to make product separation easier. Another reason is that such catalytic electrodes can be employed in entire electrolytic cells, complete with suitable counter electrodes and separators, utilising existing electrolyzer and fuel cell technologies. By immobilising molecules that have been proven to be efficient homogeneous molecular catalysts onto the electrode surface, we can predict a quick development of this sort of catalytic system. When it comes to catalyst benchmarking, it has been demonstrated that it can be efficiently operated using catalytic Tafel plots that relate the turnover frequency to the overpotential for each system under consideration. It's unclear how this method, which was developed for homogeneous molecular catalysts, can be applied to deposited molecular catalysts. In terms of the second point, analogies with surface-active site catalysis on the one hand and homogeneous molecule catalysis on the other point to the hypothesis that the stability of the intermediate created in the initial phase of the reaction may be the most important element in catalytic efficiency. Increasing the intermediate's stability should result in two opposing impacts. One is to lower the energy barrier for the production of the intermediate in a favourable way. The other is that a high level of intermediate stabilisation slows the conversion of the intermediate into products and the subsequent regeneration of the catalytic surface. This "Sabatier principle" predicts the presence of a maximum, or "volcano," on the plot of exchange current vs. adsorption free energy of the intermediate among a family of electrocatalysts for a particular process. These ideas have been around since the late 1950s, yet they are still the subject of intense discussion and controversy today. Their application to homogeneous molecular catalysis is relatively new, and we must take care in the transition to molecular film catalysis. Taking into account the numerous ways of connection between modes of transport Reactants, charges, and catalytic reactions are all discussed. We explore scenarios in which the catalytic reaction is the rate-determining factor at the expense of charge and reactant transport in order to focus on the possible occurrence of volcano plots in the context of molecular film catalysis (the film behaves as a monolayer or as a set of identically behaving monolayers). To get down to the nitty-gritty of the appearance or nonappearance of such volcanic plots, we'll first look at a simple molecular catalysis one-electron-one-step reaction scheme.