SUPPLEMENTARY MATERIAL

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Theoretical Analysis of a Surface Catalytic Mechanism Coupled with Reversible Chemical Step Under Conditions of Cyclic Staircase Voltammetry

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<u>Model Reaction</u>: Surface Catalytic EC' mechanism with reversible regenerative step:

Electrochemical step (E) $\mathrm{Ox(ads)} + n\,e^- \underset{}{\overset{k_s^{\theta}}{\rightleftharpoons}} \mathrm{Red(ads)}$

Catalytic (regenerative) reaction (C') $\mathrm{Red(ads)} + \mathrm{Y} \underset{k_b}{\overset{k_f}{\rightleftharpoons}} \mathrm{Ox(ads)}\ (+\ \mathrm{S})$

<u>Description of the file:</u>

MATHCAD File for calculation of cyclic voltammograms, with all parameters and equations given.

Mathcad - [CYCLIC-surf catalys-EC'revers-sept 2019 OK]

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$tac := 0.000[illegible] \quad \tau := 0.0005$

$el := 2 \qquad \alpha := 0.5 \qquad d := \frac{tac}{25}$

$Es := -0.6 \qquad Ef := 0.4 \qquad \Delta E := Ef - Es \qquad dE := 0.004 \qquad s := 1 .. \frac{tac}{d}$

$$m := \frac{\tau}{d} + 1 .. \frac{\Delta E}{dE}\cdot 25 + \frac{\tau}{d} \qquad n := \frac{\Delta E}{dE}\cdot 25 + \frac{\tau}{d} + 1 .. \left(\frac{\Delta E}{dE}\cdot 25\cdot 2 + \frac{\tau}{d}\right)$$

$$E_m := Es + \left(\mathrm{ceil}\left(\frac{m - \frac{\tau}{d}}{25}\right)\cdot dE - dE\right) \qquad k := 1 .. 2\cdot\left(\frac{\Delta E}{dE}\cdot 25 + \frac{\tau}{d}\right)$$

$$E_n := Ef - \left[\mathrm{ceil}\left[\frac{n - \left(\frac{\Delta E}{dE}\cdot 25 + \frac{\tau}{d}\right)}{25}\right]\cdot dE - dE\right]$$

$R := 8.314 \qquad F := 96500$

$T := 298$

$$\Phi ac := \frac{el\cdot F}{R\cdot T}\cdot Es \qquad \Phi_m := \frac{el\cdot F}{R\cdot T}\cdot E_m \qquad \Phi_n := \frac{el\cdot F}{R\cdot T}\cdot E_n$$

$$\Psi_s := \frac{KET\cdot e^{-\alpha\cdot\Phi ac}}{1 + KET\cdot e^{-\alpha\cdot\Phi ac}\cdot\left(1 + e^{\Phi ac}\right)\cdot\frac{S_1\cdot Keq}{\gamma\cdot(1 + Keq)\cdot 25} + KET\cdot e^{-\alpha\cdot\Phi ac}\cdot\left(1 + e^{\Phi ac}\right)\cdot\frac{S_1\cdot 1}{25\cdot(1 + Keq)}}$$

$$\Psi_m := \frac{KET\cdot e^{-\alpha\cdot\Phi_m}\cdot\left[1 - \frac{\left(1 + e^{\Phi_m}\right)\cdot Keq}{Kcatalytic\cdot(1 + Keq)\cdot 25}\cdot\sum_{j=1}^{m-1}\left(\Psi_j\cdot S_{m-j+1}\right) - \frac{\left(1 + e^{\Phi_m}\right)\cdot 1}{25\cdot(1 + Keq)}\cdot\sum_{j=1}^{m-1}\left(\Psi_j\cdot S_{m-j+1}\right)\right]}{1 + KET\cdot e^{-\alpha\cdot\Phi_m}\cdot\left(1 + e^{\Phi_m}\right)\cdot\frac{S_1\cdot Keq}{Kcatalytic(1 + Keq)\cdot 25} + KET\cdot e^{-\alpha\cdot\Phi_m}\cdot\left(1 + e^{\Phi_m}\right)\cdot\frac{S_1\cdot 1}{25(1 + Keq)}}$$

$ks := 1000$

$KET := ks\cdot\tau$

$KET := 1$

$Keq := 10.00$

$Kcatalytic := 10^{-1.250}$

$$S_k := e^{-\frac{Kcatalytic}{25}\cdot(k-1)} - e^{-\frac{Kcatalytic}{25}\cdot(k)}$$

SURFACE CATALYTIC ECrev mechanism

Keq - equilibrium constant of catalytic reversible chemical reaction
Kcatalytic - dimensionless rate parameter of regerenartive (catalytic) reaction
KET - dimensionless rate parameter of electron transfer step
τ - duration of potential step
el - number of electrons
dE - potential step height
α - electron transfer coefficient
S - numerical integration parameter
Φ - dimensionless potential
Em - cathodic potential ramp
En - anodic potential ramp
Ψm - cathodic dimensionless currents
Ψn - anodic dimensionless currents
ks - standard rate constant of electrn transfer
R - gas constant
T - temperature
F - Faraday constant

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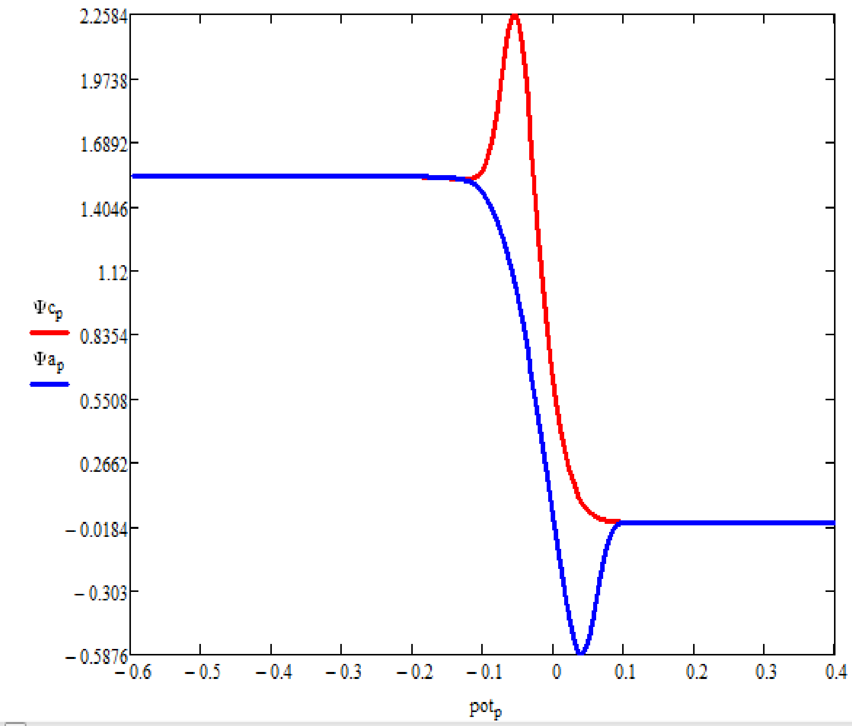
Mathcad - [CYCLIC-surf catalys-EC'revers-sept 2019 OK]

$$\Psi_n := \frac{KET\cdot e^{-\alpha\cdot\Phi_n}\cdot\left[1-\frac{\left(1+e^{\Phi_n}\right)\cdot Keq}{Kcatalytic\cdot(1+Keq)\cdot 25}\cdot\sum_{j=1}^{n-1}\left(\Psi_j\cdot S_{n-j+1}\right)-\frac{\left(1+e^{\Phi_n}\right)\cdot 1}{25\cdot(1+Keq)}\cdot\sum_{j=1}^{n-1}\left(\Psi_j\cdot S_{n-j+1}\right)\right]}{1+KET\cdot e^{-\alpha\cdot\Phi_n}\left(1+e^{\Phi_n}\right)\cdot\frac{S_1\cdot Keq}{Kcatalytic(1+Keq)\cdot 25}+KET\cdot e^{-\alpha\cdot\Phi_n}\left(1+e^{\Phi_n}\right)\cdot\frac{S_1\cdot 1}{25(1+Keq)}}$$

$p := 1..\frac{\Delta E}{dE}$ $\quad \Psi a_p := (\Psi)_{\left(\frac{\tau}{d\cdot 25}+p\right)\cdot 25}$ $\quad \Psi c_p := (\Psi)_{\left[\left[\frac{\Delta E}{dE}\cdot 2+\left(\frac{\tau}{25\cdot d}\right)\right]-p\right]\cdot 25}$ $\quad pot_p := Es + p\cdot dE$



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