Bright Solitons in the Electrooxidation of CO on Platinum Thin Film Electrode

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13, July 2018

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- The combustion of fossil fuels releases emissions into the air which causes pollution.

- W. R. Grove in 1839 [1], using the knowledge of electrolysis discovered the fuel cell.

- Fuel cells are considered as a prime candidate for the "green" energy production : clean, quiet.

- Fuel cells are electrochemical devices, that use chemical energy from a fuel (hydrogen, methanol, etc.) and an oxidant (air or oxygen) and PLATINUM catalyst to produce electrical energy.

- The 'reforming' fuels such as natural gas or methanol, introduces CO into the hydrogen gas, which poisons the platinum catalyst.

- The electrochemical oxidation of CO on Pt is an electrolytic reaction through which the CO is removed from the Pt surface.

- SOLITARY waves were first observed in this process in 1992 by Rotermund et . al [2] using PEEM spectroscopy.

- In 2005, Bauer et.al experimentally observed dissipative SOLITONS in the electrooxidation of CO on Pt using FTIR spectroscopy in the ATR configuration.

Purpose and method

- Analytical proof of bright solitons in the electrooxidation of CO on Platinum electrode observed experimentally.
- The perturbation analysis ; reductive perturbation and multiple scale expansion.

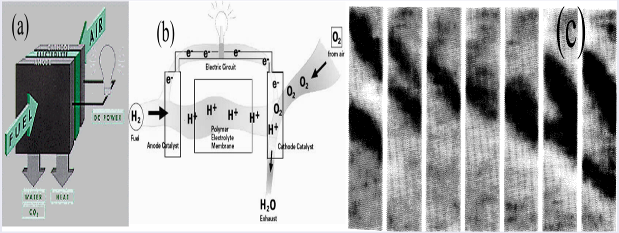

FIGURE: (a.) Fuel cell. (b) Schematic diagram .(c) Experimental observation of soliton collision of CO coverage [2].

$$\text{Anode}: \quad H_2 \longrightarrow 2H^+ + 2e^-, \tag{1}$$

$$\text{Cathode}: \quad \frac{1}{2}O_2 \longrightarrow 2H^+ + 2e^- \longrightarrow H_2O. \tag{2}$$

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The Chemistry of electrooxidation

•Mean field Langmuir−Hindshelwood (L-H) mechanism is use to describe the reaction.

• Simulations are usually based on three elementary reaction steps ; adsorption of CO, oxidative adsorption of water and reaction of adsorbed CO and OH molecules

$$Pt - CO_s \xrightarrow{V_{CO}^{ads}} Pt - CO, \tag{3}$$

$$Pt + H_2O \rightleftharpoons Pt - OH + H^+ + e, \tag{4}$$

$$Pt - CO + Pt - OH \xrightarrow{V^{reac}} 2Pt + CO_2 + H^+ + e. \tag{5}$$

•In Souradip et al. [4] it was shown that, the additional competitive adsorption of anions such as Cl^-, blocks free surface sites for OH and CO, and may induce oscillations ;

$$X^- + * \rightleftharpoons X_{ads} + e^-. \tag{6}$$

•As long as the OH coverage remains very small, the reaction rate can be expressed without taking explicitly the coverage of OH into account, Zhang et al. [5].

• The temporal evolution of the chemical subsystem, consisting of Θ_{CO}, the CO coverage of the electrode, and Θ_X, the anion coverage is then given by [4]

$$\begin{aligned} \partial_t \Theta_{CO} &= \nu_{CO}^{ads} - \nu^{reac} + D_0 \partial_{xx} \Theta_{CO}, \\ \partial_t \Theta_X &= \nu_X^{ads} - \nu_X^{des} + D_0 \partial_{xx} \Theta_X, \\ C \partial_t \phi_{DL} &= -S_{tot} F (2\nu^{reac} + (\nu_X^{ads} - \nu_X^{des})) - \sigma \partial_z \phi_{DL}|_{z=WE}, \end{aligned} \quad (7)$$

where the corresponding adsorption, desorption and reaction rates are given by the following expressions :

$$\begin{aligned} \nu_{CO}^{ads} &= k_{CO}^{ads} c_s (0.99 - \Theta_{CO} - \Theta_X), \\ c_s &= \frac{c_b D_0}{D_0 + S_{tot} k_{CO}^{ads} \delta (1 - \Theta_{CO} - \Theta_X)}, \\ \nu^{reac} &= k^{reac} (1 - \Theta_{CO} - \Theta_X) \Theta_{CO} \exp\left(\alpha \frac{F}{RT} \phi_{DL}\right), \\ \nu_X^{des} &= k_X^{des} \Theta_X \exp\left[(\alpha - 1) \frac{F}{RT} \phi_{DL}\right], \\ \nu_X^{ads} &= k_X^{ads} c_X \Re \left(0.99 - \Theta_{CO} - \frac{\Theta_X}{\Theta_X^{max}}\right) \exp\left(\alpha \frac{F}{RT} \phi_{DL}\right). \end{aligned} \quad (8)$$

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The model

• Let $\Theta_{CO} = U$, $\Theta_X = V$, $\phi_{DL} = W$ and the equations reduce to

$$U_t = \frac{\bar{\alpha}(0.99 - U - V)}{D_0 + \beta(1 - U - V)} - k_X^{ads}(1 - U - V)U\exp(\alpha f W) + D_0 U_{xx}, \tag{9}$$

$$V_t = \gamma(0.99 - U - V/V^{max})\exp(\alpha f W) - k_X^{des} V \exp(gW) + D_0 V_{xx}, \tag{10}$$

$$\begin{aligned} W_t &= -\gamma_1(1 - U - V)U\exp(\alpha f W) - \gamma_2(0.99 - U - V/V^{max})\exp(\alpha f W) \\ &+ \gamma_3 V \exp(gW) - \sigma \partial_z W|_{z=WE}, \end{aligned} \tag{11}$$

• where $\bar{\alpha} = k_{CO}^{ads} c_b D, \beta = S_{tot} k_{CO}^{ads} \delta, \gamma = k_X^{ads} c_X \mathfrak{R}, f = F/RT, g = (\alpha - 1)F/RT,$
$\gamma_1 = \frac{2S_{tot}Fk^{reac}}{C}, \gamma_2 = \gamma\frac{S_{tot}F}{C}, \gamma_3 = \frac{k_X^{ads}StotF}{C}, \sigma_1 = \frac{\sigma}{C}.$

Hypothesis

•Since the coverages are very small, we can take the Binomial expansion of the quotient.

• We assume that the coverages are very small and that the electrode potential is close to the strictly potentiostatic case ($W << 1$) and hence $\exp(W) \approx 1$.

• We neglect the surface diffusion of adsorbed CO, since it is small compared to the lateral diffusion of the bulk CO [4].

•We consider the homogeneous case of the migration coupling, where it vary linearly with the electrode potential.

• Using these assumptions and putting the equation in the Linard, we have

$$V_{tt} - \Omega_0 V_{xx} - (k_0 + k_1 V + k_2 V^2)V_t - (\bar{\Omega}_8 + \bar{\Omega}_9)V_t^2 - (\Lambda_0 V + \ell_0 V^2)V_{xx} - (\chi_2 + \chi_3)V_{xx}^2 - \eta_0$$
$$-\eta_1 V - \eta_2 V^2 - \eta_3 V^3 - (\Lambda_2 - \Lambda_3 V)V_t V_{xx} - \varpi_0 V_t V_{xx}^2 + \varpi_1 V_t^2 V_{xx} + \Gamma V_t^3 + \varpi V_{xx}^3 + D_0 Vxxt = 0,$$

$$\begin{aligned} W_t &+ (\tau_3 + \tau_4 V)V_t - (\pi_0 + \pi_1 V)V_{xx} - \pi_2 V_t^2 + \rho_0 V_t V_{xx} - \tau_4 V^2 - \tau_1 V - \tau_0 \\ &- \sigma_1(W - U_0) - D_2^2 V_{xx}^2 = 0, \\ U &= 0.99 - \beta_2 V - \bar{\alpha_2} V_t + D_0 V_{xx}. \end{aligned} \quad (12)$$

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Reductive perturbation method

- We seek for weak amplitude wave, by applying the reductive perturbation technique.

- Constraint : nonlinearity balances dispersion.

- At the order $O(\epsilon^{3/2})$, we have ;

$$-k_0 V_T + k_1 u_0 V V_S + D_0 u_0 V_{SSS} = \Omega_0 V_{SS} + u_0(\varpi_0 + \Lambda_2) V_S V_{SS}, \tag{13}$$

- where $\Omega_0 = \epsilon^{1/2}\Omega_0$, $\Lambda_2 = \Lambda_2/\epsilon$, $\varpi_0 = \varpi_0/\epsilon$.
- After applying the scaling on this equation, we have the perturbed kdV given by

$$V'_\tau - 6V'V'_\varepsilon + V'_{\varepsilon\varepsilon\varepsilon} = \gamma V'_{\varepsilon\varepsilon} - \beta V'_\varepsilon V'_{\varepsilon\varepsilon}, \tag{14}$$

where $\tau = \dfrac{u_0 k^{3/2} D_0}{k_0} T$, $\varepsilon = -\sqrt{k_1} S$, $V = -6D_0 V'$.
- This equation is known as the Modified kdV−Burger (MkdVB) equation.

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- The unperturbed equation is given by ;

$$V'_\tau - 6V'V'_\varepsilon + V'_{\varepsilon\varepsilon\varepsilon} = 0. \quad (15)$$

- This equation has a one soliton solution given by Kivshar et al. [5] which has the form

$$V' = -2k^2 sech^2 z, \quad (16)$$

where $z = k(\varepsilon - \zeta)$ and ζ is the phase.

- Then, using the perturbation theory based on the inverse scattering transform that predicts the temporal evolution of the amplitude k, and the phase ζ we have

$$\frac{dk}{d\tau} = -\frac{12\gamma k}{15}, \quad (17)$$

$$\frac{d\zeta}{d\tau} = 4k^2 - \frac{112\beta k}{105}. \quad (18)$$

- Solving these, we see that $k(\tau) = A_0 \exp(-\alpha\tau)$ which implies that the dissipation brings about the exponential decay of the amplitude.

- Also, the phase $\zeta(\tau) = 4k^2\tau + \sigma k\tau$.

Derivation of KdV

- Substituting these expression in the unperturbed solution, it then become

$$V'(\varepsilon,\tau) = -2a_0^2\exp(-2\alpha\tau)sech^2\left[a_0\exp(-\alpha\tau)\left(\varepsilon - 4a_0^2\exp(-2\alpha\tau)\tau + a_0\sigma\exp(-\alpha\tau)\tau\right)\right]. \quad (19)$$

In terms of the original variable, it is given by

$$\Theta_X = Ae^{-2\lambda t}sech^2(X), \quad (20)$$

- where $X = a_0e^{-\lambda t}(-qx + (p+qu_0)t - 4e^{-\lambda t\lambda})$ and $A = 12a_0D_2$.

- From the coupling equation, the CO coverage is given by

$$\begin{aligned}\Theta_{CO} &= 0.99 - \beta_2Ae^{-2\lambda t}sech^2(X) - 2\lambda\bar{\alpha}_2Ae^{-\lambda t}sech(X) - 2a_0\lambda cAe^{-3\lambda t}sech^2(X)tanh(X)\\ &+ 8\lambda Ae^{-4\lambda t}sech^2(X)tanh(X) + 4D_2q^2a_0^2Ae^{-4\lambda t}sech^2(X)tanh^2(X)\\ &- 4D_2a_0^2q^2Ae^{-4\lambda t}sech^4(X). \quad (21)\end{aligned}$$

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Model graphs

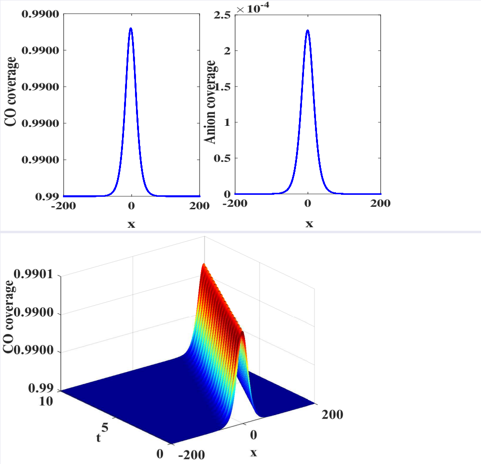

FIGURE: *CO and Anion coverage for the KdV-Burger equation :* $c = 100, \lambda = 0.005, q = 5, \alpha = 0.005$

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Multiple scale expansion method

•This is a perturbation technique in which, the wavelength of periodic oscillations of the carrier wave is comparable to the envelope width, the latter looks like it is breathing [6].

•Constraint : nonlinearity and dispersion are not balanced.

•In order to determine the order of the different terms, we introduce the variable $V = \epsilon\phi$ and $W = \epsilon\varphi$.

•We suppose that D_0 is perturbed to the order ϵ^2. This is due to the roughness of the anode which increases the process of reaction and the equations become

$$\begin{aligned}\phi_{tt} &- \Omega_0\phi_{xx} - \epsilon^2 D_0\phi_{xxt} - (k_0 + \epsilon k_1\phi + \epsilon^2\phi)\phi_t - \epsilon(\bar{\Omega}_8 + \epsilon\bar{\Omega}_9\phi)\phi_t^2 - (\epsilon\Lambda_0\phi + \epsilon^2\ell_0\phi^2)\phi_{xx} \\ &- \epsilon(\chi_2 + \epsilon\chi_3\phi)\phi_{xx}^2 - \eta_0/\epsilon - \eta_1\phi - \epsilon\eta_2\phi^2 - \epsilon^2\eta_3\phi^3 - \epsilon(\Lambda_2 - \epsilon\Lambda_3\phi)\phi_t\phi_{xx} \\ &- \varpi_0\epsilon^2\phi_t\phi_{xx}^2 + \epsilon^2\varpi_1\phi_t^2\phi_{xx} + \epsilon^3\Gamma\phi_t^3 + \epsilon^2\varpi\phi_{xx}^3 = 0, \qquad (22)\end{aligned}$$

$$\begin{aligned}\varphi_t &+ (\tau_3 + \epsilon\tau_4\phi)\phi_t - (\pi_0 + \epsilon\pi_1\phi)\phi_{xx} - \epsilon\pi_2\phi_t^2 + \epsilon\rho_0\phi_t\phi_{xx} - \epsilon\tau_2\phi^2 - \tau_1\phi - \tau/\epsilon \\ &+ \sigma_1(W - U_0) - \epsilon D_2^2 v_{xx}^2 = 0. \qquad (23)\end{aligned}$$

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- We now look for modulated solution of the form

$$\phi = A(X_1, T_1, X_2, T_2)e^{i\theta} + c.c + \epsilon[C(X_1, T_1, X_2, T_2) + D(X_1, T_1, X_2, T_2)e^{2i\theta}] + c.c + O(\epsilon^2), \quad (24)$$

$$\varphi = F(X_1, T_1, X_2, T_2)e^{i\theta} + c.c + \epsilon[G(X_1, T_1, X_2, T_2) + H(X_1, T_1, X_2, T_2)e^{2i\theta}] + c.c + O(\epsilon^2). \quad (25)$$

- At the order ϵ^0, the annihilation of terms in $e^{i\theta}$,gives the dispersion relation of linear waves of the system.

$$\omega^2 = \Omega_0 k^2 + \eta_1 \tag{26}$$

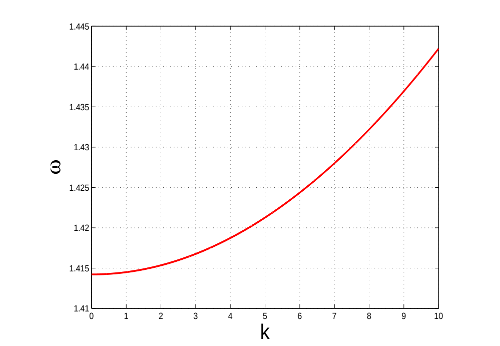

FIGURE: *The dispersion relation of the reaction pulse :* $\Omega_0 = 0.008, \eta_1 = 2.$

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- At the order ϵ^1, the cancellation of terms in $e^{i\theta}$ gives the solvability condition

$$\frac{\partial A}{\partial T_1} + V_g \frac{\partial A}{\partial X_1} = 0. \tag{27}$$

- At the order ϵ^2,the cancellation of terms in $e^{i\theta}$ and using the transformation $\xi_i = X_i - V_g T_i$ and $\tau_i = T_i$ yields

$$i\frac{\partial A}{\partial \tau_2} + P\frac{\partial^2 A}{\partial \xi_1^2} + Q|A|^2 A = i\frac{R}{2}A. \tag{28}$$

- This equation shows that, the evolution of modulated waves in this flow cell model is described by the Complex Ginzburg-Landau equation.

- where the nonlinearity and dispersion coefficients Q is complex while R and P are respectively real and are given by $Q = \bar{R} - i\bar{F}$ and $R = \frac{\bar{G}}{\omega}$, where the expressions of the coefficients are

$$\bar{G} = k_0 - D\omega k, \qquad P = \frac{\Omega_0 - V_g}{2\omega}, \qquad \bar{F} = \frac{\varpi_0\omega^4 - \omega k - 2k_2}{2}, \tag{29}$$

$$\bar{R} = \frac{3\ell_0\omega^2 - 3\eta_3 - \Omega_9\omega^- 3\chi_3 - \varpi_1 - 3\omega_2^6 + \frac{6\eta_2^2}{\eta_1}}{2\omega}. \tag{30}$$

- We look for a solution of the form $A(\tau_2, \xi_1) = B(\xi_1)e^{-i\omega\tau_2}$ using the method in Soto-Crespo et al [3], we have our solution in the original reference frame to be

$$\Theta_X = Nsech(Z_1)cos(Z_2) - \frac{\eta_2}{\eta_1}N^2 sech^2(Z_1)cos^2(Z_2), \tag{31}$$

where $Z_1 = \epsilon\sqrt{\beta}(x - x_0)$, $Z_2 = \Phi + kx - \delta\omega t$, $N = 2\epsilon\sqrt{\frac{\beta}{\alpha}}$ and $M = (k + \Phi_x)$ with $b = V_g + \epsilon V_p$ and $\delta = 1 + \epsilon^2$.

- Similarly,

$$\begin{aligned} \phi_{DL} &= \frac{-(\omega^2\tau_3 + \tau)}{\omega}Nsech(Z_1)cos(Z_2) \\ &+ 2\epsilon^2[\frac{4\tau_3 k\eta_2\beta\sqrt{\beta}}{\eta_1\alpha\omega}sech^2(Z_1)tanh(Z_1)cos(2\Phi - 2\omega\tau_2)cos(2\theta) \\ &+ (\omega^2\tau_4 - \pi_2\omega^2 - \frac{\eta_2}{\eta_1}(4\tau_3\omega^2 + \tau))cos(2\theta) + \Phi'\omega k^2\rho_0 sin(2\theta) \\ &+ \frac{\beta}{\alpha}\Phi' sech^2(Z_1)sin(2\theta)(1 + \sqrt{\beta}tanh(Z_1)sin(2\Phi - 2\omega\tau_2))]. \end{aligned} \tag{32}$$

From the coupling equation, we have

$$\begin{aligned}
\Theta_{CO} &= 0.99 - Nsech(Z_1)cos(Z_2)[\beta_2 + \bar{\alpha}_2\epsilon btanh(Z_1)] + \frac{\eta_2\beta_2}{\eta_1}N^2sech^2(Z_1)cos^2(Z_2) \\
&+ \bar{\alpha}_2N(\Phi_t - \delta\omega)sech(Z_1)[sin(Z_2) - \frac{2\eta_2}{\eta_1}Nsin(2Z_2)] + D_2[\beta\epsilon^2Nsech^3(Z_1)cos(Z_2) \\
&- \epsilon^2Nsech(Z_1)tanh(Z_1)cos^2(Z_2) + (1 - \epsilon\sqrt{\beta})Msech(Z_1)tanh(Z_1)sin(Z_2) \\
&- N\Phi_{xx}sech(Z_1)(sin(Z_2) - \frac{\eta_1}{\eta_2}sin(2Z_2)) + M^2sech(Z_1)(\frac{2\eta_2}{\eta_1}Ncos(2Z_2) - cos(Z_2)) \\
&+ \frac{2\beta\epsilon^2\eta_2}{\eta_1}N^2sech^2(Z_1)cos^2(Z_2)(2tanh(Z_1) - sech(Z_1)) \\
&+ \frac{2\beta\epsilon^2\eta_2}{\eta_1}N^2Mtanh(Z_1)sech^2(Z_1)sin(2Z_2) \\
&- \frac{\sqrt{\beta}\eta_2}{\eta_1}N^2Mtanh(Z_1)sech(Z_1)sin(2Z_2)]. \qquad (33)
\end{aligned}$$

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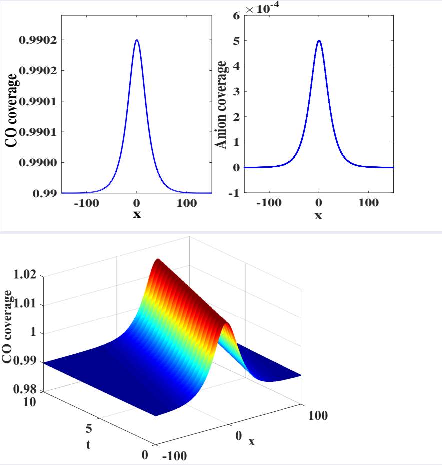

FIGURE: *CO and Anion coverage for the CGL equation* : $\epsilon = 0.1, k = 0.003, x_0 = 0.03, \delta = 0.4, \Omega = 0.005$

Conclusion

- We have analytically proven that, the reaction pulses in the electrooxidation process are solitons, a conjecture made by Krischer et al.

- We have considered the two different configurations of our system.

- We were able to show that this solitons are dissipative using the perturbation method.

Opening

- We intend to study the backfiring effect observed experimental, by doing collision studies.

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THANK YOU