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EIGHTH QUARTERLY REPORT

1 OCTOBER 1970 to 31 DECEMBER 1970

STUDIES IN FUNDAMENTAL CHEMISTRY OF FUEL CELL REACTIONS

NGR 39-010-002

UNIVERSITY OF PENNSYLVANIA

ELECTROCHEMISTRY LABORATORY

PHILADELPHIA, PENNSYLVANIA 19104

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Submitted to:

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

Washington, D. C. 20546

Submitted by:

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PROJECT PERSONNEL

Section I

Mr. R. Sen, Graduate Student

Dr. J. O'M. Bockris, Supervisor

SECTION I

Title of Project: Oxygen-Dissolution Reaction--A Theoretical Study

Long-term Aims: To calculate theoretically the rate of the oxygen dissolution reaction and then study the properties of the metal electrode on which the rate of the reaction depends most. Such a study, if successful, will be of great help in developing a catalyst for the oxygen dissolution reaction.

Specific Aims for This Period: In the previous report we had given a very crude calculation of the rate and activation energy for the oxygen dissolution reaction in a bare electrode in acid solution. The purpose ofsuch a calculation was to see, very crudely, that the rate determining step postulated from experimental studies does in fact give an activation energy in agreement with experimental facts. What we had decided to do in this period was to formulate rigorously our problem. However, before we proceed to do that, we decided to take a good look at the model proposed by the Russian group led by Levich[1] and compare this model with experimental results.

Results of the Period:

The theory developed by Levich[1] for the hydrogen evolution reaction, with the rate determining step as

$$H_2O.H^+ + M(e) \rightarrow H_2O + H_{ads}$$

predicts 3 distant overvoltage regions with different i vs. η relationships. These three distinct regions are:

(a) Low overvoltage region. - This is the region where the overvoltage satisfies the condition $e\eta < (J_{an'} - J^{\circ}_{fn}) - E_s$.

$$i(n,n') = 2eC_s\omega_o K(n,n') \left(\frac{kT\ Es}{2\pi}\right)^{1/2} \rho^* \exp \left[\frac{J^{\circ}_{fn} - J_{an'}}{kT}\right] \exp\left[\frac{e\eta}{kT}\right]$$

(b) Medium overvoltage region. - This region is characterized by the condition, $|e\eta - (J_{an'} - J^{\circ}_{fn})| < E_s$ and the i vs. η relation has the form,

$$i(n,n') = eC_s\rho^*\omega_o K(n,n')kT \exp\left[\frac{J^{\circ}_{fn} - J_{an'}}{2kT}\right] \exp\left[-\frac{E_s}{4kT}\right] \exp\left[\frac{e\eta}{2kT}\right]$$

(c) High overvoltage region. - This is the region where the overvoltage satisfies the

condition $e\eta > (J_{an'} - J_{fn}^{\circ}) + E_s$ and the current potential relationship has the form,

$$i(n,n') = 2eC_s\omega_o\rho^* K(n,n') \left(\frac{kTE_s}{2\pi}\right)^{1/2}$$

Our first aim is to evaluate these potential regions and the current-potential relations and compare them with experiments. According to Levich

$$J_{Fn}^{\circ} = \varepsilon_F - e\phi + E_{oi} - e\eta + n\omega_i(n+1/2)$$

where J_{Fn}° = minimum potential energy of the initial state.

ε_F = Fermi energy of the metal

$e\phi$ = potential of the metal at the reversible potential

= (potential in the ration scale) + (surface potential of the metal at)

= $0.2 + \chi_M$ [for hydrogen evolution on Hg]

E_{oi} = minimum PE of the H^+ ion in solution

= solution energy of H^+ + ionization energy of H

= -267 KCal/mole + 313.22 KCal/mole

So $J_{Fn}^{\circ} = \varepsilon_F - (0.2 + \chi_M e) + E_{oi} - e\eta + n\omega_i \ (n+1/2)$

$= (\varepsilon_F - e\chi_m) - 0.2 + E_{oi} - e\eta$

where we have neglected the $n\omega_i \ (n+1/2)$ term because its contribution is extremely small compared to the other terms in the equation,

So, $J_{Fn}^{\circ} = (\varepsilon_F - e\chi_M) - 0.2 + E_{oi} - e\eta$

$= o_{Hg} - 0.2 \text{ ev} + E_{oi} - e\eta$

$= -4.6 \text{ ev} - 0.2 \text{ ev} + E_{oi} - e\eta$

$= -64.4 - e\eta$

Now J_{an}, = adsorption energy of H or Hg

= -6.08 KCal/mole

So for low overvoltage,

$e\eta < (-6.08 + 64.4 + e\eta) - 46$

or $e\eta$ has to be less than 0.28 volts. Here we have considered E_s the repolarization energy of the solvent to be 2 ev. Now for high overvoltage

$e\eta > (-6.08 + 64.4 + e\eta) - 46$

i.e., $e\eta > 2.3$ volts.

The region in between fits the medium overvoltage condition. The current potential curves for these 3 distinct regions were then evaluated as follows:

(a) Low overvoltage

$$i(n,n') = 2eC_s\omega_o K(n,n') \left(\frac{kTE_s}{2\pi}\right)^{1/2} C^* \exp\left[\frac{J_{Fn}-J_{an'}}{kT}\right] \exp\left[\frac{e_\eta}{kT}\right]$$

where

$$\left(\frac{kTE_s}{2\pi}\right)^{1/2} = 1.452 \times 10^{-13} \text{ ergs.}$$

$K(n,n')$ = transmission coefficient

$$= \text{Const. } \exp[-\alpha R^2]$$

where $\alpha = \frac{m_p}{h} \cdot \frac{\omega_i-\omega_f}{\omega_i+\omega_f} = 1.058 \times 10^{16}$

Taking $R = 0.5A°$

$$K(n,n') = \text{const} \times 0.8442.$$

Since the value of K' should at the maximum be equal to 1, the value of the constant should also be of the order of unity. So,

$$K \simeq 0.8442$$

c_s = surface concentration of H_3O^+ ion

$$= 2r_{H_3O^+} N.C_{H_3O^+} = 1.26 \times 10^{15}$$ for 0.1 molar H_3O^+ sol^n.

ω_o = librational frequency $\simeq 10^{11}$

So, $i(n,n') = 2 \times 4.8 \times 10^{-10} \times 1.26 \times 10^{15} \times 10^{11} \times 0.8442 \times 1.452 \times 10^{-13} \times \rho^* \exp\left[\frac{J_{Fn}^\circ-J_{an'}}{kT}\right] \exp\left[\frac{e\eta}{kT}\right]$

$$= 2\text{x}4.8\text{x}1.26\text{x}0.8442\text{x}1.452\text{x}10^{3}\text{x}10^{23} \exp\left[\frac{J_{Fn}^\circ-J_{an'}}{KT}\right] \exp\left[\frac{e_\eta}{kT}\right]$$

assuming arbitrarily that ρ^* has a value of 10^{23} and it does not vary with potential.

or, log i (n,n') - log(2x4.8x1.26x0.8442x1.452)

$$+ 26 - \frac{(J_{an'} - J_{Fn}^{\circ})}{2.303\ kT} + \frac{e\eta}{2/303\ kT}$$

$$= 1.1708 + 26 - 42.48 + \frac{e\eta}{2.303\ kT}$$

$$= -15.309 + \frac{e\eta}{2.303\ kT}$$

It is continuously mentioned in this report that the calculations were done for Hg. However, this low overvoltage region for Hg has still not been observed. Bowden and Grew[2] has studied hydrogen evolution at low overvoltage but gets a tafel line with a slope of 120 mv not with 60 mv as predicted by Levich.[1] However, Bockris, et.al.[3] does get a slope of RT/F for Pt and some other metals. They have explained that as the situation where the migration of the H atoms on the surface is rate determining. A further study in this direction maybe is to study both the Levich model and the Bockris et.al.[3] explanation of the $\frac{RT}{F}$ slope and try to figure out which one may be correct.

(b) Medium overvoltage

$$i = e\, C_s \rho^* \omega_o K(n,n') kT \exp\left[\frac{J^{\circ}_{fn} - J_{an'}}{2kT}\right] \exp\left[-\frac{E_s}{4kT}\right] \exp\left[\frac{e_n}{2kT}\right]$$

$$\text{or } \log i(n,n') = 11126 + \frac{en}{4.606kT}$$

(c) High overvoltage

$$i = 2eC_s - \omega_{o\rho}{}^* K(n,n') \left(\frac{KTE_s}{2\pi}\right)^{1/2}$$

$$\text{or } \log i = 19.193.$$

This region has not been detected in experiments also. Thus, by plotting the current potential relations predicted by Levich it can be seen that the agreement with experiments in the medium overvoltage region is not bad. However, the other two regions are not unambiguously verified by experiments.

The next thing we tried to compare was the Arrhenius plot obtained from the Levich theory with the experimentally obtained ones; for the medium overvoltage region.

Again the

experimental ones do not differ very much from the theoretically predicted values.

Aims for the next Period:

We plan to carry out some more tests of the Levich theory.

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