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THIRD QUARTERLY REPORT

1 July 1969 to 30 September 1969

STUDIES IN FUNDAMENTAL CHEMISTRY OF FUEL CELL REACTIONS

NGR 39-010-002

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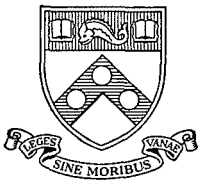

UNIVERSITY OF PENNSYLVANIA

ELECTROCHEMISTRY LABORATORY

PHILADELPHIA, PENNSYLVANIA 19104

THIRD QUARTERLY REPORT

1 July 1969 to 30 September 1969

STUDIES IN FUNDAMENTAL CHEMISTRY OF FUEL CELL REACTIONS

NGR 39-010-002

Submitted to:

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

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Submitted by:

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SECTION I

T. Emi

J.O'M. Bockris, Supervisor

Title of project: Theoretical Study of Electrocatalysis

Specific aims for this period: To establish quantum mechanical expressions for the rate of the rate-determining step and to evaluate the dominant factors affecting the electrocatalytic oxygen reduction.

Results of work in this period: The rate-determining step of the cathodic oxygen reduction reaction on the oxide-free Pt electrode in acid solution has been determined by Brusic and Damjanovic[1] to be

$$O_2 + H^+ + e(M) \rightarrow {}_sOH + {}_sO \text{ or } {}_sHO_2 \quad (1)$$

where e(M) is an electron in Pt, and s represents an adsorption site on Pt.

1. The model for the structure of the Pt electrode-solution interface:

There is some evidence[2] that the proton which takes part in the rate-determining step is located on the O.H.P. In order to make clear the position of the O_2 molecule which is involved in the rate-determining step, relative magnitude of the energies of adsorption on Pt of O_2 and H_2O is calculated.

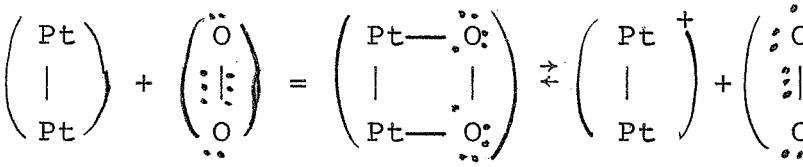
A. Adsorption of O_2 on Pt surface

The energy of physisorption, W_p, of O_2 on Pt is calculated by using the following equation, which is the sum of the attractive (W_{pa}) and repulsive contribution to the energy (W_{pr}).

$$W_p = W_{pa} - W_{pr} \simeq 0.6W_{pa} = 0.6 \left[\frac{f_1 n_0 e^4 \hbar}{32\pi^2 m^{*2} \nu_1^3 D_0^3} - \frac{Ce^2 \alpha_{(o)}^{(i)} (\perp)(0)}{16 r_s D_0^3} - \frac{\mu^2}{8D_0^3} \right] \tag{2}$$

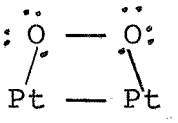
where n_0 is the actual number of free electrons per unit volume of Pt; m^*, true mass of the free electrons in Pt; ν_1, the classical frequency of the oscillator (adsorbed molecule); f_1, the oscillator strength ($= \frac{e}{2\pi}[f/\alpha m]^{\frac{1}{2}}$); $\alpha_{(o)}^{(i)}(\perp)(0)$, the static polarizability of the molecule; r_s, the radius of a sphere containing one metal electron; $C = 2.5$; μ, the dipole moment of the molecule; $\hbar$, the Planck constant divided by 2π; D_0, the distance of the molecule from Pt surface, and e is the charge of an electron. The expression for W_{pa} is due to Margenau and Pollard[3] and Bardeen.[4] Similar to Lennard-Jones,[5] a rather crude approximation of $W_{pr} \simeq 0.4W_{pa}$ is used, because explicit derivation of W_{pr} is very difficult (ref. 6). According to eq. 2, W_p of -3.0 kcal/mole is obtained for O_2 at $D_0 = 2.0$Å.

No sophisticated method seems to be available at the moment which could give quantitative evaluation of the energy of chemisorption of O_2 on Pt surface. The method proposed by Higuchi et al.,[7] therefore, is employed. Several possible versions of the resonant ionic and covalent structures including the following scheme are assumed for the surface complex of Pt and O_2, and corresponding heats of adsorption are calculated by eq. 4.



(3)

$$\frac{1}{C^2} = 1 + (E - H_{ii})/(E - H_{cc}) \quad . \qquad (4)$$

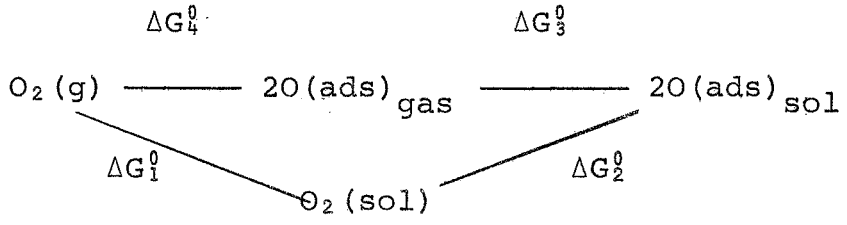
Here, the complex  is that proposed by Aston,[8]

the ion $(\ddot{O} \cdots \ddot{O})^-$ is that confirmed for NaO_2 and KO_2; E, the energy of ideal ionic bond given by $A - I + \frac{8}{9}\frac{e^2}{r}$ (A, electron affinity of O_2; I, ionization potential of Pt, and r is the distance between O_2 and Pt); H_{cc}, the energy of the ideal covalent bond; and C_i^2 is the fraction of ionic bonds in the adsorption bond. However, every assumed resonant structure gives the result that the heat of adsorption of molecular oxygen on the surface of Pt is positive. Thus, O_2 may not be chemisorbed but physisorbed on Pt surface.

B. Adsorption of H_2O on Pt surface

Since the physisorption of O_2 on Pt in aqueous solution takes place in competition with the adsorption of H_2O on Pt, the energy for the latter adsorption was calculated as follows:

Firstly, the free energy change for the dissociative adsorption reaction of oxygen, $O_2(sol) \rightleftarrows 2O(ads, sol)$ is estimated by using the following thermodynamic cycle:



Here,

$$\Delta G_1^0 = -2{,}960 + 3.18T \qquad \text{for } O_2(g) \rightarrow O_2(sol)\ ,^{9}$$

$$\Delta G_3^0 \simeq \frac{2}{3}\Delta H_4^0 \simeq -38{,}000 \qquad \text{for } 2O(ads)_{sol} \rightarrow 2O(ads)_{gas}\ ,$$

$$\Delta G_4^0 = 57{,}900 - 45.47T \qquad \text{for } 2O(ads)_{gas} \rightarrow O_2(g).^{10}$$

Hence, one gets

$$\Delta G_2^0 \simeq 16{,}900 - 13.7T \qquad \text{for } O_2(sol) \rightarrow 2O(ads)_{sol}\ . \quad (5)$$

Secondly, the free energy change ΔG_a^0 of the reaction $2H_2O(ads)_{sol} + O_2(sol) = 2H_2O(sol) + 2O(ads)_{sol}$ is determined as

$$\Delta G_a^0 = -RT \ln[X^2_{H_2O(sol)} X^2_{O(ads)_{sol}} / X^2_{H_2O(ads)_{sol}} X_{O_2(sol)}]$$

$$= -RT \ln[\frac{\theta_0}{1-\theta_0}]^2 \cdot X^{-1}_{O_2(sol)}\ , \quad (6)$$

where θ^0 is the coverage of atomic O on Pt, and is known to be 0.22 for the present situation.[11]

If one assumes that θ^0 does not change appreciably with temperature, one gets from the temperature dependence of $X_{O_2(sol)}$

$$\Delta G_a^0 = -5{,}050 - 6T\ . \quad (7)$$

By subtracting ΔG_2^0 from ΔG_a^0, one gets

$$\Delta G^0_{H_2O} = \frac{1}{2}(\Delta G_a^0 - \Delta G_2^0) = -11{,}000 + 3.9T$$

$$\text{for } H_2O(ads)_{sol} \rightarrow H_2O(sol)\ . \quad (8)$$

C. The amount of O_2 molecules adsorbed on Pt surface

The number of O_2 molecules adsorbed on the unit area of Pt surface may, therefore, be given very roughly by

$$N_{O_2} = \frac{N}{V} X_{O_2} \delta \exp(-8{,}000/RT) \simeq 5\times10^4 \quad (9)$$

where N is the Avogadro number; V, the molar volume of the solution; $\delta \simeq 2\times10^{-8}$ cm; and the free energy change of the reaction $H_2O(ads) + O_2(liq) = O_2(ads) + H_2O(liq)$ is taken to be the difference between the enthalpy part of $\Delta G^0_{H_2O}$ and the physisorption energy of O_2 on Pt. Although this approximation is crude, it may be not unreasonable to assume that the physisorption energy is not so much changed by the coexistence of H_2O molecules around the O_2 molecule adsorbed. It may also be acceptable to assume that the entropy change due to the adsorption of O_2 in H_2O on Pt is not so much different from the entropy term in $\Delta G^0_{H_2O}$.

D. The model for the structure in the vicinity of the Pt electrode-solution interface

As is considered so far, one may assume that a small portion of O_2 dissolved in the solution is physically adsorbed on the surface of Pt electrode replacing the water molecules. Further, there are hydrated protons located at the O.H.P. As far as one believes in the mechanism shown in eq. 1, any O_2 molecule which is placed beyond the first adsorbed layer on the Pt surface should be considered not reduced. The reason is that O_2, H^+ and e(M) must react by one act to form adsorbed HO_2 or OH and O. One can hardly visualize the fact that HO_2 or OH and O formed beyond the first adsorption layer will consequently be adsorbed on the Pt surface replacing the H_2O molecules previously adsorbed on the surface. The qualitative figure of the sequence of the rate-determining step may then be as follows:

$$ {}_sO_2 + H^+ + e(M) \rightarrow (HO_2^*) \rightarrow {}_sHO_2 \quad \text{or} \quad {}_sOH + {}_sO \qquad (10) $$

i.e., at the moment O_2 and H^+ are, due to the overall effect caused by thermal fluctuation, displaced from their stable position and encountered in such a way as to provide on the resultant O_2-H^+ complex an energy level which is equal to the energy of an electron which has more than the Fermi-energy of Pt, electron transfer occurs with some probability, yielding (O_2H^*) activated complex. Then, the latter complex either dissociates into OH and O or remains as HO_2 which are subsequently adsorbed on the Pt surface.

2. Qualitative expression for the current density

The current density expression for the rate-determining step may be given by

$$i = \frac{e}{h} \int_{E_1}^{E_2} \int_{0}^{x'} \frac{\partial E}{\partial K} C_{H^+} C_{O_2} n(E) g(E) P(E,x) Q(E,x) dE dx \quad .)11)$$

Here, E is the energy of an electron, k is the wave vector of an electron in Pt, C_{H^+} and C_{O_2} are the concentrations of hydrated protons and physically adsorbed oxygen molecules in the vicinity of the electrode per unit area, n(E) is the probability that an electronic state at energy E is occupied, g(E) is the number of state at energy E, P(E,x) is the tunneling probability for an electron which neutralizes the O_2-H^+ complex in the rate-determining step, and Q(E,x) is the factor which will be discussed later.

If one assumes nearly free gas behavior (in the sense of the effective mass approach) for the electron in Pt, eq. 11 turns into a simple form, i.e.,

$$i = \frac{e(2m)^{\frac{1}{2}}}{\pi^2 h^2} \int_{E_1}^{E_2} \int_{0}^{x'} \{ \frac{kC_{H^+} C_{O_2} E^{\frac{1}{2}}(E,x)}{\exp[(E-E_f)/kT]+1} \exp[\frac{2}{h} \int_{x_1}^{x_2} [2m(V-E)]^{\frac{1}{2}} dx\} dx dE \quad (12)$$

where E_f is the Fermi energy for Pt, m is the mass of an electron, $x_2 - x_1$ and V are the width and the height of the potential barrier between Pt electrode and a O_2-H^+ complex. E_1 corresponds to the lowest electronic ground state for the complex in the system, E_2 corresponds to the highest energy of the electron in Pt, and x' extends to the distance at which neutralized complex can still form adsorbed HO_2 or OH and O on the electrode surface. Q(E,x) reflects the characteristics of the reaction. This is the Boltzmann factor which gives the possibility of having a O_2-H^+ complex which is able to receive an electron from the electrode and is able to turn into adsorbed HO_2 or OH and O. Denoting the energy differenc between the ground state of the initial state [O_2 + H^+ + e(M)] and the cross point of the two energy-distance curves (one is for the energy of the initial state - the distance from the electrode surface, another is for the energy of the final state HO_2 - the distance) by $\Delta\varepsilon$, one can write

$$Q(E,x) = \exp -\Delta\varepsilon/kT \quad (13)$$

Since $\Delta\varepsilon$ is the function of the vibrational and rotational energy of the O_2-H^+ complex in the normal or distorted first adsorbed layer on the electrode surface, it may be reasonable to relate $\Delta\varepsilon$ to the electronic states E_1 and E of the complex as

$$Q(E,x) = \exp[-f(E - E_1)/kT] \quad (14)$$

where the value of $f(E - E_1)$ is determined mainly by the interaction between HO_2 and H_2O molecules as well as the interaction

between HO_2 and the electrode surface. It must be emphasized here that E_1 involves the contribution from the electric field in the double layer. According to the recent data[12] and some elementary calculations, the values shown in Figure 1 are known for HO_2 and HO_2^+. It is quite difficult, however, to estimate the above mentioned interactions because the electronic properties of these species are unclear at the moment. Although quantitative examination of the present approach is left in the future, suggested interface model (section 1) and eq. 12 (section 2) will provide useful tools for calculating the current density of the reaction.

3. Some comments

The modification of eq. 12 to give a more realistic solution of eq. 11 is the differentiation of E in eq. 11 at the surface of the Fermi sphere of the metal in question, and replacing, in the vicinity of the Fermi level, the expression for the density of the electronic state by a suitable analytic function which fits the $n(E)g(E)$ curve given by the superposition of higher order calculation. Since the curve sometimes extends (as in the case of Ni) far beyond the Fermi level with appreciable $n(E)g(E)$ value, the integration in eq. 11 may become large provided that the $Q(E,x)$ term makes many O_2-H^+ complexes available at the corresponding energy level. Thus, not only the increase in the number density of the electrons in the metal but also in the span of the integration in eq. 11 may increase the value of the current density. Investigation of the value Q as a function of E and x is awaited from this point of view.

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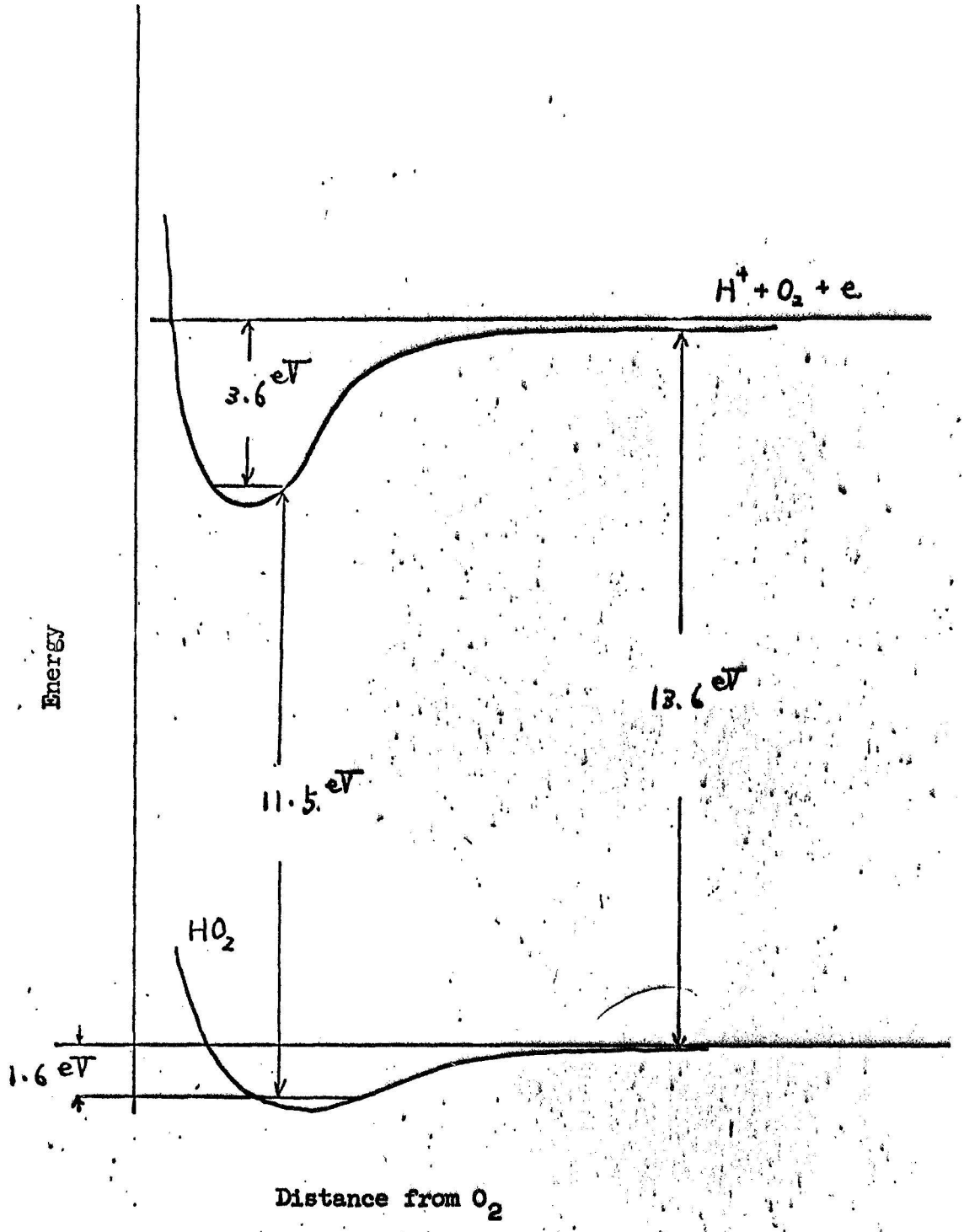

Fig. 1

SECTION II

John W. Diggle

A. Damjanovic, Supervisor

Title of project: The Study of the Dendritic Deposition of Zinc from Alkaline Solution

Long-term technological aims: To gain (a) an understanding of the formation of zinc dendrites, (b) a mechanism which adequately describes the experimental behavior, (c) the ability to apply simple equations to the growth of metal deposits in dendritic and non-dendritic forms, and (d) to obtain a basis for evaluating the effectiveness of inhibitors in the problems associated with the zinc negative electrode, i.e., in dendrites and negative capacity loss problems.

Specific aims of this report period: To extend earlier work[1] concerning inhibition to the polyethoxylated compounds, such as the Emulphogenes, Igepols and Tritons.

New evidence revealed and conclusions reached: In these initial evaluations of the polyethoxylated compounds Emulphogene BC-420, Triton X-15, and Triton X-100 were examined. Emulphogene BC-420 and Triton X-15 have approximately the same ethylene oxide

content but differ in the nature of hydrocarbon fragment--BC-420 being aliphatic and Triton X-15 aromatic. Triton X-15 and X-100 were chosen to ascertain any possible influence of the ethylene oxide content upon inhibitor efficiency.

The influence of these three compounds upon the total current during zinc deposition is typified by Figure . which shows that, as the compound (Emulphogene BC-420 in this case) is adsorbed at the zinc surface, the total current at a given potential decreases. The greater the concentration of Emulphogene, the faster the total current for zinc deposition diminishes (Figure 1). This concentration dependence would seem to imply a mass transport controlled adsorption of Emulphogene, as suggested by Payne, Tachikawa and Bard.[2]

Following adsorption of the Emulphogene upon the zinc electrode, the polarization curve shown in Figure 2 was determined. This shows that at an overpotential of -240 mV the total current increases rapidly--this apparently as a consequence of desorption. The current is then equal to the limiting zinc current density in the absence of inhibitor. Upon lowering the overpotential, substantial hysteresis is observed. The descending arrows in Figure 2 indicate that the total current was decreasing with time.

With regard to the initiation of dendrites, at point A (Figure 2) no initiation was observed in 180 min, whereas at point B (desorbed inhibitor) initiation proceeded as in the uninhibited system (10 min). Initiation was only found to occur whenever the initiation time was much shorter than the time required for inhibitor adsorption.

Following initiation in the presence of Emulphogene BC-420, dendrites were seen to propagate for a short time only before propagation was seen to be diminished--this would be in accordance with a mass transfer controlled adsorption of Emulphogene, i.e., adsorption being sufficiently slow (under linear conditions) to prevent initiation, but sufficiently fast under spherical diffusion conditions to diminish dendrite propagation.

Figure 3 shows the polarization curve for zinc deposition in the presence of Triton X-15. Desorption is seen to occur at -80 mV. Initiation and propagation of dendrites proceeds as in the uninhibited systems at η = -100 mV.

Figure 4 shows the equivalent polarization curves for the inhibitor Triton X-100. Desorption at higher negative overpotentials than Triton X-15 is experienced. Considerable hysteresis and an inhibitor concentration dependent desorption potential was observed, as in the case of Emulphogene BC-420 (cf. Fig. 2). Similar observations with regard to initiation and propagation of dendrites in the presence of Triton X-100 were obtained as were obtained with Emulphogene BC-420. In Figures 2 and 4 high overpotentials were required to promote desorption. At these high overpotentials, substantial evolution of H_2 is observed. Hence, when recharging a zinc electrode at constant current, Emulphogene BC-420 and Triton X-100 are not expected to be beneficial, and are actually detrimental by promoting increased gassing (as reported by Lander et al.[3]).

Conclusions with regard to dendrite inhibition are (cf. previous reports):

(1) The tetraalkyl ammonium salts are far superior to any of the polyethoxylated compounds examined here.

(2) Emulphogene BC-420 and Triton X-100 may actually be detrimental to battery operation due to evolved H_2.

(3) Any dendrite inhibition due to Emulphogene addition (such inhibition has been observed[4]) can be obtained only after initiation and at high Emulphogene concentrations.

Specific aims for the next report period: Theoretical analysis of the obtained results.

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LEGENDS TO FIGURES

Figure 1. Total current (representing the zinc deposition current) as a function of time during the adsorption by zinc of Emulphogene BC-420. Conditions: 10% KOH + 0.1M zincate, η = -50 mV. T = 30±1°C.; × 3.7×10^{-5}M and o 7.4×10^{-5}M Emulphogene.

Figure 2. Polarization curve (η-log i) for a zinc electrode inhibited with Emulphogene BC-420. Conditions: 10% KOH + 0.1M zincate, T = 30±1°C; ● 7.4×10^{-5}M Emulphogene--cathodic direction; × 7.4×10^{-5}M Emulphogene--anodic direction; o 3.7×10^{-5}M Emulphogene--cathodic direction.

Figure 3. Polarization curve (cathodic direction) for a zinc electrode inhibited with 1.5×10^{-4}M Triton X-15. Desorption potential -80 mV zinc overpotential. Base electrolyte and temperature as for Figure 2.

Figure 4. Polarization curve for a zinc electrode inhibited with Triton X-100. Base electrolyte and temperature as for Figures 2 and 3. o 1.76×10^{-4}M Triton X-100--cathodic direction; × 1.76×10^{-4}M--anodic direction; Δ 1.4×10^{-4}M--cathodic direction; □ 8.8×10^{-5}M--cathodic direction.

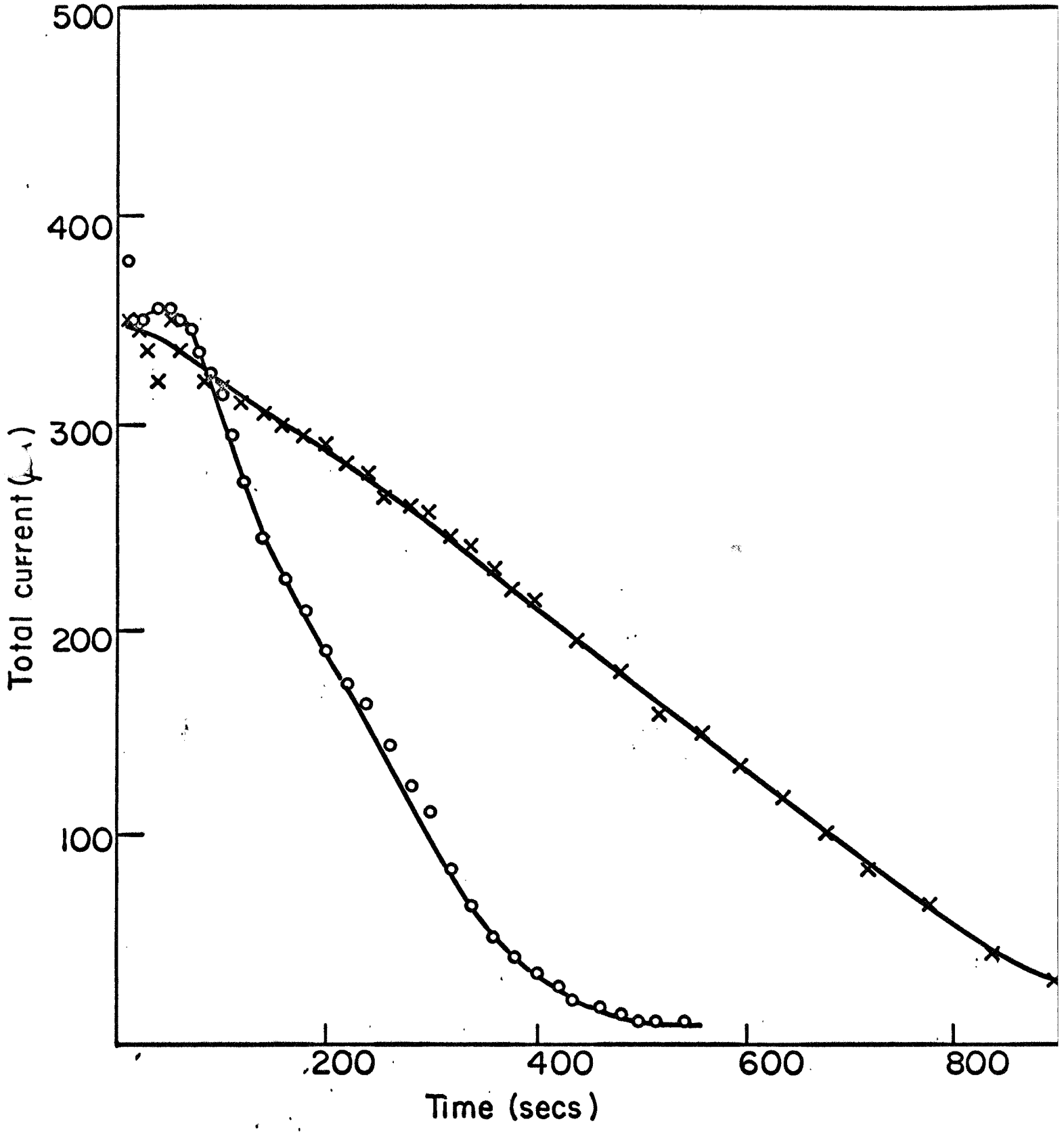
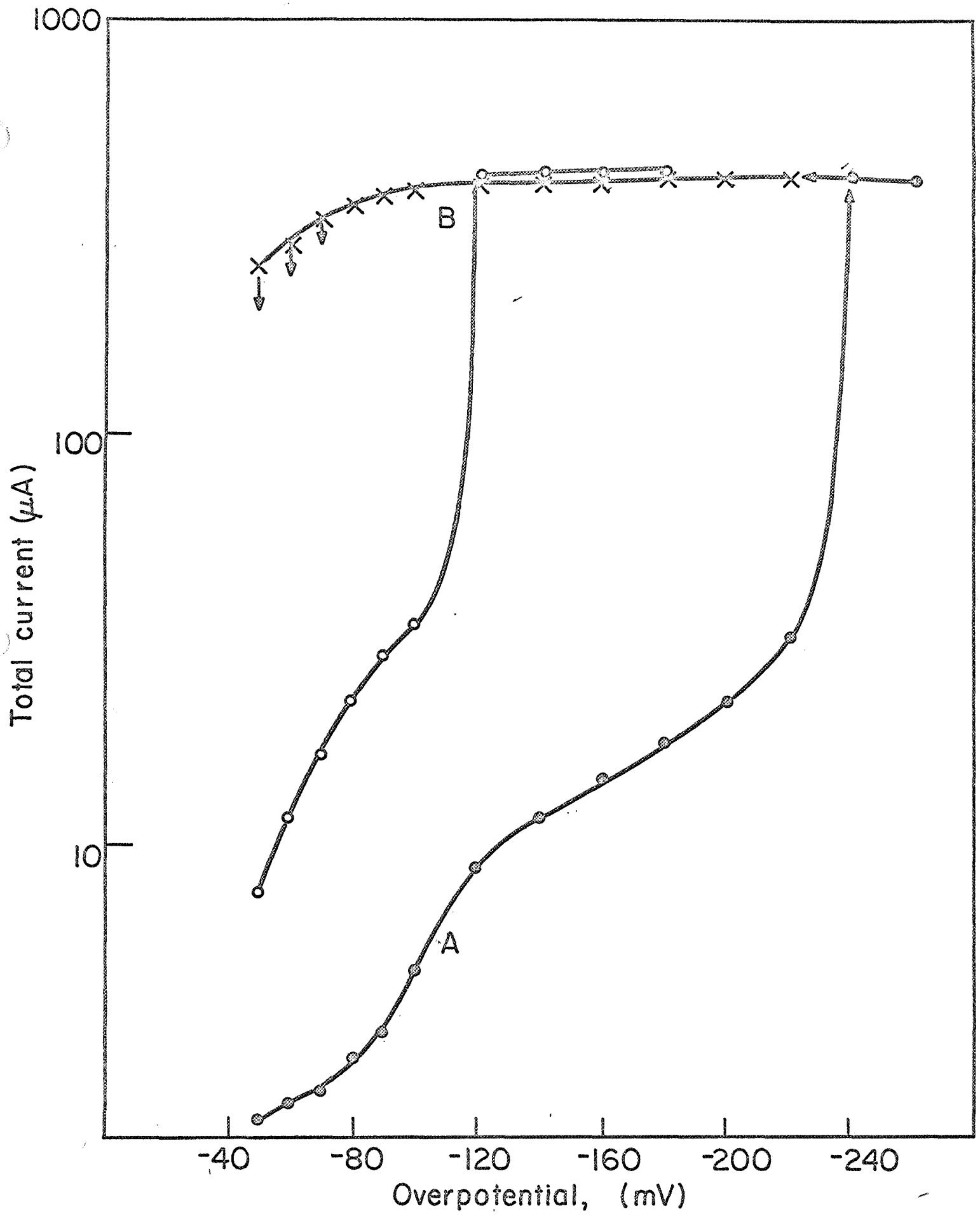


Fig. 1


1000
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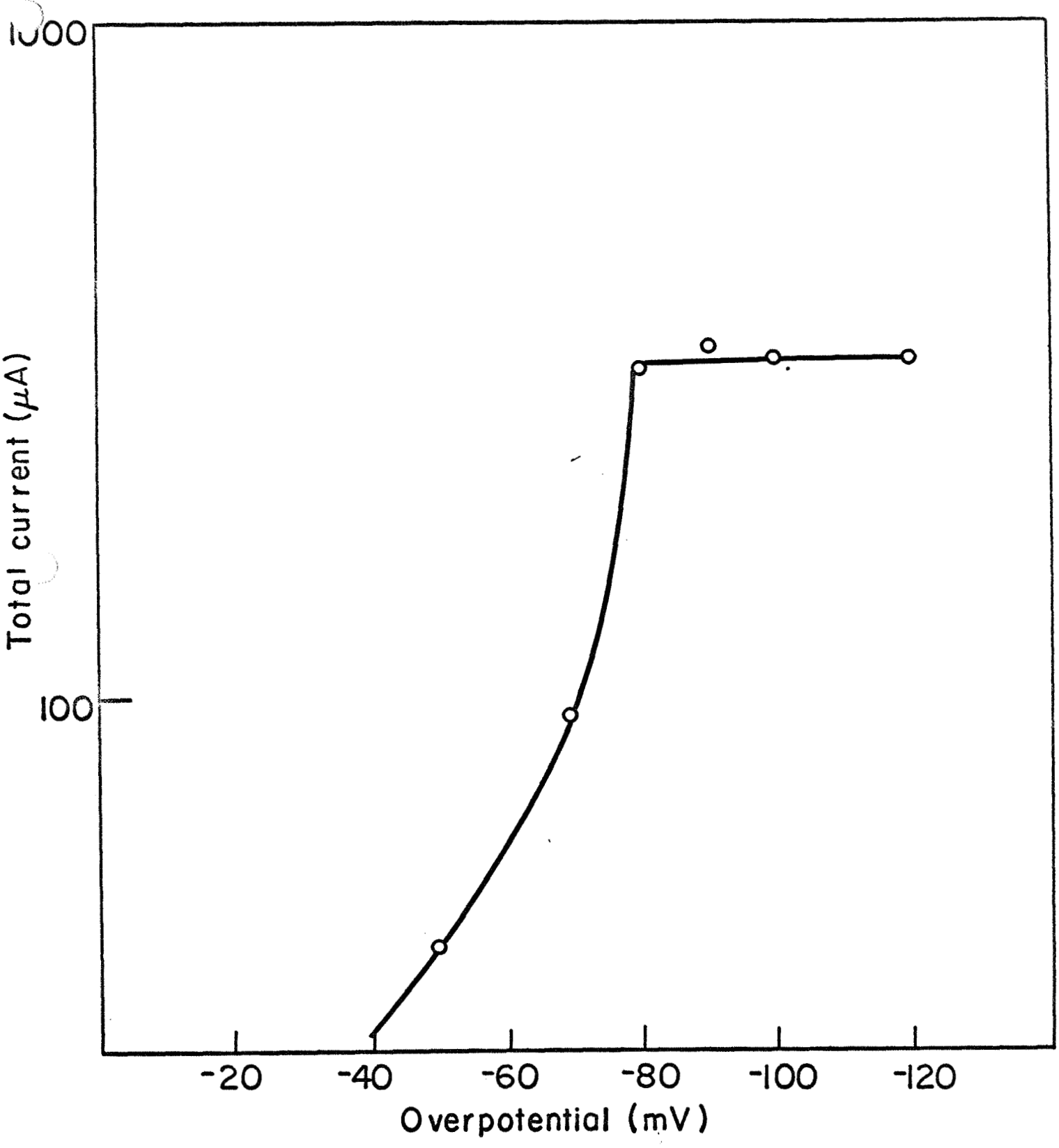
10
Total current (μA)
B
A
-40
-80
-120
-160
-200
-240
Overpotential, (mV)

Fig. 2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Total current (μA)
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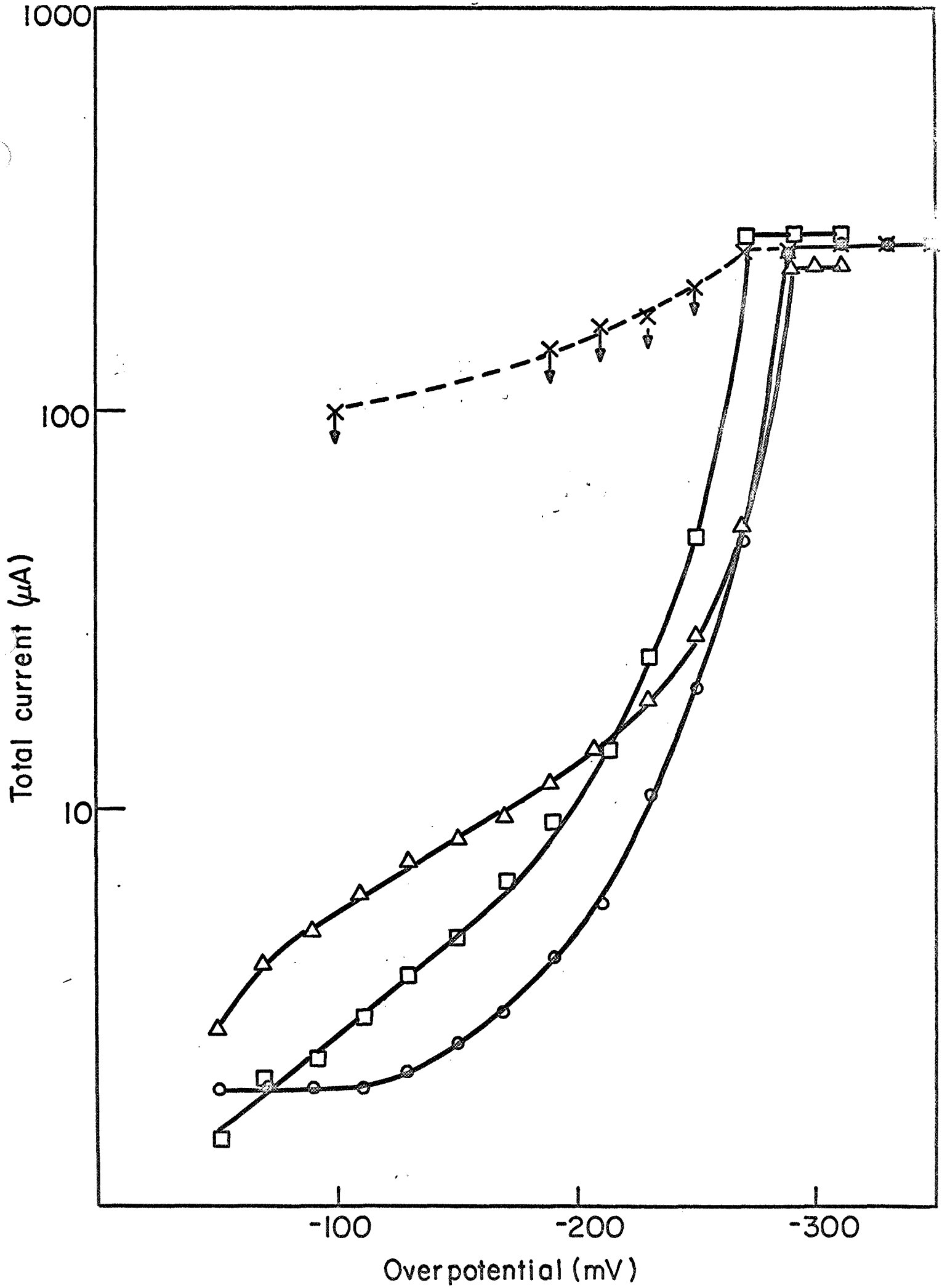
-40
-60
-80
-100
-120
Overpotential (mV)

Fig. 3


1000
100
10
Total current (μA)
-100
-200
-300
Overpotential (mV)

SECTION III

D. Cipris

A. Damjanovic, Supervisor

Title of project: Reversibility of Organic Reactions

Long-term technological aims: Investigation of capability of organic compounds for use in high energy secondary batteries.

Specific aims for this period: The kinetic study of electroreduction of rhodizonic acid (RA) to hexahydroxylbenzene (HHB), and reverse. Reaction examined at platinum and gold electrodes.

Results of work in this period: RA and HHB form reversible couple with 4 electrons transferred at the electrode according to the over-all reaction:

$$RA + 4H^+ + 4e \rightleftarrows HHB$$

The dependence of the electrode potential on concentration ratio of Ox/Red form according to the Nernst equation should be 15 mV (at the same pH). Experimentally, this dependence was found to be 17 mV (at pH 4). The pH dependence of the electrode potential at the same concentration ratio of Ox/Red is found to be 52-57 mV, compared to theoretically expected 60 mV per pH.

The set of kinetic parameters obtained by galvanostatic transients are shown in Table 1. The measurements were made at three pH's: 4, 7, and 9, but at pH 9 the solution proved to be unstable, and the conclusions are drawn on the basis of the measurements at pH 4 and 7.

It is seen from the table that the reaction is concentration independent in the cathodic direction, and only slightly dependent in the anodic direction. The Tafel slopes are the same on both metals, in both directions, and correspond to 120 mV or to 2RT/F. The pH dependence, at both metals, is observed only on the cathodic side of the reaction.

On the basis of the obtained kinetic parameters the tentative mechanism of the reaction has been postulated, according to which the hydrogen discharge reaction is rate-determining step, which is then followed by fast hydrogenation of RA in cathodic reaction, or proceeded with fast dehydrogenation of HHB in anodic reaction.

The exchange current density of the reaction is high, being equal to 1×10^{-3} A cm^{-2} on Pt, and to 5×10^{-4} A cm^{-2} on Au. The reversible potential is found to be +210 mV vs. NHE.

The long-time polarization measurements--in terms of hours--are carried out in a four-compartment cell with anodic and cathodic compartments separated with fritted glass, and with the possibility of measuring the electrode potential at each electrode separately. Gold wire electrodes, 3 cm^2 each, were chosen as working electrodes for these long-time experiments.

With 0.50 g of solid RA in approximately 1×10^{-2} M RA at pH 4 driven cathodically in one compartment, and with 0.50 g of solid HHB in approximately 10 ml 1×10^{-2} HHB driven anodically in the second compartment, the experiment was carried out for 50 hours in one direction, and then after changing polarity of the electrodes for another 50 hours in the opposite direction. Current density was 0.2 mA cm^{-2}. Electrode potentials were relatively stable during this long run (flat e vs. t curves). pH of the solutions before and after this long run shows only a difference of less than half of a pH unit.

At higher current densities (i = 1-2 mA cm^{-2}), the electrode at which HHB is oxidized becomes covered with black solid particles. The potential then increases. It is believed that black powdery particles cover the surface and reduce the active surface area. At lower current density this side effect is not observed. It is noted that at platinum electrode black powder formed more readily than at Au, and for that reason Au was chosen for long-time polarization measurements. The solid precipitates are believed to be the Na-salt of RA (or even of THQ), since RA is known to form the salts with a whole series of cations like Na, K, Li, Ba, Sn, Hg, Pb, and is used in analytical chemistry as a reagent for some of them.

The experiment with 1 mA cm^{-2} was carried out for 3 hours without much difficulty arising from the formation of black powder.

Specific aims for next report period: Preliminary testing of RA (mixed with carbon black) as a cathode, coupled with Mg and/or

Zn as an anode will be made under conditions similar to those existing in practical systems.

Table 1. The kinetic parameters for the electrode reaction $RA + 4H^+ + 4e \rightleftarrows HHB$ at Pt and Au electrodes in $0.1N\ Na_2SO_4$

	Pt		Au	
	Anodic	Cathodic	Anodic	Cathodic
Tafel slope	120	-120	120	-120
$(\frac{d \log i}{d \log c})_{V,pH}$	≤ 1	0	≤ 1	0
$(\frac{d \log V}{d pH})_{c}$	0	-120	0	-120

SECTION IV

Woon-Kie Paik

M. Genshaw, Supervisor

Title of project: Ellipsometric Studies of Ion Adsorption on Solid Electrodes

Long-term technological aims: The purpose of this project is to study the adsorption of ions on electrodes of different metals by means of ellipsometry and to investigate the nature of interaction between the ions and electrode materials.

Specific aims for this period: (1) To develop model and theory for the correlation between the optical response and ion adsorption.

(2) Numerical calculation of amount of adsorption from the experimental data.

(3) To correlate adsorption energies to the properties of metals and anions.

Results of work in this period: Theoretical calculations are made for the correction due to the "electromodulation" effect. The results can be summarized as

$$\delta\Delta_{modn} = -bdq$$

where $\delta\Delta_{modn}$ is the ellipsometrical response (relative phase retardation) due to change in electrode charge (dq) only.

The constant b has values of .0042, .0029, .0010, .0017, .0017, and .0021 deg $\mu coul^{-1}$ cm^2 for Au, Ag, Pt, Ni, Rh, and Hg, respectively.

By taking the difference between the observed optical response and the above theoretical $\delta\Delta_{modn}$, the amount of adsorption of ion is calculated. In this calculation an optical model, which yields a linear combination of Lorenz-Lorentz functions of solvent and ions to give the molar refraction of the mixed adsorption layer, was used. The results are shown in Figures 1-5.

A paper which describes experiments, results and discussions including correlation of adsorption energy and properties of metals is being written for publication.

Specific aims for next period:

Pending further analysis of the data and fuller discussion over the nature of adsorption forces, the paper will be finished and submitted for publication.

Possibly more experiments will be planned for the investigation of kinetics of ion adsorption.

CAPTIONS TO FIGURES

Figure 1. Adsorption of anions on nickel and rhodium electrodes.

Figure 2. Adsorption of Cl^- and ClO_4^- on silver electrode.

Figure 3. Adsorption of Br^- and $SO_4^=$ on silver electrode.

Figure 4. Adsorption of Cl^- and ClO_4^- on gold electrode.

Figure 5. Adsorption of Br^- and $SO_4^=$ on gold electrode.

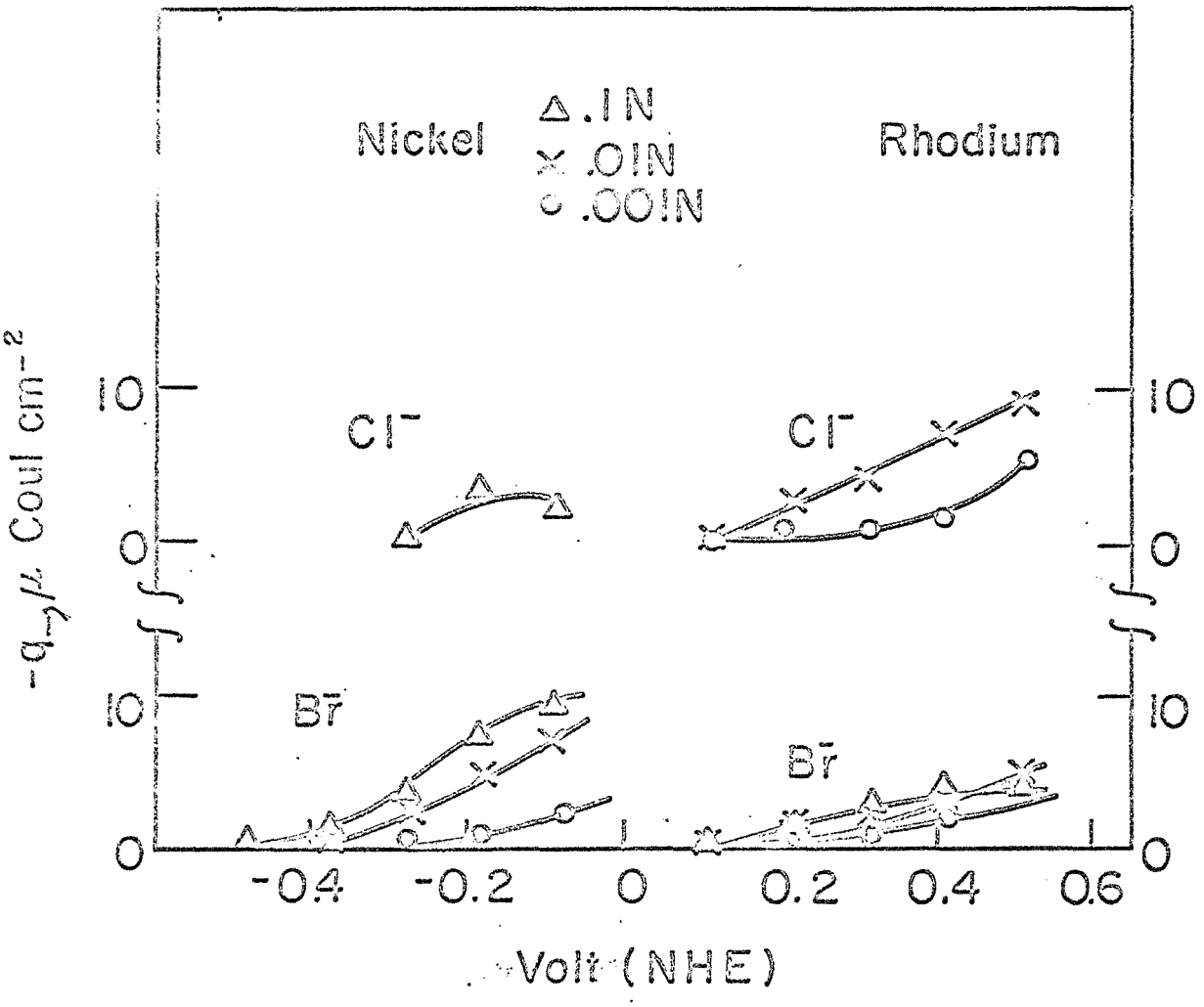

Nickel
Rhodium
△ .1N
× .01N
○ .001N
Cl⁻
Cl⁻
Br⁻
Br⁻
10
0
10
0
10
0
10
0
-0.4
-0.2
0
0.2
0.4
0.6
Volt (NHE)
$-q_{M}/\mu$ Coul cm^{-2}

FIG. 1

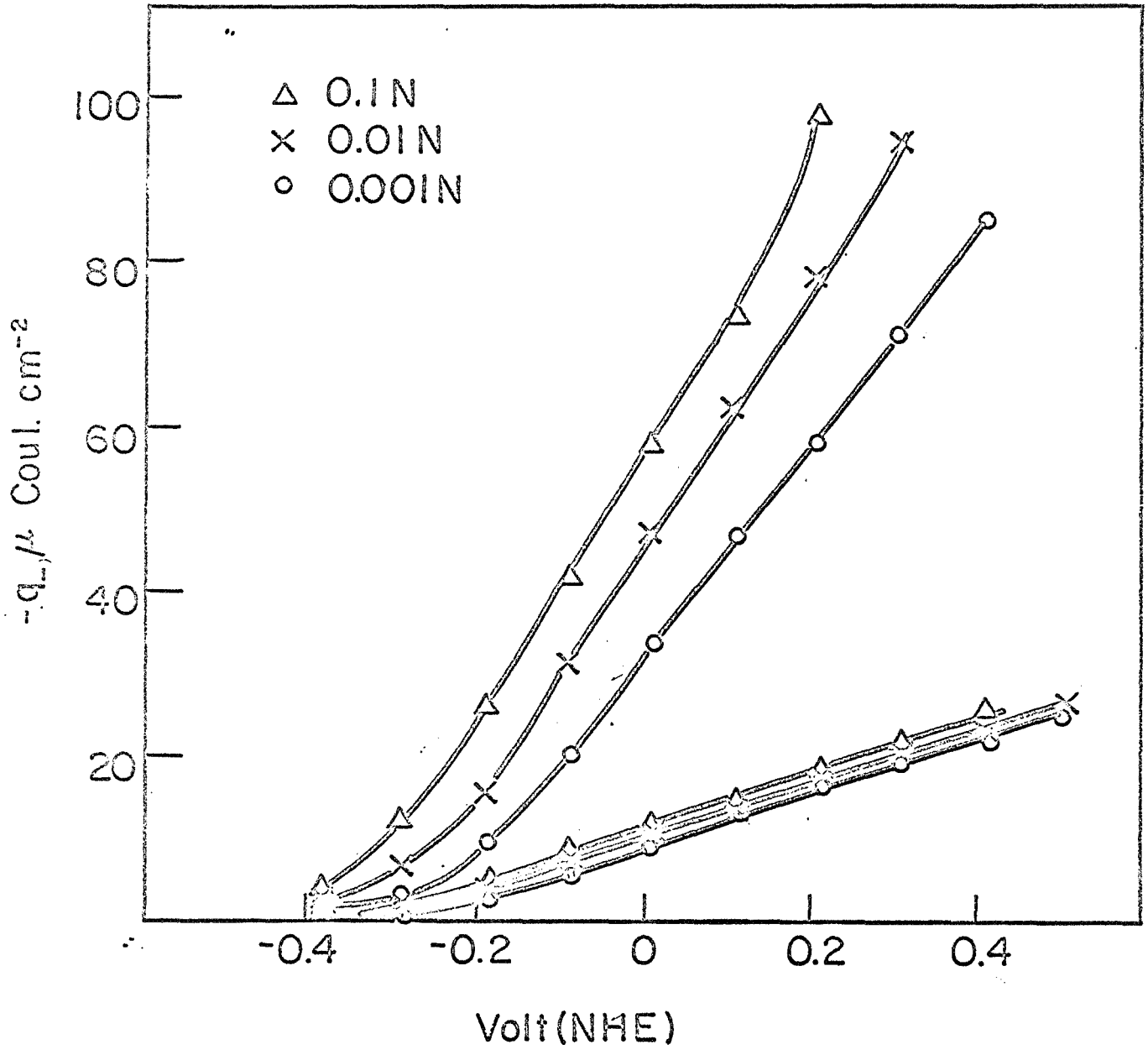


FIG. 2

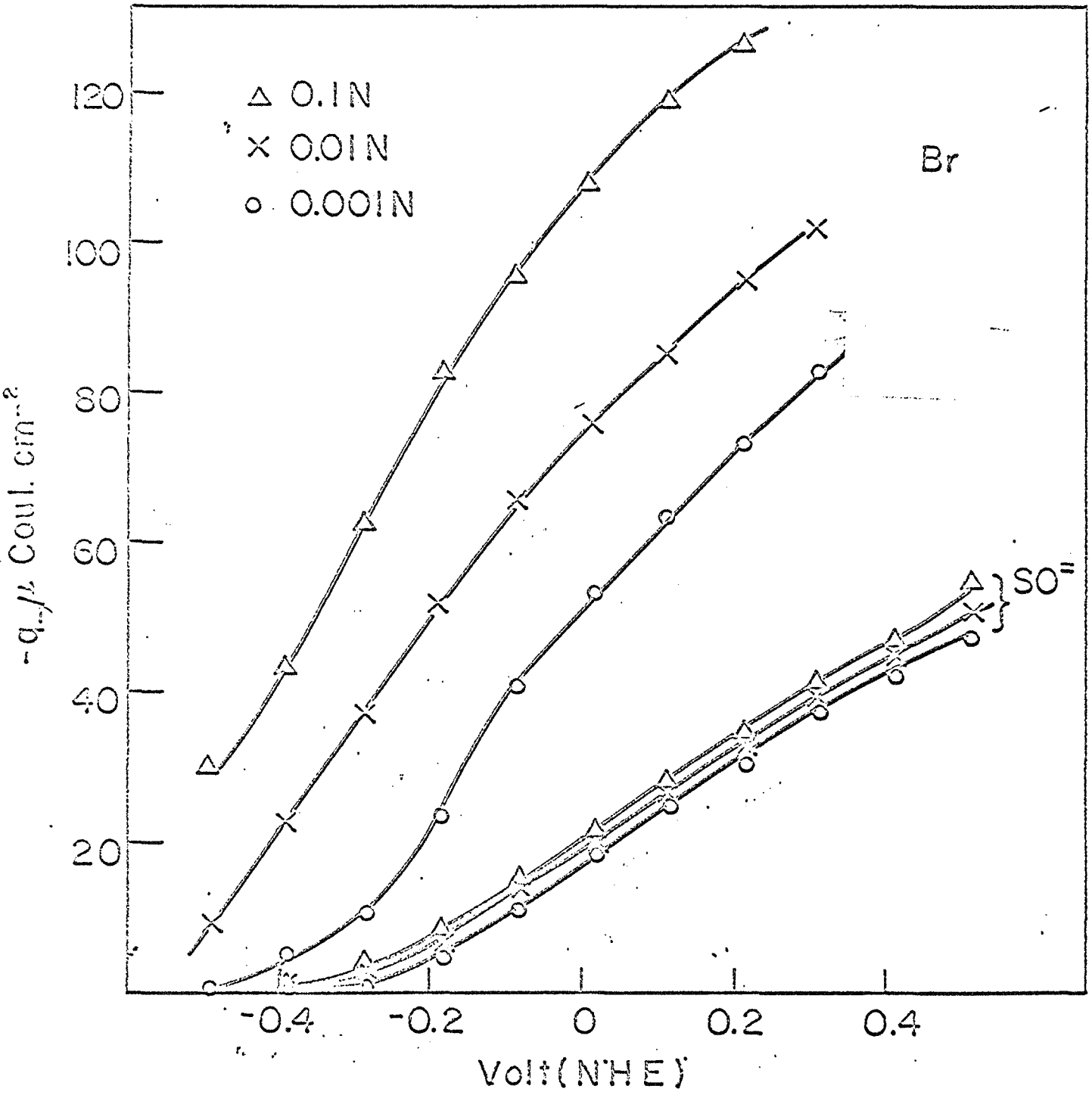

△ 0.1 N
× 0.01 N
○ 0.001 N
Br
SO=
120
100
80
60
40
20
-0.4
-0.2
0
0.2
0.4
Volt (N.H.E)
-q_M/μ Coul. cm⁻²

FIG. 3

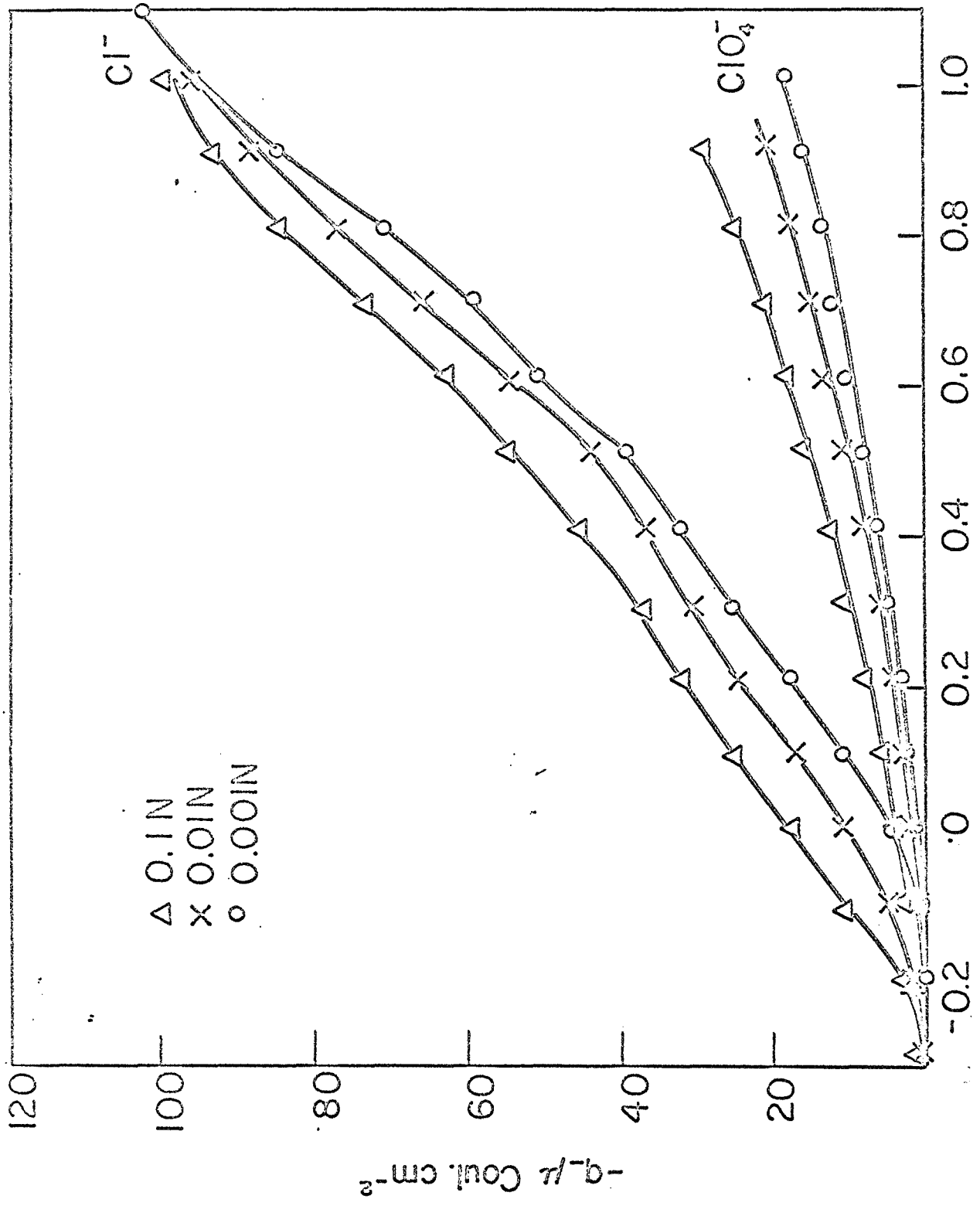

Cl⁻
ClO₄⁻
Δ 0.1 N
× 0.01N
○ 0.001N
-q_M Coul cm⁻²
120
100
80
60
40
20
-0.2
0
0.2
0.4
0.6
0.8
1.0
Volt (NHE)

FIG. 4

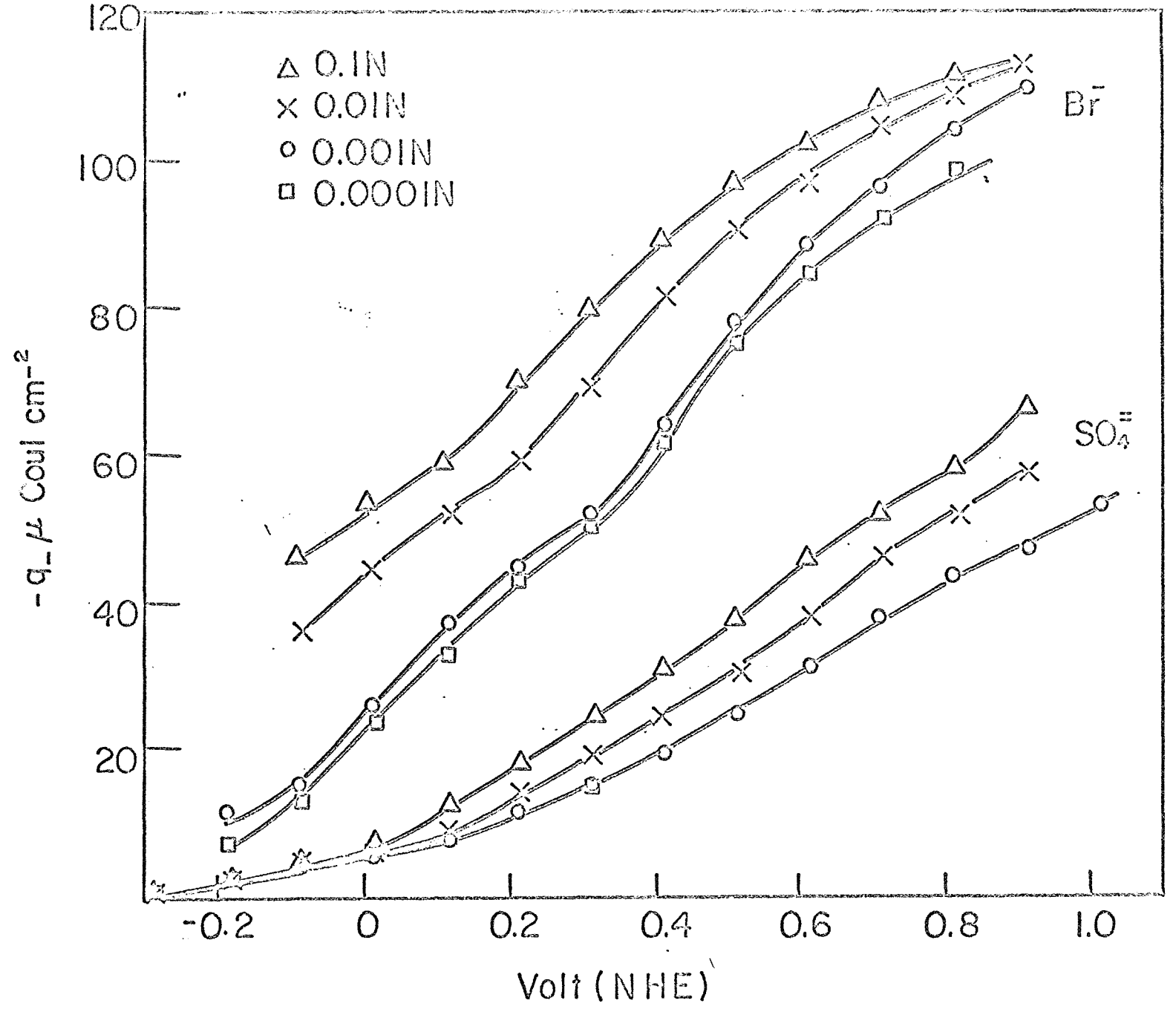

120
100
80
60
40
20
-q_μ Coul cm⁻²
△ 0.1N
× 0.01N
○ 0.001N
□ 0.0001N
Br⁻
SO₄⁼
-0.2
0
0.2
0.4
0.6
0.8
1.0
Volt (NHE)

FIG. 5

SECTION V

David LaPointe and Rajat K. Sen

J.O'M. Bockris, Supervisor

Title of project: Sodium Diffusion in Crystals

Long-term aims: The purpose of this project is to study theoretically the dependence of the activation energy for Na^+ ion diffusion through a crystal lattice, on the various structural parameters. The reason for such a study is to find out why β-alumina has such a low activation energy for Na^+ ion diffusion, and if possible, to hypothesize an ideal lattice which will have very low activation energy for Na^+ ion diffusion.

Specific aims for this period: In the previous report it was mentioned that a model was set up similar to the Burton and Jura[1] model which allowed us to calculate the activation energy for self diffusion in solids under a certain set of approximations. The aim for this period was to test this model for as many cases as possible.

Results of work in this period: It was decided that the above-mentioned method should first be applied to the simplest case of self diffusion in rare gas solids. A Lennard-Jones potential function of the type (suggested by H.R. Glyde[2])

$$V(r) = t\left(\frac{6m}{m-6}\right)\left\{\frac{1}{m}\left(\frac{r_0}{r}\right)^{m} - \frac{1}{6}\left(\frac{r_0}{r}\right)^{6}\right\}$$

was used to calculate the energy of the migrating particle as it moves from one position to the other. It was seen that contribution to the energy for sixth and higher neighbors was almost negligible. Hence in calculating the energy of the migrating particle, summation up to the fifth nearest neighbors was done. These calculations were done for both argon and krypton self diffusion and gave results which were in quite good agreement with theory.

The next case attempted was Na^+ diffusion through a NaCl lattice. The ionic crystal was assumed to behave according to the Born[2] model and consequently the potential function was supposed to be of the form[3]

$$V(r) = -\frac{q^2}{r} + \left[M\beta_{+-}b_+b_- \exp(-r/\rho) + \frac{1}{2}M'(\beta_{++}b_+ + \beta_{--}b_-) \exp(-r'/\rho)\right]$$

The first term is the coulombic attraction and the second term is the Huggins-Mayer repulsion term. It was found that the coulombic energy oscillates and dies out after a long distance from the original lattice point. So it was decided that, to use the above-mentioned potential function, we have to generate a very large NaCl crystal in the computer. Ultimately a computer program was developed which builds a crystal of 1100 ions in the computer and the coulombic energy calculated for such a crystal. It was also found that increasing the crystal size by a large amount does not alter the coulombic energy very much. So the migration energy calculation was done with such a crystal. The results obtained were fairly good. Relaxation energy calculation for such a process is still going on.

Calculation for MgO has just begun, and no results worth mentioning have yet been obtained.

Specific aims for next report period: The aim for the next period is to complete the NaCl and MgO calculations and start the β-alumina case. After completing β-alumina we propose to look into the structural parameters which can radically change the energy at any lattice point--hoping to find some parameters which when suitably altered minimize the activation energy for diffusion.

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3. Mario P. Tose, "Cohesion of Ionic Solids in the Born Model," in Solid State Physics, Vol. 13.

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