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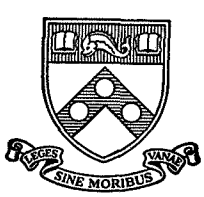
1 APRIL 1970 to 30 JUNE 1970
STUDIES IN FUNDAMENTAL CHEMISTRY
OF FUEL CELL REACTIONS

NGR 39-010-002



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ELECTROCHEMISTRY LABORATORY

PHILADELPHIA, PENNSYLVANIA 19104

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STUDIES IN FUNDAMENTAL CHEMISTRY
OF FUEL CELL REACTIONS

NGR 39-010-002

Submitted to:

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

Washington, D.C. 20546

Submitted by:

Professor John O'M. Bockris
The Electrochemistry Laboratory
University of Pennsylvania
Philadelphia 19104

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Mr. R. Sen, Graduate Student

Dr. J.O'M. Bockris, Supervisor

Section II

Mr. J. Ricodeau, Graduate Student

Dr. J.O'M. Bockris, Supervisor

SECTION I

Title of the 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 above 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 this period we decided to finalize the model to be used in the calculation, and then start doing a rough calculation on the lines of Parsons and Bockris,[1] and see what sort of activation energy values we get.

Results of the Period: The model decided was the following: In deciding the model the rate determining step found by Damjanovic and Brusic[2] was always used. It is

$$O_2 + H^+ + e \rightarrow O_2H_{ads}$$

Once this rate determining step was decided upon, the initial state was

then defined as the electron in the metal, + O_2 molecule in solution, + the solvated proton. The final state is the H_2O complex adsorbed on the metal electrode. With the above rate determining step it is obvious that the O_2 and the solvated proton, due to some interactions, must attain a particular energy when the electron from the metal will be transferred to the $O_2...H_3O^+$ complex in solution to produce either the adsorbed HO_2 or the adsorbed OH + O species on the electrode surface. So the rate of the reaction should be directly proportional to the product of the probability of attaining the particular geometry for the $O_2...H_3O^+$ complex having a particular energy and the probability of electron transfer from the metal to the $O_2...H_3O^+$ complex.

Now the problem is to calculate these two probabilities. Before we discuss the means of calculating the probability of the initial state attaining a certain configuration we should define what that needed configuration is. It is generally accepted that electron transfer in the

electrochemical reaction is generally radiation-less in nature, i.e., no energy is absorbed or liberated during the electron transfer process. So the configuration which the $O_2...H_3O^+$ complex must attain is the one to which a radiation-less electron transfer can occur. A radiation-less electron transfer can occur only at the point where the initial and final states have equal energy. So the configuration of the $O-H_3O^+$ complex must be such that its energy will be equal to the final state. This condition can be established in the following way.

Fig. 1 represents the model of the initial state. We must calculate the energy of the initial state as r_1 changes, which automatically means that r_2 is changing. And for each r_1 value the contribution to the energy due to the variation of r_3, r_4, and r_5 must be calculated. If this can be done, then we will get a curve, Fig. 2, of the electronic energy of the initial state (U) versus r_1 or r_2. Similarly, Fig. 3 represents the final state model and we can plot the energy

of the final state as a function of r_1', taking into account the variation of r_3', r_4', r_5' to get Fig. 4. This model of the final state says that the primary solvation structure of this state is very much like that of the initial state. Now if we plot Fig. 2 and Fig. 4 together, taking care of placing the minimum of the suitable curves at the correct points, we get Fig. 5. In Fig. 5 the point of intersection represents the point where the energy of the initial and final states are equal. So we now have to calculate the probability of the initial state attaining that energy.

The next problem is then to calculate the electron transfer probability using the Landau-Zener approximation. This is the complete model of the system. Right now we have started a calculation along the lines of Bockris and Parsons[1] to get a very rough estimate of the activation energy and its dependence on the M–O bond strength.

Specific Aim for Next Period:

In the next period we would like to complete the Parsons-Bockris type

calculation and start on the quantum mechanical solution of the above model.

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(1) Roger Parsons and J.O'M. Bockris, Trans. Faraday Soc. 47, 914 (1951).

(2) A. Damjanovic and V. Brusic, Electrochim. Acta, 12, 615 (1967).

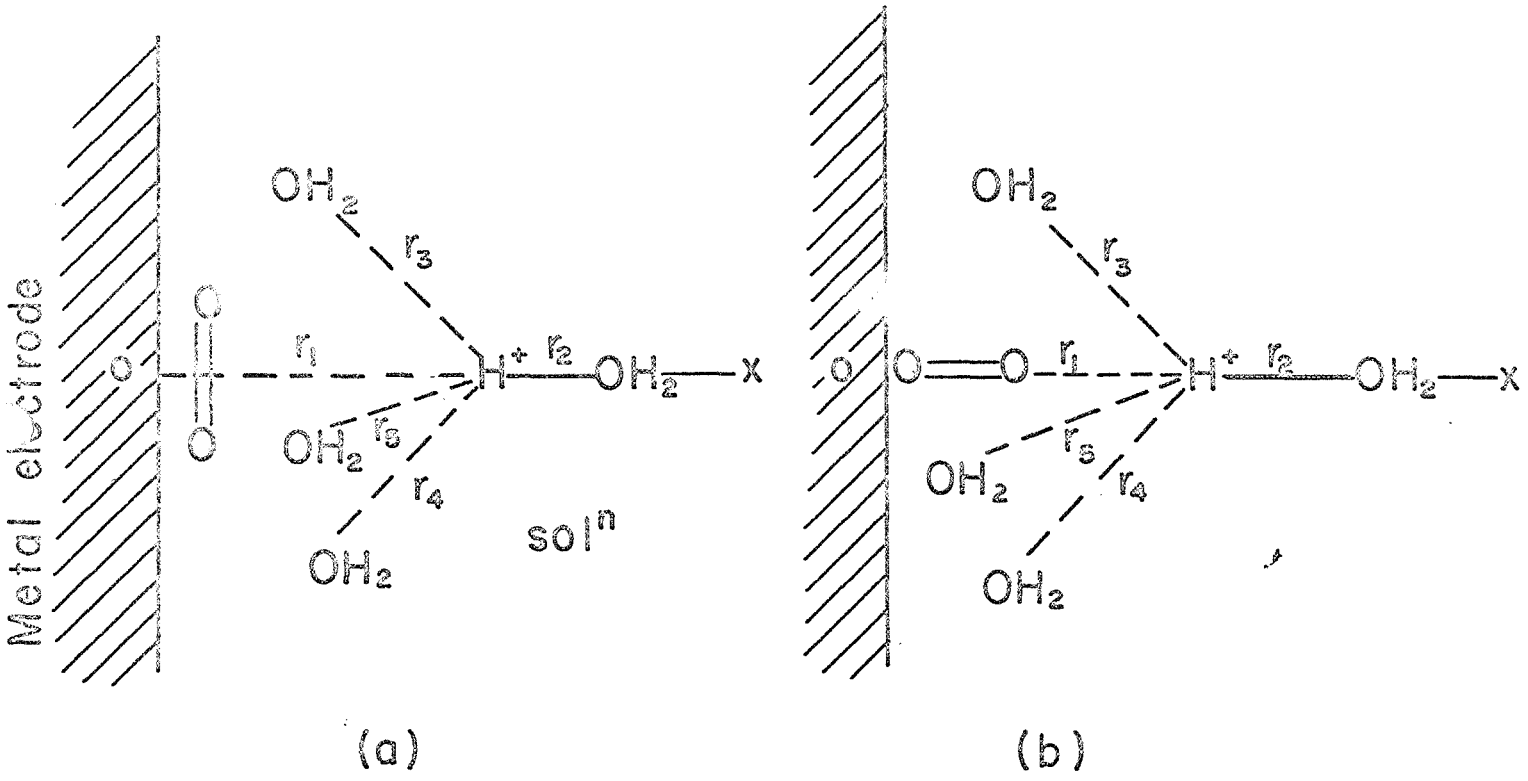

Fig. 1 The initial state model

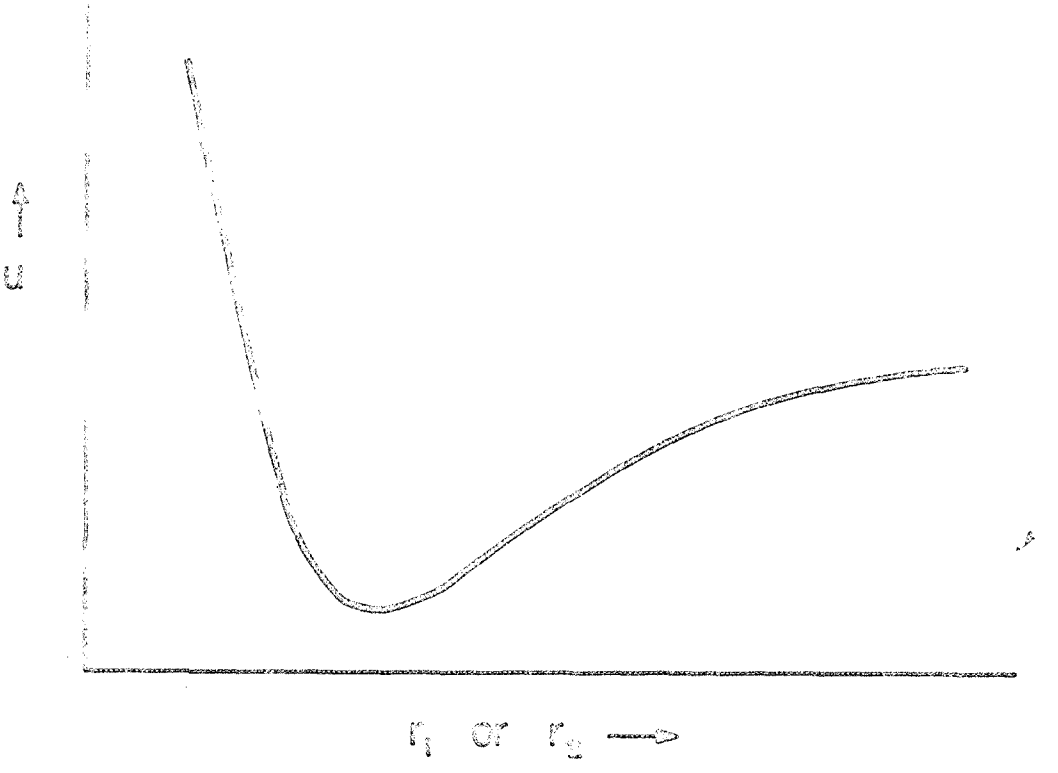

Fig.2 Variation of the energy of the initial state as a function of r_1 or r_2

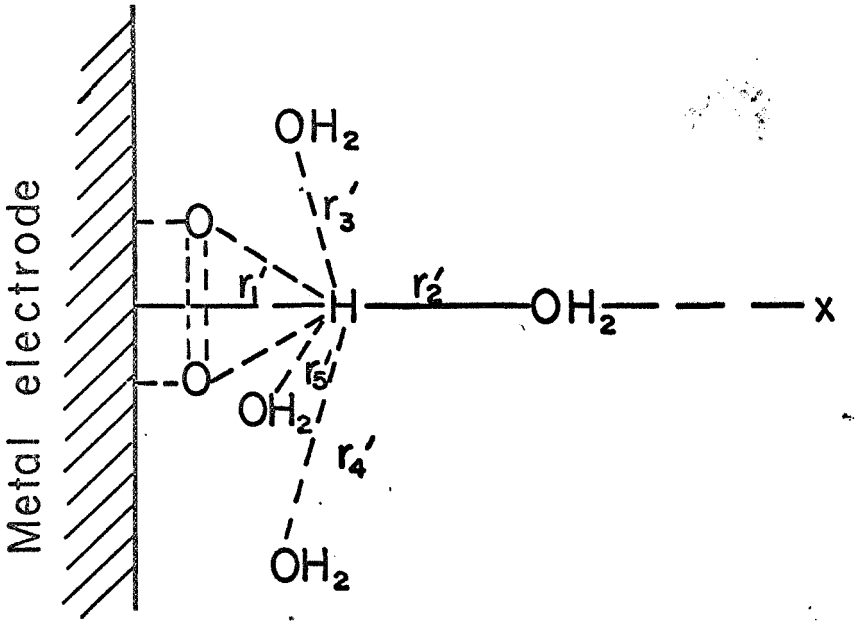

Fig.3 Model for final state

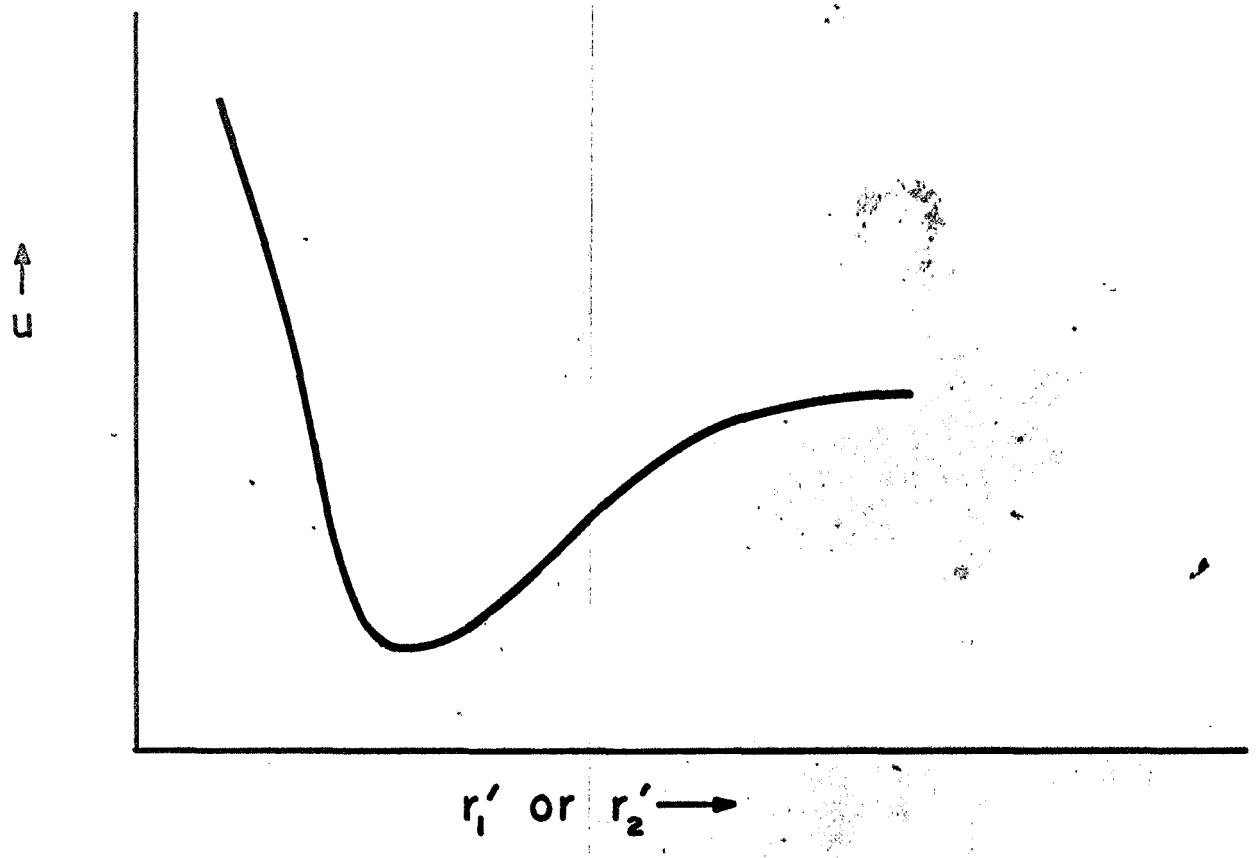

Fig.4 Variation of energy for final state as a function of r_1' or r_2'

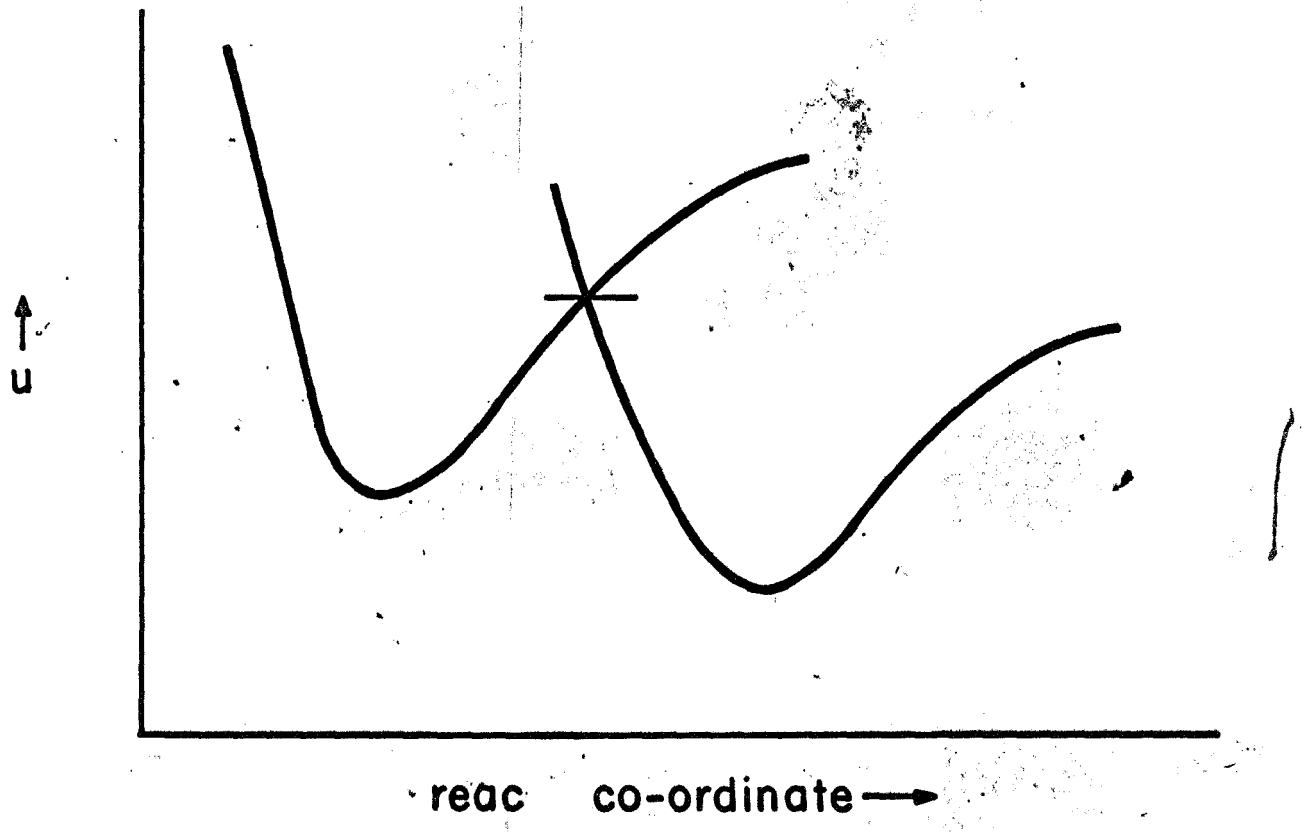

Fig. 5 Potential energy diagram for the complete system.

SECTION II

Title of the Project: Zinc Electrode Study for Silver-Zinc Batteries

Long-term Aims: Prevention of dendrite growth and of decrease of active surface on the Zn electrode.

Specific Aims for This Period: While crystallization process is also involved, we first deal with organic adsorption in order to understand dendrite growth inhibition.

Previous studies[1] invite the investigation of adsorption of large organic cations (tetraalkylammonium..., hemulphogenes, Triton...) and to select those which would adsorb on the cathodic side, Zn deposition, i.e., charging potential region, and desorb on the anodic side, Zn dissolution, i.e., discharge potential region of a battery. Dendrite growth is expected to be inhibited if we can decrease the exchange current density by adsorption of organic species in the charge potential region, while battery efficiency needs the exchange current not to be decreased in the discharge region of potential.

Most of the usual methods of adsorption studies are forbidden for our purpose:

electrocapillary, varied sweep or pulse methods, hydrogen coverage measurement--because we want to operate on a solid metal which is far from "noble," while all these methods need to avoid large currents as those expected in the potential range to be investigated.

The only convenient method appears (cf. previous reports) to be radiotracer measurements, although the need of some unusual labeled compounds makes them expensive and their high concentrations (10^{-3} - 10^{-2} M) decrease its accuracy.

Results of the Work:

We set up the apparatus needed to make the radiotracer measurements. We chose to use the same principles as those of a previous work[2] of this laboratory.

The apparatus consists basically of a metal tape which is passed through an electrolyte bath containing the tagged organic substance. The tape remains in the bath long enough for adsorption equilibrium to be attained at the electrode. The tape is then drawn out of the bath in such a way that no back diffusion occurs and so that one can know the amount of solution adhering to the tape. The total amount of radioactive material on the tape is then determined and this, when

corrected for the amount contained in the adhering solution, gives the amount adsorbed on the metal surface. The essential feature of this method is that the quantity of active material adsorbed is of the same order or larger than the amount held up in the adhering solution. The whole apparatus is mounted in an aluminum box with a plexiglass front panel to allow the work to be carried out in a controlled atmosphere.

The electrode is a 5 N Zinc foil (Cominco Electronic material, Spokane, Wash.) 0.5" wide and 0.002" thick, point-welded as an endless belt.

The cell is a two-body cell, the first one used to clean the tape electrode and the second to have the adsorption. Teflon slits allow the path of the tape and make separation between the cells and the atmosphere. The electrode potential is controlled by a Wenking potentiostat and a 350 digital multimeter of Data Technology Corporation.

The amount of solution adhering on the tape is known by measuring the capacitance between the tape and a plate, using either a Delta Decker Unit or an Autobalance universal bridge, B641, of Wayne Kerr.

The radiation counting is made with two

gas flow proportional counters (ultra-thin window) and a scaler-timer Baird-Atomic Model 135.

Specific Aims for Next Report Period:

We will have to calibrate the whole apparatus: counting and adherent solution film thickness, using a carbon-14 Na_2CO_3 standard solution of 0.2 mCi/g (Amersham/Searle).

We plan to do a preliminary measurement with hexadecultrimethylammonium bromide, long linear chain compound with small solubility which is expected to be strongly adsorbed. This would allow us to have ideas on the experimental behavior of the method while avoiding use of high concentration of the radioactive compounds. This one came from Amersham-Searle with name Trimethyl(cetyl-1-C14) ammonium bromide 6.0 mCi/mH (16.5 mCi/mg) of specific activity.

Then we will use the tetraethyl-1-C^{14} ammonium bromide 3.2 mCi/mH of specific activity (New England Corporation). But previous studies[1] may be seen to show that its adsorption does not occur at concentrations less than 10^{-2} M/C; if this is true, it would be very difficult to measure because of the too high concentration of the compound.

Then other compounds of larger molecular weight will be studied depending on their

interest and on their availability from radiocompound suppliers.

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2. Swinkels, thesis, University of Pennsylvania (1963). Green, Swinkels, and Bockris, Rev. Sci. Inst., 33, 18 (1962).

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Jet Propulsion Laboratory
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MS 198-223
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Clothing & Organic Materials Div.
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