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THE GALVANIC CORROSION OF ALUMINUM-STAINLESS STEEL AND ALUMINUM-GRAPHITE COUPLES AS A FUNCTION OF WATER COMPOSITION AND TEMPERATURE

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SERIES A

AEC RESEARCH AND DEVELOPMENT REPORT

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THE GALVANIC CORROSION OF ALUMINUM-STAINLESS STEEL AND ALUMINUM-GRAPHITE COUPLES AS A FUNCTION OF WATER COMPOSITION AND TEMPERATURE

WRITTEN BY

R. L. LOFTNESS
J. B. CHRISNEY

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REPORT APPROVED BY:

H. PEARLMAN, Group Leader, Chemistry

S. SIEGEL, Associate Director

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ABSTRACT

Experiments on galvanic corrosion of aluminum when coupled to stainless steel or graphite are described. Observations of static corrosion, at 50° C and 90° C, were made over a period of 60 days for the cases of distilled, demineralized, and simulated Hanford process water, both with and without 0.005M H_2O_2.

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The rate of aluminum pick-up by the water ranged from 0.0016 to 0.07 mg/cm^2/day at 50° C and from 0.007 to 0.11 mg/cm^2/day at 90° C, and the corrosion rate increased with time. Pit depths varied from 0 to 5 mils after 2 months in distilled or demineralized water and from 20 to 40 mils in simulated Hanford water.

Graphs of the galvanic current are presented and pictures of the corroded samples are included.

This report is based upon studies conducted for the Atomic Energy Commission under Contract AT-11-1-GEN-8.

I. INTRODUCTION

In the design of certain reactors, circumstances arise where aluminum is in contact with stainless steel or graphite in the presence of vapor or liquid water. The magnitude of the corrosion caused by the galvanic current passing through the aluminum-graphite and aluminum-stainless steel couples has been the cause for some concern about the operating life of reactor parts which are made of aluminum.

Numerous investigations have been reported of the corrosion of aluminum by water, under a wide variety of experimental conditions.[1] A previous report[1] summarized the results available through mid-1950. Although some of the studies were directed specifically at galvanic effects (Table XIII of Ref. 1), the range of conditions covered did not include those of interest here. A subsequent report[2] contains the information that no significant pitting occurs in 95 days for aluminum-stainless steel couples. The conditions of test were: H_2O temperature, 90° C; specific water resistance of 0.5×10^6 to 4×10^6 ohm-cm; pH of 6.5 to 7.8; and H_2O velocity, 25 ft/sec. A very slight weight gain is also reported.

The experiments described in this report were performed to determine the magnitude of the galvanic current and the nature of the corrosion at 50° C and at 90° C as a function of time, temperature and water composition. The water compositions and temperatures were chosen to simulate possible reactor operating conditions.

II. EXPERIMENTAL PROCEDURE

The couples to be tested were in the form of thin flat plates, spaced apart and electrically insulated by a Plexiglas block. A copper lead wire was attached to each plate of the couple. The assembly was suspended in a large pyrex test tube by means of the lead wires, which passed through the rubber stopper inserted in the test tube. The leads terminated in jacks on a panel, which facilitated periodic measurement of the galvanic current. An assembled cell is sketched in Fig. 1, and details of the couples are shown in Fig. 2. A total of 48 of such cells was utilized in the experiments, a "set" of 24 at each temperature.

The 2S aluminum, type 347 stainless steel and AGOT graphite plates were cut from sheet stock, edges smoothed on emery paper; the aluminum and stainless steel surfaces was polished with a felt pad, by use of a suspension of alumina in water.

A. Construction of Galvanic Cells

Two cells were made up with each couple (Al-SS and Al-C), immersed in each of the three types of water. A duplicate number of cells was assembled using water containing added H_2O_2. (The H_2O_2 concentration was initially adjusted to 0.005M, and no subsequent additions of H_2O_2 were made.) The resulting set of 24 cells was placed in a bath of "white" mineral oil, which was circulated by a small pump. Bath temperatures were maintained within ±0.5° C of nominal by thermostatically controlled immersion heaters.

B. Experimental Arrangement

The test tube was filled with water of the desired composition to a level such that 10 centimeters of the electrodes were immersed. The pH of the water was maintained at 4.5 to 5.5 by passing CO_2 through each test tube. The CO_2 was pre-saturated with water vapor to prevent evaporation of the water in the test tubes. Three different types of water were used: distilled (6.5 ppm solids), demineralized (8.3 ppm solids) and Hanford process (53 ppm solids). (See Table I for complete analyses.) The demineralized and Hanford process waters were made synthetically by adding salts to distilled water.

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C. Observations

During the run, galvanic currents were measured at intervals of approximately 5 days. The duration of the tests was 60 days.

The aluminum electrodes were weighed before and after the run (following light surface cleaning with an alumina suspension in water). Corrosion of the electrodes was observed visually, with the aid of a microscope. Post-run chemical analyses were made of the cell solutions.

III. EXPERIMENTAL RESULTS

The data on weight change of the electrodes (Tables II and III), chemical analyses of the solutions (Table IV), and integrated galvanic current (Figs. 3 through 5), give three separate measurements of the extent of corrosion. The results of these measurements are compared in Table V.

It is clear that distilled and demineralized water have essentially equal corrosion properties, while Hanford process water is considerably more corrosive, under the conditions of the present experiments. The initial 0.005M concentration of H_2O_2 is seen to have no effect on the corrosion rates except for the case of Hanford water at the lower temperature.

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In general, there is good agreement between the weight loss of the electrodes and post-run chemical analyses of the water (assuming the oxide film weight to be negligible compared to the weight loss). The weight loss of aluminum to be expected from the integrated galvanic current was low by a factor of four in some cases. The fluctuating nature of the galvanic current, as shown in Figs. 3 through 5, makes the time-integration of the current subject to sizable error. The plots of galvanic current vs time do indicate, nevertheless, that the corrosion rate increases with time.

The corrosion rates listed in Tables II and III were computed by assuming the corrosion to be uniform over the surface exposed and converting the weight change into mg/cm^2/day of aluminum lost from the surface. The rates range from 0.0016 to 0.07 mg/cm^2/day at 50° C, and from 0.007 to 0.11 mg/cm^2/day at 90° C, depending upon the water composition.

The severity of the corrosion is not measured, however, by the overall corrosion rate, but rather by the depth of the pits formed. Pit depths ranged from 0 to 5 mils in distilled and demineralized water and from 20 to 40 mils in Hanford process water (see Tables II and III and the photograph of various pitted surfaces in Fig. 6). The pitting was most severe at edges, corners and interfaces, especially where electrodes were clamped to the lucite spacer (see Fig. 6). Pitting was more severe on the side facing the other electrode.

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The pitting attack was more pronounced at 90° C than at 50° C and in Hanford process water rather than in distilled or demineralized water.

The appearance of the couples at the end of the run is indicated in Figs. 7 through 12. The graphite electrodes were coated with a light gray solid (hydrated aluminum oxide), but appeared otherwise unchanged. There was no discernible corrosion of any of the stainless steel used for the tests, only a slight discoloration.

IV. DISCUSSION

Since the electrode potential difference is greater for the case of aluminum-graphite (as oxygen electrode) than for aluminum-stainless steel, the rate of corrosion should be higher in the Al-C couple than in the Al-SS couple. This hypothesis is borne out by the experimental evidence (see Table V). The corrosion rates of the aluminum were 3 to 10 times higher both at 50° C and 90° C in the Al-C couple than in the Al-SS couple.

Galvanic currents in the Al-C couple of 1 to 5 $\mu a/cm^2$ at 50 and 90° C, increasing with time, do not agree with Sullivan's data[3] which report galvanic currents of 35 to 5 $\mu a/cm^2$ decreasing with time. Sullivan's experiments, however, employed a constant slow flow of process water through the cell; whereas, in the present experiments, the water was entirely static.

The relatively heavy pitting attack observed in the present experiments contrasts with the absence of pitting reported in Ref. 2. The explanation probably lies primarily in the very high purity of the water used in the latter work (resistivity, near 10^6 ohm-cm; pH, 6.5 to 7.8). Resistivity was not measured in the present experiments, but it must certainly have been one to several powers of 10 lower. The pH, also, was maintained between 4.5 and 5.5. The influence of water purity may be seen from the results of the present experiments (see Tables III and IV), where the greatest corrosion rate is observed in Hanford water, which was the least pure of the three types of water used.

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V. CONCLUSIONS

A. Corrosion Rates

The overall static corrosion rates of 2S aluminum (in aluminum-stainless

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steel and aluminum-graphite couples in distilled, demineralized and Hanford process waters) were observed to range from 0. 0016 to 0. 07 mg/cm^2-day at 50° C, and 0. 007 to 0. 11 mg/cm^2-day at 90° C. The pH was maintained at 4. 5 to 5. 5 in all tests by bubbling water-saturated CO_2 through the test cells. The duration of the tests was 60 days.

The largest corrosion rates were observed in Hanford process water. The rates were smaller, and approximately equal, in distilled and in demineralized water. The presence of H_2O_2 (initial concentration 0. 005M; no subsequent additions made) had no significant effect on the corrosion rates, except in the case of Hanford water at 50° C, where the addition of H_2O_2 reduced the severity of pitting.

Corrosion of the aluminum was more severe in the Al-graphite couples than in the Al-stainless steel couples. There was negligible corrosion of the 347 stainless steel. Except for a non-adherent surface deposit, there was no effect on the graphite.

The principal mode of aluminum corrosion was pitting. Pit depths ranged from 0 to 5 mils in distilled and demineralized water, and from 20 to 40 mils in Hanford process water. Pitting attack was most severe at edges, corners and interfaces. It was more pronounced at 90° C than at 50° C.

The corrosion rate, as indicated by the galvanic currents, increased with time.

B. Design Recommendations

Galvanic corrosion may be reduced by the following measures, which are indicated from the present experiments and verified by work at other laboratories: use of maximum purity water; use of water velocities which are at least moderately high (10 to 30 ft/sec); and avoidance of geometrical configurations which permit cavitation or stagnation of the water.

TABLE I

WATER COMPOSITIONS

Distilled Water

pH	5.87
Total solids	6.5 ppm[(a)]

Demineralized Water[4]

Dissolved CO_2	2.1
Dissolved solids	8.3
Iron	0.02
Aluminum	< 0.01
Copper	< 0.02
Nickel	< 0.05
Chromium	< 0.05
Calcium	0.12
Magnesium	0.16
Sodium	1.30
Chloride	0.30
Carbonate	2.10
Sulfate	5.00
Bicarbonate	4.20

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Hanford Process Water[5]

Total iron	0.02
Methyl orange alkalinity or $CaCO_3$	52.0
Dissolved silica or SiO_2	5.0
$Na_2 Cr_2 O_7 \cdot 2H_2C$	1.9
Calcium	21.0
Magnesium	4.5
Chloride	1.7
Sulfate	18.0
Manganese	0.002
Sodium	2.0
Copper	0.02-0.005
Aluminum	< 0.02

(a) All concentrations are in ppm.

TABLE II

ALUMINUM WEIGHT CHANGE DATA AT 50° C

Aluminum[(a)] Sample	Original Weight, gm	Weight after Test, gm	Loss of Weight, gm	Metal Loss[(b)] mg/cm^2/day	Water Composition	Post-Run Surface Appearance
1-1	19.6476	19.6406	0.0070	0.001	distilled	1 or 2 pits,
1-2	19.6939	19.6856	0.0083	0.005		1 to 5 mils deep
1-3	19.6090	19.5973	0.0117	0.003	distilled +	1 or 2 pits,
1-4	19.5968	19.5830	0.0138	0.004	0.005 M H_2O_2	1 to 5 mils deep
1-5	19.0345	19.0323	0.0022	0.006	demineralized	no pitting
1-6	19.3027	19.2937	0.0090	0.003		
1-7	19.5417	19.5311	0.0106	0.003	demineralized +	1 or 2 pits,
1-8	19.5141	19.5007	0.0134	0.004	0.005 M H_2O_2	1 to 5 mils deep
1-9	19.6117	19.5342	0.0775	0.021	Hanford	scattered pitting
1-10	19.9540	19.8615	0.0925	0.025		10 to 20 mils deep
1-11	19.5005	19.4565	0.0440	0.012	Hanford +	light pitting
1-12	19.5700	19.5031	0.0669	0.019	0.005 M H_2O_2	1 to 5 mils deep
1-13	19.2886	19.2321	0.0565	0.016	distilled	scattered pitting
1-14	19.2545	19.1983	0.0562	0.016		5 to 10 mils deep
1-15	19.3100	19.2332	0.0768	0.021	distilled +	moderate pitting
1-16	19.5661	19.4130	0.1531	0.041	0.005 M H_2O_2	10 to 20 mils deep
1-17	19.9029	19.8650	0.0379	0.011	demineralized	scattered pitting
1-18	19.4170	19.3872	0.0298	0.008		5 to 10 mils deep
1-19	19.7380	19.6534	0.0846	0.023	demineralized +	scattered pitting
1-20	19.8408	19.7648	0.0760	0.021	0.005 M H_2O_2	5 to 10 mils deep
1-21	19.8385	19.5509	0.2876	0.080	Hanford	severe pitting
1-22	19.6294	19.4170	0.2124	0.059		20 to 40 mils deep
1-23	19.7976	19.6256	0.1720	0.048	Hanford +	moderate pitting
1-24	19.4822	19.2161	0.1661	0.046	0.005 M H_2O_2	5 to 20 mils deep

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(a) Samples 1-1 to 1-12 were aluminum-stainless steel couples
Samples 1-13 to 1-24 were aluminum-graphite couples

(b) Each rate given for the metal loss is based on the assumption that the weight of the final oxide film is negligible compared to the weight loss of the sample.

TABLE III

ALUMINUM WEIGHT CHANGE DATA AT 90° C

Aluminum[(a)] Sample	Original Weight (gm)	Weight After Test (gm)	Loss of Weight (gm)	Metal Loss[(b)] $mg/cm^2/day$	Water Composition	Post-run Surface Appearance
2-1	19.6432	19.3641	0.2791	0.078	distilled	scattered pitting
2-2	19.6082	19.5961	0.0121	0.003		1 to 5 mils deep
2-3	19.5663	19.5434	0.0229	0.006	distilled +	light pitting
2-4	19.2961	19.2346	0.0615	0.017	0.005 M H_2O_2	1 to 5 mils deep
2-5	19.6426	19.5604	0.0822	0.023	demineralized	scattered pitting
2-6	19.4719	19.4167	0.0552	0.016		1 to 5 mils deep
2-7	19.3617	19.3444	0.0173	0.005	demineralized +	light pitting
2-8	19.2619	19.2262	0.0357	0.010	0.005 M H_2O_2	1 to 5 mils deep
2-9	19.8030	19.4793	0.3237	0.089	Hanford	severe pitting
2-10	19.3376	18.9955	0.3421	0.093		20 to 40 mils deep
2-11	19.2801	18.9342	0.3459	0.096	Hanford +	scattered pitting
2-12	19.5781	19.2438	0.3343	0.091	0.005 M H_2O_2	20 to 40 mils deep
2-13	19.3016	19.1271	0.1745	0.048	distilled	scattered pitting
2-14	19.8715	19.6840	0.1875	0.052		1 to 5 mils deep
2-15	19.2168	19.0120	0.2048	0.057	distilled +	scattered pitting
2-16	19.3789	19.0461	0.3328	0.091	0.005 M H_2O_2	1 to 5 mils deep
2-17	19.7410	19.5518	0.1892	0.052	demineralized	scattered pitting
2-18	19.3847	19.1623	0.2224	0.062		1 to 5 mils deep
2-19	19.1244	18.9253	0.1991	0.055	demineralized +	scattered pitting
2-20	19.3636	19.1152	0.2484	0.068	0.005 M H_2O_2	1 to 20 mils deep
2-21	19.1367	18.7172	0.4195	0.114	Hanford	moderate pitting
2-22	19.7871	19.3452	0.4419	0.123		1 to 10 mils deep
2-23	19.9785	19.6100	0.3685	0.102	Hanford +	moderate pitting
2-24	19.7845	19.4636	0.3209	0.089	0.005 M H_2O_2	1 to 10 mils deep

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(a) Samples 2-1 to 2-12 were aluminum-stainless steel couples
Samples 2-13 to 2-24 were aluminum-graphite couples

(b) Each rate given for the metal loss is based on the assumption that the weight of the final oxide film is negligible compared to the weight loss of the sample.

TABLE IV

POST-RUN WATER ANALYSES

Cell	Aluminum (ppm)	Iron (ppm)	Nickel (ppm)	Chromium (ppm)	Cell	Aluminum (ppm)	Iron (ppm)	Nickel (ppm)	Chromium (ppm)	Initial Water Composition
Al -- SS at 50° C					Al -- SS at 90° C					
1-1	23.6	1.11	1.39	< 0.005	2-1	124	47.2	0.94	< 0.005	distilled
1-2	19.1	3.46	0.35		2-2	46.0	312	2.4		
1-3	31.8	0.86	0.58		2-3	50.8	1.81	0.56		distilled +0.005M H_2O_2
1-4	35.4	134	2.00		2-4	329	3.48	0.50		
1-5	8.0	1.15	0.74		2-5	64.6	1.31	1.00		demineralized
1-6	19.0	1.09	0.82		2-6	128	1.65	0.99		
1-7	23.8	0.86	0.98		2-7	58.2	2.52	0.68		demineralized + 0.005M H_2O_2
1-8	27.5	0.96	0.37		2-8	70.9	140	1.00		
1-9	167	1.49	0.56		2-9	274	31.7	0.89		Hanford
1-10	148	2.06	0.75		2-10	64.5	131	0.88		
1-11	90.4	49.0	2.80		2-11	219	43.4	0.87		Hanford + 0.005M H_2O_2
1-12	115	1.51	1.78		2-12	224	6.24	1.06		
Al -- C at 50° C					Al -- C at 90° C					
1-13	134				2-13	199				distilled
1-14	113				2-14	295				
1-15	143				2-15	250				distilled + 0.005M H_2O_2
1-16	211				2-16	399				
1-17	57.2				2-17	147				demineralized
1-18	111				2-18	182				
1-19	294				2-19	268				demineralized + 0.005M H_2O_2
1-20	182				2-20	320				
1-21	322				2-21	504				Hanford
1-22	345				2-22	438				
1-23	475				2-23	826				Hanford + 0.005M H_2O_2
1-24	305				2-24	712				

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TABLE V

COMPARISON OF CORROSION DATA

(a) Aluminum Sample	Weight Loss of Aluminum Electrode (grams)	Weight of Aluminum in Solution (grams)	Weight of Aluminum from Integrated Current (grams)	Initial Water Comp.	(a) Aluminum Sample	Weight Loss of Aluminum Electrode (grams)	Weight of Aluminum in Solution (grams)	Weight of Aluminum from Integrated Current (grams)
1-1	0.007	0.014	0.007	distilled	2-1	0.28	0.11	0.023
1-2	0.008	0.009			2-2	0.012	0.028	
1-3	0.012	0.016	0.006	distilled + 0.005M H_2O_2	2-3	0.023	0.032	0.017
1-4	0.014	0.018			2-4	0.062	0.194	
1-5	0.002	0.004	0.007	demineralized	2-5	0.082	0.043	0.022
1-6	0.009	0.009			2-6	0.055	0.079	
1-7	0.011	0.012	0.003	demineralized + 0.005M H_2O_2	2-7	0.017	0.034	0.009
1-8	0.013	0.014			2-8	0.036	0.038	
1-9	0.078	0.084	0.03	Hanford	2-9	0.32	0.18	0.058
1-10	0.093	0.074			2-10	0.34	0.036	
1-11	0.044	0.045	0.012	Hanford + 0.005M H_2O_2	2-11	0.35	0.17	0.086
1-12	0.067	0.058			2-12	0.33	0.16	
1-13	0.057	0.046	0.025	distilled	2-13	0.17	0.070	0.050
1-14	0.056	0.028			2-14	0.18	0.13	
1-15	0.077	0.064	0.039	distilled + 0.005M H_2O_2	2-15	0.20	0.14	0.077
1-16	0.15	0.061			2-16	0.33	0.20	
1-17	0.038	0.020	0.016	demineralized	2-17	0.19	0.087	0.059
1-18	0.030	0.027			2-18	0.22	0.11	
1-19	0.085	0.074	0.030	demineralized + 0.005M H_2O_2	2-19	0.20	0.13	0.047
1-20	0.076	0.055			2-20	0.25	0.17	
1-21	0.29	0.12	0.060	Hanford	2-21	0.42	0.17	0.093
1-22	0.21	0.12			2-22	0.44	0.22	
1-23	0.17	0.13	0.057	Hanford + 0.005M H_2O_2	2-23	0.36	0.48	0.066
1-24	0.17	0.13			2-24	0.32	0.24	

(a) Samples 1-1 to 1-12 and 2-1 to 2-12 were from Al-S.S couples.
Samples 1-13 to 1-24 and 2-13 to 2-24 were from Al-C couples.
Cells maintained at 50° C are prefixed by 1; those at 90° C, by 2.

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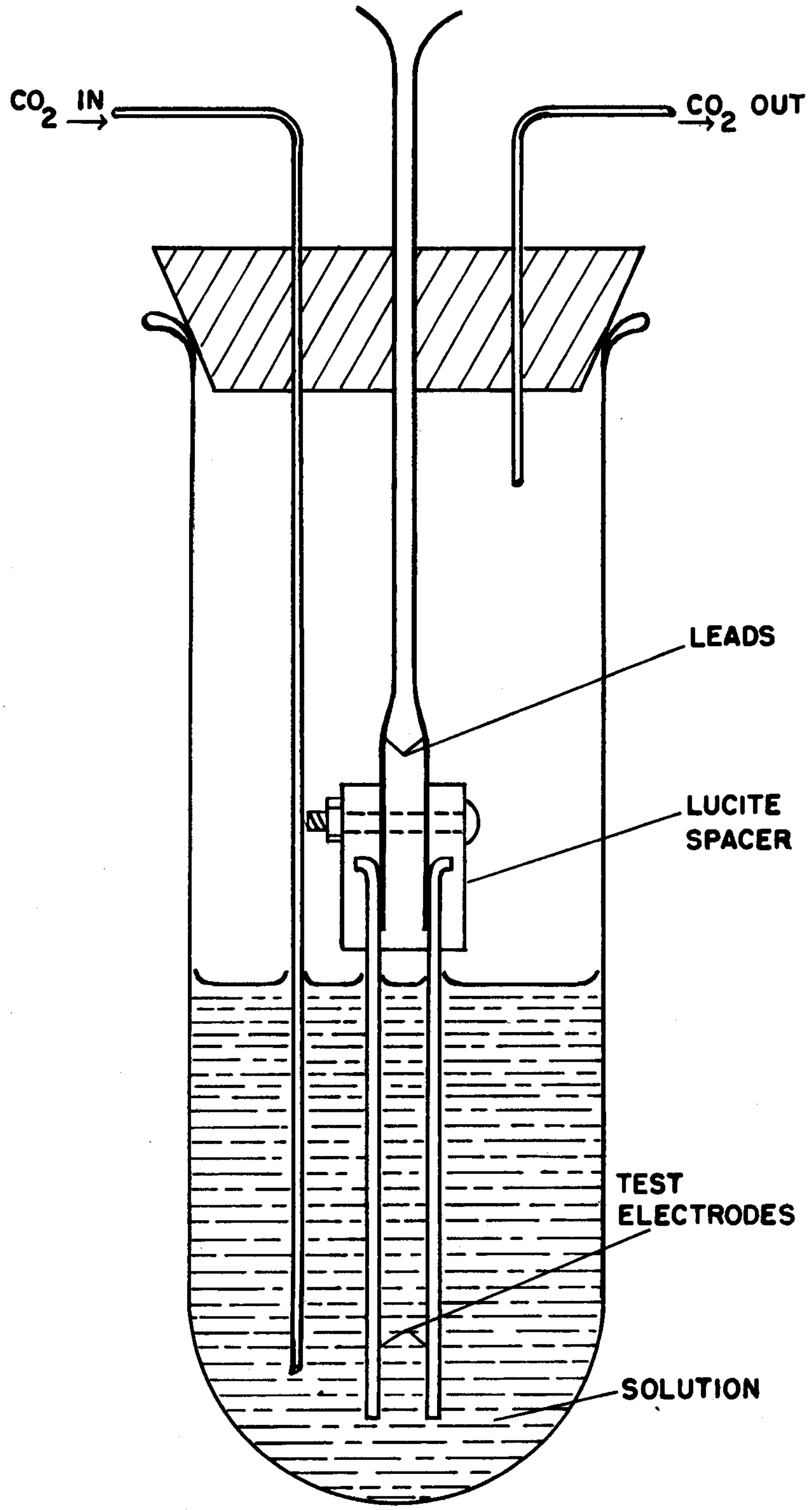

Figure 1. Assembled Galvanic Cell

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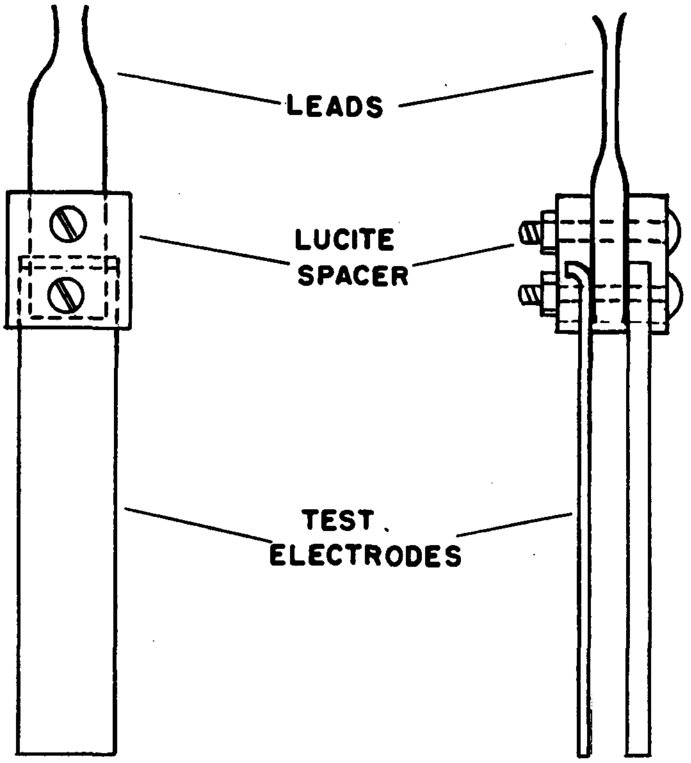
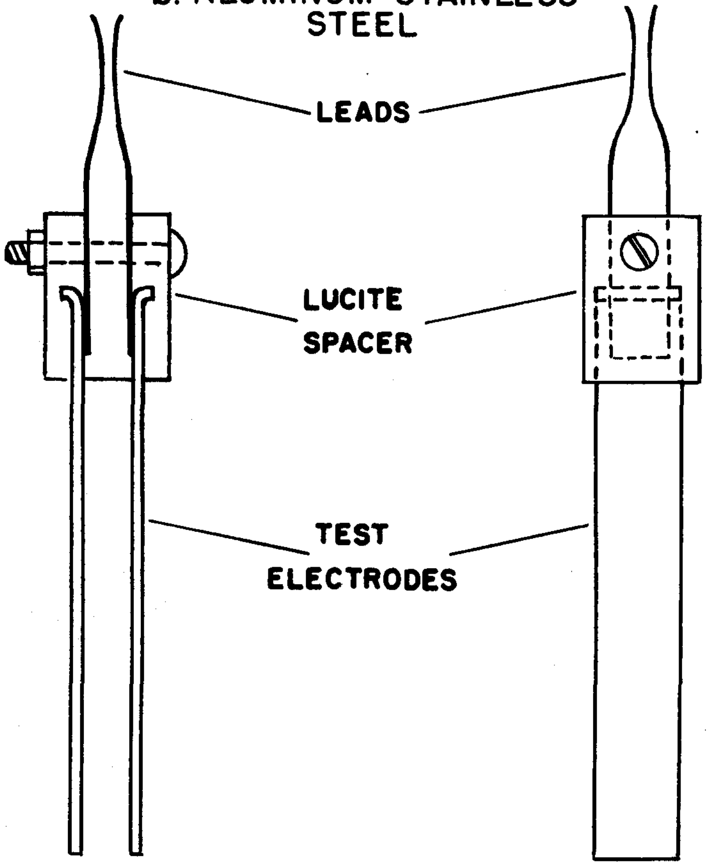


a. ALUMINUM-GRAPHITE



b. ALUMINUM-STAINLESS STEEL

Figure 2. Construction of Couples

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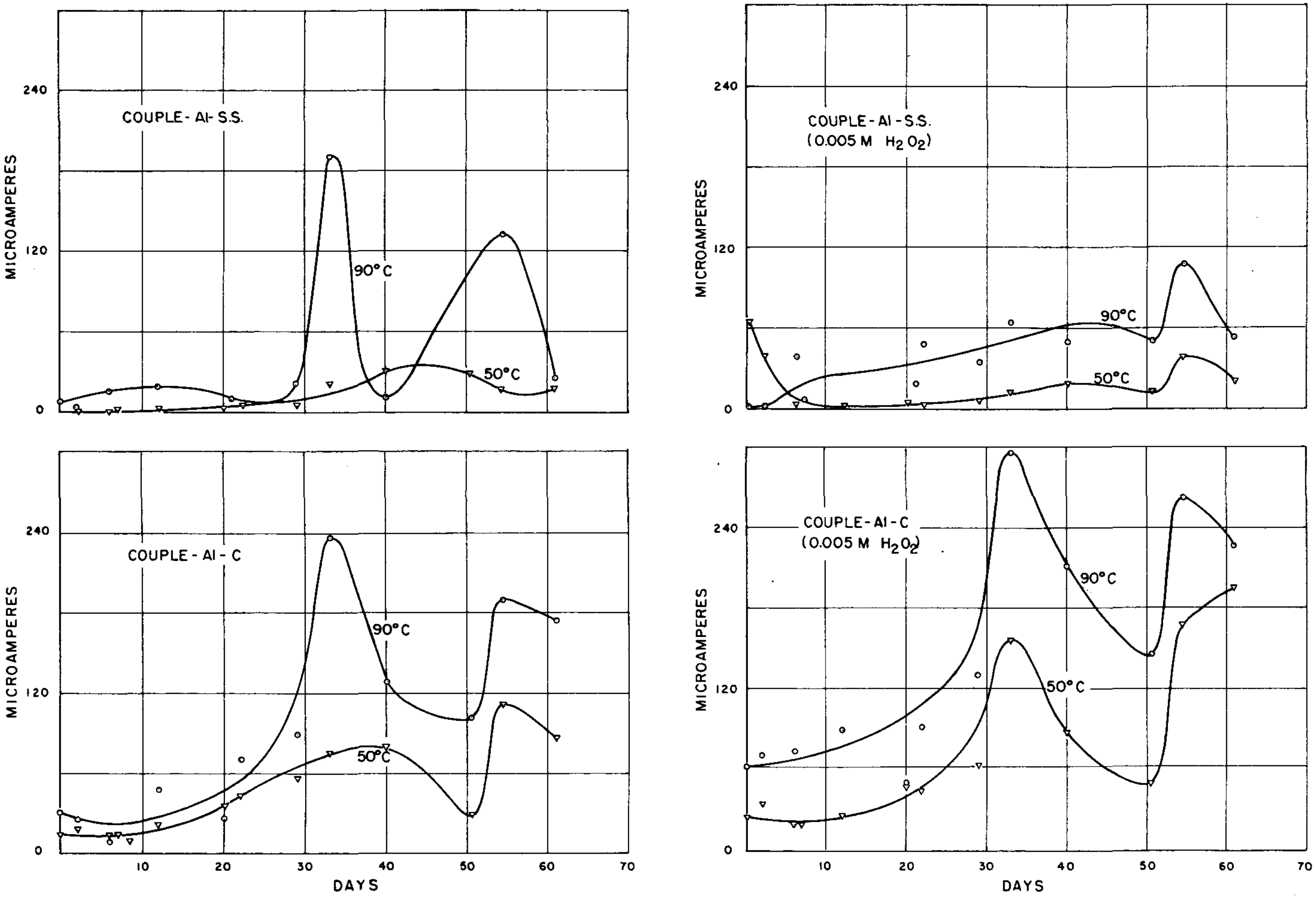

Figure 3. Galvanic Current vs Time (Distilled Water)

[redacted]

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[redacted]

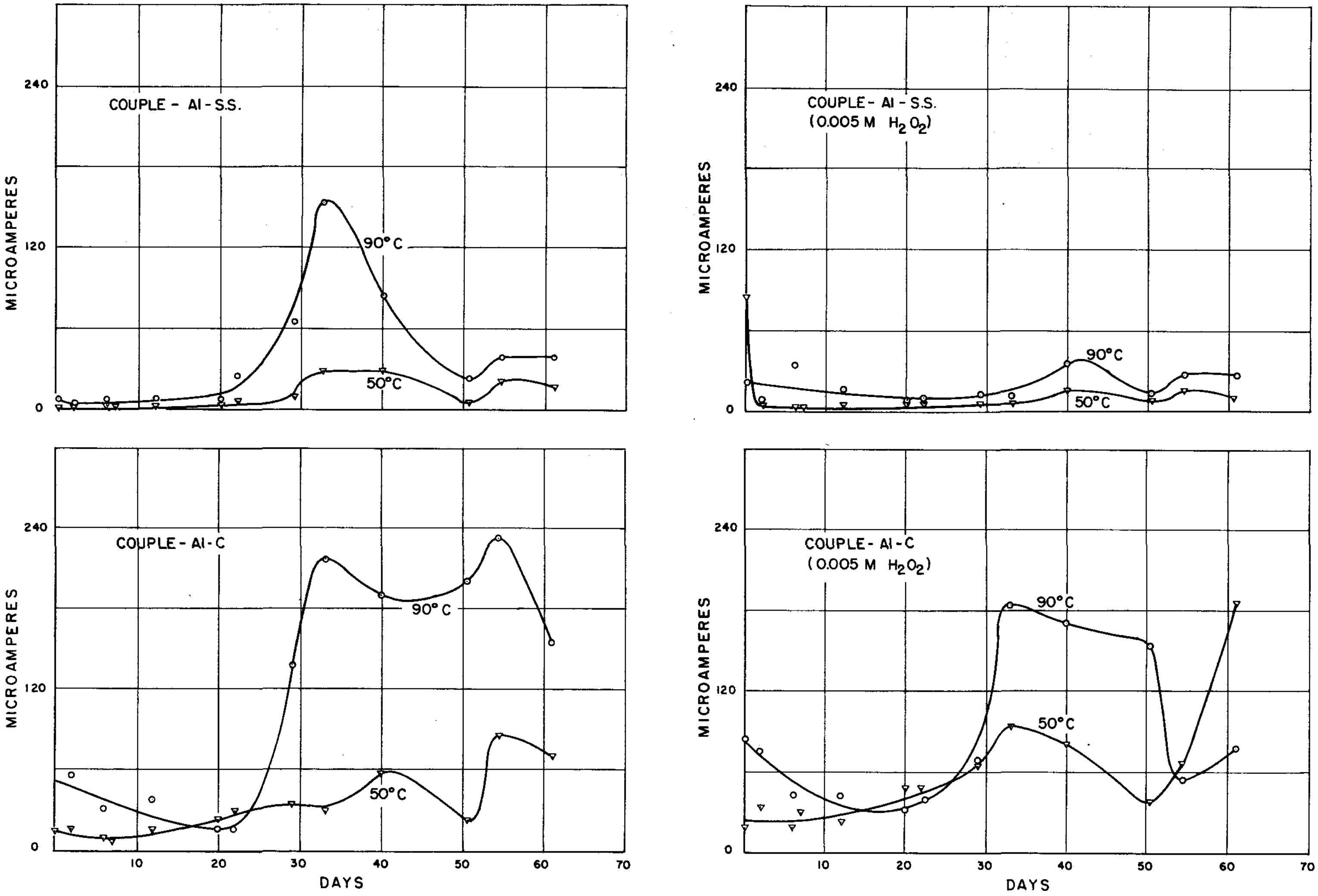

Figure 4. Galvanic Current vs Time (Demineralized Water)

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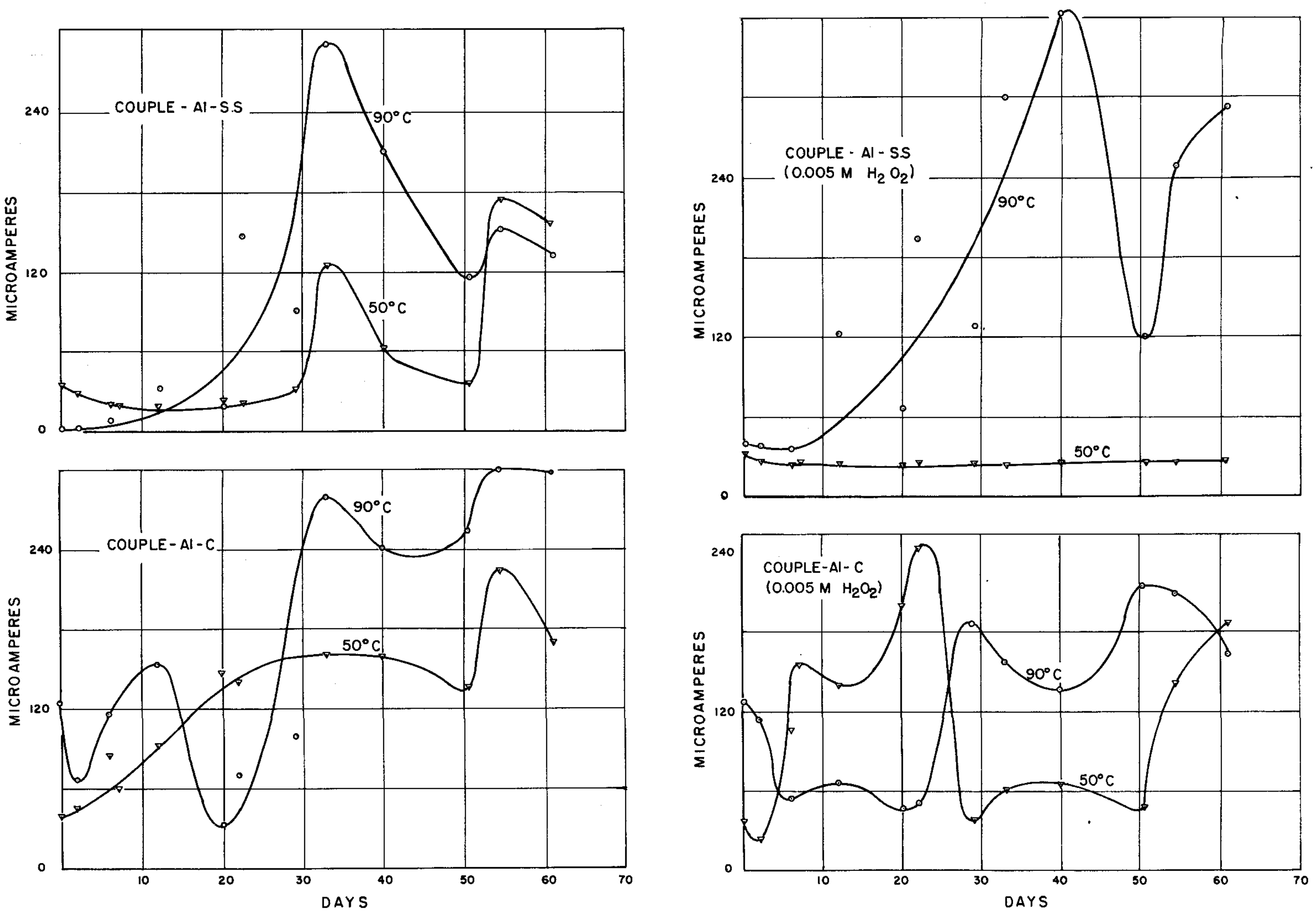

Figure 5. Galvanic Current vs Time (Hanford Water)

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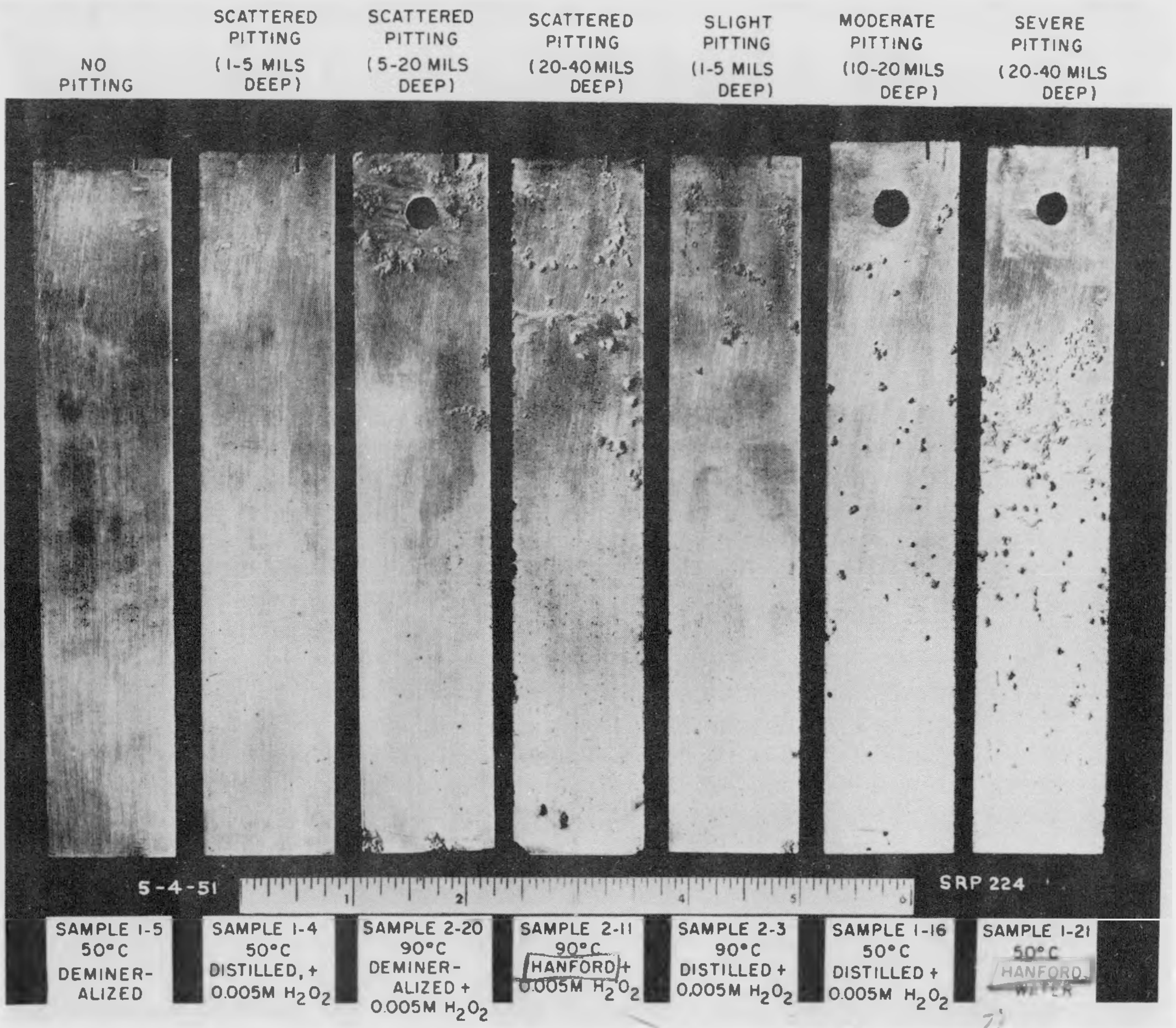

Figure 6. Types of Pitting Attack.

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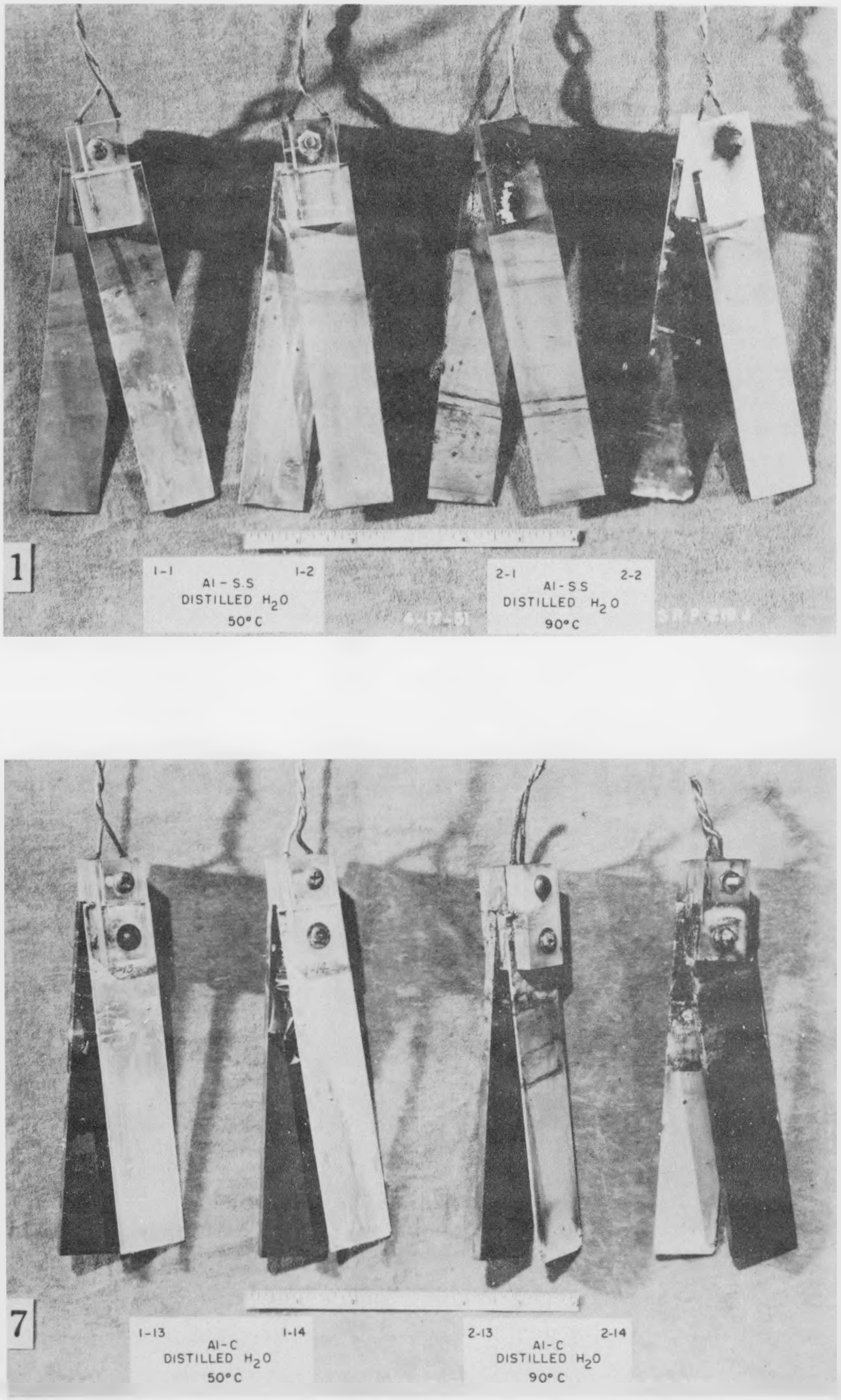

Figure 7. Electrode Assemblies After Exposure.

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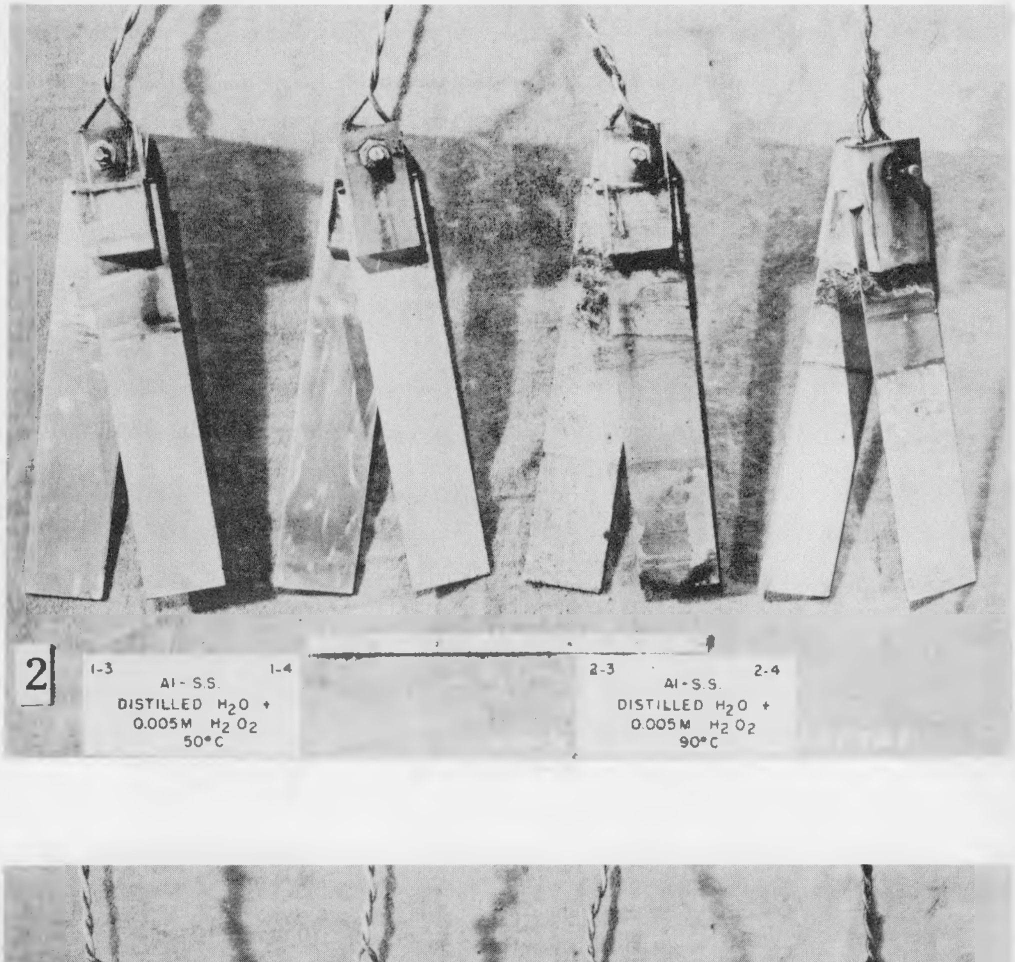

Figure 8. Electrode Assemblies After Exposure.

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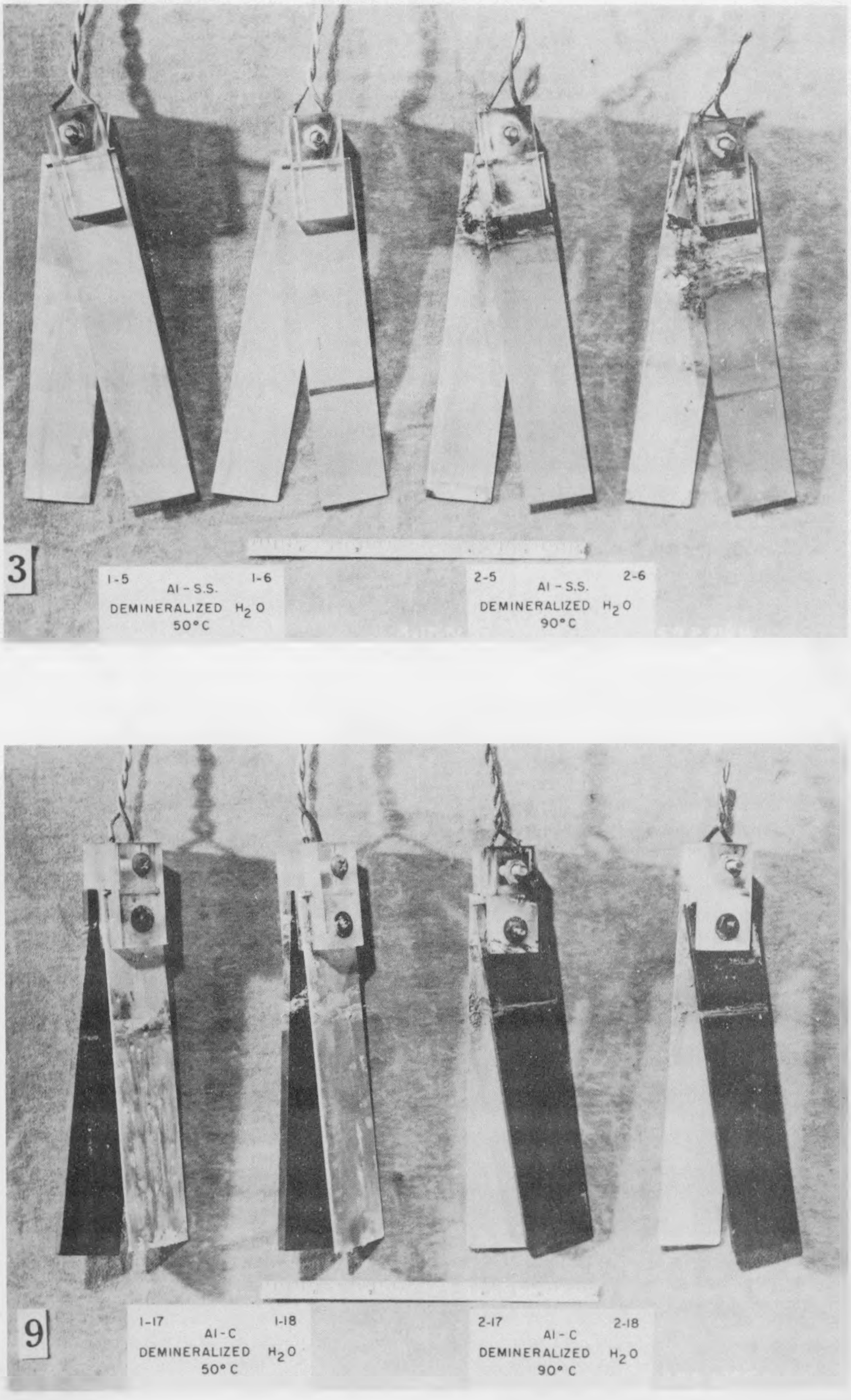

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Figure 9. Electrode Assemblies After Exposure.

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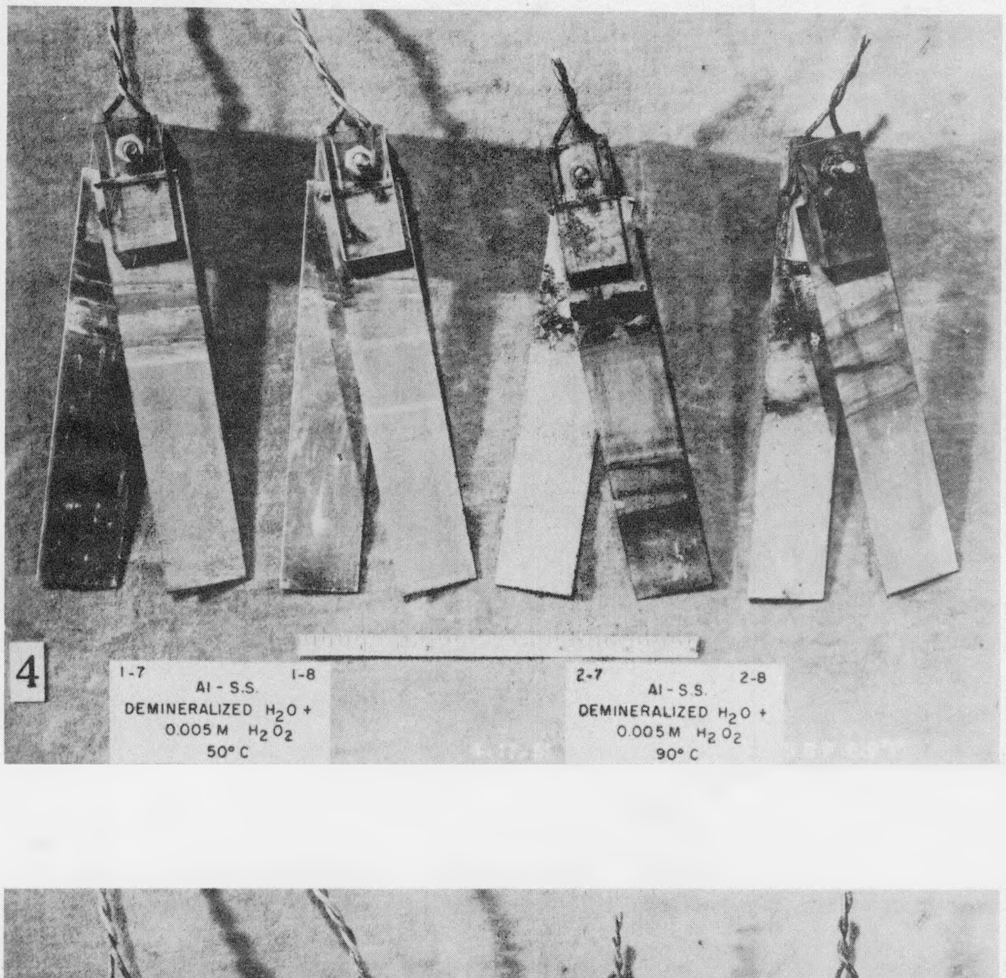

Figure 10. Electrode Assemblies After Exposure.

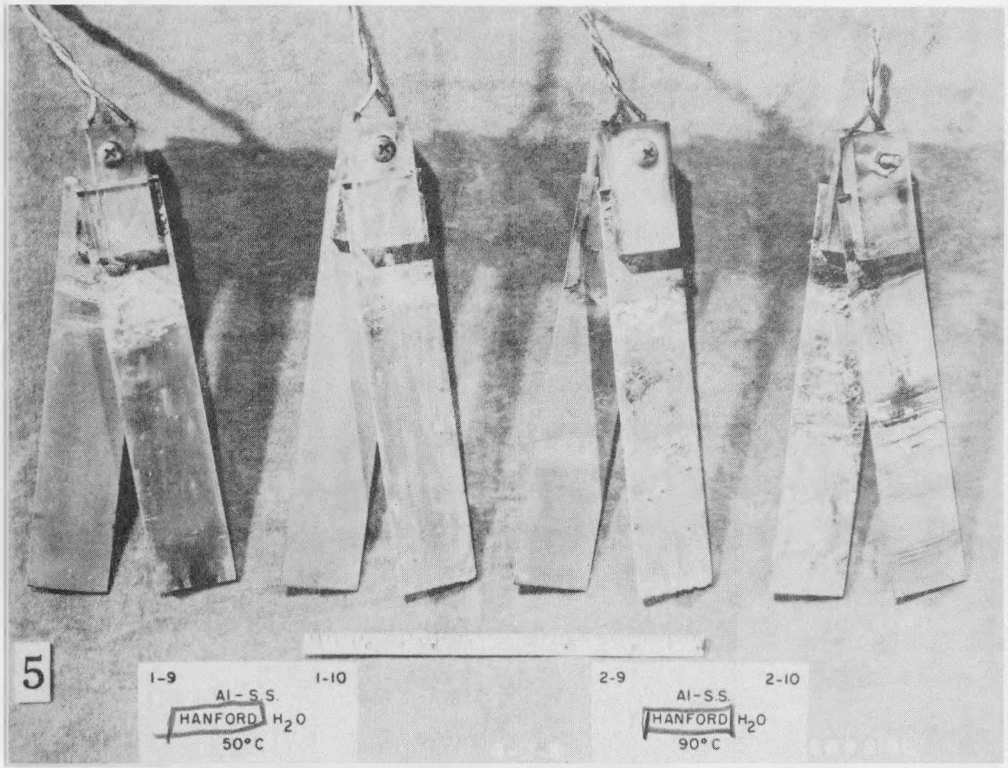

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Figure 11. Electrode Assemblies After Exposure.

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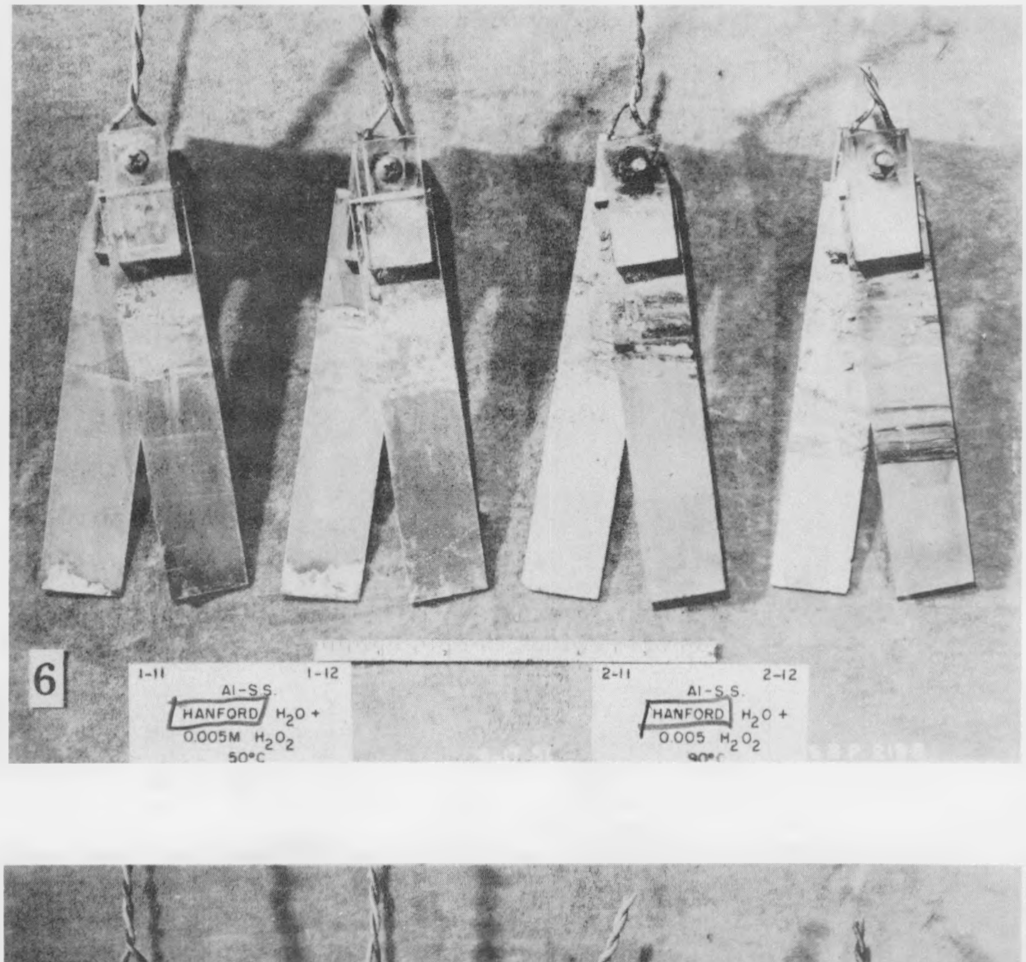

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Figure 12. Electrode Assemblies After Exposure.

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2. Huffman, J. R., Technical Progress Letter, ANL-4718, October, 1951.
3. Sullivan, J., Personal Communication, Pile Technology Division, HEW, August 11, 1950.
4. English, J. L., "The Corrosion of 356 Aluminum in Simulated Cooling Water for the Proposed MTR," ORNL-681, July 20, 1950.
5. Fromm, L., Personal Communication, Pile Technology, Hanford Engineering Works, August 11, 1950.

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