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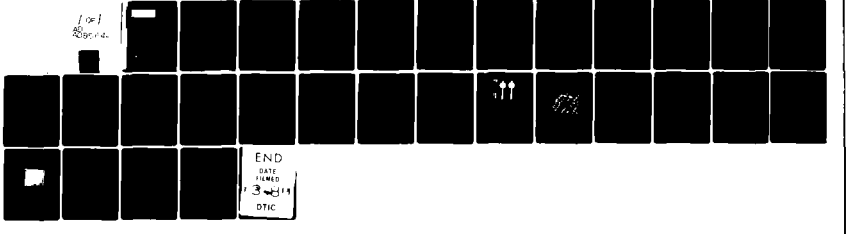

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CORROSION OF BUTTERFLY VALVES IN SEA WATER SERVICE

D. R. Lenard L. C. MacLeod

September 1980

Approved by J. R. Brown A/ Director / Technology Division

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ABSTRACT

Electrochemical techniques were used to compare the corrosion resistance in sea water of butterfly valve components made from an alloy which did not meet compositional requirements to components made from the specified Monel alloy 400. Polarization resistance studies indicated that the alloy which did not meet the specification had corrosion rates several times higher than Monel in both aerated and de-aerated sea water. Anodic polarization studies also showed that this alloy had a much higher probability for propagation of localized corrosion than Monel. Thus components made from this alloy can be expected to provide poorer service than those made of Monel.

RESUME

L'utilisation de techniques électrochimiques a permis de comparer, pour ce qui est de la résistance à la corrosion dans l'eau de mer, des éléments de robinets à papillon fabriqués en alliage non conforme aux exigences de composition et des éléments fait de l'alliage prescrit monel 400. Des études de résistance de polarisation ont montré que l'alliage non conforme aux spécifications présentait des taux de corrosion plusieurs fois supérieurs au monel lorsqu'il était mis en présence d'eau de mer aérée et deaérée. Des études de polarisation anodique ont également révélé, pour cet alliage, une probabilité de propagation de la corrosion localisée beaucoup plus élevée que pour le monel. Par conséquent, on peut s'attendre que les éléments fabriqués à partir de cet alliage donneront un rendement inférieur aux éléments de monel.

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NOTATION

E_r	corrosion potential
E_{pp}	primary passive potential
E_p	critical pit potential
I	polarization current
b_a	anodic Tafel slope
b_c	cathodic Tafel slope
R_p	polarization resistance
$\Delta\phi$	difference between applied potential and corrosion potential
i_{corr}	instantaneous corrosion rate
SCE	saturated calomel electrode

1. INTRODUCTION

Butterfly valves which were used in a sea water application as part of a ship service piping system on several HMC Ships have recently shown instances of failure due to severe localized corrosion at the interface between the disc and shaft areas (Figure 1). A chemical analysis of these valve components, which should have met the chemical requirements of the standard specification[1] for Monel alloy 400 (ASTM B164), indicated a composition which did not conform to this specification or to that of any other known commercial alloy. Table I compares the chemical compositions of the specified material and the valve alloy.

Nickel and its alloys, such as Monel, have excellent resistance to corrosion in aerated, flowing sea water. This resistance results from the formation of a passive film on the alloy surface which imparts to the alloy the electrochemical behaviour of an appreciably more noble metal (that is, one with a more positive corrosion potential and a lower corrosion rate). The nature of this passive film is not yet fully understood. It has been described either as a protective, self-repairing film of oxide or hydrated oxide[2] or as a chemisorbed film of oxygen[3]. In circumstances where there is insufficient oxygen available to maintain the film, such as stagnant sea water trapped in the crevice between the disc assembly and the Buna N rubber seat of the butterfly valves, pitting may develop.

A study was initiated in order to establish the relative corrosion susceptibilities of Monel and the valve alloy in a sea water environment. The response of the alloys to both aerated and de-aerated sea water was studied in order to simulate the operating conditions of the valve disc and shaft areas. The study was part of a larger investigation of localized corrosion damage experienced by Monel butterfly valves in various aqueous environments on HMC Ships.

2. BACKGROUND

Electrochemical methods have proven to be valuable in determining the susceptibility of alloys to crevice corrosion[4,5]. At the corrosion potential (E_r), the rates of anodic and cathodic processes on a specimen surface are equal.

However, if the potential of the specimen is forced away from its corrosion potential, the partial processes are no longer equal and an external current flows. A plot of the resulting current versus the applied potential produces a polarization curve.

If the potential of the specimen is forced away from the corrosion potential in the cathodic (negative) direction, the anodic areas on its surface are extinguished and the specimen behaves essentially as a cathode. In sea water the principal cathodic reactions are oxygen reduction and hydrogen evolution, with the former being predominant as long as sufficient oxygen is available. The polarization resistance technique, which will be discussed in more detail later, provides information concerning the cathodic reaction involved in a corrosion process.

If the potential of the specimen is forced away from the corrosion potential in the anodic (positive) direction, the sample behaves essentially as an anode, the primary anodic process being metal dissolution. The potentiodynamic anodic polarization technique, in which the potential is increased above the corrosion potential at a constant rate, provides a method for evaluating the effects of various environments on the rate of metal dissolution.

A typical anodic polarization curve for an alloy which can be repassivated is shown in Figure 2. Above the corrosion potential, the curve exhibits an increase in current with increasing potential until the passive film is formed. The potential at which the current ceases to increase is called the primary passive potential (E_{pp}). As the passive film develops, it protects the alloy surface from further corrosion. Thus the current decreases with increasing potential until the film covers the the entire surface. Once the film is completely formed there is no substantial change in current until the potential reaches a sufficiently noble value that the passive film begins to break down and localized corrosion is initiated. The potential at which localized corrosion is first observed is called the critical pit potential (E_p). The region above this potential is called the transpassive region and is characterized by an exponential increase in current with increasing potential. The proximity of the pit potential to the corrosion potential provides an indication of the susceptibility of the alloy to initiation of localized corrosion.

If the direction of the potential scan is reversed in the transpassive region, hysteresis is observed in the

back scan. This hysteresis occurs because the specimen surface has been irreversibly altered by the localized corrosion. The value of the current at which the potential is reversed is chosen to be sufficiently large to ensure appreciable hysteresis yet must also be low enough to avoid additional polarization effects that can occur with high current densities. Furthermore, it is very important that the same current value be used for each specimen in an experiment. As the reverse scan is continued, a potential will be attained at which all localized corrosion is extinguished and the current returns to a passive value. This potential is known as the repassivation potential (E_{rp}). The degree of hysteresis provides qualitative information concerning the probability that existing localized corrosion will propagate.

Another technique, the polarization resistance technique, provides numerical values for important kinetic parameters of corrosion reactions. In this technique, a specimen is polarized over a narrow potential range in the order of ±30 mv from the corrosion potential. Specimen surface changes which may result from high current densities are consequently eliminated.

This method, recently discussed in detail by Mansfeld[6], is based on the derivation of Equation (1) from mixed-potential theory:

$$I = \frac{1}{2.3} \cdot \frac{b_a b_c}{b_a + b_c} \cdot \frac{1}{R_p} \left(\exp\left(\frac{2.3\Delta\phi}{b_a}\right) - \exp\left(\frac{-2.3\Delta\phi}{b_c}\right)\right) \quad \ldots(1)$$

where:

I = polarization current

b_a, b_c = anodic and cathodic Tafel slopes

R_p = reciprocal of the slope of the polarization curve at the corrosion potential (polarization resistance)

$\Delta\phi$ = difference between the applied potential and the corrosion potential

A manual or computer fit of polarization data to equations derived from Equation (1) yields the polarization resistance (R_p) and the anodic and cathodic Tafel slopes. The Tafel slopes b_a and b_c are dependent on the mechanisms of the anodic and cathodic reactions, respectively. The instantaneous corrosion rate (i_{corr}) can then be determined from Equation 2:

$$i_{corr} = \frac{b_a b_c}{2.303\ (b_a + b_c)} \cdot \frac{1}{R_p} \qquad \ldots(2)$$

3. EXPERIMENTAL PROCEDURE

3.1 Materials

Cylindrical specimens were sectioned from shafts of the appropriate valves to a length of $\frac{1}{2}$ inch and machined to a diameter of $\frac{1}{4}$ inch. One end was drilled and tapped for 3-48 N.C. threads. The specimen surface was gound through a series of silicon carbide abrasive papers from 120 to 400 grit. Prior to immersion in the electrolyte, the surface was given a final 600 grit polish to remove any oxide films, degreased in isopropanol and rinsed with distilled water.

The electrolyte used was natural sea water at room temperature. Aerated or de-aerated conditions were ensured by passing either compressed air or nitrogen through the electrolyte continuously, commencing one hour before immersing the specimen.

The corroded butterfly valve components examined by the Dockyard Laboratory exhibited extensive porosity in the castings (Figure 3). The cavities resulting from this porosity would tend to enhance crevice corrosion susceptibility above that for a properly cast component of the same alloy. However, all specimens, whether made of Monel or of the unusual alloy, were machined from valve components obtained from HMC Ships. Thus this study provided a direct comparison of the electrochemical properties of the components found in service.

3.2 Apparatus

A schematic diagram of the circuit used for the polarization experiments is shown in Figure 4. The system

utilized the following apparatus:

Wenking potentiostat (Model 70HC3)
Wenking scanning potentiometer (Model SMP 69)
Standard polarization cell[7] and Luggin probe
salt bridge
Fisher calomel reference electrode
Hewlett-Packard - XY recorder (Model 7004B)
Programmable calculator (Model 9810A)
Plotter (Model 9862A)
Decade resistance box

3.3 Calibration

The Wenking potentiostat had a feature which permitted the recording of a wide range of current values, for full anodic scans, without the inconvenience of changing the recorder range switch. A pair of diodes in parallel with the meter resistance would conduct a current after a certain potential drop across the meter was exceeded. This resulted in a logarithmic output signal above this potential value as part of the total current was shunted across the diodes. Thus it was necessary to determine the relationship between the current flowing in the cell and the corresponding output voltage to the X-Y recorder in order to calibrate the pen response.

The calibration was performed with the circuit shown in Figure 5. The potentiostat sensitivity switch was set to the 0 - 100 μamp range. A constant current flow through the external circuit was regulated by adjustments of the counter electrode potentiometer control and/or the decade resistance box during which the corresponding output potential was measured.

The calibration curve had three distinct regions - linear, parabolic (transitional) and logarithmic. Curve fitting routines obtained the appropriate coefficients which defined the three regions. A program was written for the Hewlett-Packard calculator-plotter combination which would output the calibration curve of "Output Potential vs Current" from an input of the coefficients defining the three regions. The calibration curve generated by this program for the Wenking 70HC3 potentiostat is shown in Figure 6.

3.4 Procedure

Before the specimen was mounted and immersed in the electrolyte, the exposed specimen area was determined. A 24

hour period was allowed for the specimen to reach its steady-state corrosion potential in the electrolyte before polarization was performed. The tip of the Luggin probe salt bridge was placed 1 - 2 mm from the surface of the specimen. It was found convenient to obtain the polarization resistance data (within ±30 mv of the corrosion potential) before proceeding with the destructive full anodic scan. External potentials were applied by the scanning potentiometer at a constant rate of 5 mv/min in 5 mv increments for both methods. In the full scan, the direction of the potential steps was reversed when the current reached a value of 100 microamps/cm^2.

Initially a condition of zero current flow was created by matching the counter electrode potential to the specimen corrosion potential. The specimen was then polarized by scanning to 30 mv away from the corrosion potential in the cathodic direction. The specimen was then allowed to return to its original corrosion potential before being polarized to 30 mv away from E_r in the anodic direction. Several of these experiments were performed over a 24 hour period for each specimen before full anodic polarization scans were made.

The polarization curves were analyzed by the method of linear least squares using a computer program initially described by Mansfeld[8]. The output from this program provided the polarization resistance, anodic and cathodic Tafel slopes, the instantaneous corrosion current and several statistical parameters.

4. RESULTS

4.1 Polarization Resistance Measurements

A typical polarization curve obtained within ±30 mv of the corrosion potential is shown in Figure 7. The kinetic parameters obtained from the polarization resistance experiments in each of the metal-electrolyte systems are given in Table II. The reported values are average results obtained over a 24 hour period following the attainment of steady-state conditions. The least squares analysis using a DECsystem-20 computer involved no less than 25 experimental current-potential pairs for each curve and fits to the data were obtained with standard deviations ranging from 2 percent to 7 percent.

If the only electrochemical reactions occurring in a system are metal dissolution and the corresponding reduction of a single oxidizing species, then the Tafel slopes depend

on the rate determining step in the reaction mechanism. In sea water, oxygen reduction and hydrogen evolution are the most common cathodic reactions. The infinite cathodic Tafel slopes observed in aerated sea water for both alloys indicate that the dominant cathodic reaction is oxygen reduction occurring under diffusion control[9]. In de-aerated sea water, the cathodic Tafel slope and corrosion potential determined for Monel alloy 400 are consistent with hydrogen evolution as the cathodic reaction[10]. The corrosion potential of the valve alloy in de-aerated sea water was too noble for hydrogen evolution and no oxygen was present so the cathodic reaction could not be determined.

Even without knowledge of the reaction mechanism, the Tafel slopes can be used to determine the instantaneous corrosion rate. In both aerated and de-aerated sea water, the average corrosion rate of the valve alloy was found to be several times that of Monel alloy 400 in the same environment (Table II). Thus the valve alloy was found to be inferior to Monel by the polarization resistance method.

4.2 Anodic Polarization Curves

The anodic polarization curves used to determine crevice corrosion susceptibilities for Monel and the valve alloy in de-aerated sea water are shown in Figures 8 and 9. The critical potentials extracted from these curves are summarized in Table III.

The Monel specimen displayed typical behaviour for an alloy with an active-passive transition (Figure 8). From the active corrosion potential of -333 millivolts versus the saturated calomel electrode (SCE) the dissolution rate increased with increasing applied potential. Above the primary passive potential of -250 millivolts (SCE) the dissolution rate decreased and then remained independent of potential for 100 millivolts. Above -60 millivolts (the pitting potential), the current increased dramatically, accompanied by the initial appearance of pits on the specimen surface. Finally, the repassivation potential, below which no localized corrosion can occur, was found to be -130 millivolts (SCE).

The valve alloy had an initial corrosion potential (-103 millivolts vs. SCE) that was 230 millivolts more noble than that of Monel. Furthermore, it did not display the S-shaped anodic scan typical of an active-passive metal

(Figure 9). There was, however, an inflection in the anodic scan at -45 millivolts (SCE) which corresponded to the appearance of pits on the specimen surface. The repassivation potential of -240 millivolts (SCE) was much lower than that of Monel.

The corrosion potential of Monel in aerated sea water (-75 millivolts vs. SCE) was within the passive region shown in Figure 8 but was only 15 millivolts below the pitting potential. Thus Monel should have a marked tendency for the initiation of localized corrosion under conditions where the passive film cannot be maintained. This tendency is well known and has been demonstrated in this laboratory (Figure 10). However, the repassivation potential is well above the active corrosion potential (203 millivolts more noble) and only 70 millivolts below the passive corrosion potential, indicating that Monel can be readily repassivated.

The corrosion potential of the valve alloy in aerated sea water was only 10 millivolts above that in de-aerated water and 49 millivolts below the pitting potential. Thus this alloy should show a marked tendency for localized corrosion which is apparently not dependent on the presence of oxygen in the water. Furthermore, the repassivation potential is so far below the rest potential that repassivation is very unlikely. Thus there is a very high probability for propagation of any localized corrosion which occurs.

5. CONCLUSIONS

Valve components made from the alloy which did not meet specifications were found to have a corrosion rate several times that of Monel components. Furthermore, they were found to have a much higher probability for propagation of crevice corrosion than their Monel counterparts. This study has demonstrated that the nickel-copper alloy containing excessive concentrations of chromium and molybdenum is not suitable for use in a sea water environment. Any components made from this alloy can be expected to exhibit severe localized corrosion.

ACKNOWLEDGEMENT

The authors wish to express their thanks to Ms. B. Trenholm and Mr. P. Gergely of the Applied Mathematics Section at Defence Research Establishment Atlantic for development of the computer program used in this investigation.

TABLE I

CHEMICAL COMPOSITION OF BUTTERFLY VALVES

Element	Composition, percent	
	A.S.T.M. B164	Valve
Nickel	63.0 - 70.0	69.4
Copper	Remainder	19.6
Iron	2.5 (max)	1.6
Manganese	2.0 (max)	-
Carbon	0.3 (max)	-
Silicon	0.5 (max)	1.1
Sulphur	0.024 (max)	-
Chromium	-	3.5
Molybdenum	-	4.8

TABLE II

CORROSION PARAMETERS FOR MONEL AND VALVE ALLOY IN AERATED AND DE-AERATED SEA WATER

Alloy	Sea Water Medium	E_r (mv vs. SCE)	R_p (KΩ)	b_a (mv)	b_c (mv)	i_{corr} (μa/cm²)
Monel	Aerated	- 74	97.8	81	∞	0.10
	De-Aerated	-337	35.9	53	179	0.11
Valve Alloy	Aerated	- 94	21.3	104	∞	0.54
	De-Aerated	-104	26.3	161	∞	0.69

Legend

E_r corrosion potential

R_p polarization resistance

b_a anodic Tafel slope

b_c cathodic Tafel slope

i_{corr} instantaneous corrosion rate

TABLE III

CRITICAL POTENTIALS FOR MONEL AND VALVE ALLOY IN DE-AERATED SEA WATER

Alloy	Sea Water Medium	Corrosion Potential E_r (mv vs. SCE)	Pitting Potential E_p (mv vs. SCE)	Repassivation Potential E_{rp} (mv vs. SCE)
Monel	De-Aerated	-333	-60	-130
Valve Alloy	De-Aerated	-103	-45	-240

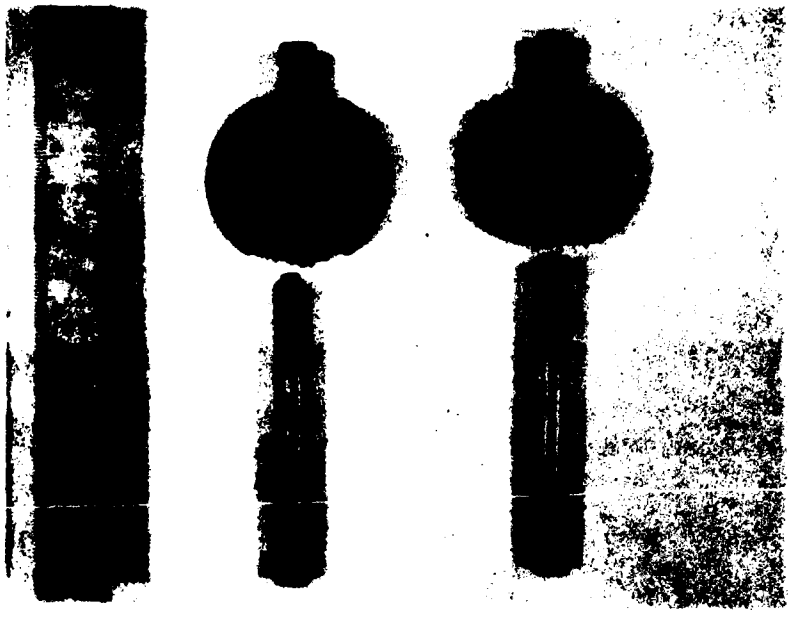

Figure 1: Corroded butterfly valve components in sea water service.

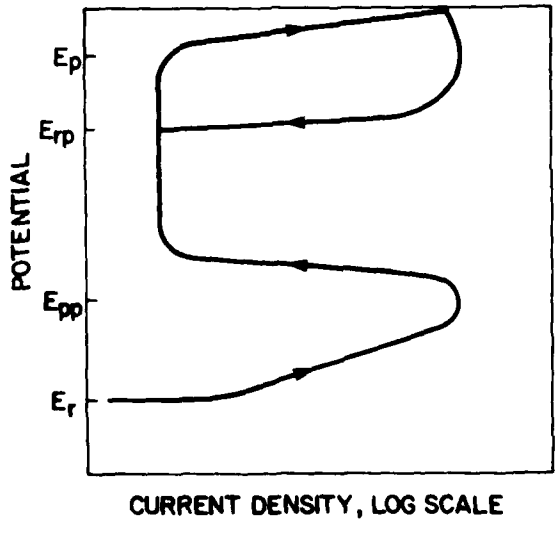

Figure 2: Typical anodic polarization curve for an active-passive alloy.

Figure 3: Porosity (dark areas) in a valve shaft made of the unusual composition. (250X), unetched.

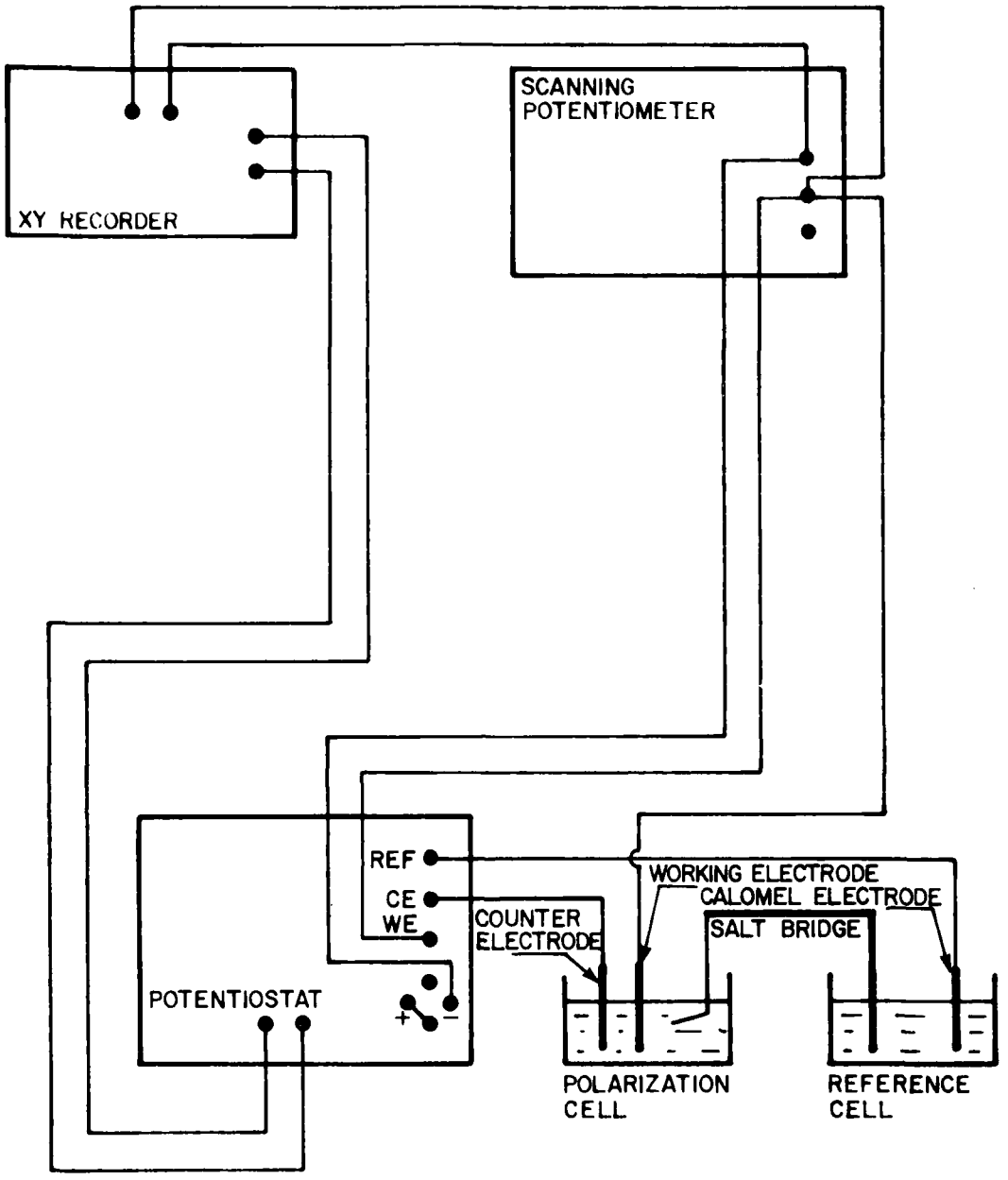

Figure 4: Schematic diagram of polarization circuit.

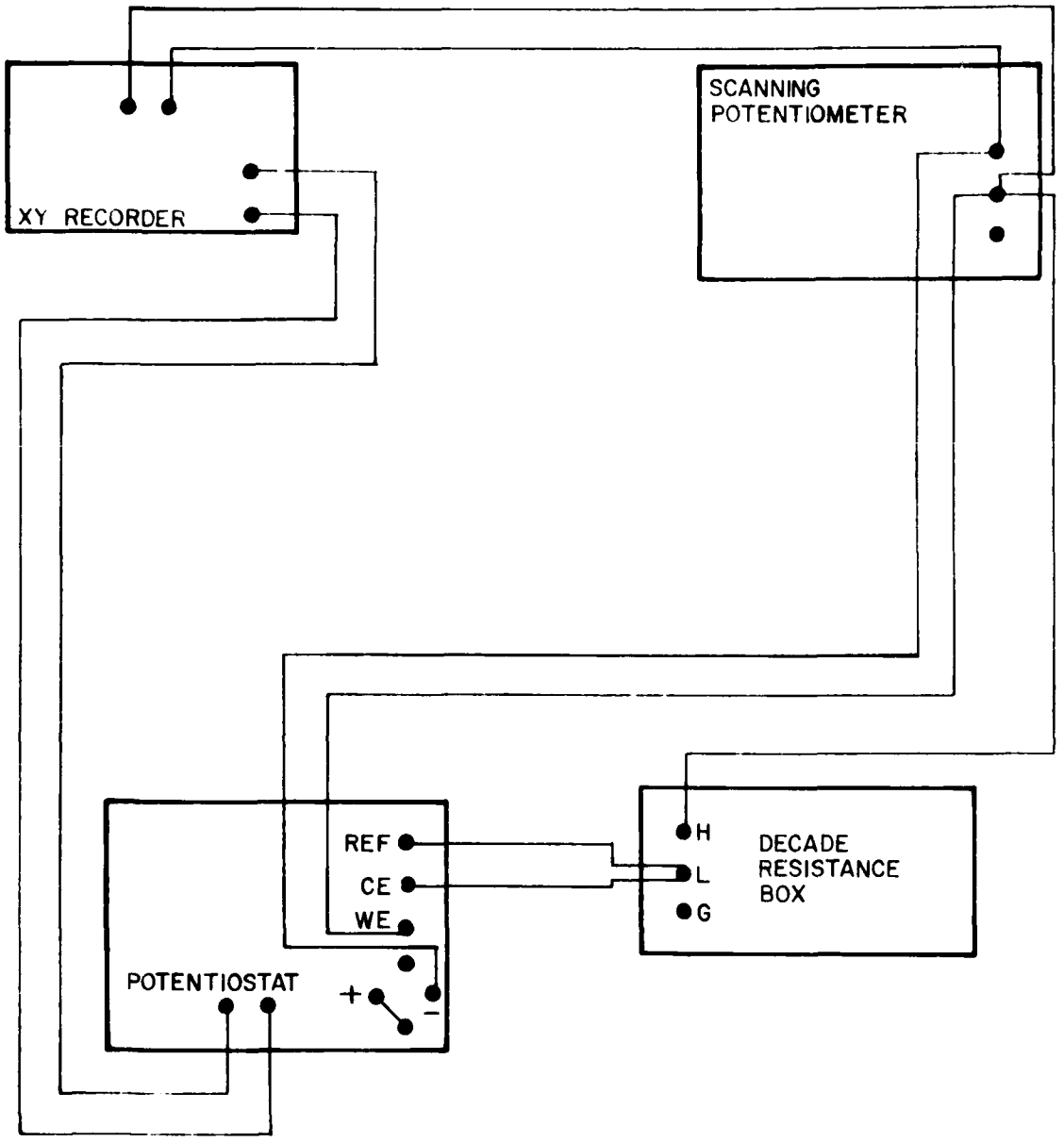

Figure 5: Schematic diagram of calibration circuit.

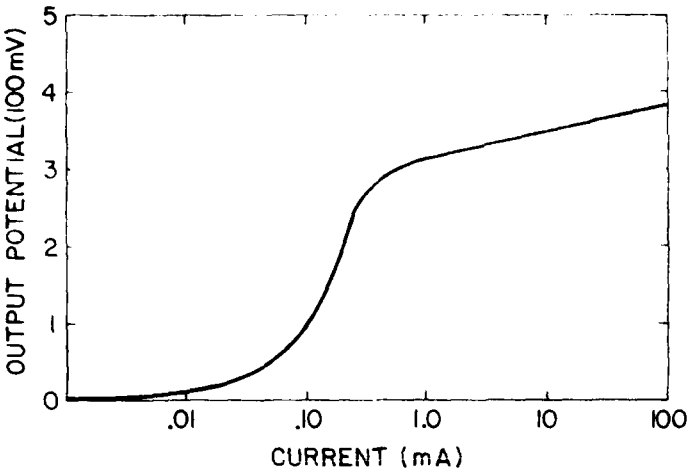

Figure 6: Calibration curve for Wenking 70HC3 potentiostat.

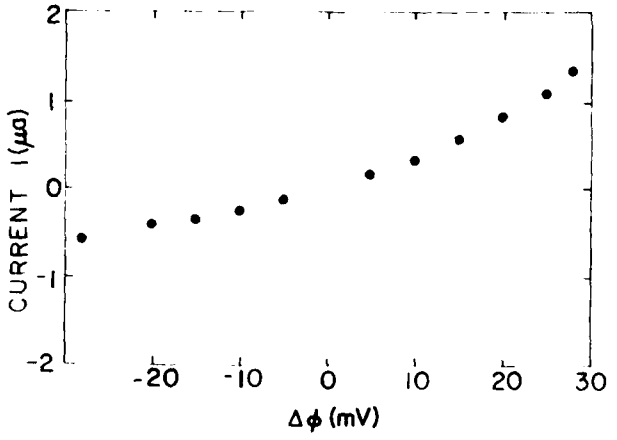

Figure 7: Experimental polarization resistance curve for Monel in de-aerated sea water.

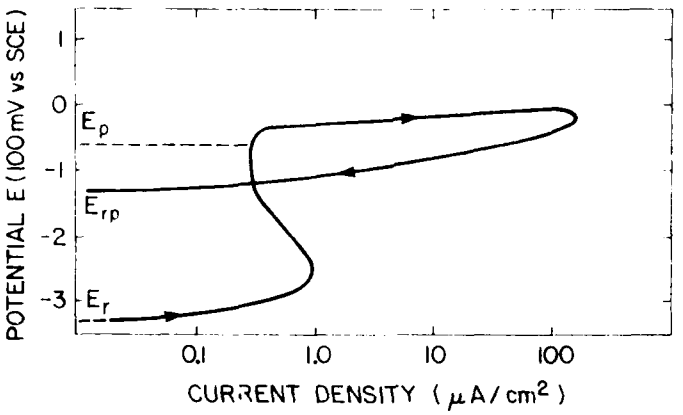

Figure 8: Anodic polarization curve of Monel in de-aerated sea water.

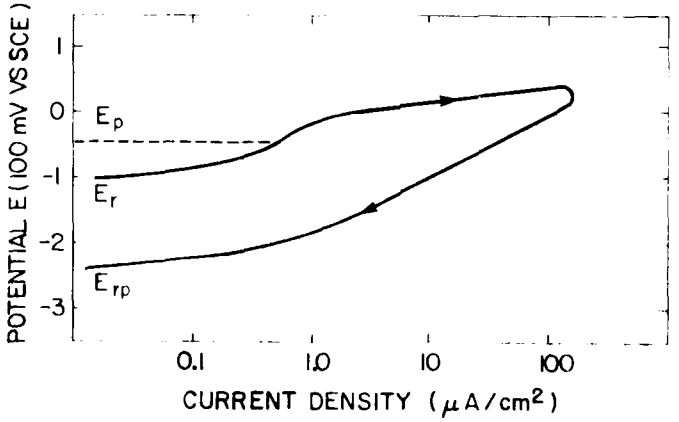

Figure 9: Anodic polarization curve of valve alloy in de-aerated sea water.

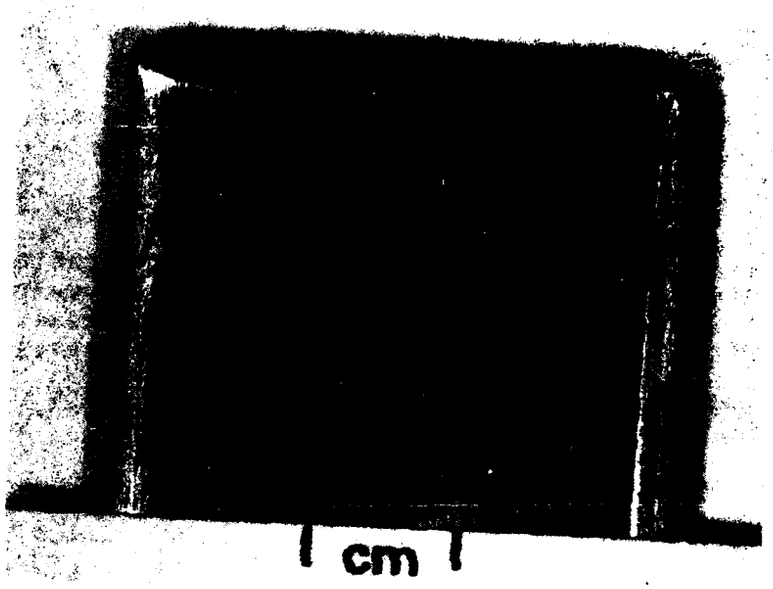

Figure 10: Crevice corrosion occuring under nylon retaining screw on Monel alloy 400 immersed in sea water for 40 days.

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13 ABSTRACT

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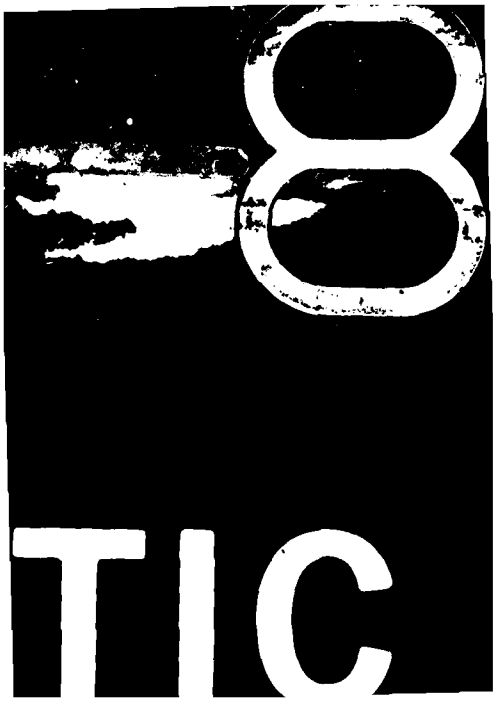
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