

TECHNICAL REPORT

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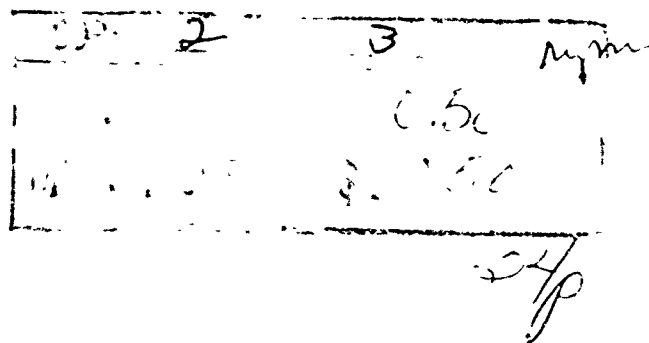
**ACTION OF SODIUM CITRATE AND SODIUM BORATE ON
THE POLARIZATION CHARACTERISTICS OF A
URANIUM-STEEL COUPLE**

by

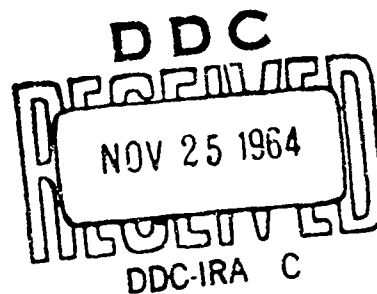
MILTON LEVY

and

PHILIP WONG



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OCTOBER 1964

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ACTION OF SODIUM CITRATE AND SODIUM BORATE ON THE POLARIZATION
CHARACTERISTICS OF A URANIUM-STEEL COUPLE

Technical Report AMRA TR 64-33

by
Milton Levy
and
Philip Wong

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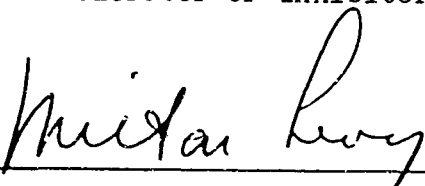
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ACTION OF SODIUM CITRATE AND SODIUM BORATE ON THE
POLARIZATION CHARACTERISTICS OF A URANIUM-STEEL COUPLE


ABSTRACT

The action of sodium citrate and sodium borate in aqueous media on the polarization characteristics of uranium and steel was studied. The effect of citrate and borate concentration on these polarization characteristics was also studied.

Steady-state corrosion currents developed by the uranium-steel couple were measured using zero-resistance ammeter circuitry, and polarization measurements were made using galvanostatic techniques. Sodium citrate and sodium borate were classified as either accelerator or inhibitor, dependent upon concentration.

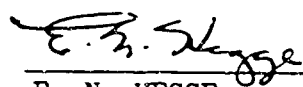


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INTRODUCTION

Army materials development has been directed primarily toward constructional materials having a high strength-to-weight ratio. However, there is also the need for high-density engineering materials having high strength, a reasonable amount of ductility and toughness, and resistance to corrosion. These high-density material applications vary from those requiring sufficient mass to enable matching of ballistic trajectories (spotting rounds) to those requiring combined resistance to ballistic penetration and gamma shielding (armor). Wrought uranium alloys (unclassified, depleted) are being developed to meet this requirement.

In many ways uranium is similar to iron in its corrosion behavior. It is quite resistant to corrosion in dry air but, like iron, corrodes rapidly in the presence of moisture. Various inorganic inhibitors which are known to be effective with iron have been tested with uranium. Aerated solutions containing chromate, molybdate, tungstate, and nitrite were used. These were only moderately effective, the nitrite ion being best.¹ Ward and Waber,² in a later study, reported that the nitrate ion appeared to be a better inhibitor for uranium than does nitrite ion.

A survey of the literature reveals that there is little published data concerning the galvanic action produced by coupling uranium with dissimilar metals in aqueous media. There is no published data available for the uranium-steel couple and the action of inhibitors on the polarization characteristics of this couple.

The current flow between dissimilar metals immersed in aqueous media is a promising means for studying the inhibition and acceleration of corrosion in relation to galvanic attack. The magnitude of this current is a measure of the influence of the couple on the attack of the anodic member of the pair. This method permits the growth of protective films to be followed readily and is amenable to polarization studies. Polarization studies yield basic and significant information about the processes associated with corrosion inhibition and acceleration.

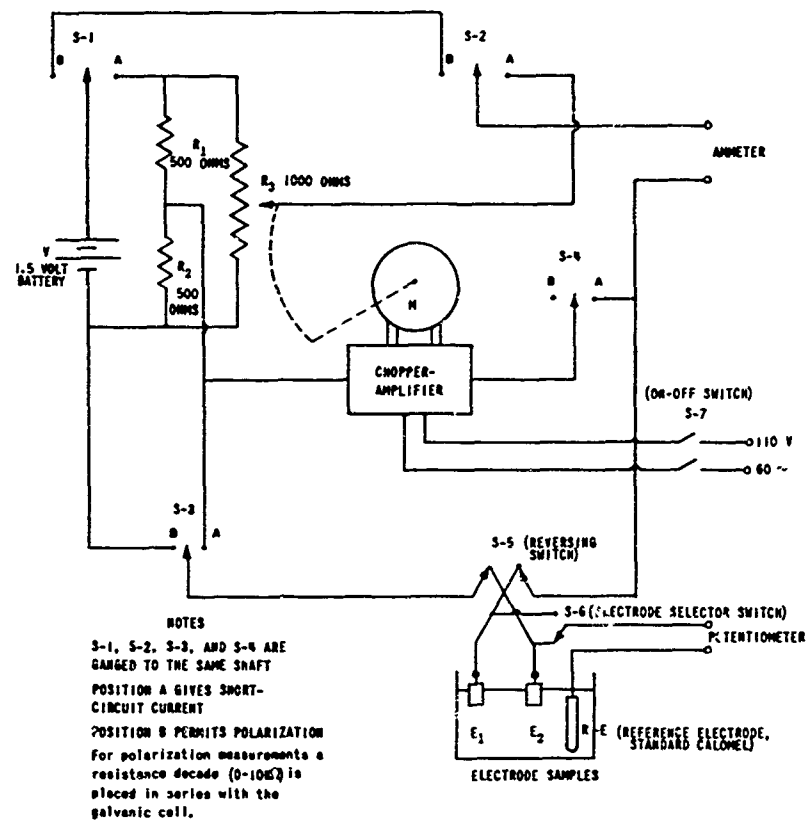
This study deals with the action of sodium citrate and sodium decaborate in aqueous media on the polarization characteristics of uranium and steel. The effect of citrate and borate concentration on these polarization characteristics was also studied. A prior study reported by Levy³ showed that citrate accelerated corrosion of iron in the iron-copper couple, while the borate behaved as an inhibitor.

APPARATUS AND EXPERIMENTAL PROCEDURE

The test electrodes were metal strips, 2.15 cm x 1.25 cm x 0.3 cm, uranium-8% molybdenum alloy and 1015 mild steel, held parallel to and at a fixed distance of 1/2 inch from each other by an insulating spacer.

Each electrode surface area exposed to electrolyte was 7.0 square centimeters. The electrodes were cleaned by polishing with 2/0 emery paper, rinsing with ethyl alcohol, and air drying prior to use.

The apparatus, basically that described by Levy³ and Hatch,⁴ was modified to automatically and continuously record current measurements and still maintain a short-circuited cell. Instead of a direct short circuit across the cell terminals, a source of current equal to the short-circuit current was interposed. This was accomplished by sensing the voltage across the cell terminals and adjusting automatically the output of the current source to maintain the voltage across the cell effectively at zero. The effect of the ammeter resistance is compensated by this self-adjusting current source. Figure 1 is a schematic representation of the apparatus.



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Figure 1. SCHEMATIC DIAGRAM FOR CURRENT FLOW AND POLARIZATION MEASUREMENTS

Current-flow measurements were made between the uranium-steel couple in untreated tap water, sodium citrate-tap water solutions, and sodium decaborate-tap water solutions. The concentrations were varied between 0.1 percent and 1.5 percent (weight-volume). Anodic and cathodic polarization measurements were made after the electrodes had been immersed in the desired medium for 24 hours, so that any protective film would become well established. The potentials of the individual electrodes of the couple were measured against a saturated calomel electrode and as a

function of current. Plots of this potential versus current represent the polarization characteristics of the anode and cathode metals in the desired environment. The electrolytic test cells were maintained at 20 C throughout the test, without aeration of solution. The tap water used in the electrolytes had the following chemical analysis:

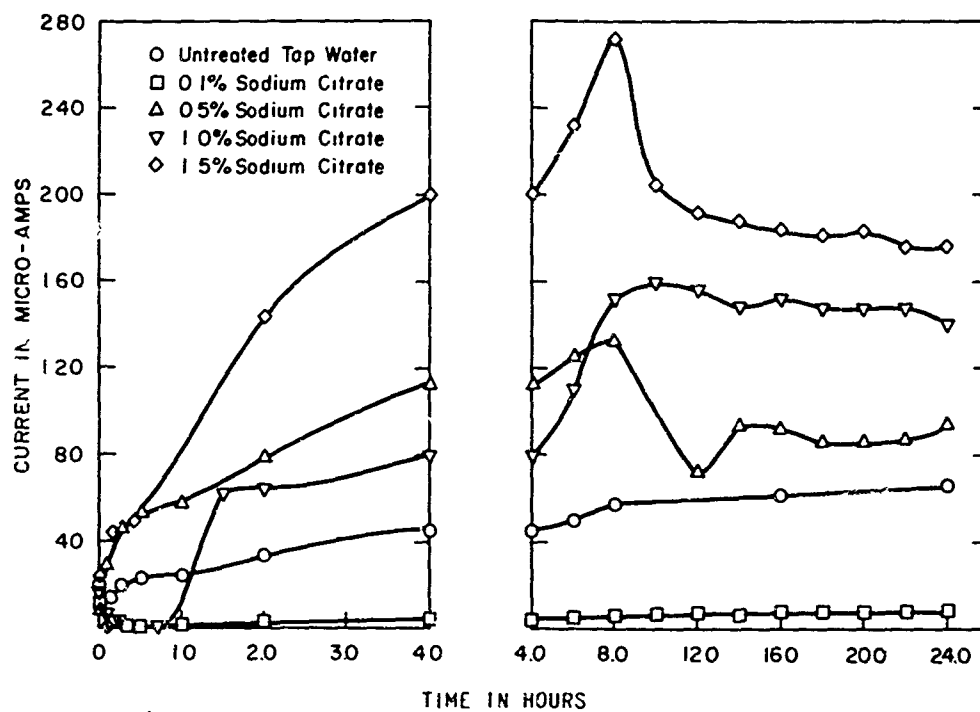
Chloride	- 7 ppm	Magnesium	- 2 ppm
Total Hardness, CaCO ₃	- 18 ppm	Sulfate	- 10 ppm
Calcium	- 6 ppm	Total Solids	- 40 ppm
pH	- 6.5		

All chemical used were of reagent grade.

RESULTS AND DISCUSSION

Sodium Citrate

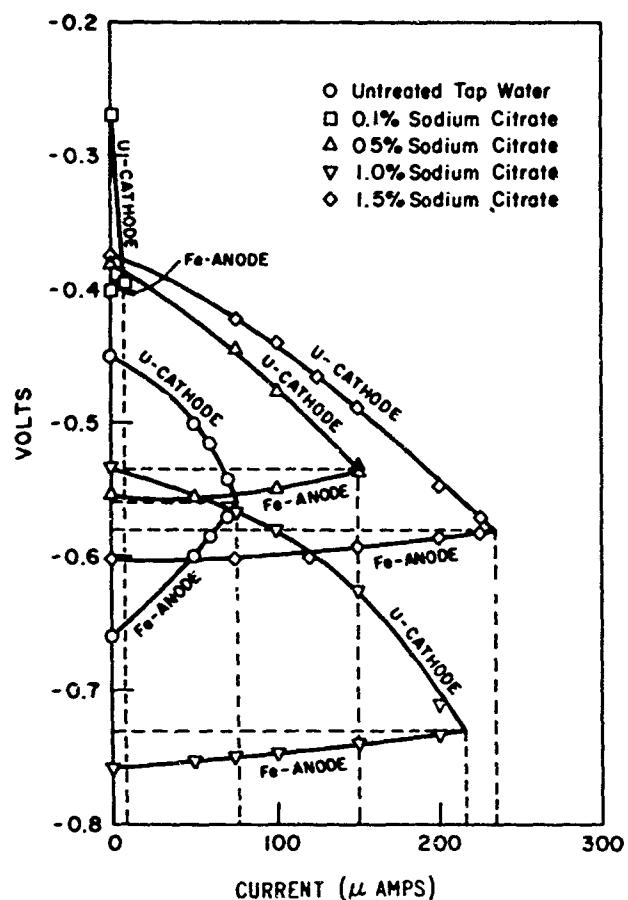
The current flow-time curve for uninhibited tap water (Figure 2) is in a relatively high current range, an indication of acceleration of the anodic member (steel) of the uranium-steel couple as a result of the couple. Initially, the current dropped, indicative of oxide film formation, followed by a gradual rise as destruction of the oxide film proceeded on the steel anode.



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Figure 2. CURRENT FLOW CHARACTERISTICS FOR URANIUM-STEEL COUPLE IN TAP WATER TREATED WITH SODIUM CITRATE

The current flow decreased to 0 microamperes initially for the couple in tap water treated with 0.1 percent sodium citrate. Uranium was the anodic member of the couple, and steel the cathodic member. At 0 μ amps the polarity reversed (uranium-cathode, steel-anode) and subsequently the flow rose to a relatively low level. Sodium citrate in concentration of 0.1 percent considerably decreased the galvanic attack of the anodic member of the couple.



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Figure 3. EFFECT OF SODIUM CITRATE ON THE POLARIZATION CHARACTERISTICS OF URANIUM-STEEL

of uranium as cathode and as a function of current; the lower arms those of steel as anode. The intersection of the anodic and cathodic potentials represent the potentials of the short-circuited couple. The currents corresponding to these intersections are those generated by the short-circuited couples.

The addition of 0.1 percent sodium citrate to tap water markedly increased the slope of the cathodic polarization curve. The anodic slope decreased (see Table I). This increased cathodic polarization caused a

The current flow curve for tap water treated with 0.5 percent sodium citrate rose sharply as destruction of the oxide film on the steel anode proceeded, fell sharply, indicative of some repair, and leveled off at a relatively high value. The steady-state current was greater than that of the untreated tap water. There was no reversal of polarity at this concentration.

The current flow curves for tap water treated with 1.0 percent and 1.5 percent sodium citrate were similar to that of the 0.5 percent concentration. Steel was the anodic member of the couple, uranium the cathodic member. The steady-state current increased with increasing concentration and was considerably greater than the untreated system.

Figure 3 shows the results of polarization measurements of steel as anode and uranium as cathode after 24-hour immersion in untreated tap water, tap water treated with 0.1, 0.5, 1.0, and 1.5 percent sodium citrate. These data are from the same series of tests as the current flow data. The upper arms of the curves represent the potential

Table I. POLARIZATION RESISTANCE $\left(R = \frac{E}{I}\right)$ FOR URANIUM AND STEEL IN SEVERAL AQUEOUS MEDIA

Environment	Polarization Resistance (ohms)	
	Uranium (Cathode)	Steel (Anode)
Untreated Water	2,250	1750
0.1% Na Citrate	22,000	778
0.5% Na Citrate	1,067	250
1.0% Na Citrate	1,300	100
1.5% Na Citrate	1,040	134

marked reduction in the current flow between the anodic and cathodic members. Conditions which increase the slopes of polarization curves, i.e., shift the convergence point of anodic and cathodic potential to lower current values, inhibit corrosion. Thus, the inhibition of current flow at this concentration appears to be due primarily to a marked increase in the polarization of the cathode.

The slopes (polarization resistance) of both anodic and cathodic polarization curves decrease for tap water treated with 0.5 percent sodium citrate. However, there is a greater decrease in the anodic slope (Table I). Conditions which decrease the slopes of polarization curves, i.e., shift the convergence point of anodic and cathode potential to higher current values, accelerate corrosion. If polarization of the cathode or anode is prevented, convergence occurs at a higher current value and corrosion is increased. Sodium citrate in concentration of 0.5 percent behaves as a corrosion accelerator primarily by anodic depolarization in tap water.

The polarization curves for both the 1.0 percent and 1.5 percent concentrations are similar to those obtained with the 0.5 percent concentration. The convergence point of anodic and cathodic potential increased with increasing concentration, resulting in increased acceleration of corrosion. Sodium citrate in concentrations of 1.0 percent and 1.5 percent accelerates corrosion primarily by preventing polarization of the anode. Table II contains potentials of the short-circuited couples and current values for the short-circuited couples in untreated tap water and tap water treated with sodium citrate as determined by polarization curves. The inhibitor action of the 0.1 percent sodium citrate is reflected in both potential and current values. The current has decreased to 8 μ amps and the potential is more electropositive than the untreated system and the systems containing the higher concentrations of sodium citrate. Similarly, the acceleration of corrosion by the higher sodium citrate concentrations was reflected by the increasing current values. Table III compares the current generated by the short-circuited couple as determined by polarization data with the experimental maximum current noted in the current-time curves. In general, they are in good agreement.

Table II. POTENTIALS AND CURRENT VALUES DETERMINED BY POLARIZATION CURVES

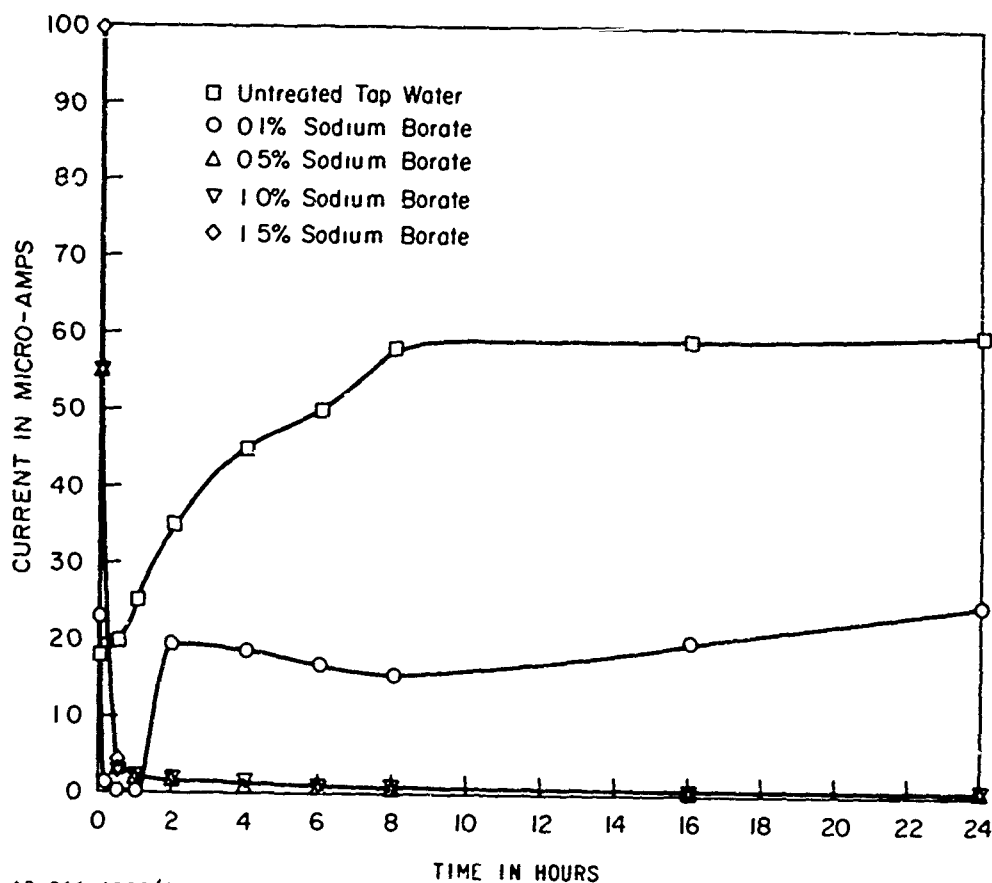
Environment	Potential (Volts) Versus S.C.E.	Current (μ amps)
Untreated Tap Water	-0.560	76
0.1% Na Citrate-Water	-0.395	8
0.5% Na Citrate-Water	-0.535	150
1.0% Na Citrate-Water	-0.730	216
1.5% Na Citrate-Water	-0.580	234

Table III. COMPARISON OF CURRENTS GENERATED BY THE SHORT-CIRCUITED COUPLES (POLARIZATION DATA) WITH EXPERIMENTAL MAXIMUM CURRENTS NOTED IN CURRENT-TIME CURVES

Environment	Current (μ amps)	
	Polarization Data	Experimental (max.)
Untreated Tap Water	76	71
0.1% Na Citrate-Water	8	8.5
0.5% Na Citrate-Water	150	133
1.0% Na Citrate-Water	216	161
1.5% Na Citrate-Water	234	272

Sodium Decaborate

Figure 4 represents the current-flow characteristics of tap water treated with sodium decaborate in concentrations of 0.1, 0.5, 1.0, and 1.5 percent. Initially, the current flow decreased sharply to 0 μ amps for the couple in 0.1 percent sodium borate solution, uranium behaving as the anodic member of the couple. This was followed by a reversal of polarity (steel anodic) and accompanied by a sharp rise in current flow to 20 μ amps, where it leveled off. However, the steady-state current was considerably lower than that obtained in untreated water, indicative of the inhibitor action of the sodium decaborate at this concentration. The current-flow curves for tap water treated with sodium decaborate in concentrations of 0.5, 1.0, and 1.5 percent were characterized by initial sharp decreases followed by gradual decreases, finally leveling off at very low current values. At these concentrations, uranium remained anodic throughout the entire run and the sodium borate effectively reduced the galvanic attack of the anodic member of the couple. Sodium borate is most effective at concentrations greater than 0.1 percent where uranium is the anodic member of the couple. The reversal of polarity noted in the 0.1 percent



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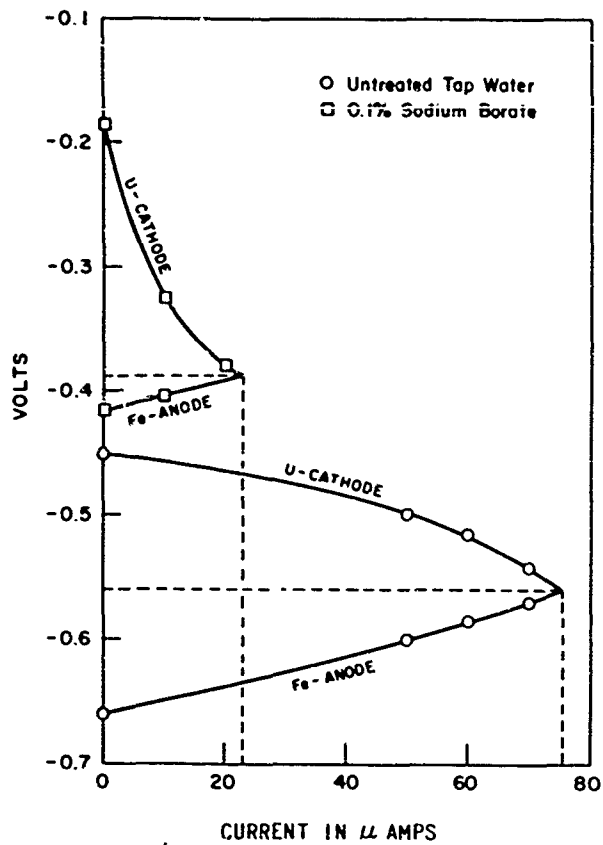
Figure 4. CURRENT FLOW CHARACTERISTICS FOR URANIUM-STEEL COUPLE IN TAP WATER TREATED WITH SODIUM BORATE

concentration solution was accompanied by an increase in current flow and a steady-state current value of approximately 40 times greater than those noted at the higher concentrations.

Figure 5 shows results of polarization measurements of uranium as cathode and steel as anode in untreated tap water and tap water treated with 0.1 percent sodium decaborate. The upper arms of the curves represent the potential of uranium as cathode and as a function of current; the lower arms, those of steel as anode.

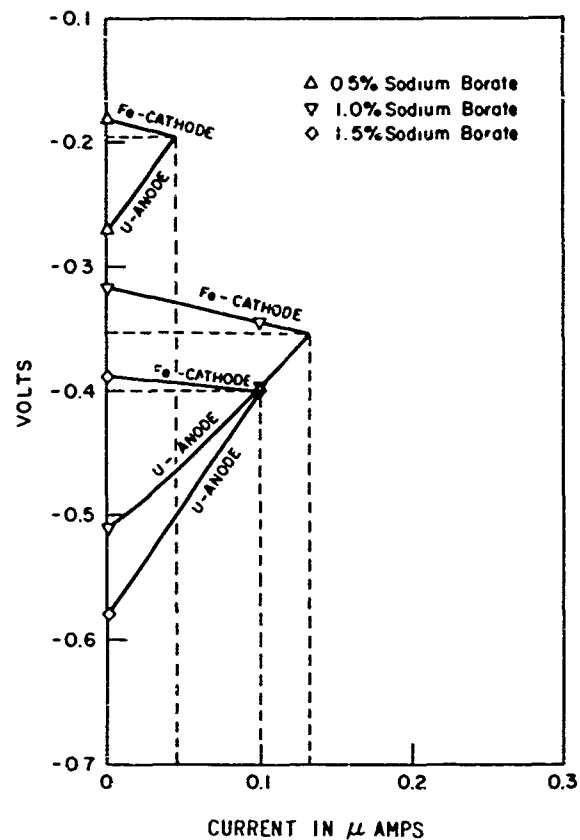
The addition of 0.1 percent sodium decaborate caused little change in the slope of the anodic polarization curve, but markedly increased the slope of the polarization curve of the cathode. Thus, the inhibition of current flow at this concentration appears to be due to cathodic polarization.

Polarization curves for uranium and steel in tap water treated with 0.5, 1.0, and 1.5 percent sodium borate are contained in Figure 6. The upper arms of the curves represent the potential of steel as cathode and as a function of current; the lower arms, those of uranium as anode. At these concentrations, uranium was the anodic member of the couple, steel the cathodic member.



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Figure 5. EFFECT OF SODIUM BORATE ON POLARIZATION CHARACTERISTICS OF URANIUM-STEEL



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Figure 6. EFFECT OF INCREASING SODIUM BORATE CONCENTRATION ON POLARIZATION CHARACTERISTICS OF URANIUM-STEEL

Table IV shows that for the 0.5 percent concentration the potential of the short-circuited couple was -0.195 volts and the current was 0.045 μamps. At this concentration, the greater inhibition over the 0.1 percent concentration is reflected in the decrease of current from 23 μamps to 0.045 μamps and the more electropositive potential from -0.390 volts to -0.195 volts.

Table IV. POTENTIALS AND CURRENT VALUES DETERMINED BY POLARIZATION CURVES

Environment	Potential (Volts) Versus S.C.E.	Current (μamps)
Untreated Tap Water	-0.560	76
0.1% Na Borate-Water	-0.390	23
0.5% Na Borate-Water	-0.195	0.045
1.0% Na Borate-Water	-0.353	0.132
1.5% Na Borate-Water	-0.400	0.100

The current generated by the short-circuited couple in tap water treated with 1.0 percent sodium borate was 0.132 μ amps, the potential -0.353 volts. This shows marked inhibition when compared to the untreated tap water system, the current decreasing from 76 to 0.132 μ amps. However, the inhibitor action at 1.0 percent concentration was not as effective as at 0.5 percent concentration. The currents generated by the short-circuited couples were 0.132 and 0.045 μ amps, and the potentials -0.353 volts and -0.195 volts, respectively (Table IV). Table V contains the values for the slopes of anodic and cathodic polarization curves. Comparing slopes for the 0.5 and 1.0 percent concentrations, it is evident that the cathodic slopes are unchanged, while the anodic slope for the 1.0 percent concentration has decreased. Thus, the greater current generated by the short-circuited couple in tap water treated with 1.0 percent sodium borate is due, in part, to this depolarization of the anode. Also, the more electronegative shift of potential is a contributing factor.

Table V. SODIUM BORATE

Polarization Resistance $\left(R = \frac{E}{I}\right)$ for Uranium and Steel
in Aqueous Sodium Borate Media

Environment	Polarization Resistance (ohms)	
	Uranium	Steel
Untreated Water	2,250 (cathode)	1,750 (anode)
0.1% Na Borate	9,360 (cathode)	1,167 (anode)
0.5% Na Borate	1,660,000 (anode)	289,000 (cathode)
1.0% Na Borate	1,190,000 (anode)	288,000 (cathode)
1.5% Na Borate	1,800,000 (anode)	120,000 (cathode)

The current generated by the short-circuited couple in tap water treated with 1.5 percent sodium borate was 0.100 μ amps, the potential of the short-circuited couple was -0.400 volts (Table IV). This demonstrates the marked inhibition of sodium borate in concentration of 1.5 percent. The inhibitor effectiveness at this concentration was greater than the 1.0 percent concentration, but less than the 0.5 percent concentration. Comparing the anodic and cathodic slopes for tap water treated with 1.0 and 1.5 percent sodium borate (Table V), it is seen that there was a greater change in the anodic slope which increased with increasing concentration. Thus, the greater inhibitor effectiveness of the 1.5 percent concentration over the 1.0 percent concentration appears to be due to this increased anodic polarization of uranium.

Table VI compares the current generated by the short-circuited couples determined by polarization data with the experimental maximum current noted in the current-time curves. In general, they are in good agreement.

Table VI. COMPARISON OF CURRENTS GENERATED BY THE SHORT-CIRCUITED COUPLES (POLARIZATION DATA) WITH EXPERIMENTAL STEADY-STATE CURRENTS NOTED IN CURRENT-TIME CURVES

Environment	Current (μ amps)	
	Polarization Data	Experimental (max)
Untreated Tap Water	76	71
0.1% Na Borate-Water	23	24
0.5% Na Borate-Water	0.045	0.23
1.0% Na Borate-Water	0.132	0.36
1.5% Na Borate-Water	0.100	0.20

CONCLUSIONS

Sodium citrate in concentration of 0.1 percent in tap water behaves primarily as a cathodic polarizer, although some anodic depolarization was observed. This increased cathodic polarization caused a marked reduction in the current flow between the anodic and cathodic members. The more electropositive potential of the short-circuited couple is also a contributing factor. The maximum reduction in current as noted in current-flow data occurred when uranium was the anodic member of the couple. Upon reversal of polarity (steel anodic), the current flow increased, although it leveled off at a value considerably lower than that associated with the untreated system. From this data, it appears that sodium citrate is more effective on uranium than on steel.

Sodium citrate in concentrations greater than 0.1 percent behaves as corrosion accelerator in tap water primarily by anodic depolarization. At these concentrations, steel was the anodic member of the couple, uranium the cathodic member.

Sodium decaborate in concentration of 0.1 percent exhibited a marked reduction in current flow. This inhibition of current flow is attributed to cathodic polarization where steel was the anodic member of the couple, uranium the cathodic member.

At concentrations greater than 0.1 percent, steel was the cathodic member of the couple and uranium the anodic member. The polarization curves showed that the cathodic slopes were relatively flat and unchanged by increasing concentration, while the anodic slopes were steep and increased or decreased with increasing concentration.

Therefore, sodium decaborate in concentrations greater than 0.1 percent behaves as a corrosion inhibitor in tap water by anodic polarization.

ACKNOWLEDGMENT

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