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Technical Note N-950

CORROSION RATES OF URANIUM ALLOYED STEEL IN MARINE HYDROSPACE

by

C. V. Brouillette

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CORROSION RATES OF URANIUM ALLOYED STEEL IN MARINE HYDROSPACE

Y-F015-21-05-001A

Technical Note N-950

by

Carl V. Brouillette

ABSTRACT

Corrosion rates in the 3 hydrospace environments were primarily controlled by the dissolved oxygen concentrations and to a lesser extent by temperature.

Segregation of uranium in the steels, which possibly could have occurred during production, could not be detected.

Profuse micro-pitting over the surface of the uranium alloyed stainless steel was attributed to the uranium.

Corrosion rates of steel in deep ocean were low.

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INTRODUCTION

An inquiry of Atomic Energy of Canada Limited revealed that Eldorado Mining and Refining of Ottawa, Ontario, Canada, had produced a uranium alloyed steel. Information received from the Department of Mines and Technical Surveys, Mines Branch, Physical and Metallurgy Division of Ottawa, Canada, indicated that this steel had been shown to have a greatly reduced incidence of pitting when exposed to an immersed ocean environment. Such a steel could find unlimited uses in an ocean environment.

The uranium content of the alloys was 0.10 percent for carbon steels and 0.23 for 17 chromium steels. The Department of Mines and Technical Surveys furnished the following information on these steels: these low levels of uranium did not affect the usual physical properties appreciably, however, alloys of higher levels of uranium can show marked loss of impact resistance; the uranium used in these alloys was natural uranium (e.g., the isotope U-235 is about 0.7 percent of the added uranium); the β -radiation levels from the type of alloy used in these tests were so low that no health hazards were involved from proximity; and the alloys can be machined in the usual way without any particular care regarding disposal of chips, etc.

However, the uranium bearing test specimens prepared by the Department of Mines and Technical Surveys for exposure in an ocean water environment by the U. S. Naval Civil Engineering Laboratory, were handled with the precautions deemed necessary for a low level radiological material.

TEST PROCEDURE

The uranium alloyed steel test specimens, as received, were approximately 6 x 12 x 0.2 inches in size. The specimens exhibited no visible rust. They were cleaned with a soft cloth and mineral spirits, dried and weighed.

The steels are identified by reference to Table 1.

A space and weight problem limited the number of replicate specimens of each alloy which could be placed on each submersible test unit (STU).

Four exposure sites were utilized for these tests;

- (1) STU I-4 at 6800 feet deep for 13 months;
- (2) STU II-1 at 2500 feet deep for 6 months;
- (3) STU II-2 at 2500 feet deep for 13 months; and
- (4) in the harbor at Port Hueneme at 3 feet below mean low water (MLW).

Table 1. Steel Identify Supplied by Manufacturer.

Steel* Number	Percent							
	C	Mn	Si	U	Cr	S	P	
(2261-1)*	0.11	0.37	0.25	0.00	NI1	0.002	0.010	
2261-2	0.11	0.37	0.25	0.10	NI1	0.002	0.009	
(2267-1)*	0.36	0.72	0.19	0.00	NI1	0.005	0.006	
2267-2	0.36	0.72	0.10	0.09	NI1	0.006	0.009	
2252-1	0.12	0.67	0.20	0.00	17.1	0.010	0.010	
2252-2	0.12	0.67	0.20	0.23	17.1	0.010	0.010	
1010	SAE 1010 mild steel							

* Laboratory tests at NCEL have shown that these identify numbers should be interchanged; 2267-1 conforms with the 0.11C steel (2261-1 above) and 2261-1 conforms with the 0.36C steel (2267-1 below). The corrected numbers appear in the text below.

In addition, replicates of the test specimens were placed on the STU I-4 in such a way as to be exposed imbedded in the mud on the ocean floor, the stainless steel specimens were not available at the time that the STU I-4 was implaced.

Upon retrieval from the ocean exposure sites the test specimens were cleaned to remove rust. The loose rust and scale were removed by wet wire brushing and scraping. Then the panels were placed in a 10 percent aqueous solution of ammonium citrate, heated to 140°F and left for 45 minutes. Upon removal from this cleaning bath the panels were rinsed in tap water and brushed with a stiff bristle brush. The panels were dipped into alcohol to remove water, dried in warmed air and weighed. Recleaning was performed on replicate panels to determine cleaning loss. Using the weight loss data densities and linear dimensions of the panel the total rust penetration per year was calculated and expressed as mils penetration per year (mpy). One mil is 0.001 inch.

The relative incidence to pitting was examined through visual observation. Maximum depth of pits was measured using a needle penetrometer.

Test Site I was located about 85 nautical miles west of Port Hueneme, California, latitude 30° 46' North and longitude 120° 37' West. Test Site II was about 75 nautical miles west of Port Hueneme, latitude 36° 06' North and longitude 120° 42' West. The Port Hueneme harbor site was near the harbor entrance. A large fresh water drainage ditch emptied into the harbor at the opposite side from the ocean entrance and caused slight fluctuations in salinity and pH of the harbor water in the proximity of the test site.

RESULTS

Table 2 gives data descriptive of the various ocean environments used to expose the steel test specimens.

The data in Table 2 for the 3 environments show only a slight variation in velocity of water, pH and salinity when compared one with the other. The change in salinity from 28 to 34.56 cc/l would have no measureable affect on the electrolytic properties of the sea water which could affect corrosion rates. These slight variations in velocity could not be expected to vary the corrosion rates of iron or steel in the three environments more than very slightly. Higher velocities of sea water more rapidly replenish oxygen to a steel surface and also removes soluble corrosion products. Lower velocities approaching stagnation were expected to produce comparatively low weight losses, but may introduce pitting.¹

The temperature change from 6800 feet to the surfaces was about 27°F. The higher temperature at the surface could increase the corrosion rate of steel and iron by about 25 percent. However, the corrosion rate of iron and steel within these temperature and pH ranges was expected to be essentially uniform.¹

Table 2. Test Site Environments

Location	Depth in ft. below MLM ^{1/}	Temp. °F	Velocity knots	pH	Oxygen ^{2/} cc/l ^{2/}	Salinity ppt ^{3/}	Pressure psi	Fouling
Site I	6800	37	<0.5	7.25	0.83	34.56	2330	trace
Site II	2340	45	~0.3	7.46	0.42	34.37	1040	trace
Harbor	3	58-64	~0.2	7.8-8.2	~5.6	28-3	4/	moderate to heavy

1/ MLM = mean low water

2/ cc/l = cubic centimeters per liter

3/ ppt = parts per thousand

4/ near atmospheric

The oxygen concentration varied from 0.42 cc/l at the 2500 foot depth to 0.83 cc/l at the 6800 foot depth and was about 5.6 cc/l near the surface. The corrosion of iron and steel immersed in unpolluted sea water has been shown to be dependent primarily on the supply of oxygen to the metal.^{1,2,3} The limiting rate is controlled by the rate of oxygen replenishment.²

The pH range of 7.15 to 8.2 is considered relatively neutral, and its influence, in this range, on the corrosion rates of iron and steel would be of minor importance.^{1,4,5} In this pH range pitting could be the major form of corrosion.⁴

Thus the velocities and salinities of water at the various depths were found to be very similar. The pH was that of relatively neutral sea water. The temperature rise near the surface would tend to slightly increase the corrosion of iron and steel. However, the oxygen concentrations appear to be the distinguishing property for the 3 environments. Pressure effects on corrosion were expected to be very slight if discernable at all.⁶

The moderate to heavy fouling found in the shallow environment would tend to lower the pH and oxygen content and promote pitting of the steel.

In Table 1 the compositions of the various test steels are shown. The carbon steels vary in carbon content from about 0.10 to 0.36 percent and in uranium content from 0.00 to 0.10 percent. The 2252 series is a 17 percent chromium stainless steel. Replicate specimens of the 2252-2 stainless steel contain 0.23 percent uranium. With respect to corrosion in sea water, little difference in corrosion rates were predicted for the 0.10 and 0.36 carbon steels because of the carbon content.^{1,4,5} Although, theoretically, a slight increase in corrosion rate may be expected with increase in carbon content of steels exposed in sea water, the observed corrosion rates in any one environment would be essentially the same.⁵

Table 3 gives the observed corrosion rates of the steel test specimens on the STU's and in the harbor at Port Hueneme (California). The lowest rates were at Site II (near the ocean floor at about 2500 feet depth) and varied from 0.5 mpy to 1.7 mpy. At Site I (a depth of 6800 feet) the rates varied from 1.8 to 2.7 mpy. The oxygen content for the two environments was measured at 0.42 and 0.83 cc per liter, respectively. The higher oxygen level giving proportionally higher corrosion rates.

In a previous study (reference 7) corrosion rates were determined for in-place steel sheet piling at 5 naval harbors. Of particular interest were the corrosion rates found at one foot above the floor of the 5 harbors. Table 4 gives the evaluated corrosion rate data for sheet steel piling at these 5 harbors for the environment 1 foot above the mud line. These corrosion rates are calculated from weight loss and are expressed in mils penetration per year. The corrosion rates, found 1 foot above the mud line, were about 2.5 to 3.0 mpy. This would indicate a low oxygen concentration at these environments. The corrosion rates for all steel specimens exposed near the deep ocean (6800 feet) floor are shown to be only slightly less than that found for steel near the floor of the 5 harbors.

Table 3. Corrosion Rates in the Ocean (express in mpy).

Number	Months Exposed	Rates at Indicated Depths Below Surface (feet)		
		5300	2500	3
2267-1 0.11 carbon	6		1.6	
	12			5.8
	13	2.5	1.1	
	-in mud 24	1.6		5.7
2261-2 0.11 carbon 0.1 uranium	6		1.4	
	12			5.7
	13	2.2	1.0	
	-in mud 24	1.5		5.4
2261-1 0.36 carbon	6		1.7	
	12			5.3
	13	2.0	1.2	
	-in mud 24	1.4		5.6
2267-2 0.36 carbon 0.09 uranium	6		1.7	
	12			6.4
	13	1.8	1.3	
	-in mud 24	1.5		5.5
2252-1	6			
	12			(1.7)*
	13		(0.25)*	
	-in mud 24			(3.1)*
2252-2 0.23 uranium	6			
	12			(2.5)*
	13		(0.26)*	
	-in mud 24			(2.0)*
SAE 1010 Mild steel	6		0.5	
	12			7.1
	13	2.7	1.0	
	-in mud	1.7		

* Corrosion due to deep pits generally perforating panel and to elongated pits.

On STU I-4 at the 6800 foot depth, replicate steel specimens exposed on the bottom of the STU so as to be imbedded in the mud, showed lower corrosion rates than did those in the sea water near the bottom. The rates in the mud were 1.5 to 1.7 mpy and in the free moving ocean water immediately above the bottom from 1.8 mpy to 2.7 mpy.

The oxygen concentration in the surface ocean water near Port Hueneme was about 5.6 cc per liter and the corrosion rates were found to be 5.3 to 7.1 mpy. Reference to a study of the corrosion rates in Port Hueneme harbor in 1958 showed an average of 6.8 mpy for mild steel for a 30 month period.⁸

Thus, as expected, the corrosion rates of all carbon steels were primarily dependent on oxygen concentrations. Evidences of any effects of pressure were not observed.

Pitting

As stated previously, the uranium alloyed steel was purported to have more resistance to pitting than other steels when immersed in sea water. The damage from pitting could not be determined from weight losses. Table 5 shows the approximate and maximum pit depths for the steel test specimens. The frequency or incidence of pitting, was appraised visually.

The deep pitting of the stainless steels (2252-1 and 2252-2) was a type of localized pitting, and included elongated pits, Figures 3 and 4. The localized pits frequently perforated the steel specimens, Figures 5 and 6. As a result the pitting data for the stainless steels was not included in Table 5. The stainless steel, 2252-2 containing 0.23 uranium showed possibly fewer perforations, Figure 6, during 2 years exposure in shallow sea water than did the steel 2252-1 containing no uranium, Figure 5. The deep ocean exposures showed only the elongated pits typical of stainless steel in sea water, Figures 3 and 4.⁸

The average pit depths of the uranium alloyed 0.11 carbon steel 2261-2 in shallow water was shown to be the same as for 0.11 carbon steel 2267-1 containing no uranium, Table 5. Equivalent pit depths occurred in the 0.36 carbon steel 2267-2 containing uranium and the 0.36 carbon steel 2261-1 without uranium. In the shallow sea water environment after 2 years exposure, no decrease in pit depth was observed when either steel was alloyed with uranium. In the deep ocean environments, at 2500 and 6800 feet, no pitting was evident in these steels after 1 year of exposure. The corrosion appeared to be uniform over the surface.

The uranium alloyed steel 2252-2 (stainless) after 2 years exposure in shallow sea water, exhibited profuse micro-pits, Figures 7 and 8. Since this type of corrosion was found only in the steel containing uranium, the phenomenon was attributed to its presence.

Table 4. Corrosion of Steel One Foot Above the Mudline⁷

Harbor	Puget Sound, Washington	San Diego, California	Norfolk, Virginia	Key West, Florida	Coco Solo, Panama
Corrosion rate - mpy	2.5	3.0	2.5	2.5	3.0
Temperature	48-52°F	61-78°F	49-86°F	71.5-87°F	80-84°F
Water Depth	20 ft	10 ft	15 ft	2 ft	--
Harbor depth below MLW	40 ft	12 ft	20 ft	15 ft	10-25 ft

Table 5. Pitting in Shallow Environment (mpy)*

Steel	% Carbon	Avg.	Max.	Min. Measured
2267-1	0.11	0.033	0.064	0.015
2261-2	0.11	0.034	0.057	0.013
2261-1	0.36	0.049	0.064	0.033
2267-2	0.36	0.043	0.059	0.026

* Pitting rate is the average from about 14 pits for each specimen.

Uranium forms a carbide which is heat stable but is attacked by water.⁹ Its presence in the surface lattice could affect the corrosion processes.

The slight radioactivity at the interface between the steel and the sea water is sufficient to promote ionization of water and sea water salts and thereby possibly affect the corrosion processes.

During manufacture of the stainless steel (2252-2) the manufacturer reported a tendency towards segregation of the uranium during cooling of the steel.

To check these possibilities in the stainless steel (2252-2) the following techniques were used; (1) preparation of an autoradiograph using X-ray film, and (2) using NTA films, (3) X-ray spectroscopy, (4) counting α -particles with a thin window Geiger tube, and (5) metallographic examination of 2261-1, 2261-2, 2267-1 and 2267-2.

Also, the open circuit potential of each steel was measured in 0.5 molar sodium chloride versus the saturated calomel electrode. The measurements were made using a Keithley model 610B electrometer, with an input impedance of 10^{14} ohms; the output was monitored as a Leeds and Northrup Speedomax recorder for a period of 5-10 minutes to obtain a steady potential.

The potentials at indicated time intervals are shown in Table 5.

The uranium alloyed steels, after 19 hours in salt solution without aeration, show about 20 mv greater negativity than the steels containing no uranium, but after 91 hours the polarity in the 2252-1 and 2252-2 had reversed.

The 2261-1 and 2261-2 steels show consistent changes in open circuit potential with time, either with or without aeration. The uranium bearing steel 2261-2 producing a very slightly more negative (or active) potential, However, the 2251-1 and 2251-2 stainless steels show a converging towards the same potential in the passive range during continuous aeration.

X-ray spectrographs were made on the 2252-2 uranium alloyed stainless steel. One set of readings was taken at a highly corroded area and the second set at a relatively unattacked area. Both areas were cleaned of all rust before the readings were taken. The accuracy of these readings was ± 10 percent and were not conclusive, although they indicated less uranium in the corroded area.

An α -counter was then used to determine uranium content in these same 2 areas and also on the back side of these same specimens. The results showed, for the four surfaces integrated over a circular area 1 inch in diameter, that the content of uranium does not differ by more than 1 percent for each of these four surfaces. Thus the highly corroded anodic area of the 2252-2 and the relatively unattacked area showed uniform uranium distribution.

Exposure of these same 2 specimens was made in contact with NTA Eastman emulsion film, 10 microns thick, used in film badges and suitable for recording α -particles. For an exposure time of 12 hours, the number of α -tracks observed in visual fields at 600 X magnification varied from 0 to 1, 2 and 3 tracks for all areas examined. Thus for all surfaces, relatively no difference could be observed. A much longer exposure time could show differences in α -emission if it did exist.

Table 6. Potential vs. Saturated Calomel Electrode
(0.5 molar NaCl; 25C)

Steel	Without Aeration			Contiruous Aeration	
	at 2 hours	at 19 hours	at 91 hours	16 hours	21 hours
2261-1	-631	-672 mv	-699	-660	-669
2261-2	-648	-690 mv	-707	-669	-683
2252-1	-214	-189 mv	-280	-187	-175
2252-2	-147	-208 mv	-268	-162	-178

The developed autoradiograph film showed generally uniform radiation exposure over its entire surface and indicated no pronounced segregation of the uranium. The film density from the stainless steel 2252-2 (0.23 uranium) was much greater than that from steels 2261-2 (0.10 uranium) or 2267-2 (0.09 uranium). However, several small shadow like areas were present in the developed film. These areas were slightly more pronounced in film exposed to the stainless steel than when exposed to the other two steels, 2261-2 or 2267-2. The location of these areas on the steels did not always occur in the area of greatest corrosion attack. The shadowy areas on the film was interpreted as caused by very slightly more radioactivity in these areas and consequently as areas of very slightly higher uranium density.

The examination of the microstructures of the steels were made by Mr. F. M. Reinhart, Metallurgist, NCEL, and reported by him as follows.

"1. The carbon and uranium contents of the subject steels were reported by the supplier to be as follows:

2261-1, 0.11C
2261-2, 0.11C and 0.10U
2267-1, 0.36C
2267-2, 0.36C and 0.09U

2. The microstructure types of inclusions of the steels were determined to see if the difference in the corrosion pattern between the steels containing no uranium and those containing uranium could be explained on the basis of the types of inclusion or differences in microstructure.

3. Examination of the microstructures of 2261-1 and 2261-2 showed that they did not have the same carbon content; that of 2261-1 was approximately 0.4 carbon and that of 2261-2 was about 0.1 carbon. Because of this the microstructures of 2267-1 and 2267-2 were also examined and were found to vary in their carbon contents; that of 2267-1 was about 0.1C and that of 2267-2 was about 0.4C. This showed that the specimens had been interchanged by the producers.

Radiological counts showed that specimens numbered 2261-2 and 2267-2 did contain uranium.

These examinations showed that 2267-1 and 2261-2 contained about 0.1C with 2261-2 containing the uranium and that 2261-1 and 2267-2 contained about 0.4C with the 2267-2 containing the uranium.

4. Examinations of polished, and polished and etched longitudinal and transverse sections of the four subject specimens showed that:

(a) 2267-1 and 2261-2: (0.1C)

There was no difference in the type or characteristics of the non-metallic inclusions. The grain size of 2267-1 was non-uniform while the grain size of 2261-2 (containing the uranium) was larger and more uniform. There were no variations which would indicate differences in corrosion behavior of the two steels.

(b) 2261-1 and 2267-2:

There was no difference in the types or characteristics of the non-metallic inclusions. The grain size and uniformity of 2261-1 was about the same as 2267-1. The microstructures, in addition, were banded to about the same degree. There were no peculiarities which would indicate differences in the corrosion behavior of the two steels.

5. Examinations of the microstructures of the steels revealed no metallurgical differences which would explain a difference between the types of corrosion of the uranium and non-uranium bearing steels."

DISCUSSION

Corrosion of steel in sea water is essentially an electrochemical process. In sea water variations of pH, velocity, oxygen content, fouling, pollution, salinity, temperature and for metals of alloying constituents, physical condition or shape, all combine to produce varying corrosion rates. Sea water pressure alone did not demonstrate any affect on corrosion rate of iron or steel.

Analyses of the measured parameters of the three exposure sites, (1) shallow sea water, (2) 2500 feet deep and (3) 6800 feet deep, showed an unpolluted sea water differing primarily in fouling and oxygen content. The pH was that of relatively neutral sea water. The velocities at each site were not different enough to produce any appreciable variation in removal of soluble corrosion products. Moderate to heavy fouling in the shallow sea water environment was not found in the deep hydrospace. The slightly higher water temperature at the surface could slightly increase corrosion rates, possibly by about 25 percent. However, the highest corrosion rates observed in the deep sea environments were low. The variations in salinity were not sufficient to measurably affect an electrochemical process such as corrosion in a natural sea water environment.

Thus the only environmental parameters which varied significantly were fouling and oxygen content. The fouling in the shallow environment did contribute to pitting.

The corrosion rates of all steels exposed in the 3 environments were the least at 2500 feet deep, slightly greater at 6800 feet deep and several times higher near the surface in the shallow water environment. The rates in the deep environments reached a maximum of 2.7 mpy at 6800 feet and a minimum of 0.5 mpy at 2500 feet. The average rate at 6800 feet was 2.2 mpy and at 2500 feet was about 1.4 mpy. The oxygen concentration at 5700 feet depth was about double that for 2500 feet and the average corrosion rate was nearly double. The maximum rate for all steels exposed in the shallow ocean environment was 7.1 mpy; the average was about 5.8 mpy.

The uranium content of the steels did not affect, significantly, the weight losses or the pitting due to corrosion. Profuse micro-pits were observed in the uranium alloyed stainless steel and their occurrence was attributed to the presence of the uranium.

No positive benefit from the uranium was observed by visual evaluation of pitting damage in the stainless steels, although there appeared to be fewer holes corroded through the 2252-2 steel than through the 2252-1 steel, Figures 5 and 6.

CONCLUSIONS

The addition of uranium to the 0.10 or the 0.36 carbon steels had little or no effect on the pitting depth caused by corrosion.

Uranium alloying did not prevent elongated pitting or perforation of stainless steel (17.0 chromium) in hydrospace.

Profuse micropitting in the surface of uranium alloyed stainless steel was attributed to the presence of the uranium.

No significant segregation of uranium in the alloyed steels, which may have occurred during production of the steels, could be detected.

Corrosion rates in the various hydrospace environments were primarily controlled by the oxygen concentrations and to a much lesser extent by temperature; effects on corrosion by high hydrostatic pressure were not evident.

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The contribution of Mr. F. M. Reinhart of NCEL in metalographic examinations of the steel specimens and preparation of the paragraph on this examination was appreciated.

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Figure 1. STU I-4, 6800 feet, 13 months.

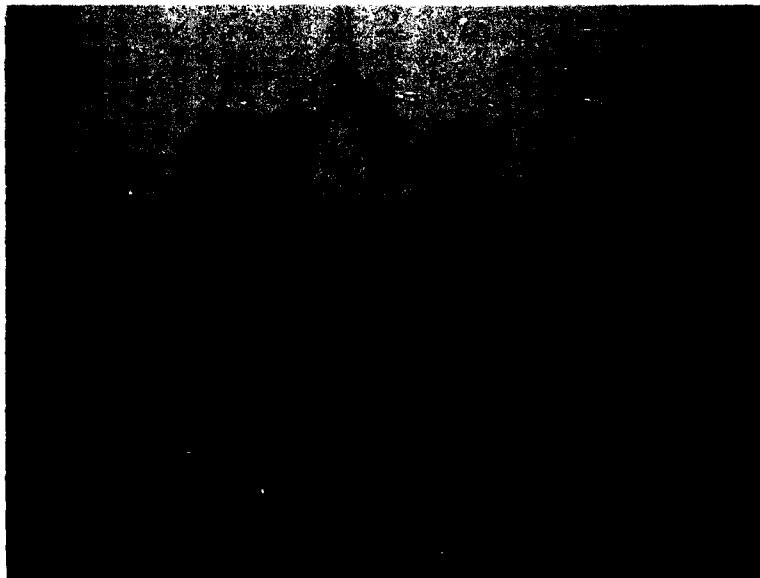


Figure 2. STU II-1, 2500 feet, 6 months.

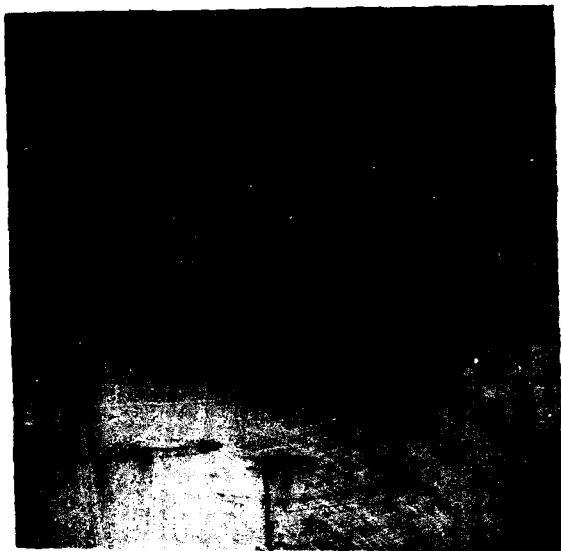


Figure 3. Stainless steel, 2500 feet, 13 months, no uranium, showing elongated pits.

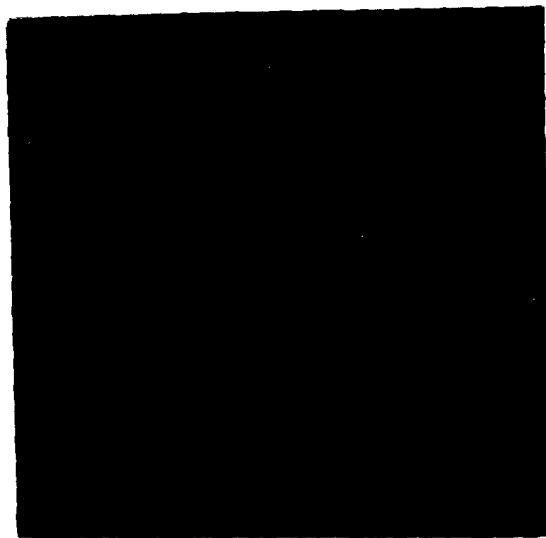


Figure 4. Stainless steel, 2500 feet, 13 months, 0.23 uranium, showing elongated pits.

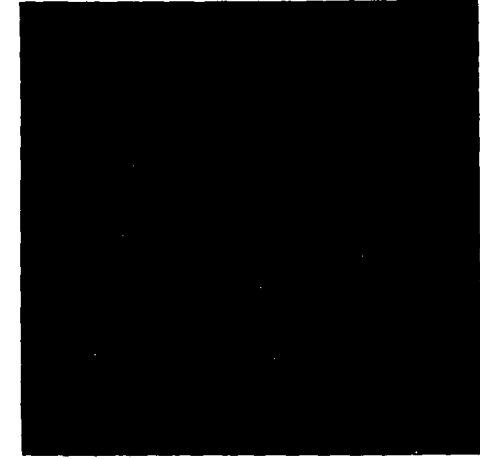


Figure 5. Stainless steel, 3 feet immersion, 24 months
no uranium, showing many deep pits.

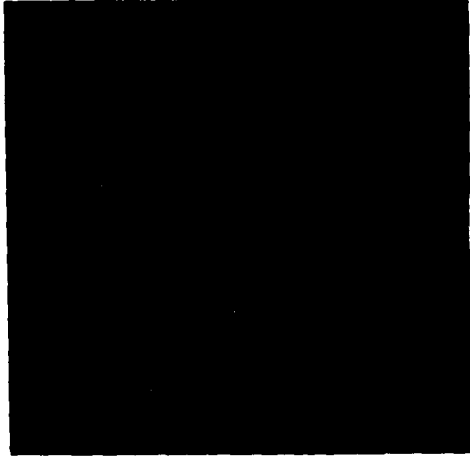


Figure 6. Stainless steel, 3 feet immersion, 24 months, 0.23
uranium, showing few deep pits, elongated pits at
edges, and profuse micro-pits.

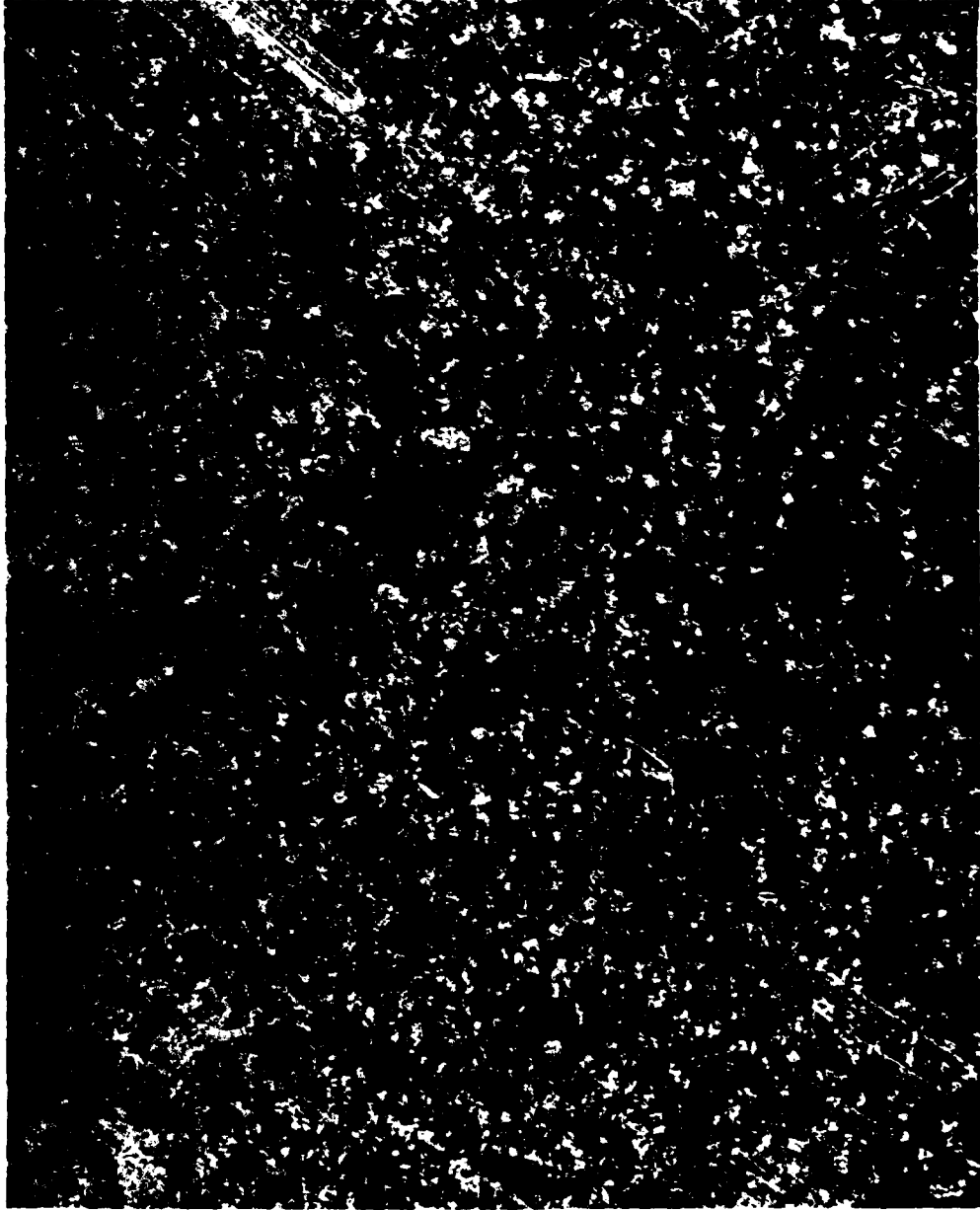


Figure 7. Stainless steel, 0.23 uranium, 3 feet immersion, 12 months, showing profuse micro-pits, magnification about 8X.



Figure 8. Stainless steel, no uranium, 3 feet immersion, 12 months, showing relatively smooth surface, magnification about 8X.

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13. ABSTRACT			
<p>Corrosion rates in the 3 hydrospace environments were primarily controlled by the dissolved oxygen concentrations and to a lesser extent by temperature.</p> <p>Segregation of uranium in the steels, which possibly could have occurred during production, could not be detected.</p> <p>Profuse micro-pitting over the surface of the uranium alloyed stainless steel was attributed to the uranium.</p> <p>Corrosion rates of steel in deep ocean were low.</p>			

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14 KEY WORDS	LINK A		LINK B		LINK C	
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