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**EROSION-CORROSION OF HAYNES-25 ALLOY;
RADIOACTIVE TRACER TECHNIQUES**

by

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ABSTRACT

The erosion-corrosion caused by the washing of a mixture of seawater and ocean-bottom material across a specimen of Haynes-25 at ambient room temperature has been studied with radioactive tracer techniques. The results obtained under the conditions of this study showed that the corrosion rate in such an environment increased 4 orders of magnitude over that observed in seawater alone.

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SUMMARY

The Problem

Haynes-25 alloy has been selected as the fuel container material in several SNAP systems for space applications. Because of possible accident situations which could place the fuel container in an ocean environment, it has been necessary to examine the corrosion behavior of this material in seawater to fully assess the radiological hazard from the radionuclide fuel. An exposure parameter which has not been examined to date is the erosive-corrosive nature of a mixture of seawater and ocean-bottom material flowing over the fuel container.

Findings

The present report describes an experiment in which a mixture of seawater and ocean-bottom material is allowed to wash across the face of a specimen of Haynes-25. The erosion-corrosion rate, determined by radioactive tracer techniques, was found to be 4 orders of magnitude greater than the corrosion rate observed in seawater alone.

INTRODUCTION

For the past several years the U. S. Naval Radiological Defense Laboratory has been carrying out studies to determine the nature and extent of the potential release of radionuclide fuel material from proposed and operational SNAP (Systems for Nuclear Auxiliary Power) units to the seawater environment.

In addition to examination of the solubility behavior of the fuel materials themselves in seawater,¹⁻⁶ experiments have been carried out for determination of the corrosion rates of various fuel container materials in seawater.⁷⁻⁹

It is conceivable that a SNAP unit could become exposed to ocean-bottom conditions which will cause erosion (scouring) of the container material (e.g. relatively high current velocities). Since many metals owe their corrosion resistance to protective films or layers of corrosion products,¹⁰ erosive removal of these films or layers may lead to significantly increased corrosion rates.

Because of these considerations a study has been carried out on Haynes-25 with an apparatus which allowed a mixture of ocean-bottom material and seawater to wash across radioactive specimens. The subsequent measurement of the activity in the bottom material-seawater mixture permitted calculation of the amount of Haynes-25 removed by the erosive-corrosive action of the mixture. The present report describes the apparatus used and presents the results of the erosion-corrosion experiments.

EXPERIMENTAL

A. Test Apparatus

The principal considerations in the design of the erosion-corrosion apparatus were: (a) complete electrical isolation of the specimen from other metallic components (test chamber); (b) control of the velocity of the seawater-bottom material mixture across the face of the specimen (motor, speed reducer, tilting platform); radiological shielding of the experimenters from radioactive specimens (lead cave).

The test chamber, which contained the specimen and the seawater-bottom material mixture, was a stainless steel box having internal dimensions of 42 cm length, 9 cm width and 5 cm depth. The interior of the chamber was coated with Teflon. The specimen, mounted in a square cavity in the bottom of the chamber and held in place with a silicone rubber adhesive, was positioned such that its exposed surface was flush with the bottom surface of the chamber. The top of the chamber had a removable pyrex window assembly to permit placement of the specimen and addition and removal of the seawater-bottom material mixture. Figure 1 shows one of the three such test chambers used.

The drive mechanism which tilted the platform on which the three test chambers were mounted consisted of a 1/8 HP AC motor coupled to a variable-output (0-35 rpm) speed reducer. The output shaft of the speed reducer was attached to a flywheel-and-rod assembly which translated the rotary motion to a reciprocating motion that tilted the platform. The degree of tilting, or pitch, of the platform was made variable by (1) slotting the flywheel along its radius to permit one end of the rod to be positioned at various distances from the center of the flywheel and (2) making the effective length of the rod variable

Fig. 1 Stainless Steel Exposure Chamber.

by drilling several mounting holes (to which the platform was attached) along its length.

The lead shield, required for minimizing the radiation hazard to personnel, consisted of a cave (large enough to enclose the entire apparatus) made up of 2-in.-thick interlocking lead bricks. A wooden box enclosed the lead cave. The hinged top of the wooden enclosure had a 1/2 in. lead plate to provide shielding in the upward direction. Figure 2 shows an overall view of the test apparatus in the lead shield.

B. Seawater

The seawater used in this study was collected on a biweekly schedule. Collection was made at least one mile from the nearest land mass (Farallon Islands, off San Francisco) by dip-filling of the seawater into 5-gallon polyethylene carboys. Prior to use the seawater was filtered through an 0.8 μ Millipore filter for removal of particulate matter.

C. Ocean-Bottom Material

The ocean-bottom material used in this study was obtained at a depth of 150 feet off the Wilson Cove area of San Clemente Island. The material is a feldspathic sand containing large amounts of calcium carbonate (in the form of shell fragments) and minor amounts of pyroxene.

D. Specimens

The radioactive specimens of Haynes-25 used were the same ones used in a previous study.⁸ Details of specimen preparation, neutron activation and other pertinent information are described in Ref. 8. Surface profile measurements of the specimens, taken before and after exposure, were performed on a Proficorder (Micrometrical Division, Bendix Corporation, Ann Arbor, Michigan). This instrument has the capability of measuring vertical surface irregularities as small as 0.5 microinches.

Fig. 2 Erosion-Corrosion Exposure Apparatus.

E. Exposure Procedure

The parameters of the drive mechanism of the test apparatus were set such that the platform had a pitch of 45° from the horizontal at a repetition rate of 20 times a minute. This pitch and repetition rate produced a smooth transfer of the seawater and bottom material from one end of the test chamber to the other.

The approximate average velocity of the seawater as it washed past the specimen was determined by consideration of the time it took for the given volume of seawater to pass through the cross sectional area defined by the width of the test chamber and the depth of the seawater when the chamber was at rest. Admittedly, the value of the velocity obtained, 0.9 ft/sec, is very crude since there is some turbulence within the chamber when it is in motion and the cross sectional area does not remain constant during a cycle.

Three specimens of Haynes-25 were studied, one in each of the three test chambers. A mixture of 500 ml of seawater and 50 g of ocean-bottom material was placed in two of the chambers. The third chamber, used as a control, contained 500 ml of seawater. After addition of the seawater and bottom material, the pyrex window assembly was screwed in place on the top of the test chambers and the apparatus set in motion. At the end of a sampling period, nominally 7-14 days, the apparatus was stopped and the contents of each test chamber removed and assayed for cobalt-60 activity.

F. Counting Procedure

Analysis for cobalt-60 in the seawater and seawater-bottom material mixture was made by direct counting in a gamma pulse height analyzer system. The amount of cobalt-60 present in the seawater and seawater-bottom material mixture was determined by comparison with standard samples (seawater and seawater-bottom material mixture) containing known amounts of cobalt-60. The amount of cobalt found in the samples

was converted to the corresponding amount of Haynes 25 by use of the known cobalt content of Haynes-25.

Counting errors for the erosion samples were such that a deviation of one sigma was less than 0.5 %. For the control samples the one-sigma deviations were on the order of 40 % due to much lower counting rates.

RESULTS AND DISCUSSION

Figure 3 shows the amount of Haynes-25 removed as a function of exposure time (in mils per year (mpy)) due to the erosive-corrosive action of a mixture of seawater and ocean-bottom material. Each point represents the average of two replicate determinations averaged over a sampling period; the vertical lines indicate average deviation. Although the scatter of the points is large, there seems to be a trend toward lower values of the erosion-corrosion effect at longer times. The important factor to note, however, is the value of the erosion-corrosion rate, nominally 1.4×10^{-2} mpy. This value is about 4 orders of magnitude greater than the 4×10^{-6} mpy observed for the control specimen. Table 1 shows a comparison of the corrosion rates obtained in this work with those obtained previously.

A combination of three possible effects is responsible for the increased rate of metal deterioration in the seawater-bottom material erosion-corrosion system. The first is the increase in the surface area of the specimens (corrosion rates reported here are based upon geometric surface area). The surface profile of the specimens before and after exposure to the seawater-bottom material system is shown in Fig. 4. It can be seen that the surface profile is significantly rougher after exposure. The surface area effect was found to account for a factor-of-10 increase in the corrosion rate (see a and c in Table 1). The second effect is the removal of any corrosion-protective surface film from the specimen by the erosive action of the seawater-bottom material system. This exposes a continuously fresh metal surface to the seawater; hence an increase in the corrosion rate would be expected (see b in Table 1). The third effect is the "wearing away" of the metal by physical abrasion by the seawater-bottom material system (see b in Table 1).

Fig. 3 Erosion-Corrosion Rate of Haynes-25 in a Moving Mixture of Seawater and Ocean-Bottom Material.

TABLE 1

Comparison of Haynes-25 Corrosion Rates in Seawater

Exposure Conditions	Method	Corrosion Rate (mpy)	Reference
6°C	Radioactive tracer	1×10^{-6}	8
6°C	Radioactive tracer	3×10^{-6}	11
Room temperature	Electrochemical	3.5×10^{-3}	7
90°C	Electrochemical	2.6×10^{-3}	9
a. Room temperature, seawater washing over specimen	Radioactive tracer	4×10^{-6}	this work
b. Room temperature, seawater and bottom material washing over specimen	Radioactive tracer	1×10^{-2}	this work
c. Room temperature, seawater washing over specimen which had previously been exposed to both sea- water and bottom material washing over specimen	Radioactive tracer	4×10^{-5}	this work

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Fig. 4 Representative Surface Profile of Haynes-25 Before and After Exposure to Erosion-Corrosion Environment.

It is of particular significance to compare the corrosion rates measured by electrochemical methods to those measured by radioactive tracer methods (Table 1). The fact that the erosion-corrosion rate is in agreement with the electrochemically determined corrosion rate (taking into account the above-mentioned surface area effect) and not with the rates determined by radioactive tracer methods suggest that the same mechanism (i.e., removal of protective film, or loss of passivity) may be responsible for the apparent disagreement between the corrosion rates determined by these two methods. This suggestion has strong support from the fact that when metals such as chromium, stainless steels and passive iron (chromium is one of the major components of Haynes-25) are cathodically polarized, passivity is destroyed.¹² This occurs by reduction of the passive oxide or chemisorbed oxygen film. In addition, hydrogen atoms, discharged on transition metals (e.g., cobalt and chromium) during cathodic polarization, tend to dissolve to a certain extent within the metal. The hydrogen so dissolved is partly dissociated into protons and electrons. These electrons preclude the metal from chemisorbing oxygen or becoming passive by filling vacancies in the d band of the transition metal. If this explanation is valid, it would indicate that the third possible effect for metal deterioration mentioned above, i.e., physical abrasion, would not be an important cause for erosion in an erosive ocean environment.

Because of the above considerations, it is felt that corrosion rates determined by electrochemical means represent an upper limit.

CONCLUSIONS

The results obtained in this study show that if the fuel container of a SNAP unit is accidentally placed in an ocean-bottom environment which can cause erosion (scouring) effects, such as high current velocities which can wash ocean-bottom material over the container, the corrosion rate of the fuel container material can be significantly increased. The extent of this increase, of course, will be dependent on several factors such as current velocity, type of bottom material and bottom terrain.

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