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A Limited Evaluation of the Corrosion
of Uranium and Weapon-Associated Metals
in Substitute Ocean Water

(Title Unclassified)

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ABSTRACT

Corrosion specimens of uranium, aluminum, beryllium, and stainless steel were exposed alone and in various galvanic couples to 'substitute' ocean water for 31 days at 75°F.

The rate of evolution of gases was monitored, and representative gases were analyzed. Hydrogen and air were present in every case. The relationship between the corrosion and quantity of evolved gases remains obscure.

Uranium was attacked at a rate of 0.3 to 1.0 mg/in.²/day. This is about fiftyfold less than rates reported for plutonium in sea water. Deep pitting occurred in the cases of uranium exposed alone or in contact with stainless steel to substitute ocean water or to distilled water. Penetration rates of about 1/8-in./year were recorded.

The corrosion rate of uranium was reduced and the type of corrosion altered when uranium was galvanically coupled to aluminum, beryllium, or beryllium plus stainless steel. The attack became much less localized and changed to the formation of a general scale or coating. Coupling uranium with stainless steel did not affect the rates for either metal, but it may alter the mechanism as considerably more gases were evolved.

Aluminum, beryllium, and stainless steel by themselves

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corrode at much lower rates than uranium, viz., 0.01 to 0.02 mg/in.²/day.

The anodic and cathodic couples and the approximate factors of increase or decrease in corrosion rates were:

<u>Couple</u>	<u>Anodic Member</u>	<u>Cathodic Member</u>
U-Al	Al(4)	U(0.5)
Be-SS	Be(14)	SS(0.5 to 1)
U-Be	Be(2)	U(0.5)
U-SS	?	?
U-Be-SS	Be(13)	U(0.4)

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I. INTRODUCTION

A limited study of the corrosion of uranium by "substitute" ocean water, synthesized in the laboratory, was conducted. This study was performed with uranium specimens alone and in contact with other metals with which uranium might be associated in a weapon. Consequently, the corrosion of these metals - aluminum, beryllium, and stainless steel - in substitute ocean water was also studied. The evaluations were made with the possibility in mind of furnishing data which would allow the assessment of the criticality hazard arising from accidental exposure of uranium weapons or devices to sea water. A related study of plutonium corrosion in sea water has appeared in AWRE Report No. O-11/63 by H. C. Barlow and J. F. Sackman.

II. EXPERIMENTAL TECHNIQUES

A. Materials and Construction of Specimens


Uranium coupon-type specimens were exposed by them-

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selves in both the unwelded condition and with a transverse weldment. Coupons of aluminum, beryllium, and stainless steel were all in the unwelded condition. All of the coupons were machined from hemishells of uranium, aluminum, beryllium, or AISI 304 stainless steel which had been fabricated originally as parts for various weapons or devices. Hemishells of nearly equal diameter were selected so that coupons of each of the metals would fit snugly against coupons of the others. Composite specimens were constructed by bolting together coupons of uranium and aluminum; uranium and beryllium; uranium and stainless steel; beryllium and stainless steel; and uranium, beryllium, and stainless steel.

The coupons were 2.0 in. long, 0.50 in. wide, and of the following thicknesses: uranium, 0.040 in.; aluminum, 0.281 in.; beryllium, 0.082 in.; and stainless steel, 0.029 in. The coupons were sawed, sheared, or turned oversize, and then the edges were shaper finished or milled to the desired dimensions at as low a working temperature as possible to minimize edge effects. A 1/4-in. hole was drilled near one end of each specimen and 1/4-20 nylon machine screws with nylon nuts held the composite specimens tightly together at one end. This simulated the galvanic coupling and the crevices between shells that would be present under normal conditions. Single specimens were also equipped with a nylon screw and nut so that any unpredicted effect arising from the use of nylon would be imposed equally upon every specimen and would be, in effect, cancelled out.

The welded coupons were machined from a hemishell that had previously received a single latitudinal weld bead. This was welded by members of Group CMB-6 using the

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tungsten arc-inert gas process with a nonconsumable electrode. No filler metal was added.

B. Substitute Ocean Water

The substitute ocean water used for the exposures was prepared according to ASTM Designation D 1141-52. The final composition was calculated to be that shown in Table 1. The final mixing and pH adjustment required vigorous stirring and dissolved an appreciable quantity of air. Therefore, the solutions were allowed to stand for 4 days in their individual beakers before the exposures were started so that some of this air might be expelled.

C. Method of Exposure

Graduated cylinders (50 cc) were filled to the top with the substitute ocean water and each carefully inverted and placed in a beaker completely filled with solution, with the mouth just slightly above the bottom of the beaker. Each metal coupon, which had been carefully cleaned, dried, measured, and weighed, and assembled into corrosion specimens, was inserted into the open mouth of a graduated cylinder so that the tightly bolted ends rested on the beaker's floor and the opposite ends - and crevices in the case of composite specimens - extended upward into the cylinder. Each beaker and cylinder contained approximately 900 cc of substitute ocean water. During the introduction of the specimens care was taken to wet all of the metal surfaces and shake the air bubbles from crevices before placing the specimens beneath the graduated cylinders.

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TABLE 1
 CALCULATED CHEMICAL COMPOSITION OF SUBSTITUTE OCEAN
 WATER PREPARED ACCORDING TO ASTM D 1141-52

Compound	Concentration, g/liter
NaCl	24.41
MgCl ₂	5.17
Na ₂ SO ₄	4.07
CaCl ₂	1.15
KCl	0.692
NaHCO ₃	0.200
KBr	0.101
H ₂ BO ₃	0.027
NaOH	0.015
SrCl ₂	0.025
NaF	0.003
Ba(NO ₃) ₂	0.0000989
Mn(NO ₃) ₂	0.0000338
Cu(NO ₃) ₂	0.0000307
Zn(NO ₃) ₂	0.0000096
Pb(NO ₃) ₂	0.0000066
AgNO ₃	0.00000049

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The beakers were covered with cork sheet cut to enclose the cylinder and stored in an air-conditioned room where the temperature remained within a few degrees of 75°F. The final solution levels at the start of the exposure were marked on the sides of the beakers, and periodically the solutions were restored to these levels by additions of distilled water. Very little evaporation occurred.

Every two days, or so, the cylinders were shaken to dislodge gas bubbles adhering to their walls or in the crevices of the specimen. Following this, the amounts of gases evolved and captured in the closed ends of the inverted cylinders were carefully recorded.

D. Final Specimen Treatment

After exposure, the corrosion coupons were removed from the solution, the nylon bolts were disassembled, and the coupons were cleaned by brushing with a stiff bristle brush beneath the surface of water. Examination of the cleaned specimens with a low-power microscope revealed considerable corrosion deposits still adhering to many of the coupons, especially inside pits. It was decided that some chemical cleaning of the corroded specimens would be necessary before the measured weight losses would be meaningful. The uranium coupons were cleaned by submerging them for 30 min in an aqueous 10 v/o H₂SO₄ solution held at 44 to 54°C.* Similar uncorroded coupons were exposed simultaneously and weight losses of these coupons were in

* A cleaning method given by V. H. Troutner, Corrosion, 16, 283t (June 1960).

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the order of 0.1 to 0.2 mg - within the experimental error of weighing - so corrections were not necessary. This treatment cleaned the coupons of all but tightly adherent oxide scales which probably did not contribute greatly to the coupon's weight.

The coupons of the other materials were carefully cleaned and weighed. The corrosion products were stripped from the stainless steel coupons by immersing them in aqueous 10 v/o HNO_3 at 60°C for 30 min. Again, the correction of 0.1 mg on uncorroded specimens was neglected.

The aluminum specimens were given a 30-min soak in aqueous 5 v/o HCl at room temperature. It was necessary to scrub the loosely adhering corrosion products from the coupons following this treatment and also to apply a correction of 1.1 mg per coupon.

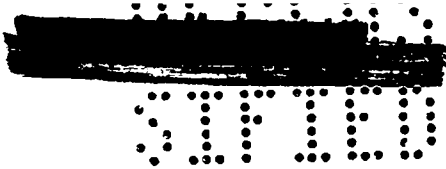
No good chemical cleaning method was found for the beryllium coupons which would not attack the uncorroded metal. Consequently the only cleaning these coupons received was the underwater brushing. However, microscopic examination showed no signs of localized attack nor any heavy deposits of corrosion products. All the coupons were rinsed in distilled water followed by chemically pure acetone, then air dried and weighed.

III. DISCUSSION OF RESULTS AND CONCLUSIONS

A. Gas Evolution

It was thought for some time that the amount of gas evolved was a good measure of the amount of reaction

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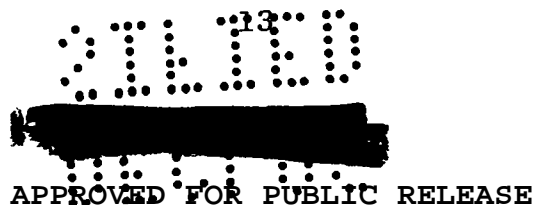
occurring. Plots of the gas evolution versus time are shown in Figures 1 and 2. Later it will be shown that the correlation between gas evolution and corrosion - especially that of uranium - is not as good as might be expected.

B. Uranium Coupons

The results of the weighings and microscopic examinations of the uranium coupons are presented in Table 2. The corrosion rates have been expressed as weight losses in terms of milligrams per square inch per day. These units can be converted to units of penetration expressed in mils per year - a common term in corrosion literature - by multiplying by 0.0929/specific gravity, or, in the case of uranium, by 0.00492. However, because of the large amount of localized attack in the form of pitting on many of the coupons, such values of average penetration are not very accurate. In fact, measurement of two of the deepest pits found - in this case on uranium coupons exposed to distilled water - gave dimensions, in one case, of 7 by 12 mils by 10 mils deep and, in another, 25 by 30 mils by 9 mils deep. This is approximately one-quarter penetration of the 40-mil sheet, or, roughly equivalent to complete penetration of 1/8-in. sheet in a year's time.

The highest dissolution rate observed for uranium was 1.000 mg/in.²/day. This occurred on a uranium coupon coupled to stainless steel. For comparison purposes, the dissolution rate for plutonium exposed to sea water at 30°C - a slightly higher temperature - was 1.1×10^{-5} g/cm²/min.*

*Barlow, H. C., and Sackman, J. F., AWRE Report No. 0-11/63, p.4.



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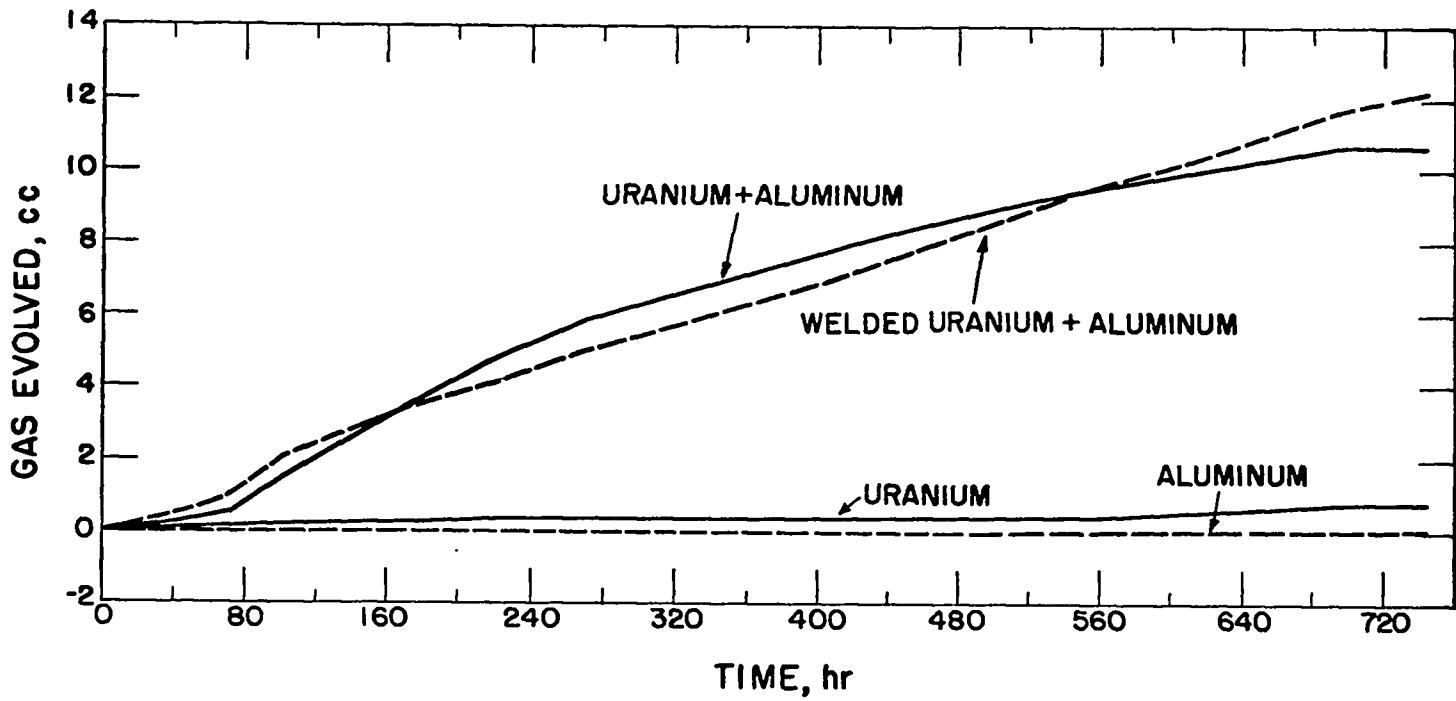


FIGURE 1. GAS EVOLUTION VS. TIME FOR THE URANIUM - ALUMINUM SYSTEM

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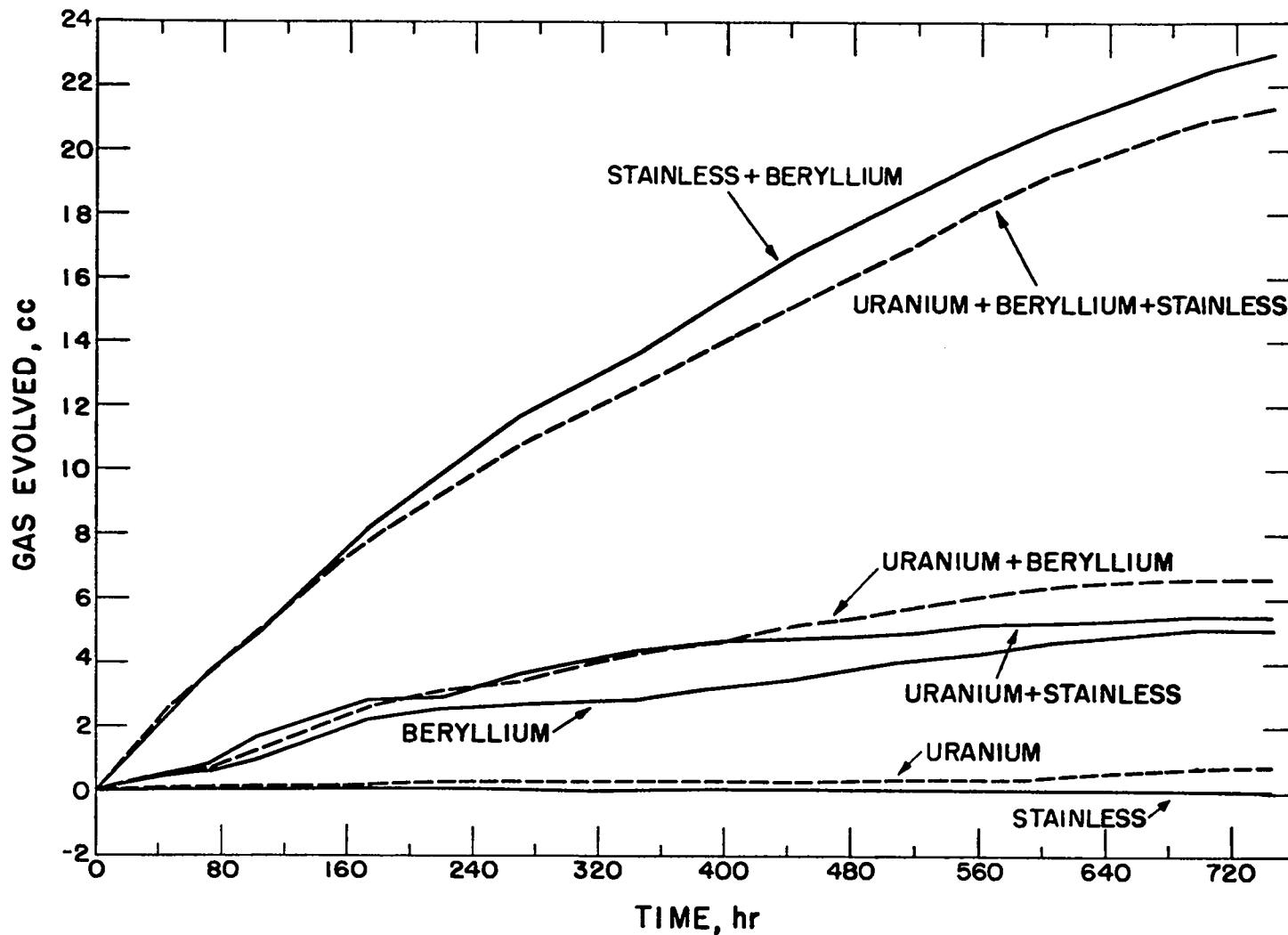


FIGURE 2. GAS EVOLUTION VS. TIME FOR THE URANIUM-BERYLLIUM-STAINLESS STEEL SYSTEM

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TABLE 2
CORROSION RATES AND DESCRIPTIONS OF URANIUM SPECIMENS
EXPOSED TO SUBSTITUTE OCEAN WATER

Specimen	Coupled with:	Weight Loss, mg	Corrosion Rate mg/in. ² /day	Gas Evolved μ l/day	Description of Specimens After Exposure
U3	---	39.6	0.636	29.)	dark oxide coating, several pits, some deep
U4	---	43.0	0.689	23.)	
W1*	---	33.4	0.535	16.)	
W2*	---	61.0	0.977	19.)	moderate oxide coating, large areas of shallow pits, few deep pits, weldments unattacked
U5	Al	23.3	0.373	313.)	
U6	Al	25.3	0.403	377.)	heavy oxide coating, few shallow pits
W3*	Al	16.5	0.262	419.)	
W4*	Al	11.7	0.187	368.)	heavy oxide coating, no pitting, weldments unattacked
U7	Be	19.8	0.315	206.)	
U8	Be	23.5	0.374	226.)	heavy black scale, no pitting
U9	SS**	62.5	1.000	187.)	
U10	SS	60.4	0.964	168.)	moderately heavy oxide coating, large areas of shallow pits, few deep pits
U11	Be+SS	20.9	0.334	700.)	
U12	Be+SS	13.0	0.210	677.)	heavy black scale, no pitting
U1***	---	56.4	0.919	6.)	
U2***	---	60.1	0.963	10.)	many deep pits, some measured 9 to 10 mils deep


* Specimen with transverse weldment

** SS= Stainless steel (AISI 304)

*** Exposed to distilled water only

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This converts to 102 mg/in.²/day or a factor of 100 more soluble. Extrapolating the rates for plutonium to 24°C to compare with our figures would still show plutonium to be roughly fiftyfold more soluble than uranium.

Also included in Table 2 is a column giving values for the average gas evolution in units of microliters per day. It can be quickly ascertained that very little correlation exists between the uranium weight losses and the gas evolution although either value has been duplicated quite well within the duplicate exposures. This indicates that more than one mechanism of corrosion may be operative.

The important conclusions to be drawn from these results concerning the corrosion of uranium are:

- (1) There is little difference between the corrosion rates for unwelded and welded uranium in substitute ocean water.
- (2) Uranium weldments are not preferentially attacked either in the weld bead or in the heat-affected zone.
- (3) Localized attack in the form of shallow and deep pits occurs with great frequency on uranium exposed alone or in contact with stainless steel to substitute ocean water.
- (4) Pitting and weight losses of plain uranium coupons are as large or larger with distilled water than with substitute ocean water.
- (5) Coupling uranium with aluminum, beryllium, or beryllium plus stainless steel appears to afford galvanic protection to the uranium, thereby lowering its corrosion rate. Conversely coupling uranium to stainless steel did not afford this protection and may have promoted the corrosion slightly.

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- (6) Not only was the weight loss diminished by the galvanic couples mentioned above, but also the form of attack was altered so that fewer pits and more black scale formed.
- (7) There is no direct correlation between the volume of gas evolved and the amount of attack occurring on the uranium.

C. Composite Specimens

In Table 3, the weight losses of all of the coupons are tabulated, expressed as micromoles. The total micromoles of metal dissolved is shown in the seventh column, and the number of hydrogen microequivalents of the metal in the eighth. These are based on the following equivalences: U/3, Al/3, Be/2, and SS/3, with a synthetic atomic weight of 55.39 being used for 18 w/o Cr - 8 w/o Ni stainless steel.

In column 10 the total amount of gas evolved has been converted, arbitrarily, to microequivalents of hydrogen using 24374 cc as the molar volume of hydrogen at 75°F. Column 11 then contains the ratio of column 8 to column 10. If the corrosion mechanism consisted simply of the metal atoms ionizing and liberating hydrogen as they enter solution, this ratio should be unity - taking into consideration small deviations caused by dissolved air leaving solution or hydrogen bubbles escaping the confines of the graduated cylinder. Many ratios are in the neighborhood of unity; but ratios from some specimens, especially uranium alone, are much higher, indicating the amount of gas expected from the weight loss never materialized. In this case, the corrosion mechanism either is different or

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TABLE 3
COMPARISONS OF METAL WEIGHT LOSSES WITH EVOLVED GASES

Beaker	Specimen	Metal Weight Losses, μ-moles				Total	Metal H ₂ μ-equiv- alent	Evolved Gas, cc	Gas H ₂ μ-equiv- alent	Ratio Metal Gas
		U	Al	Be	SS					
1	U*	237	---	---	---	237	711	0.2	16	43
2	U*	252	---	---	---	252	757	0.3	25	31
3	U	166	---	---	---	166	499	0.9	74	7
4	U	181	---	---	---	181	542	0.7	57	9
21	U-welded	140	---	---	---	140	421	0.5	41	10
22	U-welded	256	---	---	---	256	769	0.6	49	16
5	Al	---	74 (0.019)**	---	---	74	222	0.0	---	---
6	Al	---	67 (0.018)	---	---	67	200	0.0	---	---
13	U+Al	98	278 (0.073)	---	---	376	1128	9.7	796	1
14	U+Al	106	315 (0.083)	---	---	421	1264	11.7	960	1
23	U-welded+Al	69	300 (0.079)	---	---	369	1109	13.0	1067	1
24	U-welded+Al	49	256 (0.067)	---	---	305	914	11.4	935	1
7	Be	---	---	44 (0.006)	---	44	89	5.1	419	0.2
8	Be	---	---	gain	---	---	---	5.0	410	---
9	SS	---	---	---	7 (0.007)	7	22	0.1	8	3
10	SS	---	---	---	9 (0.009)	9	27	0.1	8	3
11	Be+SS	---	---	577 (0.078)	2 (0.002)	579	1154	23.0	1887	0.6
12	Be+SS	---	---	610 (0.083)	7 (0.007)	617	1220	23.1	1896	0.6
15	U+Be	83	---	111 (0.015)	---	194	472	6.4	525	0.9
16	U+Be	99	---	89 (0.012)	---	188	474	7.0	574	0.8
17	U+SS	263	---	---	5 (0.005)	268	804	5.8	476	2
18	U+SS	254	---	---	5 (0.005)	259	777	5.2	427	2
19	U+Be+SS	88	---	632 (0.086)	4 (0.003)	724	1539	21.7	1781	0.9
20	U+Be+SS	55	---	499 (0.068)	7 (0.007)	561	1184	21.0	1723	0.7

* Exposed to distilled water, all others exposed to substitute ocean water.


** Numbers in parentheses are corrosion rates in mg/in.²/day.
See Table 2 for comparable values for uranium.

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- (5) Galvanic coupling of beryllium with stainless steel results in little change in the corrosion rate of the stainless, but the corrosion rate of the beryllium increases about fourteenfold to a value of 0.08 mg/in.²/day. Considerable quantities of gases are evolved.
- (6) Galvanic coupling of beryllium with uranium results in the corrosion rates of beryllium being about doubled while those of uranium are halved.
- (7) Stainless steel coupled to uranium apparently has little effect on the corrosion rate of either metal; but the quantity of evolved gases is greatly increased, indicating a possible change in mechanism.
- (8) The galvanic coupling of uranium, beryllium, and stainless steel results in a reduction in the corrosion rate of uranium by a factor of 2 to 3, about a thirteenfold increase in the corrosion rate of beryllium, and little change in the rate for stainless steel. A considerable quantity of gas is evolved similar to the case when beryllium and stainless are coupled.

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(9) The anodic members of the couples are:

<u>Couple</u>	<u>Anodic Member</u>
U-Al	Al
Be-SS	Be
U-Be	Be
U-SS	?
U-Be-SS	Be

D. Analyses of Evolved Gases

In the cases where sufficient quantities were collected during the exposures, the gases were transferred from the graduated cylinders to evacuated gas sample bulbs and then analyzed with the CEC-620 mass spectrometer.

The gas analyses showed hydrogen and air to be present in all cases. There was no clear evidence that any component of air such as oxygen or nitrogen was preferentially consumed by any of the corrosion processes.

Although precautions against accidental introduction of air were taken during the sample transfer, and in most cases, there was no reason to think that air was introduced, this is a possible source. More probably, the source of the air was that which slowly came out of solution or that which was entrapped in the crevices of composite specimens.

All of the gases were saturated with water vapor, and this may have influenced the analyses. However, since

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they appear to be hydrogen and air, exact compositions would not elucidate the corrosion mechanisms.

E. Residual Solutions

In the same vein, the residual solutions were not analyzed for corrosion products. In the presence of so many interfering ions, the analyses would most certainly be difficult, and the information to be gained has not seemed to justify the expenditure of the effort required.

IV. SUMMARY

Welded and unwelded coupons of uranium; unwelded coupons of aluminum, beryllium, and AISI 304 stainless steel; as well as composite specimens composed of combinations of these metals as might occur in weapons or devices were exposed to "substitute" ocean water for 31 days at 75°F.

The evolution of gases from the specimens was followed at intervals, and many of the gases were analyzed after the exposure period was complete. Hydrogen and air were present in each. There were no good correlations between the degree of corrosion and the volume of gas evolved indicating that different corrosion mechanisms were controlling the reactions with different specimens.

Based upon the losses in weight of the various coupons

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and microscopic observations of their surfaces after exposure it was concluded that:

In the case of uranium, both unwelded and welded coupons were attacked by substitute ocean water. The attack was localized in the form of deep and shallow pits and generalized in the form of an oxide coating. Weldments were not preferentially attacked. Corrosion rates of 0.3 to 1.0 mg/in²/day were calculated, based on weight losses. Equally severe corrosion rates occurred upon exposure of uranium coupons to distilled water. Penetration rates of roughly 1/8-in./year were measured in pits on these coupons.

Coupling uranium with aluminum, beryllium, or beryllium plus stainless steel appears to afford galvanic protection to the uranium, thereby lowering its corrosion rate and altering the form of corrosion from localized attack to formation of a more general scale or coating. Coupling uranium with stainless steel may increase the rate of corrosion slightly. The highest rate observed was still some fiftyfold lower than has been calculated for plutonium at the same temperature.

Aluminum, beryllium, and AISI 304 stainless steel, by themselves, corrode at much lower rates of 0.01 to

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0.02 mg/in.²/day. Coupling uranium with aluminum results in about a fourfold increase in the rate of aluminum corrosion while that of uranium is cut by a factor of 2 or 3.

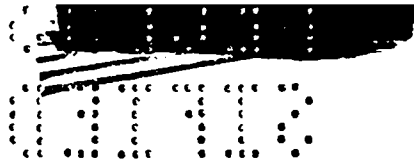
Galvanic coupling of beryllium with uranium doubles the rate for beryllium and halves the rate for uranium. Coupling beryllium with stainless steel does not greatly change the rate for stainless but increases the rate of beryllium about fourteenfold. Coupling uranium with stainless steel does not affect the rates for either metal but may alter the mechanism as considerably greater quantities of gases are evolved. Coupling all three metals, viz., uranium, beryllium, and stainless steel, results in a reduction in the corrosion rate of uranium by a factor of 2 to 3, a thirteenfold increase in the corrosion rate of beryllium, and little change in the rate for stainless steel. A considerable volume of gas is evolved from this combination, being of the same order as evolved by the stainless steel-beryllium couple.

The anodic and cathodic couples and the approximate factors of increase or decrease in corrosion rates of their members, as disclosed by this

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evaluation, are summarized below:

<u>Couple</u>	<u>Anodic Member</u>	<u>Cathodic Member</u>
U-Al	Al (4)	U (0.5)
Be-SS	Be (14)	SS (0.5 to 1)
U-Be	Be (2)	U (0.5)
U-SS	?	?
U-Be-SS	Be (13)	U (0.4)

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