Program: Sodium Technology

DOE Task: 27-Sodium Removal and Decontamination of Components

Program Manager: R. W. Keaten

Project Manager: L. Baurmash

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Category: HN-10-04-01.5

General Order: 09286

Subaccount: 36360-36380

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I. PROJECT OBJECTIVES

This project has several tasks, each with its own specific objectives. One of these tasks is the Sodium Removal Process Development. The first objective of this task was to develop and evaluate techniques for sodium (Na) removal from components by the reaction of sodium with alcohol. The culmination of this program was the incorporation of the Sodium Removal Process in the DOE Standard F 5-9, and a Topical Report presenting the pertinent sodium-alcohol reaction rate data. The second objective of this task is to determine the process parameters for the removal of Na from test specimens and components by the use of evaporation techniques.

A second task of this project is the Decontamination Process Development. The objective of this effort is to develop a well-characterized chemical decontamination process for application to the LMFBR primary system components that subsequently permits contact maintenance and allows requalification of the components for reuse in reactors.

A new task, decontamination by sputtering, was initiated last fiscal year. The objective of this task is to determine the feasibility of using argon ion sputtering as a method for removing diffused radioisotopes from LMFBR components.

Another new task, Metal Removal Studies on Type 316 Stainless Steel, was initiated last fiscal year.

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II. MAJOR ACCOMPLISHMENTS DURING FISCAL YEAR 1978

The sodium evaporation apparatus for cleaning failed fuel pin assemblies was built and checked out. A simulated failed fuel pin assembly was made.

Metallographic examination of mechanically tested tensile specimens was completed. It was concluded that the Evaporative Removal of Sodium (ERNA) Process causes no deleterious effects on Type 304 stainless steel, even though many of the specimens were exposed to conditions that would not be encountered during normal processing.

A Test Report, N707TR830013, "Evaporative Removal of Sodium: Stressed Type 304 Stainless steel Tensile Specimens," was issued.

A Test Report, N707TR810012, "Evaporative Removal of Sodium from the IHX Tube Bundle Mockup," was issued. It was concluded, based upon this successful demonstration test and the previously generated parametric data on variously sized crevice specimens* that the Evaporative Removal of Sodium (ERNA) Process is extremely promising for the cleaning of both small and large Na-wetted components.

Decontamination studies consist of the following:

- 1) Development of a short-term sensitization method for Type 316 stainless steel (AI-DOE-13220).
- 2) Initiation of traveler study (AI-DOE-13220).
- 3) A method of restarting a decontamination run that has been individually contaminated with oxygen was evaluated. By thoroughly rinsing the component and replacing the contaminated GCA solution with fresh solution, the original metal removal rate can be obtained (AI-DOE-13220 and this report).
- 4) The rate of metal removal of both wrought metal and castings of Type 316 stainless steel was evaluated using the GCA process at temperatures up to 90°C. The rate is too low to be practical unless the metal removal requirements is lower than that for Type 304 stainless steel (AI-DOE-13220).

*AI-DOE-13208

- 5) Because of the significance of the results of Item (4) above, a deposition loop run was started to allow the evaluation of ^{54}Mn diffusion during long-term sensitization of Type 316 stainless steel (this report).
- 6) Evaluation of a Polarographic Oxygen Monitor (this report).
- 7) The effect of carbon in Type 304 stainless steel on the deposition of ^{54}Mn in a sodium loop was reported for the first time (this report).
- 8) A study of glow discharge sputtering in argon as a means of decontamination of LMFBR components was continued (AI-DOE-13220 and this report).

III. PROGRESS DURING REPORT PERIOD

A. ALCOHOL PROCESS DEVELOPMENT

No work is funded for this activity in FY 1978.

B. SODIUM EVAPORATION PROCESS DEVELOPMENT

1. Simulated Failed Fuel Pin Assembly

Using the CRBR Fuel Assembly as a guide (Figure 1), a simulated failed fuel pin assembly was fabricated (Figure 2). The assembly consists of a Calrod type heater in a central tube, surrounded by 8 spiral wirewrapped fuel tubes and one heater lead-return tube. One of the dummy fuel tubes has a small slot milled through the wall to simulate a cracked tube.

Figure 1. CRBR Fuel Assembly

Figure 2. Simulated Failed Fuel Pin Assembly (3-ft Length)
for Sodium Removal Tests

The assembly has been wetted with sodium at 565°C (1050°F) and stored under vacuum until start of testing.

2. Test Apparatus Assembly

The test apparatus for the removal of sodium from the simulated failed fuel pin assembly has been constructed.

The apparatus consists of:

- 1) A heated argon supply system
- 2) An evaporation chamber that contains the fuel pin assembly
- 3) A water-cooled argon exit line
- 4) A scrubber to remove sodium particles and vapor
- 5) A large Stokes vacuum pump to rough pump the system
- 6) A Turbomolecular pump (1600 liters/s) to evacuate the system to 1.3×10^{-3} Pa (10^{-5} torr) or less
- 7) Associated controllers, recorders, flowmeter, vacuum gauges, etc.

The system checkout is nearly complete and has proceeded smoothly, except for operation of the Turbomolecular pump, which required the services of the vendor service technician.

The test procedure, "Evaporative Removal of Sodium-Test Procedure for Sodium-Wetted Fuel Assemblies," N707DTP830013, was issued.

C. DECONTAMINATION STUDIES

The major activities during the quarter were:

- 1) Statistical study of the effect of carbon content of Type 304 stainless steel on ^{54}Mn deposition.
- 2) Evaluation of the reproducibility of the metal removal rate from Type 304 stainless steel using the GCA process
- 3) Evaluation of a Polarographic type oxygen monitor for the GCA process

4) Initiation of a deposition test using Type 316 stainless steel castings and wrought stock

5) Continuation of sputtering experiments.

1. Statistical Study - Effect of Carbon Content of Type 304 Stainless Steel on ^{54}Mn Deposition

A statistical study was made of the ^{54}Mn activity on some of the specimens taken from the bottom holder in the hot leg of the deposition loop.

The hot leg specimens removed from the loop had accumulated 4700 h at the specified temperature range and had a bright metallic appearance with grain grooving. Most of the hot leg specimens consisted of two heats, 74966 (0.08%C) and 55181 (0.03%C). They were in two separate specimen holders in the hot leg, one at the bottom and one at the top. Sodium entered at the bottom and the activity level of both heats was highest in this holder. The specimens were randomly distributed in each holder.

About two-thirds of the specimens from the bottom holder of the hot leg of the deposition loop have been counted with a 3-by-3-in. sodium iodide crystal sensor. This much counting was done to see if there is a real difference between the amount of ^{54}Mn absorbed by a low-carbon heat of Type 304 stainless steel and a high-carbon heat which have otherwise similar metallurgical and chemical characteristics. The bottom holder specimen was analyzed because these specimens have a higher level of radioactivity.

The distribution of the activity of equivalent specimens for both the high- and low-carbon heats is shown in Figure 3. The low-carbon specimens have a narrow distribution except for two specimens, which distort the distribution curve to the right. The high-carbon specimens show a much greater dispersion of activity values, but these also appear to be mildly distorted to the right.

Tables 1 and 2 give statistical data for the high- and low-carbon specimens, respectively. These values might be different if all specimens had been counted. Table 3 gives the analysis of the data according to the "t" test.

The results shown in Table 3 indicate that there is a real difference in the activity level of the high- and low-carbon specimens.

The total chemical analysis in weight percent of the two samples is given as follows:

Figure 3. Frequency Distribution of Radioactivity, Specimens from Hot Leg, Bottom Holder

TABLE 1
STATISTICAL DATA

[Hot Leg Specimens (1000 +50°F), Bottom Holder,
Heat 74966, 0.08%C, Type 304 Stainless Steel]

Specimen No.	Count/Min	f	$\bar{X} - x$	$(\bar{X} - x)^2$
28	16033	3	-1777	3.16×10^6
29	13548	7	-706	0.50×10^6
30	16624	3	+2368	5.61×10^6
31	15276	3	+1020	1.04×10^6
32	13848	M	-408	0.17×10^6
33	13691	7	-565	0.32×10^6
34	11392	3	-2864	8.23×10^6
35	12557	7	-1699	2.89×10^6
37	11821	3	-2435	5.93×10^6
38	14637	3	+381	0.15×10^6
39	19342	H	+5086	25.87×10^6
40	13495	7	-761	0.58×10^6
41	14078	3	-178	0.03×10^6
42	11312	L	-2944	8.67×10^6
44	15547	3	+1291	1.67×10^6
45	13378	7	-908	0.82×10^6
46	12751	7	-1505	2.27×10^6
47	17258	3	+3002	9.01×10^6
N = 18	$\bar{X}_H = 14256$		$S_H = 2127$	

f = frequency of occurrence based on intervals of 2000 (6-8, 8-10, etc.)

M = median H = high L = low

$\bar{X}_H$ = average of all measurements

x = individual measurement

S_H = standard deviation

N = number of specimens counted

TABLE 2
STATISTICAL DATA

[Hot Leg Specimens (1000 ± 50°F), Bottom Holder
Heat 55183, 0.03%C, Type 304 Stainless Steel]

Specimen No.	Count/Min	f	$\bar{X} - x$	$(\bar{X} - x)^2$
23	12573	1	+2359	5.56×10^6
24	8571	13	-1643	2.70×10^6
25	9325	13	-889	0.79×10^6
26	12004	3	+1790	3.20×10^6
27	15471	H	+5257	27.64×10^6
28	9824	13	-390	0.15×10^6
30	9524	13	-690	0.48×10^6
34	9632	13	-582	0.39×10^6
35	9012	13	-1202	1.44×10^6
37	9863	13	-351	0.12×10^6
38	9915	13	-299	0.09×10^6
39	9884	13	-330	0.11×10^6
42	9878	M	-336	0.13×10^6
43	8922	13	-392	0.15×10^6
44	8794	13	-1420	2.02×10^6
46	10316	3	+102	0.01×10^6
49	8584	L	-1530	2.34×10^6
50	11775	3	+1561	2.44×10^6
N = 18	$\bar{X}_L = 10214$		$S_L = 1711$	

f = frequency of occurrence based on intervals of 2000 (6-8, 8-10, 10-12, etc)

M = median H = high L = low

$\bar{X}_L$ = average of all measurements

x = individual measurement

S_L = standard deviation

N = number of specimens counted

TABLE 3
DATA ANALYSIS FOR SIGNIFICANCE

The assumption is made that there is a real difference in radioactivity between the low- and high-carbon specimens after 4700 h in the deposition loop.

Then using the "t" test,

$$t = \frac{\bar{X}_H - \bar{X}_L}{\left(S_L^2/N + S_H^2/N \right)^{1/2}} \quad \dots (1)$$

$$\text{d.f.} = (N + 1) \left(1 + \frac{2}{\frac{S_L^2}{S_H^2} + \frac{S_H^2}{S_L^2}} \right) - 2 \quad \dots (2)$$

where

d.f. = degree of freedom

$$t = \frac{14,256 - 10,214}{\left[\frac{(1711)^2}{18} + \frac{(2127)^2}{18} \right]^{1/2}} = \frac{4042}{(1.63 \times 10^5 + 2.51 \times 10^5)^{1/2}} = 6.3$$

$$\text{d.f.} = 19 \left[1 + \frac{2}{(0.65 + 1.54)} \right] - 2 = 34.3$$

For a d.f. of 34 a "t" value of over 1.7 is significant at the 95% confidence level

	<u>C</u>	<u>Cr</u>	<u>Ni</u>	<u>Mn</u>	<u>Si</u>	<u>P</u>
74966	0.08	18.47	8.65	1.60	0.66	0.023
55183	0.03	18.25	9.92	1.83	0.51	0.022

Nickel is higher in the low-carbon heat, but this would be expected to increase ⁵⁴Mn pickup, if any change would be expected.

Since carbon appears to be the only element to which the difference can be attributed, it appears that the pickup of ⁵⁴Mn by Type 304 stainless steel from sodium at 1000°F (538°C) increases as the carbon content of the metal increases.

2. Evaluation of Metal Removal Rate – Reproducibility Using the GCA Decontamination Process

Several objectives were established for the major metal removal tests run this period:

- 1) To evaluate the reproducibility of metal removal rates between two specimens from the same heat in the same container. This was done for both a high-and low-carbon heat.
- 2) To evaluate the reproducibility of metal removal rates using the GSC process at 70°C and 80°C with two different reaction containers. Two different metal heats were used, high and low carbon.
- 3) To compare the metal removal rate for specimens sensitized for 1 h at 1250°F (677°C) with the rate for specimens from the same heat sensitized in the sodium deposition loop for 4700 h at 1000° ±50°F (538°F ±28°C). Two heats were used, a high and low carbon.
- 4) To measure the decontamination achieved by the GCA process using high-and low-carbon specimens from the hot leg of the deposition loop.

The two heats of Type 304 stainless steel which have become the main standards for this program are McLouth 74966 (0.08%C) and J and L 55183 (0.03%). They are designated as HC and LC, respectively, in Table 4 which summarizes the major metal removal results for this report period.

These results show that excellent reproducibility is possible using the GCA process for equivalent specimens either within the same container or in different containers. A maximum 10% variation in metal removal is shown at 70°C,

TABLE 4
METAL REMOVAL AND DECONTAMINATION RESULTS

	Metal Removal in mils (microns)/day at 70°C				Decontamination, Activity in Counts per minute, of ⁵⁴ Mn			
	Container 1		Container 2		Container 1		Container 2	
	Specimen 1	Specimen 2	Specimen 1	Specimen 2	Original Activity	After Decon	Original Activity	After Decon
HC, Sensitized 1250°F, 1 h	0.024 (0.61)	0.027 (0.69)	0.025 (0.64)	0.026 (0.66)	-	-	-	-
LC, Sensitized 1250°F, 1 h	0.021 (0.53)	0.020 (0.51)	0.023 (0.58)	0.021 (0.53)	-	-	-	-
HC, Sensitized in Loop, 1000°F, 4700 h	0.037 (0.94)	-	0.036 (0.91)	-	13550	1371	13690	1950
LC, Sensitized in Loop, 1000°F, 4700 h	0.026 (0.66)	-	0.024 (0.61)	-	9824	2342	12573	2941

	Metal Removal in mils (microns)/day at 80°C				Decontamination, Activity in Counts per minute, of ⁵⁴ Mn			
	Container 1		Container 2		Container 1		Container 2	
	Specimen 1	Specimen 2	Specimen 1	Specimen 2	Original Activity	After Decon	Original Activity	After Decon
HC, Sensitized 1250°F, 1 h	0.041 (1.04)	0.049 (1.24)	0.036 (0.91)	0.037 (0.94)	-	-	-	-
LC, Sensitized 1250°F, 1 h	0.041 (1.04)	0.041 (1.04)	0.035 (0.89)	0.037 (0.94)	-	-	-	-
HC, Sensitized in Loop, 1000°F, 4700 h	0.082 (2.08)	-	0.079 (2.01)	-	13378	412	14951	414
LC, Sensitized in Loop, 1000°F, 4700 h	0.050 (1.27)	-	0.058 (1.47)	-	9325	996	8571	948

which would result in very much lower variation in percent decontamination, since the permeation profile of ^{54}Mn is quite flat beyond a depth of about 1/2 mil. The 80°C results also show excellent reproducibility. These results reinforce prior data showing that excellent control of metal removal is possible, using the GCA process, if the significant process parameters are properly controlled.

The data also offer additional confirmation of the relationship of metal removal to specimen time-temperature exposure. The loop samples show significantly higher metal removal rates than the short-term sensitized specimens, as expected. This relationship is the reason why it was considered necessary to include a requirement for travelers in the decontamination process specification.

Another metal removal test made during this period had the objective of attempting to recover from a run that had been inadvertently exposed to oxygen. It was hoped that by adding hydrazine to reduce the oxidized solution, it would be possible for metal removal to resume.

A regular metal removal test was run with the GCA process at 70°C and it was then terminated with a 1-day air sparge which resulted in the passivation of the specimens and the oxidation of the iron salts in the acid solution.

One-tenth mole of hydrazine was added which should have been sufficient to reduce the solution. The test was then run for an additional period of time with no evidence that metal removal would resume.

Since hydrazine is generally not effective as a reducing agent in acid solutions, its failure to reduce the solution was not entirely unexpected.

3. Evaluation of a Polarographic Type Oxygen Monitor for the GCA Process

The rate of metal removal that can be obtained with most acceptable acidic LMFBR decontamination process, including the GCA process, is critically dependent upon maintaining a very low oxygen concentration in the acid solution at all times. The actual oxygen concentration required for optimum metal removal by the GCA process is not yet known, but could be below 1 part per billion (ppb) by weight. Thus it is necessary to qualify an instrument capable of measuring oxygen in acidic solutions in this concentration range. Following this, the

instrument will be used to determine the maximum allowable concentration of oxygen in the GCA process using metal removal as a criterion of acceptability.

The instrument selected for evaluation is based on the polarographic principle and is made by Orbisphere Laboratories. This instrument was developed for measuring the oxygen concentration of boiler feedwater where an oxygen concentration of 1 ppb is a typical operating condition. Its lowest oxygen range is 0 to 10 ppb where it gives a continuous reading.

Another type of instrument is available based on a galvanic principle. It has a consumable electrode and does not have a 0 to 10 ppb range, so it seems to be less desirable for use in the decontamination process. However, it will be evaluated along with the polarographic type in the HEDL pilot plant.

Considerable difficulty has been experienced in developing an experimental system suitable for accurately measuring oxygen in the ppb range. Several of the difficulties experienced will be typical of plant operation, but many are peculiar to small-scale laboratory apparatus and will not appear in a well constructed decontamination plant.

Two of the difficulties which are likely to be found in a large plant are trapped air and stagnant bubbles of gas which have not reached the same oxygen concentration as the bulk of the acid solution. These bubbles become detached in an erratic manner and pass through the sensor circuit with attendant surges in the instrument output when operating in the low-concentration ranges.

The laboratory apparatus uses polypropylene containers in a water bath and it may be that some oxygen diffuses through the walls from the water bath. However, sparging the water bath with argon has not shown a significant effect in reducing the equilibrium level of oxygen that is reached. Glass containers are on order to see if any difference is observed.

The change in oxygen concentration with time at argon sparging rates above and below the specified rate are shown in Figure 4. It is clear that a first-order rate of reduction in concentration applies down to the 100-ppb range and then some other factor(s) predominates and reduces the rate of oxygen removal.

From the results to date, it would appear that the specification should be modified to require a high sparge rate for about 1 h to help eliminate gas

Figure 4. Effect of Argon Sparge Rate on Reduction in Oxygen Concentration in GCA Solution at 70°C

pockets, followed by a much slower rate to maintain a suitable oxygen concentration.

The laboratory is now being modified so that the oxygen monitor can be used during a regular metal removal test with the ultimate objective of determining the maximum oxygen level that can be tolerated in the GCA process and the sparging procedures required to achieve and maintain this level most economically.

4. Initiation of Type 316 Stainless Steel Deposition Test

Metal removal tests to date on both castings and wrought specimens of Type 316 stainless steel which have been given a short-time sensitization indicate that the metal removal rate is about an order of magnitude less than the rate for equivalent Type 304 stainless steel using the GCA process.

Two factors may tend to counteract this difference for reactor components. One is the diffusion profile for ⁵⁴Mn in Type 316 stainless steel compared to Type 304 stainless steel, and the other is the relative degree of sensitization achieved for the two alloys in the short-time sensitization procedures used.

The radioactivity results on the deposition specimens discussed in a preceding section of this report show the effect of one composition variable, carbon, on radioactivity deposition in Type 304 stainless steel. Molybdenum may also have an effect. The metal removal results in Section 2 on long-time sodium sensitized specimens vs the short-term sensitized specimens illustrate the importance of the variable on the time required to achieve a given degree of decontamination.

In order to develop some quantitative information on how these factors may influence the decontamination of Type 316 stainless steel components, the deposition loop was loaded with Type 316 specimens during the first week of January. Both castings and wrought specimens are included, covering a range of carbon concentrations. These samples should be ready to evaluate in the early part of FY 1979.

In another long-term sensitization test to see if vacuum sensitization can substitute for reactor "traveler" specimens, the first evaluation will be made in May when the specimens have accumulated about 4700 h. This time is the same as the exposure time of the sodium deposition loop specimens discussed in this report.

5. Sputtering Experiments

The sputtering experiments are carried on at a low level of effort to provide a preliminary evaluation of the feasibility of this new technique for decontamination.

So far some boundary conditions have been established for the process, but the main effort has been to develop equipment configurations that can be used to produce quantitative data on voltage, current density, electrode spacing, and argon pressure relationships.

The main design problem has been related to the electrical feedthrough where large power losses have occurred, resulting in unknown power input values to the sputtering electrodes. The remedy is to keep the positively and negatively charged equipment components closer together than the cathode dark space distance or else keep them very far apart, a difficult problem when the electrode spacing must be varied over a considerable range.

A good sputtering run with an electrode spacing of 2 in. is nearing completion with little power loss. The conditions suitable for the spacing is 1000 V, 0.1 A, and an argon pressure of 40 Pa for a 50-cm² specimen. The weight of stainless steel deposited per day at this power level has not yet been determined, but appears to be fairly high based on visual observation.

IV. IMPACT ON LMFBR PROGRAMS

This program addresses itself specifically to: (1) problems of cleaning surfaces exposed to sodium, (2) investigation and development of the alcohol sodium removal process and the associated process control, and (3) preparation of procedures and specifications for the decontamination of sodium-wetted components. This information is directly applicable to the FFTF and CRBRP activities and the LMFBR Target Plant design program.

V. NEXT REPORT PERIOD ACITIVITIES

A series of tests will be conducted on the removal of sodium from simulated failed fuel pin assemblies.

The major activities of the Decontamination Studies Task during the next report period will include:

- 1) Continuation of oxygen monitor evaluation
- 2) Continuation of metal removal tests on Type 304 castings and radioactive deposition loop specimens
- 3) Continuation of deposition loop run with Type 316 stainless steel
- 4) Continuation of traveler study
- 5) Continuation of sputtering study
- 6) Assistance to HEDL in the decontamination pilot plant startup.

Program: Cover Gas Seals

DOE Task: 27-LMFBR Seal Test Program

Program Manager: R. W. Keaten

Project Manager: L. Baurmash

Reporting Period: January-March 1978

Category: HN-10-04-02.2

General Order: 09294

Subaccount: 14504, 14505

189a Number: SA 007

Principal Investigators: O.P. Steele III, F. H. Welch

I. PROJECT OBJECTIVES

The objective of the Cover Gas Seal Material Development Program is to perform the engineering development required to provide reliable seals for LMFBR application. Specific objectives are to verify the performance of commercial solid cross-section and inflatable seals under reactor environments including radiation, to develop advanced materials and configurations capable of achieving significant improvement in radioactive gas containment and seal temperature capabilities, and to optimize seal geometry for maximum reliability and minimal gas permeation.

II. MAJOR ACCOMPLISHMENTS DURING FISCAL YEAR 1978

Permeation measurements were completed on the UCARSIL Carborane-Siloxane and the Horizons Research Polyphosphazene Formulation 2511-24.

The final report for Phase I of the polyphosphazene compounding study was received and evaluated. Goals of obtaining compression set values of less than 20% were not achieved.

The Parker Seal report on improved EPDM elastomers was received, and released as report N707TI810022, "EPDM Compounding Study, Parker Seal Test Report No. R10, 478U". The two formulations, XE2362-28 and XE2362-41 exhibited compression sets of less than 20% in the standard 70-h 150°C (302°F) test.

Static leakage tests on the same elastomers for 2970 h at 66°C (150°F) resulted in compression set data of 15% or less.

Evaluation of the Armstrong epoxy, A-66, showed the cement to be capable of bonding EPDM and steel at 177°C (350°F) for 238 h with no evidence of deterioration.

The four Presray EPDM inflatable seals were received and preliminary evaluation was begun. The seals were observed to lose inflation pressure gradually. The leak rate of the seals was measured to be 2.7 to 6.0×10^{-6} cc/s/linear in. of seal/atm Δ P at room abient.

III. PROGRESS DURING THE REPORT PERIOD

A. ADVANCED SEAL GEOMETRIES

No work is scheduled under this activity during FY 1978.

B. ADVANCED SEAL MATERIALS

1. Gas Permeation

Gas permeation rates ($\bar{P}$) are shown in Figure 5 for the Union Carbide Carborane-Siloxane at 41°C (106°F), 93°C (200°F), and 149°C (300°F). Other test specimens split during testing, and only data at 93°C (200°F) were obtained. These values agree fairly closely with the data reported in the last quarterly report.

Similar $\bar{P}$ data were obtained on the new formulation of the Horizons Research Polyphosphazene (No.2511-24), as illustrated in Figure 6. These rates agreed very well with the No.2333-19B formulation.

Neither elastomer presents any improvement in $\bar{P}$ compared to commercially available materials.

2. Compression Set

a. Kalrez No.335 Elastomer

The compression set test of a Kalrez No.335 elastomer (DuPont) and a comparison elastomer (Parker silicone rubber S684) completed 4 weeks at 149°C (300°F).

The Kalrez has suffered a 100% set, while the silicone rubber exhibited a 46% set. This value compares well with an earlier value of $\sim 45\%$ at 121°C (250°F) for the silicone rubber.

Figure 5. $\log \bar{P}$ vs $1/T(K)$ of H_2 , Kr, Xe, and Ar for UCARSIL (Union Carbide, Carborane-Siloxane)

Figure 6. Log $\bar{P}$ vs $1/T(K)$ of H₂, Kr, Xe, and Ar for Horizons Research Polyphosphazene Formulation 2511-24

b. Parker Seal EPDM Elastomers

The data from the four Parker Seal EPDM elastomers which had undergone 2970 h of static leakage tests at 66°C (150°F) were evaluated for compression set and changes in weight and hardness. Table 5 shows the reduced data.

The E692 compound agrees quite well with the earlier AI-measured compression set data on this compound. The E740 compound shows the lowest set, and both of the new XE-compositions are about equal at intermediate compression set values.

The vendor-derived data, shown in Table 6, from the ASTM test at 150°C (302°F) for 70 h shows the new compounds to have the lowest set, followed by E740, and E692 exhibiting the highest set.

TABLE 5
COMPRESSION SET, WEIGHT CHANGE, AND HARDNESS CHANGE
OF PARKER SEAL EPDM COMPOUNDS AT 66°C (150°F) AND 2970 h

EPDM Compound	Compression Set	Weight Change (g)	Hardness Change Shore A
E692-75*	15.3	+0.0087	-1
E692-75*	15.0	+0.0113	-1
E692-75 [†]	13.8	+0.0113	-2
E740-75*	5.4	+0.0056	-1
E740-75*	7.9	-0.0007	+2
E740-75 [†]	6.4	-0.0061	+1
XE2362-28*	2.2	+0.0062	-2
XE2362-28*	10.0	+0.0058	-3
XE2362-28 [†]	11.5	+0.0078	-4
XE2362-41*	10.2	+0.0156	-1
XE2362-42*	10.2	+0.0280	-5
XE2362-41 [†]	6.8	+0.0080	-2

*20% compression during test

[†]45% compression during test

TABLE 6
PROPERTIES OF THE EPDM COMPOUNDS

Property	Parker Compounds		Parker Experimental	
	E692-75	E740-75	XE2362-28	XE2362-41
2-214 O-Rings <u>Original Physical Properties:</u>				
Hardness, Shore A, (pts)	74	70	74	75
Tensile Strength, psi (MPa)	1680 (11.58)	1810 (12.48)	1740 (12.00)	1230 (8.48)
Elongation (%)	177	162	175	189
Modulus 100%, psi (MPa)	825 (5.69)	822 (5.67)	919 (6.34)	779 (5.37)
<u>Compression Set:</u>				
70 h 150°C (302°F) 25% Deflection				
Original Deflection	37.6	18.1	16.2	14.3

c. Parker Seal Compounding Study

The Parker Seal Co. report, submitted at the end of the compounding study to develop improved EPDM compositions, was released as N707TI810022, "EPDM Compounding Study, Parker Seal Test Report No. R10, 478U."

The Parker report SUMMARY and POSSIBLE FUTURE WORK were presented as follows:

SUMMARY

Parker experimental compound XE2362-28 was developed in fulfillment of the contract, encompassing the findings of this study. With a compression set value of 16.2% after 70 hours at 302°F on 2-214 O-rings, this experimental formulation is a substantial improvement over the currently supplied material, Parker compound E692-75, with a compression set value of 37.1%. This experimental compound combines the high carbon black loading level of an E692-75 type compound for good permeation properties, with the good compression set resistance of an E740-75 type formulation

In addition to fulfilling the major requirements of the contract, the experimental compound XE2362-41 was developed because one of the ingredients in XE2362-28, medium thermal (MT) carbon black, is predicted to become increasingly more expensive and harder to obtain. This second experimental compound has an improved compression set value of 14.3%. The tensile strength, however, is slightly lower than the standard, but not felt to be detrimental to the product.

The improvement in the compression set resistance of these two experimental compounds indicates a tighter state of cure. This condition should be beneficial to the permeation characteristics of the compound. As agreed, Rockwell International will perform the permeation testing.

POSSIBLE FUTURE WORK

Establish the performance of these ethylene propylene compounds by long term compression set or stress relaxation tests, which could predict seal life.

Optimize the formulation by using the computer designed experiment capabilities currently in-house.

Study the antioxidant optimization for improved long term compression set resistance.

Study the reasons for and possible solutions for the cracking under stress of the Opal black formulation which had the excellent compression set resistance.

Investigate the tensile strength of the experimental compound XE2362-41 while maintaining the good compression set resistance.

3. Static Leakage Tests

The static leakage tests on the four EPDM elastomers (Parker Seal E692, E740, XE2362-28, and XE-2362-41) were terminated at 2970 h. Calculation of the leakage is under way. The H₂ leakage appears normal, but the Kr leakage is higher than normal on the preliminary analysis.

The computer program for calculating the leakage values is being reviewed and revised.

4. Material Properties

Selected mechanical properties of five formulations of Polyphosphazene (Horizons Research) are shown in Table 7. The three new formulations appear

less desirable than the two earlier formulations, exhibiting higher compression set and lower tensile strength.

TABLE 7
COMPARISON OF PROPERTIES - POLYPHOSPHAZENE

Property	Formulation									
	2333-12C		2333-16D		2511-16D		2511-23		2511-24	
	MPa	psi	MPa	psi	MPa	psi	MPa	psi	MPa	psi
Tensile Strength	14.27	2070	14.89	2160	11.17	1620	11.51	1670	10.96	1590
100% Modulus	7.51	1090	13.24	1920	10.82	1570	11.13	1615	5.65	820
Elongation (%)		135		120		100		100		155
Hardness Shore A		65		70		60		65		52
Compression Set (%)		15		19		24		25		32
ASTM D395										
Method B, Type II										
70 h 150°C (302°F)										

C. GAMMA IRRADIATION

A small quantity of the new Armstrong A66 high-temperature epoxy cement was procured, and several test specimens of the inflatable seal EPDM + nylon cloth material were cemented to steel tabs.

The assemblies were air-cured over night, baked at 82°C (180°F) for 1 h, and then heated in air at 177°C (350°F). The strips were examined after heating and then tensile tested.

The cemented joint between the nylon-cloth-reinforced EPDM and the seal appear good and could not be peeled by flexing of the elastomer. Table 8 shows the results of the tensile tests.

All samples broke in the rubber strip, leaving the epoxy cement unaffected.

TABLE 8
TENSILE TESTS ON EPDM/STEEL CEMENTED
WITH A-66 EPOXY CEMENT
AFTER HEATING AT 177°C
(350°F)

Time of Exposure (h)	Tensile Strength	
	MPa	psi
120	2.02	293
238	1.12	163
238	1.89	274
238	2.10	304
238	1.69	245

Additional tests will be conducted on the effect of lubricant on the epoxy; but based on these initial tests, the A-66 was used to cement the inflatable seals into the test fixture, since the planned tests at 121°C (250°F) are above the recommended temperature for the Chemlok epoxy used on earlier tests.

1. Inflatable Seal Assembly

The Test Plan (N707TP810016, "Inflatable Seal Gamma Radiation Testing – A Test Plan") and the Test Procedure (N707DTP810019, "Inflatable Seal Gamma Radiation Testing – A Test Procedure") were issued.

All four EPDM seals were delivered. The seals were visually inspected, photographed, and weights and hardness determined. Figure 7 shows a typical seal, and Figures 8 and 9 show two observed defects, a typical area of poor fabric coverage, and a crease across the grooved sealing surface, respectively. The former defect could lead to leakage of inflation pressure, while the latter might permit across-the-seal leakage.

The EPDM seals were installed in the test fixture initially without being cemented in place (Figure 10). Across-the-seal face leakage occurred at inflation pressures up to 0.44 MPa (50 psig). The fixture was opened and no apparent reason was found for the leakage. The seals then were cemented in place to eliminate the possibility of leakage across the bottom of the seal.

Figure 7. EPDM Inflatable Seal (Inner) Serial No. 03-02

Figure 8. EPDM Inflatable Seal, Serial No. 03-02 Poor Fabric Coverage

Figure 9. EPDM Inflatable Seal, Serial No. 03-01 Crease Across Grooves

9294-10

Figure 10. Inflatable Seal Installed in Fixture Plate Prior to Cementing

Across-the-seal face leakage still occurred after cementing, and it was concluded that the seal face was not properly contacting the upper plate.

A plastic plate was made and washers were used on the bolts to simulate the 4.75-mm (0.187-in.) gap which the seal was built to expand across. When the two seals were pressurized to 0.26 MPa (25 psig), a between-the-seal pressure of 0.130 MPa (5 psig) could not be maintained, nor could the pressure be maintained across a 3.3-mm (0.130-in.) gap. But at a spacing of about 1.65 mm (0.065 in.), the between-the-seal pressure was maintained.

However, when the seals were inflated to 0.267 MPa (25 psig) with 0.095 MPa (0 psig) between-the-seal pressure, it was observed that the seals steadily lost inflation pressure. In 24 h the pressure dropped about 0.011 MPa (1.8 psig); in 48 h the pressure dropped 0.022 MPa (3.2 psig). During the same time, the between-the-seal pressure increased to 0.117 MPa (3.2 psig) and 0.133 MPa (5.5 psig), respectively.

These pressure changes indicated that the seals were leaking very slowly. To confirm this conclusion, a dam was built on each side of the seals, and the seals covered with distilled water.

Rapid bubbling was not observed at 0.267 MPa (25 psig), but several small bubbles were observed to form slowly. In addition to what appeared to be four very small leaks, numerous extremely slow-forming bubbles were noted on the inner seal. On the outer seal, only extremely slow-forming bubbles were noted.

The inflation pressure was increased to 0.302 MPa (30 psig) and then to 0.336 MPa (35 psig), but no change in the leakage pattern was observed.

It was concluded that the seals were porous, and the leak rate so high that permeation-type measurements would be meaningless.

A pressure decay test was conducted on the four seals starting with an initial inflation pressure of about 0.267 MPa (25 psig). The seal pressures were recorded for 6 days, and the results of the test are shown in Figure 11. As may be seen, one of the seals showed a much greater pressure drop.

9294-13

Figure 11. Pressure Decay Test, EPDM Inflatable Seals

The seal volumes were measured in the inflated mode, and the leak rate (L) calculated in cc/s/linear in. of seal/atm Δ P:

$$LO_F = 3.9 \times 10^{-6}$$

$$LO_P = 3.9 \times 10^{-6}$$

$$LI_F = 2.7 \times 10^{-6}$$

$$LI_P = 6.0 \times 10^{-6}$$

The leak rates of two earlier-tested inflatable seals were calculated from similar pressure decay rate to provide comparison data. These earlier seals also were porous and of poor quality. The L values were:

$$L_{\text{Butyl}} = 0.8 \times 10^{-6}$$

$$L_{\text{Nitrile}} = 5.8 \times 10^{-6}$$

The poorest of the EPDM seals compares well with the nitrile seal, and the three remaining EPDM seals are intermediate to the butyl and the nitrile.

A representative of the seal manufacturer (Presray Corp.) visited AI and observed the seal pressure decay tests. His analysis of the problem was that the inflation gas may be leaking from the inflation stem and migrating along the fabric and through the thin outer layer.

The new seals (nitrile) should be available in a few weeks, and the vendor will try to reduce the leakage by adjusting the fabrication technique.

To obtain some data from the seals until the new nitrile seals are delivered, a new top plate with a more shallow recessed region [1.65 mm (0.065 in.) instead of 4.75 mm (0.187 in.)] will be fabricated.

D. GAS TRANSPORT THROUGH SEALS

No work is scheduled under this activity during FY 1978.

IV. IMPACT ON LMFBR PROGRAMS

The initial phases of this program have guided selection of materials and seals for the FFTF and CRBRP from information on permeability, compression set, and gland design published in progress and technical reports. Designs of the FFTF IVHM inflatable and primary seals were verified. Other phases have verified the instrument tree inflatable seal design and evaluated the use of inflatable seals for CRBRP. Effects of sodium vapors and aerosols and radiation on the sealing efficiency of elastomers will be delineated. Advanced seal geometries and materials will be developed to overcome the commercial seal deficiencies of excessive compression set (short maintenance cycle) and permeation. Specifications and a design guide for use in the LMFBR industry will become available and will be updated as new information is generated.

V. NEXT REPORT PERIOD ACTIVITIES

Testing of the EPDM inflatable seals will continue.

Permeation measurements on slab specimens will continue.