Analysis of the Three Mile Island Submerged Demineralizer System Vessel Burial Data

Fiscal Year 1989

Prepared for the U.S. Department of Energy
Assistant Secretary for Nuclear Energy

Westinghouse
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ANALYSIS OF THE THREE MILE ISLAND SUBMERGED DEMINERALIZER SYSTEM VESSEL BURIAL DATA FISCAL YEAR 1989

W. G. Jasen
S. J. Amir

ABSTRACT

Long-term monitoring of a Submerged Demineralizer System vessel buried in its concrete overpack was undertaken at the Hanford Site to determine actual burial conditions during a proposed long-term test period. Parameters of interest were moisture, fission product particulates, pressure, and temperature. An evaluation of the data obtained to date from the Submerged Demineralized System vessel shows no abnormalities. Further monitoring of each parameter is expected to continue for at least the next two years. This document updates WHC-EP-0083, incorporates 1988 and 1989 data, analyzes the results, and presents recommendations.

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ANALYSIS OF THE THREE MILE ISLAND SUBMERGED DEMINERALIZER SYSTEM VESSEL BURIAL DATA FISCAL YEAR 1989

1.0 INTRODUCTION

The Submerged Demineralizer System (SDS) was used during the Three Mile Island (TMI) nuclear reactor cleanup to remove cesium and strontium from contaminated water. The SDS vessels are 2-ft-in diameter and 4-ft tall stainless steel cylinders containing up to 60 kCi of radioactive cesium and strontium loaded on damp zeolite. The water in the damp zeolite absorbs some of the ionizing radiation and decomposes to hydrogen and oxygen by a process called radiolysis. Gas generation rates approaching 1 L/h (Quinn et al. 1984) have been calculated and measured for some of these loaded vessels. Each of the SDS vessels contains a catalyst bed to recombine the available hydrogen and oxygen back to water. Tests have proven this hydrogen control method to be highly effective, even under very wet (but unsubmerged) conditions.

Nineteen SDS vessels, packaged one at a time in a shielded and licensed shipping cask, were shipped to Rockwell Hanford Operations (Rockwell). Collectively, these vessels contain approximately 7,500 kCi of radioactive material. This work has been previously reported (Quinn et al. 1984 and Henrie and Murry 1987). Sixteen vessels were transloaded into concrete overpacks and buried at the Hanford Site. The contents of the other three vessels were vitrified at Pacific Northwest Laboratory.

Subsequent to placement of the SDS vessels in the burial grounds, DOE Order 5820.2A (DOE 1988) was issued in September 1988. This order requires wastes to be evaluated against 10 CFR 61.55 for radioactivity above greater-than-class C (GTCC) limits. Fourteen of the sixteen vessels buried at the Hanford Site have been determined to be GTCC waste.

Long-term monitoring of SDS vessel No. D10011 in its concrete overpack was undertaken to determine actual burial conditions during the proposed test period of 20 to 50 yr. These burial conditions are used to identify any problems with the vessel during the test period. Parameters of interest are moisture, fission product particulates, pressure, and temperature.

Monitoring of the vessel pressure began in June 1984; the vessel was placed in its concrete overpack in September 1984. Temperature, moisture, and particulate monitoring began in November 1984, after burial of the vessel and its overpack had been completed. The instrumented SDS vessel and concrete overpack are shown in their buried condition in Figure 1.

Recommendations and rationale for continued monitoring of the instrumented SDS vessel are presented in Section 4.0.

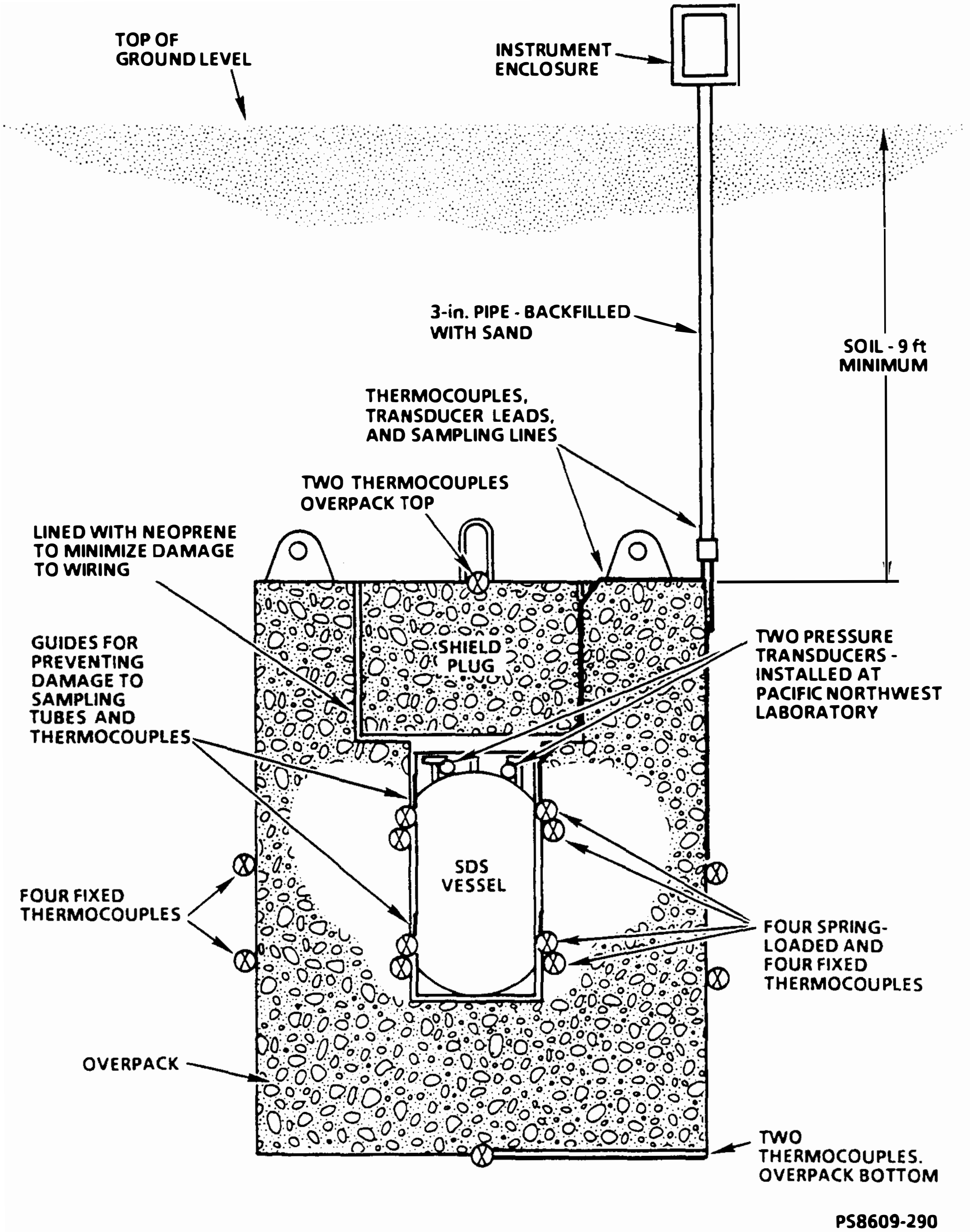

Figure 1. Instrumented Submerged Demineralized System Vessel and Overpack in Their Burial Position.

2.0 SUMMARY

An evaluation of the moisture, particulate, pressure, and temperature data obtained to date from SDS vessel No. D10011 shows no abnormalities. The humidity of the air between the vessel and its overpack is relatively low, indicating dry, ideal storage conditions. Suspended particulate materials in the air surrounding the vessel are very low and no radioactivity has been detected.

The pressure rise rate in the vessel decreased from approximately 9.4 lbf/in^2-yr to approximately 3.5 lbf/in^2-yr as its pressure rose from a few pounds per square inch (absolute) to one atmosphere. Therefore, it appears that approximately two-thirds of the initial pressure rise was caused by a slow leak of air into the vessel and that one-third was caused by gas generation. The net gas generation rate plus the air inleakage rate initially totaled approximately 70 L/yr. The gas generation rate when the gas pressure was at one atmosphere (leakage essentially zero) was approximately 25 L/yr.

The system pressure has stabilized at approximately 16 lbf/in^2 (absolute) $\pm$ 0.5 lbf/in^2, which is approximately 1.5 $\pm$ 0.5 lbf/in^2 above atmospheric pressure. The pressure readings for September 1988 and April 1989 have risen to 17.6 lbf/in^2 (absolute) and 17.9 lbf/in^2 (absolute), respectively. The future readings will be compared to analyze the reason for pressure increase. The average gas leakage is equal to the net gas generation rate, which is approximately 25 L/yr. The pressure and gas leak rate are highest during the summer months and lowest during the winter months as a result of thermal expansion and contraction.

An analysis of the temperature data shows that temperatures are at or near their maximums and the SDS vessel shell and internal temperature are decreasing slowly due to radioactive decay. The calculated peak temperature of the zeolite was 376 °F (February 1986) and is now 372 °F. The maximum recorded vessel temperature, 136.3 °F, was reached in September 1985. The temperature at the same location was 122 °F in September 1988 and 107 °F in April 1989. It is apparent that the temperatures have peaked and will continue their cooling trend.

It is recommended that data measurements be taken semiannually, each winter and summer, for the next 2 yr and documented biennially by a revision of this report. The last report will be issued in September of 1991 unless otherwise warranted.

3.0 DATA AND ANALYSIS

Analysis of the monitoring data of SDS vessel No. D10011 is presented in the following sections.

3.1 MOISTURE

A void space of approximately 171 L exists between the SDS vessel and the concrete overpack. The accumulation of moisture in this region is a concern; however, fission product heating should dry this region and prevent a saturated moisture condition from developing, particularly in the semiarid Hanford environment.

An Atkins psychrometer of calibration quality was used to measure the humidity within the overpack void space. The psychrometer provides wet and dry bulb temperatures of the air as it is withdrawn from the cavity. These data are used to determine the humidity in pounds of water per pound of dry air. The relative humidity in the overpack void space is determined using the above data, measured temperatures in the void space, and a psychrometric chart.

The low relative humidity verifies the existence of an unsaturated condition within the overpack (Table 1). However, there may be some condensation of water in the cold lines and equipment during sampling in the winter months; therefore, the humidity may be higher than indicated at those times. The data strongly indicate that moisture is not accumulating in the void space. The highest measured relative humidity has been 53%. It therefore appears that corrosion of the SDS vessel external stainless steel surfaces caused by moisture in the overpack cavity is minimal.

3.2 PARTICULATE

Gas samples are drawn from the void between the SDS vessel and the overpack. A battery-operated vacuum pump is used as the vacuum source; a 0.45-µm filter medium is used to entrain particulate drawn from the void space.

As shown in Table 2, the particulate on the filters has been near zero throughout the monitoring phase. No contamination above normal background has been found on the filter cassettes when scanned for beta-gamma activity. Therefore, no significant leakage of particulates from the SDS vessel is occurring. The data for 1988 is missing and considered lost.

Table 1. Moisture Data.

Date	Dry bulb °C cavity average	Dry bulb °C Atkins	Wet bulb °C Atkins	Humidity pounds of water per pound of air	Relative humidity
2/7/85	34.6	7.8	7.0	0.006	17
2/26/85	35.9	21.0	19.9	0.014	37
3/4/85	34.8	12.4	12.0	0.0088	25
5/7/85	37.2	20.6	20.3	0.015	37
5/28/85	38.6	28.9	27.7	0.023	53
6/18/85	40.3	28.7	25.8	0.020	42
7/16/85	42.1	37.1	29.1	0.022	43
8/16/85	44.2	22.5	19.9	0.014	25
10/31/85	45.2	11.4	10.8	0.0078	13
1/3/86	43.4	1.5	1.49	0.0042	8
4/30/86	40.5	15.5	14.6	0.0098	20
8/14/86	44.4	30.8	29.0	0.025	43
4/7/87	40.4	24.0	20.8	0.014	29
8/28/87	44.0	31.7	28.2	0.023	40
3/9/88	35.7	14.2	13.2	0.001	10
9/29/88	42.0	30.1	27.8	0.017	36
4/13/89	34.2	20.1	18.1	0.0065	20

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Table 2. Particulate Data.

Date	Particulate (g)	Air volume (L)	μg/L	Radioactivity
2/7/85	0.0149	36.4	410	a
2/26/85	0.0017	129.2	13	a
3/4/85	0.0001	114.1	1	a
5/7/85	0.0002	15.2	13	a
6/18/85	0.0009	22.8	40	a
7/16/85	<0.0001	29.1	<3	a
8/16/85	<0.0001	7.7	<13	a
10/31/85	0.0001	14.3	7	a
1/3/86	0.0001	12.0	8	a
4/30/86	0.0002	60	3	a
8/14/86	0.0002	45	4	a
4/7/87	<0.0001	35	<3	a
8/28/87	0.0001	40	3	a
3/19/88	b	b	b	b
9/29/88	b	b	b	b
4/13/89	0.0002	135	2	a

[a]None detected above normal background.
[b]Data missing.

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3.3 PRESSURE

The SDS vessel is equipped with two strain gage pressure transducers (0 to 500 lbf/in^2 and 0 to 50 lbf/in^2) to measure the pressure inside the vessel. When purchased, these transducers represented the latest development in radiation-resistant pressure transducer technology. For unknown reasons after the SDS vessel was buried the 0 to 500 lbf/in^2 transducer failed and is not operating. As a result, the pressure data throughout this report were obtained from the 0 to 50 lbf/in^2 transducer that is monitored by a portable, hand-held, battery-powered, digital-readout electronic instrument.

The 0 to 50 lbf/in^2 readout instrument experienced two periods of abnormal operation since the burial of the SDS vessel and overpack. The causes for the abnormalities were investigated, and operating procedures were implemented to prevent future abnormalities and assist in the evaluation of the data (Henrie and Murry 1986). The stable pressure data are shown in Figure 2 as a function of time after the vessel was evacuated and sealed.

The earlier data (May 1985 to February 1986) show that the system pressure approached atmospheric pressure at a decreasing rate. The initial pressure rise rate was approximately 9.4 lbf/in^2-yr. This pressure buildup resulted from air inleakage and gas generation. From the pressure rise rate and the previously determined 129-L void volume (Henrie et al. 1986), the initial gas buildup rate was calculated to be 70 L/yr. The pressure rise rate at the time the SDS vessel pressure reached standard atmospheric pressure was approximately 3.5 lbf/in^2-yr. At that time, the air inleakage was essentially zero. The net gas generation at that time was calculated to be approximately 25 L/yr. Therefore, the initial air inleakage rate was approximately 70 - 25 = 45 L/yr.

The later data (May 1986 to April 1989) show that the vessel pressure is now exceeding standard atmospheric pressure. The pressure plateaued at 16.8 lbf/in^2 (absolute) in August 1986, dropped to 15.6 lbf/in^2 (absolute) the following winter, and returned to 16.7 lbf/in^2 (absolute) at the end of June 1987. Thus, the pressure appears to be cycling on an annual basis with the pressure and leak rate lower in the winter months than during the summer months. The data taken for September 1988 and April 1989 show a pressure rising of 17.6 and 17.9 lbf/in^2 (absolute), respectively, which is about 1 lbf/in^2 above the 2-yr plateau. Future data will indicate the stabilization of pressure inside the vessel. The average leak rate so far seems to be equal to the net gas generation rate.

The catalytic hydrogen-oxygen recombiner is performing well. The measured net radiolytic gas generation rate in SDS vessel No. D10011 prior to installing the catalyst was greater than 0.4 L/h (Quinn et al. 1984; Henrie et al. 1986), which is more than 3,500 L/yr and is 140 times more than the calculated current net generation rate (leak rate) of 25 L/yr. Therefore, more than 99% of the gas being generated by radiolysis in the SDS vessel is being recombined. The gas in the SDS vessel is essentially all nitrogen from air inleakage, hydrogen from radiolysis, and carbon monoxide and carbon dioxide from the oxidation of organic contaminants (Quinn et al. 1984). The hydrogen gas cannot be recombined due to a lack of oxygen. The oxygen from air inleakage and some of the radiolytic oxygen has been used up in competing oxidation processes. However, due to very low oxygen concentrations in the vessel, certainly less than 1%, the gas mixture is not flammable.

3.4 TEMPERATURE

The fission product heating inside the SDS vessel increases temperatures in the vessel, concrete overpack, and surrounding soil. Therefore, to monitor temperature changes, thermocouples were placed on the vessel, inside and outside the overpack, and in the soil.

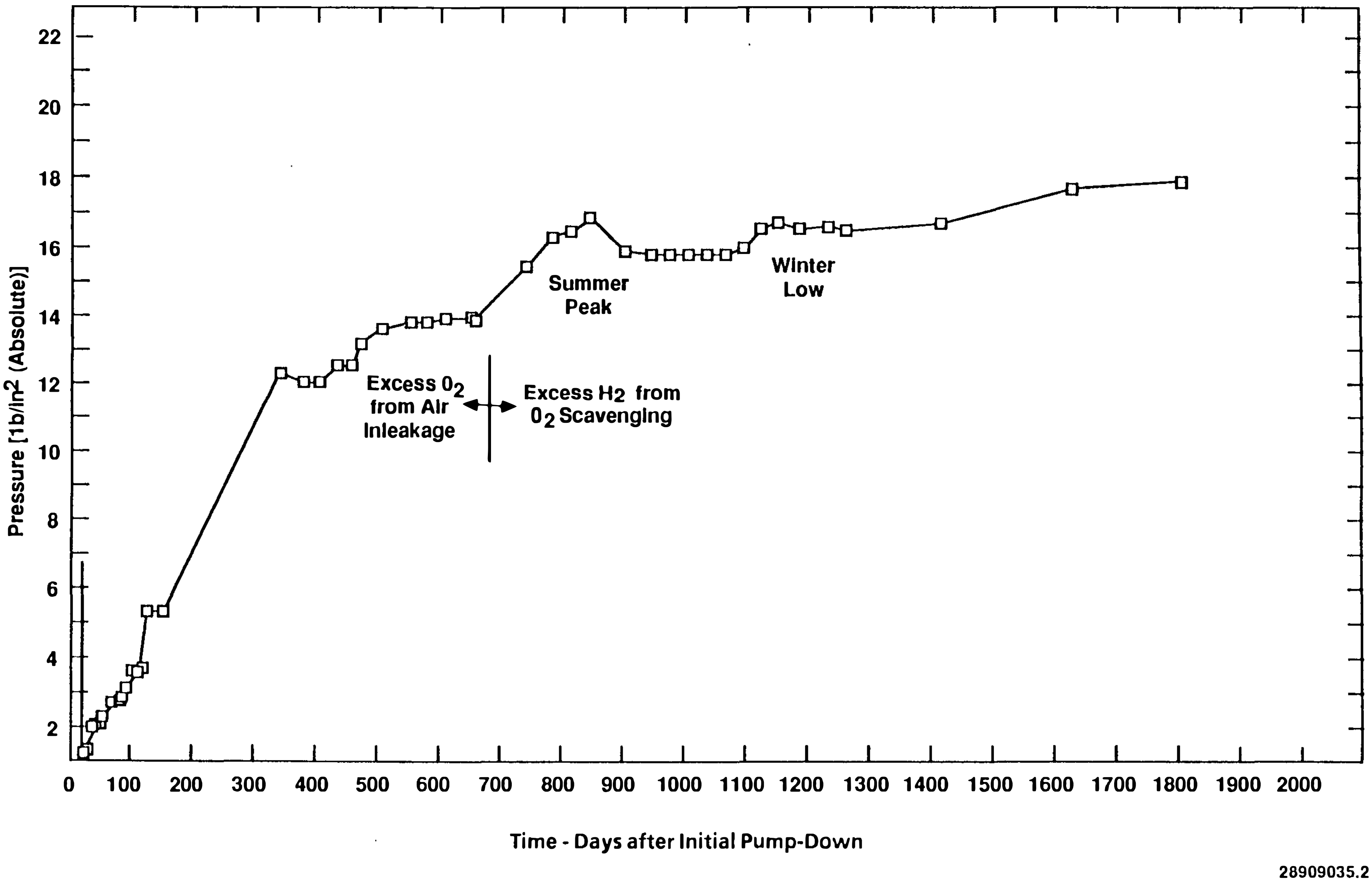

Figure 2. Instrumented Submerged Demineralized System Vessel Pressure Versus Time.

The buried SDS vessel was modeled using the HEATING5 heat transfer computer code to compute the maximum centerline temperature and other temperatures. The computer model assumes an outside ambient air temperature of 51 °F, which is near the Hanford Site average. The properties used in the final HEATING5 model are listed in Table 3.

Table 3. Thermal Constants Used in HEATING5 Model.

Material	Thermal conductivity (Btu/h-ft-°F)	Volumetric heat capacity (Btu/ft^3-°F)
Damp zeolite	0.092	10.48
Air	0.016	0.017
Concrete	1.2	30.24
Soil	0.35	24.86

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The latest temperature data and computer results are shown in Figure 3. Due to the high heat capacity of the overpack and the soil surrounding the overpack, it was originally calculated that peak temperatures might not be reached until 3 yr after burial of the vessel and overpack. Also, a peak temperature of just under 300 °F was calculated. Because the measured data show that the temperature of the SDS vessel shell peaked much earlier, the original modeling and results were reevaluated. A determination was made that the original modeling did not properly account for approximately 40% of the heat being generated [223 W in September 1983, as calculated from the 44,317 Ci of cesium plus the 2,061 Ci of strontium reported by Quinn et al. (1984)]. Making this correction and adjusting thermal conductivities for the concrete overpack and for the surrounding sand and gravel, decreased calculated peaking times and increased calculated temperatures. The latest modeling (no seasonal temperature corrections) indicated that the temperature of the zeolite near its vertical centerline and below its upper surface peaked at 376 °F in February 1986 and was 372 °F in August 1987. The maximum recorded vessel temperature, 136.3 °F, was reached in September 1985. The temperature at that same location was 134 °F in August 1986 and 133 °F in August 1987. In 1988 and 1989, there was a cooling trend as shown in Figure 3.

The latest computer modeling indicates that these temperatures should still be slowly increasing. This modeling also shows temperatures slightly higher than those measured.

The apparent reason for these temperature discrepancies is that the modeling does not take into consideration (1) all of the convection heat transfer that is occurring in the void space between the SDS vessel and the shield plug, or (2) the extra heat conduction paths provided by the reinforcing steel, lifting lugs, and steel liners in and around the upper shield plug. These heat paths apparently remove enough heat to measurably reduce the amount of heat moving laterally through the overpack. Correcting for this vertical heat shunt would reduce calculated temperatures in the lateral directions.

Because the thermal data and computations show no significant anomalies, any further refinements would be only to improve modeling techniques and are not justified under the current program.

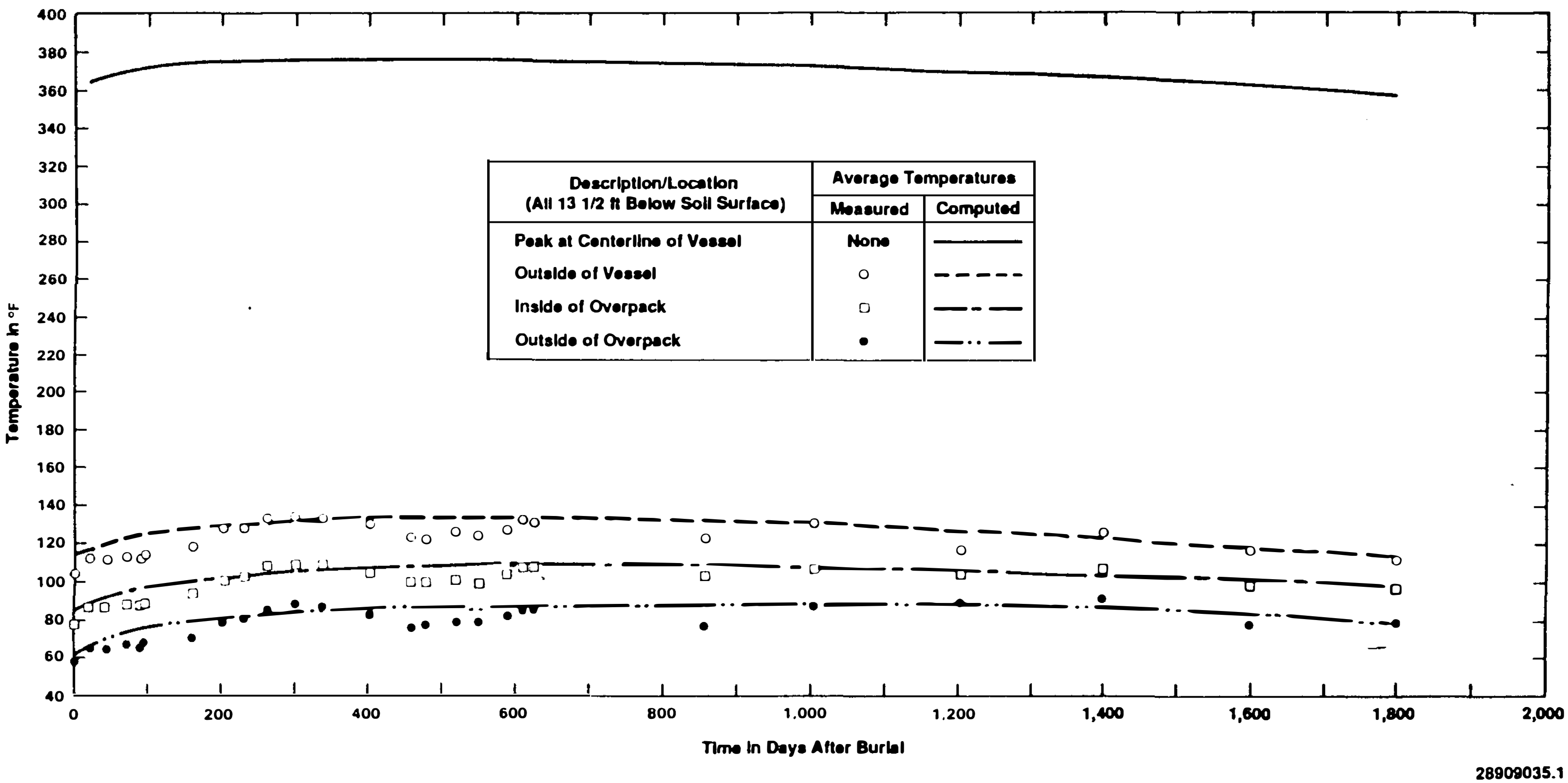

Figure 3. Calculated and Measured Temperatures In and Around a Buried, Instrumented Submerged Demineralized System Vessel.

4.0 FUTURE MONITORING RECOMMENDATIONS

Pressure monitoring of SDS vessel No. D10011 began in June 1984, a few months before the instrumented SDS vessel was placed in its overpack in September 1984, and buried in November 1984. Temperature monitoring started immediately after the overpack was buried. Moisture and particulate monitoring started in January 1985. Conclusions, recommendations, and rationale for further testing are discussed for each data type in the following sections.

4.1 TEMPERATURE

It is apparent from Figure 3 that peak temperatures have been reached. To more accurately determine long-term temperature conditions, it is recommended that temperatures be monitored on a semiannual basis, each February and August, to determine annual minimums and maximums for the next 2 yr. It is doubtful that annual temperature monitoring will be beneficial and cost effective after that time.

4.2 PRESSURE

The pressure data shown in Figure 2 are reasonably consistent and appear to be correct. The pressure in the vessel has apparently reached its plateau (August 1986). Continued pressure monitoring is important to determine (1) if leaks in the vessel are plugging or enlarging and (2) how long the pressure transducer will continue to function in its very high-radiation environment. Therefore, it is recommended that the pressure data be obtained each February and August for the next 2 yr.

4.3 PARTICULATE

Airborne particulate monitoring is occurring on a semiannual basis. Particulate monitoring is important because it would detect a major breach of the SDS vessel containment boundary. Therefore, continued semiannual particulate monitoring is recommended for the next 2 yr.

4.4 MOISTURE

The data from humidity monitoring are presented in terms of humidity and relative humidity. The measurements (see Table 1) show that moisture conditions in the overpack cavity atmosphere are far below saturation.

Moisture level measurements in the overpack cavity are important for two reasons: (1) corrosion rates increase under high moisture conditions and (2) high relative humidity measurements would be indicative of water percolation into the cavity or a breach of the SDS vessel containment. Since the humidity data can be acquired with minimum effort when the particulate sampling is done, it is recommended that the same semiannual sampling frequency be used for the next 2 yr.

4.5 SUMMARY

As stated in Sections 4.1 through 4.4, it is recommended that all four systems be monitored semiannually, during February and August, for the next 2 yr. To minimize cost, formal reporting by updating this report should be required biennially (1991), assuming that no significant abnormalities occur. The need for subsequent monitoring should be determined in 1991.

5.0 REFERENCES

DOE, 1988, "Radioactive Waste Management," DOE Order 5820.2A, U.S. Department of Energy, Washington, D.C., September 1988.

Henrie, J. O., and G. D. Murry, 1987, *Analysis of the Three Mile Island (TMI) Submerged Demineralization System (SDS) Vessel Burial Data*, WHC-EP-0083, Westinghouse Hanford Company, Richland, Washington.

Henrie, J. O., and G. D. Murry, 1986, *Analysis of the Three Mile Island (TMI) Submerged Demineralizer System (SDS) Vessel Burial Data*, RHO-RE-ST-49 P, Rockwell Hanford Operations, Richland, Washington.

Henrie, J. O., D. J. Flesher, G. J. Quinn, and J. Greenborg, 1986, *Hydrogen Control in the Handling, Shipping and Storage of Wet Radioactive Waste*, GEND-052, EG&G Idaho, Inc., Idaho Falls, Idaho.

Quinn, G. J., J. O. Henrie, and J. Greenborg, 1984, *Submerged Demineralizer System Vessel Shipment Report*, GEND-035, EG&G Idaho, Inc., Idaho Falls, Idaho.

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