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**Volume Reduction System for Solid and
Liquid TRU Waste from the Nuclear Fuel
Cycle: October-December 1977**

Don F. Luthy and William H. Bond

April 21, 1978

Monsanto

MOUND FACILITY

Miamisburg, Ohio

operated by

MONSANTO RESEARCH CORPORATION

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for the

U. S. DEPARTMENT OF ENERGY

Contract No. EY-76-C-04-0053

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Foreword

Under the sponsorship of the DOE Division of Waste Management, Production and Reprocessing, Mound is responsible for the development of design criteria for a volume reduction system to treat combustible solid and liquid waste generated in the nuclear industry.

This report is submitted by W. T. Cave, Director, Nuclear Operations and B. R. Kokenge, Manager, Nuclear Technology, from contributions prepared by members of the Nuclear Waste Technology Section: K. V. Gilbert, Manager, The Volume Reduction Technology group: D. F. Luthy, Leader and the Incinerator Technology project: W. H. Bond, Leader.

Previous reports of this and the related project, TRU Waste Cyclone Drum Incinerator and Treatment System, are listed below:

Volume Reduction System for Solid and Liquid TRU Waste from the Nuclear Fuel Cycle:

July-September 1977	MLM-2498
April-June 1977	MLM-2463
January-March 1977	MLM-2436

TRU Waste Cyclone Drum Incinerator and Treatment System:

July-September 1977	MLM-2496
April-June 1977	MLM-2482
January-March 1977	MLM-MU-77-64-0004

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Abstract

A bench-scale incinerator for the investigation of unusual particulate and gaseous radioactive material in the incinerator off-gas was assembled and equipment checkout was begun.

Modifications to a glovebox to be used for the demonstration phase of incinerator-ash immobilization are approximately 80% completed with demonstration to begin next quarter.

The curing time for ash-cement pressed pellets was optimized by periodic crush-strength testing of open-air and underwater cured samples.

Leach tests were performed at 70°C and 100°C to simulate normal and extreme storage conditions. Long-term leach tests were initiated on plutonium-238 doped ash-cement pressed pellets in distilled water at ambient temperature. Pressed pellets of bone char, ash, and sludge-ash in several mixtures were fabricated and tested to establish pressing and curing parameters. Leach tests were also performed on bone char-cement pressed pellets.

Curing studies have been conducted on the pressed pellet matrix to define differences between curing in the open atmosphere and under water. Leachability studies have been initiated on the pressed pellet ash/cement matrix in distilled water at ambient, 70°C and 100°C. Also, leachability studies on doped plutonium-238 pressed ash/cement pellets and pressed bone char/cement pellets in distilled water at ambient temperature have been conducted. Compressive strengths of bone char/cement and sludge/cement pressed matrix have been investigated.

Introduction

This project is primarily concerned with the development of design criteria for a volume-reduction system to treat combustible solid and liquid waste generated in the nuclear industry.

The identification and removal of radioactive gases and particles from the incinerator off-gas, different from those encountered in burning plutonium-contaminated wastes, are required for complete design criteria.

The demonstration of an incinerator-ash-immobilization process is an intermediate step toward defining the process description and the design criteria. Complete design criteria for the entire volume reduction system will include those determined during the ash-immobilization demonstration phase.

Effects of burning beta and gamma contaminated waste

The bench scale incinerator system has been assembled and is ready for preliminary experimental runs (Figure 1). The existing system includes a combustion chamber and off-gas cleaning equipment designed to control emissions of particulate material and hydrochloric acid vapor formed by the incineration of plastic. Capacity of the combustion chamber is approximately 1.5 ft³, and the off-gas flow rate is anticipated to be 15-20 std. ft³/min.

Hot off-gases from the burn chamber, passing through the scrubber tank, will be sprayed with a cold caustic solution for removal of particulates and hydrochloric

acid mist, as well as cooling the off-gas. The gas will then pass through a mist eliminator packed with polypropylene mesh to remove any large particles remaining in the gas stream and any entrained droplets of scrubber solution. The off-gas then passes through Balston Microfibre filter tubes capable of retaining 99.99% of 0.6 μ m particles. Finally, the filtered off-gas passes to the building exhaust system.

Other equipment may be added to the incinerator system in the future to enhance removal of radioisotopes which may be found in low-level commercial wastes. Experimentation on the laboratory system will begin in CY-1978.

Ash immobilization

Demonstration

Modifications to the glovebox will be completed early next quarter and equipment installation and check-out will be accomplished during the quarter.

This glovebox will be used for demonstrating the ash immobilization technique recommended under Task II of the TRU Waste Cyclone Drum Incinerator and Treatment System project.

FIGURE 1 - Bench scale incinerator.

Curing time of pressed pellets

Two groups of pressed pellets were prepared to determine the time required for curing 1/2-in. diameter pellets and to define the difference between curing in the open atmosphere and under water. Table 1 lists the cement-ash pellets (30% Ash III [1], 70% Portland 1A cement). Approximately 97% of the total strength of the open-cure pellets was reached after one day. For the underwater, or slow-cure process, the pressed pellets reached maximum strength after four days. The higher crush-strength values in Table 1 are representative of the underwater cure samples (approximately 5% increase).

Crush strengths of pressed bone char and sludge pellets

Bone char-cement and sludge-cement pellets were prepared and tested to determine a crush strength range for this type of waste. Also, leachability and mechanical shaking tests were initiated on the 1/2-in. diameter pressed pellets. Tables 2 and 3 provide data indicating the average crush strength and pellet densities for the various mixtures. As can be seen, the Type II bone-char pellets had a higher crush strength and higher density and were more homogeneous throughout than the corresponding Type I bone-char pellets. The underwater or slow-cure process produced slightly higher crush strength values for each pellet mixture when compared to the dry-cure process. (The higher crush strength and higher density for the Type II pellets are probably due to better packing of the bone char and cement.)

The two mixtures used for sludge pressed pellets [2] showed that the wet-cure method again produced higher crush strengths. The heating of the four sludge samples at 110°C for 1 hr appears to have increased the crush strength slightly.

Leach tests at elevated temperatures

Pressed pellets, 1/2-in. in diameter by 1/2-in. long, were immersed in distilled water at different temperatures for 24 hr to determine leach rates. The temperatures used in these experiments were ambient, 70°C, and 100°C. Figure 2 is a plot of average leach rate* as a function of temperature (°C). As can be seen, the leach rate increased only slightly (~14%) between ambient and 70°C. However, at 100°C, the leach rate was much higher (99%) than that at ambient temperature. Since the temperature in the burial location is expected to be 70°C or less, these experiments show that temperature will have very little effect on the leach rate in water.

Plutonium-238 doped pressed pellets

Several 1-in. diameter pellets of various composition were pressed at 13,000 psi. It should be noted that the plutonium content of each pellet charge was identical (1.12 mg of plutonium-238). These pellets were composed of Type I incinerator ash [1] and Portland 1A cement. The water to

*Surface area used for calculation was the geometric area of cylinder. This leach rate was calculated by dividing the total amount of material leached from the pellet by the product of surface area and total leach time.

Table 1 - EFFECT OF CURE TIME ON CRUSH STRENGTH
OF PRESSED PELLETS (ASH-PORTLAND 1A CEMENT)

Curing Time (hr)	Open Atmosphere Cure Average Crush Strength (psi)	Underwater Cure Average Crush Strength (psi)
24	3510	3691
48	3605	3744
72	3599	3771
96	3590	3808

Table 2 - AVERAGE CRUSH STRENGTH OF PRESSED
PELLETS (BONE CHAR-PORTLAND 1A CEMENT)

Mixture Bone Char/ Cement	Bone Char ^a	Crush Strength (Dry Cure) (psi)	Density (g/cm ³)	Crush Strength (Wet Cure) (psi)	Density (g/cm ³)
20/80	Type I	3615	2.25	3826	2.49
	Type II	3726	2.30	3992	2.50
30/70	Type I	3650	2.22	3736	2.42
	Type II	3665	2.31	3836	2.48
40/60	Type I	3620	2.13	3656	2.31
	Type II	3630	2.27	3776	2.48
50/50	Type I	3600	2.12	3661	2.29
	Type II	3640	2.27	3786	2.38
60/40	Type I	3590	2.09	3636	2.07
	Type II	3595	2.19	3766	2.36
70/30	Type I	3554	2.03	3585	2.22
	Type II	3615	2.20	3701	2.33
80/20	Type I	2517	2.00	3524	2.30
	Type II	3590	2.15	3610	2.38

^aType I - Denotes sintered bone char (25% smaller than 500 μ m)

Type II - Denotes crushed sintered bone char (86% smaller than 500 μ m)

Table 3 - AVERAGE CRUSH STRENGTH OF PRESSED
PELLETS (SLUDGE-PORTLAND 1A CEMENT)

Mixture Sludge/Cement	No. of Samples	Crush Strength (Dry Cure) (psi)	Density (g/cm ³)	Crush Strength (Wet Cure) (psi)	Density Average (g/cm ³)
20/80	9	3631	2.28	4185	2.34
		3651 H	2.24	4198 H	2.26
15/85	10	3696	2.45	4118	2.52
		3761 H	2.36	4197 H	2.40

H - Denotes that the sample was heated at 110°C for 1 hr prior to
crush strength test.

FIGURE 2 - Leach rate as a function of temperature.

cement weight ratio* was constant at 0.2 in all samples prepared. The pellets were placed in distilled water at ambient temperature. Each week, samples of the leach solutions were analyzed for plutonium-238 content. The total leach solution volume (250 ml) was changed weekly. Figure 3 is a plot of average plutonium-238 leach rate as a function of pellet composition. The highest leach rate occurred for pellets made of 60% ash, 40% cement, and the lowest rate was for pellets of 30% ash, 70% cement. The cement-ash pelletization appears to be an excellent method of immobilizing incinerator ash containing plutonium. All leach rates are of the order 10^{-11} to 10^{-12} g/cm²-hr. Brine leach tests have been done, but the data are inconclusive at this time. Figure 4 is a graphical representation depicting the slow leaching of plutonium-238 from a representative

*This was the ratio before pressing. The ratio after pressing was lower since water was released from the pellet charge during forming of the pellet.

FIGURE 3 - Plutonium-238 leach rate as a function of pellet composition (0.2 water/cement ratio, wet cured).

FIGURE 4 - Percent of plutonium-238 remaining as a function of time leached.

pellet (30% ash, 70% cement). As can be seen, significant leaching occurred during the first week (0.03%). During the next 11 weeks, less than 0.01% leached out of the pellet. This is the typical leaching behavior; rapid initially, then slow thereafter.

Bone-char pellet leach tests

Several 1/2-in. diameter pellets of various composition were pressed at 25,000 psi. These pellets were composed of Type I and Type II bone char* and Portland 1A cement. The pellets contained no radioactive isotopes. The water-to-cement ratio** varied from 0.20 to 0.24 with the various bone char-cement mixtures. The pellets were leached in 80 ml of distilled water at ambient temperature. The leach water was changed weekly for the first month and monthly thereafter. The total amount of material leached from a pellet was determined by boiling the leach solution to dryness and weighing the residue. Table 4 contains the data obtained from these experiments. Analysis of these data will show that, in general, the wet-cure method produced pellets having lower leach rates. These lower leach rates are probably caused by the wet-cure pellets experiencing some initial leaching during the two-week under-water curing process. Figure 5 is a plot of average leach rate as a function of pellet composition (percent bone char).

*See Table 2 notes

**These were the ratios before pressing of the pellets. The ratios are lower after pressing because water is released from the pellet charge upon pressing.

This plot is for pellets of Type II bone char and 0.2 water/cement ratio (before pressing) which were dry cured. It should be noted that this curve is similar to Figure 3.

Additional tests will be done next quarter using higher bone-char percentages (50, 60, 70, and 80%) and a 0.2 water/cement ratio. It should also be noted that, in general, the density of a certain type pellet (Type I, wet-cured, 0.2 water/cement ratio, for example) decreases with increasing weight percent bone char. This is probably because the density of bone char is lower than that of cement. It is not known presently if increasing leach rate is related to decreasing density but the phenomenon will be also studied next quarter.

FIGURE 5 - Leach rate as a function of pellet composition (Type II bone char, 0.2 water/cement ratio, and dry cured).

Table 4 - AVERAGE LEACH RATE OF PRESSED BONE CHAR-PORTLAND 1A CEMENT PELLETS

Sample Number	Cure Method	Type Bone Char	Water/Cement Ratio	Bone Char ^a (%)	Density (g/cm ³)	Average Leach Rate (g/cm ² /hr)
BCP-6	Dry	I	0.24	50	2.12	1.73×10^{-5}
BCP-8	Wet	I	0.24	50	2.29	7.24×10^{-6}
BCP-10	Dry	II	0.24	50	2.27	2.33×10^{-5}
BCP-12	Wet	II	0.24	50	2.38	9.46×10^{-6}
BCP-14	Dry	I	0.20	40	2.13	1.60×10^{-5}
BCP-16	Wet	I	0.20	40	2.31	5.68×10^{-6}
BCP-18	Dry	II	0.20	40	2.27	1.65×10^{-5}
BCP-20	Wet	II	0.20	40	2.48	8.83×10^{-6}
BCP-22	Dry	I	0.2-	30	2.22	1.62×10^{-5}
BCP-24	Wet	I	0.20	30	2.42	5.60×10^{-6}
BCP-26	Dry	II	0.20	30	2.31	1.39×10^{-5}
BCP-28	Wet	II	0.20	30	2.48	3.85×10^{-6}
BCP-30	Dry	II	0.20	20	2.30	1.21×10^{-5}
BCP-32	Wet	II	0.20	20	2.50	5.60×10^{-6}

^aThis is weight percent bone char. The remaining percentage is cement.

References

1. TRU Waste Cyclone Incinerator and Treatment System: January-March 1977,
MLM-MU-77-64-0004, p. 17.
2. Ibid, p. 19.

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