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**Pelletized Waste Form
Demonstration Program:
October 1980 - March 1981**

Edward L. Lewis and Robert F. Herbert, Jr.

May 14, 1981

Monsanto

MOUND FACILITY
Miamisburg, Ohio 45342

operated by
MONSANTO RESEARCH CORPORATION
a subsidiary of Monsanto Company

for the
U. S. DEPARTMENT OF ENERGY
Contract No. DE-AC04-76-DP00053

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Abstract

During the last six months, performance testing of waste/cement pellets was continued. These evaluations included leachability tests and compressive strength tests of "cold" soil/cement pellets of various compositions. Fractional leach rates (g/cm²/day) after 21 months of testing were, in all cases, $< 1 \times 10^{-5}$ g/cm²/day (Mound Acceptance Value). Based upon these recent data, the pressed waste/cement pellets appeared to be a suitable matrix for the immobilization of low-level transuranic wastes.

The installation of the Carver custom pellet press was completed. Plutonium-238 contaminated (< 100 nCi/g) ash/cement pellets were produced at a rate of 360 pellets/hr. Pellets of two different compositions were produced, 50% ash/50% cement and 65% ash/35% cement. The compressive strength of sample pellets was slightly lower than expected. Static MCC-1 leachability testing as well as long-term radiolysis testing of sample pellets are scheduled.

Introduction

The main objective of this program is to evaluate TRU waste immobilization methods. Wastes such as Mound Facility cyclone incinerator ash, sludge, and process residues (salts, etc.) will be immobilized by mixing with Portland 1A cement, water, and special additives, if necessary, and pressing the mixture into pellets. Sample pellets are tested for radiolytic gas generation, leachability, thermal stability, and compressive strength to determine whether the waste matrix meets Mound Facility present Acceptance Criteria. A project Milestone Work Plan for this program is included in Figure 1.

Performance Testing

The leachability and compressive strength of waste/cement pellets were tested during the last six months. Because contaminated soil now presents a realistic waste disposal problem, simulated pellets of "cold" soil/cement were

INTERMEDIATE EVENTS/MILESTONES	O	N	D	J	F	M	A	M	J	J	A	S
1.2 Performance Measurements												
1.2.2 Long-Term Testing												
1.2.3 MCC-1 Short-Term Testing												
1.5 Demonstration												
1.5.1 Cold Waste Pelletization												
1.5.2 Hot Waste Pelletization												
1.5.3 Final System Report												

Figure 1 - FY-1981 Milestone Report.

manufactured and tested for compressive strength. These soil/cement pellets were $\sim \frac{1}{2}$ in. in diameter, $\sim \frac{1}{2}$ in. in length, and were pressed at 25,000 psi. Six random soil samples were collected from various locations within Mound Facility, and these samples were then blended to form one representative soil sample for the preliminary screening experiments. The soil used in these experiments was calcined at approximately 1000°C for 1½ hr to remove organic matter. In some pellets, soil of <25 mesh/in. was used (Table 1), and in other pellets, soil having a particle size <80 mesh/in. was used. All pellets were dry-cured in the open atmosphere at ambient temperature (20°C). As can be seen in Table 2, the compressive strength of soil/cement pellets increased, as expected, with an increase in cement percentage. The 40% soil/60% cement pellets (L/D = 0.89) had an average compressive strength of 3570 psi after 20 days of dry-curing and almost met the present Mound Facility Acceptance Criteria for compressive strength (>3500 psi, L/D = 1.0). The density of the pellets increased with increasing cement content. This was as expected since cement has a higher density than calcined soil. Preliminary data also indicate that by decreasing the soil particle size the strength of the soil/cement pellets can be increased.

Leachability testing of waste/cement pellets at both ambient temperature (22°C) and 70°C has now been in progress for 21 months. The general shape of a plot of fractional leach rate as a function of time is shown in Figure 2 [1]. The low

Table 1 - PARTICLE SIZE DISTRIBUTION OF CALCINED SOIL (<25 mesh/inch)

Particle Size (mesh/in.)	Fraction of Total (%)
> 25	0.0
< 25 > 45	53.0
< 45 > 80	26.3
< 80 > 120	4.7
< 120	16.0

leach rates measured are encouraging. The average fractional leach rate of 65 wt % ash/35 wt % cement pellets (containing 0.3 wt % plutonium-238 oxide) in saturated NaCl brine solution at 70°C after ~ 14 months of leaching is $\sim 3.5 \times 10^{-8}$ (g)(cm)⁻²(day)⁻¹. As a comparison, the average fractional leach rate of identical pellets after 14 months of leaching in saturated brine at ambient temperature was $\sim 1.8 \times 10^{-9}$.

The fractional leach rates of ash/cement pellets in deionized water were also determined. The average fractional leach rate of 65 wt % ash/35 wt % cement pellets (0.3 wt % plutonium-238 oxide) in deionized water at ambient temperature (22°C) after 14 months of leaching is $\sim 7.6 \times 10^{-8}$ (g)(cm)⁻²(day)⁻¹. This is approximately 40 times higher than the fractional leach rate found previously for similar pellets in saturated brine under similar conditions. The higher rate is thought to result from the thin surface layer of NaCl crystals that have

Table 2 - PROPERTIES OF CALCINED SOIL/CEMENT PELLETS

Pellet Composition		Avg. L/D	Avg. Compressive Strength (psi)	Avg. Density (g/cm ³)	Time Dry-Cured (days)
(wt % soil < 80 mesh/in.)	(wt % cement)				
40	60	0.89	3570	2.08	20
50	50	0.92	2890	1.98	14
60	40	0.95	1875	1.94	14
50 ^a	50	0.93	1624	1.96	6

^aCalcined soil was <25 mesh/in.

See Table 1 for particle size distribution.

been seen on pellets in brine solution.

The leachability testing of sludge/cement pellets has been in progress for 19 months. Pellets containing 50 wt % sludge solids/50 wt % cement and 0.3 wt % plutonium-238 oxide were leached in saturated brine solution at both ambient temperature and an elevated temperature of 70°C. The average fractional leach rate after 19 months of leaching was 1.5×10^{-9} (g)(cm)⁻²(day)⁻¹ at 70°C, a factor of 550 greater than the rate found at ambient temperature [$\sim 8.3 \times 10^{-11}$ (g)(cm)⁻²(day)⁻¹].

Leach data are now available for pellets manufactured at two pressures, 10,000 psi and 25,000 psi. Pellets composed of 65% ash/35% cement and 0.3 wt % plutonium-238 oxide were leached in saturated brine solution at ambient temperature. After 165 days of leaching, the low pressure pellets had an average fractional leach rate of 9.9×10^{-11} (g)(cm)⁻²(day)⁻¹, while the high-pressure pellets had an average fractional leach rate of 2.7×10^{-10} (g)(cm)⁻²(day)⁻¹. These rates are within experimental error of the counting technique and therefore the effect of manufacturing pressure (in the range studied) on leachability rate appears to be extremely small. Since pressed waste/cement meet Mound Facility Leachability Criteria [leach rate of $< 1 \times 10^{-5}$ (fraction leached)(g)(cm)⁻²(day)⁻¹], these pellets

Figure 2 - A typical plot of the fractional leach rate as a function of time.

appear at present to be a suitable matrix for some transuranic-contaminated wastes based on the Leachability Test Procedure used to measure the leach rates.

Production Waste Pelletization Line

Installation

The installation of the new custom pellet press manufactured by Carver Company was completed in March 1981. This press replaces the semiautomatic laboratory press which was used for five months to produce ash/cement pellets. The custom pellet press was modified by the substitution of a small, rectangular hopper for the large, circular one. The addition of an adjustable relay now permits the pressing dwell time to be varied from 0.5 to 100 sec.

Demonstration

The pelletization process was demonstrated using ash contaminated with plutonium-238 from the Mound Facility cyclone incinerator. The first "hot" demonstration of the semiautomatic press was performed using calcined, plutonium-238 contaminated ash (< 100 nCi/g) from the pilot line. Several pellets were manufactured by pressing calcined ash, cement, and water at 25,000 psi in a 1-in. diameter die. The pellets had an L/D range of 0.5-0.8. The 65% ash/35% cement pellets were dry-cured for one week and then tested for compressive strength. The average compressive strength of pellets with a L/D ratio of 0.82 was 3560 psi.

The second demonstration of the pelletization process was conducted using plutonium-238 contaminated sludge from the waste treatment facility in the semiautomatic press and a 1-in. die. Several pellets were manufactured having a 65% uncalcined sludge solids/35% cement composition. A water/cement ratio of 0.6 was used, and the pellets were dry-cured one week before testing. The average compressive strength of the pellets was approximately 750 psi for an L/D of 1.02. The low strength is primarily the result of the high carbon content of the sludge.

The third demonstration of the pelletization process was conducted using the new Carver custom pellet press. The ash pelletized was from the calcination of two batches of plutonium-238 contaminated ash (< 100 nCi/g) in the new calcination furnace. The ash had been calcined

in an Inconel retort at 850°C for 3 hr. The total weight loss was approximately 10%, and the carbon content was reduced from 7.2% to an acceptable 0.25%. After calcination the ash was blended with Portland 1A cement and water, and pellets were manufactured at approximately 20,000 psi. The press produces 360 pellets/hr, each pellet weighing approximately 20 g. Pellets of two different compositions were produced, one type containing 65% ash/35% cement and the other type 50% ash/50% cement. Sample pellets were tested for compressive strength after dry-curing in the open atmosphere for 5 days.

Figure 3 - Pellet compressive strength as a function of L/D ratio.

The compressive strengths for various length/diameter pellet ratios are shown in Figure 3. Pellets with L/D ratios of 1.0 should have compressive strengths of 2500 psi (estimated by extending curves) and therefore will not meet the present Mound Facility Acceptance Criteria of >3500 psi, L/D = 1, 7-day dry-cure. The low compressive strengths may result from improper water/cement ratios. The general shape of a plot of compressive strength as a function of water/cement ratio is shown in Figure 4 [2]. As can be seen, if a water/cement ratio less or greater than the optimum is used, a compressive strength less than the optimum will also be attained. Since the exact chemical composition of the ash is not known, one or more compounds present in the ash could be responsible for retarding the rate of strength development. For example, the presence of small amounts of lead oxide or zinc oxide can very seriously retard the hardening of Portland cement concrete. [3]

During May 1981, the MCC-1 static leach tests will be initiated using production-line pellets. The equipment and supplies necessary to perform this task are now available. Long-term radiolysis testing of 1-in. diameter production line pellets will also be performed in the near future. This will require a small modification of the existing radiolysis system to accommodate larger pellets.

Figure 4 - A generalized plot of the compressive strength as a function of water/cement ratio.

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