MLM-2717

Pelletized Waste Form Demonstration Program: October 1979-March 1980

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

June 6, 1980

MOUND FACILITY
Miamisburg, Ohio 45342

operated by
MONSANTO RESEARCH CORPORATION
a subsidiary of Monsanto Company

Monsanto

for the
U. S. DEPARTMENT OF ENERGY

Contract No. DE-AC04-76-DP00053

NOTICE

Printed in the United States of America
Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161

NTIS price codes
Printed copy: A02
Microfiche copy: A01

MLM-2717
UC-70

Pelletized Waste Form Demonstration Program: October 1979-March 1980

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

Issued: June 6, 1980

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

Foreword

Under the sponsorship of the DOE Division of Waste Management, Mound is responsible for the development and demonstration of a pelletized waste form for treatment and fixation of incinerator ash and process sludge generated at DOE sites. The demonstration of this pelletized waste form will be realized during FY-1979 through FY-1981. Milestones for the project are shown on page 5.

This report is submitted by W. T. Cave, Nuclear Operations, and B. R. Kokenge, Manager, Nuclear Technology, from contributions prepared by members of the Nuclear Waste Technology Section, K. V. Gilbert, Manager, and the Solid Volume Reduction Technology Group, J. W. Doty, Leader.

Previous reports in this series are:

MLM-2510	MLM-2575
MLM-2516	MLM-2614
MLM-2541	MLM-2670

Contents

	Page
ABSTRACT	4
INTRODUCTION	4
PERFORMANCE TESTING	4
Radiolysis Experiments	4
Leachability Experiments	6
DESIGN CRITERIA	7
EQUIPMENT PROCUREMENT AND INSTALLATION	9
REFERENCES	9
DISTRIBUTION	10

Abstract

The main objective of this task is to evaluate several transuranic waste immobilization methods. During the first half of FY-1980, ash/cement and sludge/cement pellets were performance tested. In short-term radiolysis experiments with sludge/cement pellets, a very low hydrogen generation rate of 0.01 molecule/100 eV was obtained. Leachability testing of ash/cement and sludge/cement pellets containing plutonium-238 oxide in both brine and deionized water yielded a very low leach rate of $<10^{-5}$ (fraction leached) $(g)(cm)^{-2}$ $(day)^{-1}$. Pressed waste/cement pellets, therefore, appear to be an excellent final waste form based on leachability and radiolysis results. Design criteria were developed for the production waste pelletization line (PWPL) and documented. Equipment procurement for the PWPL was started, and installation will be continuing for several months.

Introduction

The main objective of this task is to evaluate several TRU waste immobilization methods. Wastes such as cyclone incinerator ash, sludge, and salts will be immobilized by mixing each with Portland 1A cement, water, and special additives, if necessary, and pressing the mixture into pellets.

Technical support is continuing for the defense cyclone incinerator system to ensure that incinerator ash and sludges from the aqueous offgas scrubbing system can be effectively immobilized. The immobilized waste matrix under development at Mound Facility is a pressed, high-density, low-porosity pellet. A Project Milestone Work Plan for this technical support is included in Figure 1.

Performance testing

Radiolysis and leachability testing of waste/cement pellets was started in late FY-1979. Short-term experiments have been completed and long-term experiments are still in progress.

Radiolysis experiments

In radiolysis experiments conducted in late FY-1979 on 65% Mound ash/35% cement pellets (dry basis), a hydrogen generation value, $G_{(H_2)}$, of 0.013 molecules/100 eV was obtained. During the past several months, radiolysis experiments were performed using 50% Mound sludge/50% cement pellets. The sludge was obtained from the cyclone incinerator vertical leaf filter unit and was calcined at 750°C for approximately 3 hr. The sludge was then mixed with Portland cement (Type 1A) and water and pressed into pellets 1/2 in. in diameter by 1/2 in. long. A manufacturing pressure of approximately 25,000 psi was used in the fabrication of the sludge/cement pellets. Table 1 contains the data for two radiolysis experiments. The average $G_{(H_2)}$ value of 0.010 molecule/100 eV was slightly less than that found previously for ash/cement pellets. These $G_{(H_2)}$ values are very small, and it appears that pressed pellets are an excellent final waste form. The calculated ratio of $G_{(oxygen)}/G_{(H_2)}$ was 0.38. Thus, a less than stoichiometric amount of oxygen was generated, and hence, oxidation products of water such as H_2O_2 are possibly being formed.

Title: Pelletized Waste Form Demonstration Program
W/E No.: AL 2.4.7.1 D

Date: February 1980

INTERMEDIATE EVENTS/MILESTONES	O	N	D	J	F	M	A	M	J	J	A	S	
1.2 Performance Measurements	XXX	XXX	XXX	XXX	XXX	XXX	XXX	XXX	XXX	XXX	XXX	XXX	a
1.2.1 Short-Term Testing	XXX	XXX	XXX	XXX	XXX	XXX							
1.2.2 Long-Term Testing	XXX	XXX	XXX	XXX	XXX	XXX	XXX	XXX	XXX	XXX	XXX	XXX	a
1.3 Design Criteria	XXX	XXX	XXX										
Criticality Review	XXX												
Health Physics Review	XXX												
Engineering Specifications	XXX	XXX											
QC Requirements	XXX	XXX	XXX										
Publish Design Criteria Report			XXX										
1.4 Procurement and Installation		XXX	XXX	XXX	XXX	XXX	XXX	XXX					
1.4.1 Vendor Selection		XXX	XXX										
Purchase Reqs Prepared		XXX											
Final Orders Placed			XXX										
1.4.2 QC/QA Requirements		XXX	XXX	XXX	XXX								
Equipment Specifications Review		XXX	XXX	XXX									
Equipment Verified for Receipt				XXX	XXX								
1.4.3 Equipment Installation				XXX	XXX	XXX	XXX						
Engineering Work Order Prepared				XXX									
Initiate Installation						XXX	XXX						
Complete Installation							XXX						
1.4.4 Process Review								XXX					
Safety Review								XXX					
Process Operation Checkout								XXX					
System Approved for Use								XXX					

Note: Lower case letters are used for FY-1979 intermediate events.

Upper case letters are used for milestones and events which also appear in the Work Element Plan.

[a]Extend beyond FY-1980; See Work Element Plan

FIGURE 1 - FY-1980 milestone report.

Table 1 - EFFECT OF ALPHA RADIOLYSIS ON PRESSED SLUDGE/CEMENT PELLETS

Run No.	Pu-238 (Ci)	Irradiation Time (hr)	$G_{(H_2)}$ (molecules/100 eV)	Number of Pellets Tested	Main Sludge Components
1	0.55	187	0.009	2	Na, Fe, Si
2	0.55	630	0.011	2	Na, Fe, Si

Leachability experiments

Leach tests, in progress now for almost one year, have been conducted with incinerator ash/cement and incinerator sludge/cement pellets at both ambient temperature and 70°C. Two types of leachants have been used, saturated NaCl brine and deionized water. Two test procedures have been used: (1) WIPP (Waste Isolation Pilot Plant) and (2) ANS (American Nuclear Society). The pellets, which are approximately 1/2 in. in diameter by 1/2 in. long, contain approximately 0.25 wt % plutonium-238 oxide. Tests are conducted by suspending the pellets in the leachant for the desired time period and then determing the plutonium-238 content of the entire leachate by liquid scintillation counting.

A comparison of the leach rates of 65 wt % incinerator ash/35 wt % Portland cement pellets in saturated brine solution and in deionized water is shown in Figure 2. The WIPP test procedure was employed in these two experiments which were conducted at ambient temperature (∿22°C). The leach rate is expressed in (fraction leached) $(g)(cm)^{-2}(day)^{-1}$, and the time is expressed in average days leached. Both terms are defined in the leachability section of a previous report [Lewis, 1979]. In brine the leach rate decreases with time, and in deionized water the leach rate is cyclic (10^{-6} to 10^{-7} range). Initially

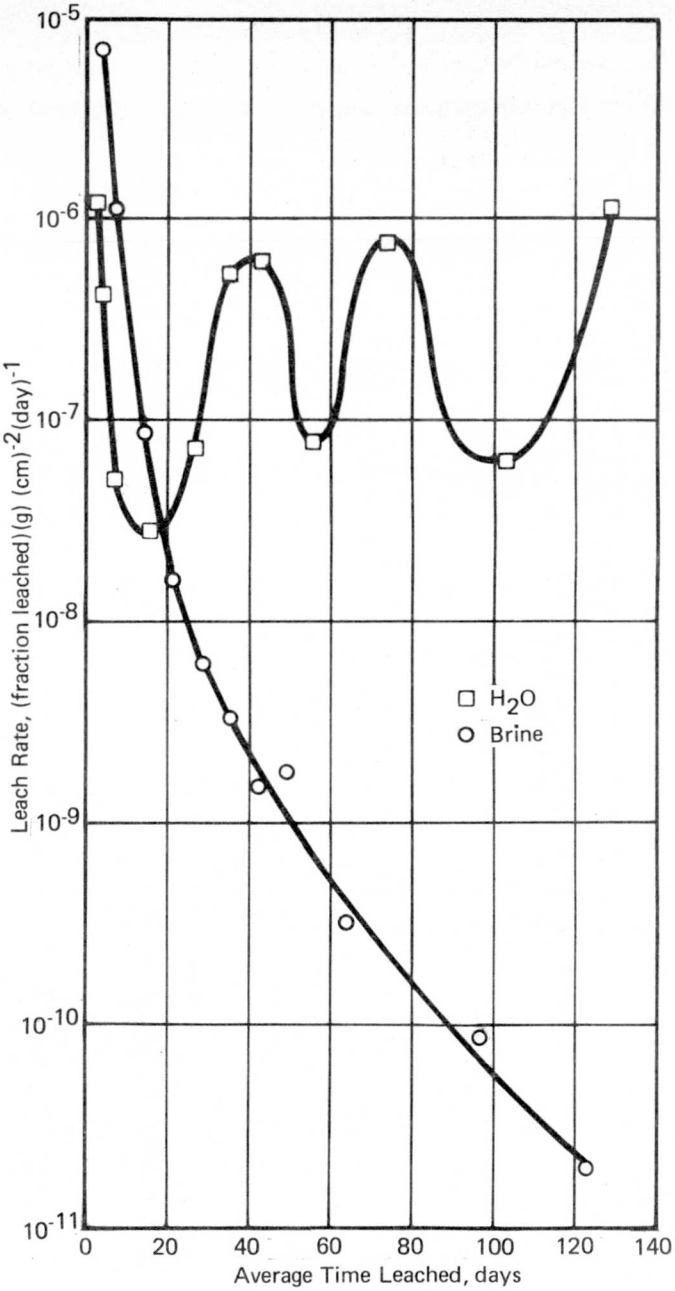

FIGURE 2 - Comparison of leach rates of ash/cement pellets in saturated brine solution and deionized water.

(<18 days) the leach rate is less in deionized water than in brine. The two leach rates are equal (3×10^{-8}) at ∿18 days, but after 18 days, the leach rate in deionized water is greater than that in brine. After 125 days there is a great difference in the leach rate (almost five magnitudes). The leach rates for plutonium-238 oxide are, in general, very small ($<10^{-5}$ g/cm^2/day), and the pressed pellets appear to be a good waste form based on short-term testing data. Long-term leachability testing of the waste forms is now in progress.

The ANS leachability test procedure was also employed in a separate experiment. The leach rates found in deionized water at ambient temperature were almost identical to those determined by the WIPP procedure. This was expected since the procedures are almost identical except for initial differences in leachate sampling frequency.

Leachability testing of ash/cement pellets in saturated brine solution at 70°C was repeated because of failure of the pellets in the first experiments. The initial two pellets broke apart after 75 days and 99 days of leaching time. Two identical ash/cement pellets used in the repeat experiments have now been in solution for 190 days, and the pellets are still intact with no signs of deterioration. The average leach rate for the two pellets after 151 days is 2.6×10^{-8} g/cm^2/day. It now appears that the first two pellets tested were defective in their manufacture.

The leachability studies of sludge/cement pellets is also continuing. These pellets contain 50 wt % incinerator sludge/50 wt % Portland cement and are spiked with 0.25 wt % plutonium-238 oxide. Leach studies are being conducted at ambient temperature and at 70°C. The average leach rate of sludge/cement pellets after 151 days of leaching at ambient temperature was 7×10^{-11} g/cm^2/day (see Figure 3). For comparison, the average ambient leach rate of ash/cement pellets after an identical time period was approximately the same (6×10^{-11} g/cm^2/day). The sludge/cement pellets appear to have a surface layer of crystals, but no such phenomenon has been observed on the ash/cement pellets. After 20 days of leaching, the leach rates for ash/cement and for sludge/cement pellets were 2×10^{-8} and 2×10^{-10} g/cm/day, respectively. It is possible that the surface layer of crystals is partly responsible for the low leach rates of sludge/cement pellets. It should be noted that the leach rate of ash/cement pellets is increasing after reaching a minimum at 122 days while the leach rate of sludge/cement is still decreasing.

Design criteria

Final design criteria for the production waste pelletization line (PWPL) was published in March [Lewis, 1980]. This report includes background information on the use of cement for immobilization of low-level and intermediate-level radioactive waste materials. Also included are a design basis for the pelletization process, process description including a flow diagram, and performance data for process equipment (sizing equipment, calciner, grinder, and press).

Health physics and safety criteria were determined for the PWPL. Equipment enclosures will be metal, welded gloveboxes and must be sufficiently leak-tight to pass a helium "sniffing" leak test. The gloveboxes housing the calciner and the

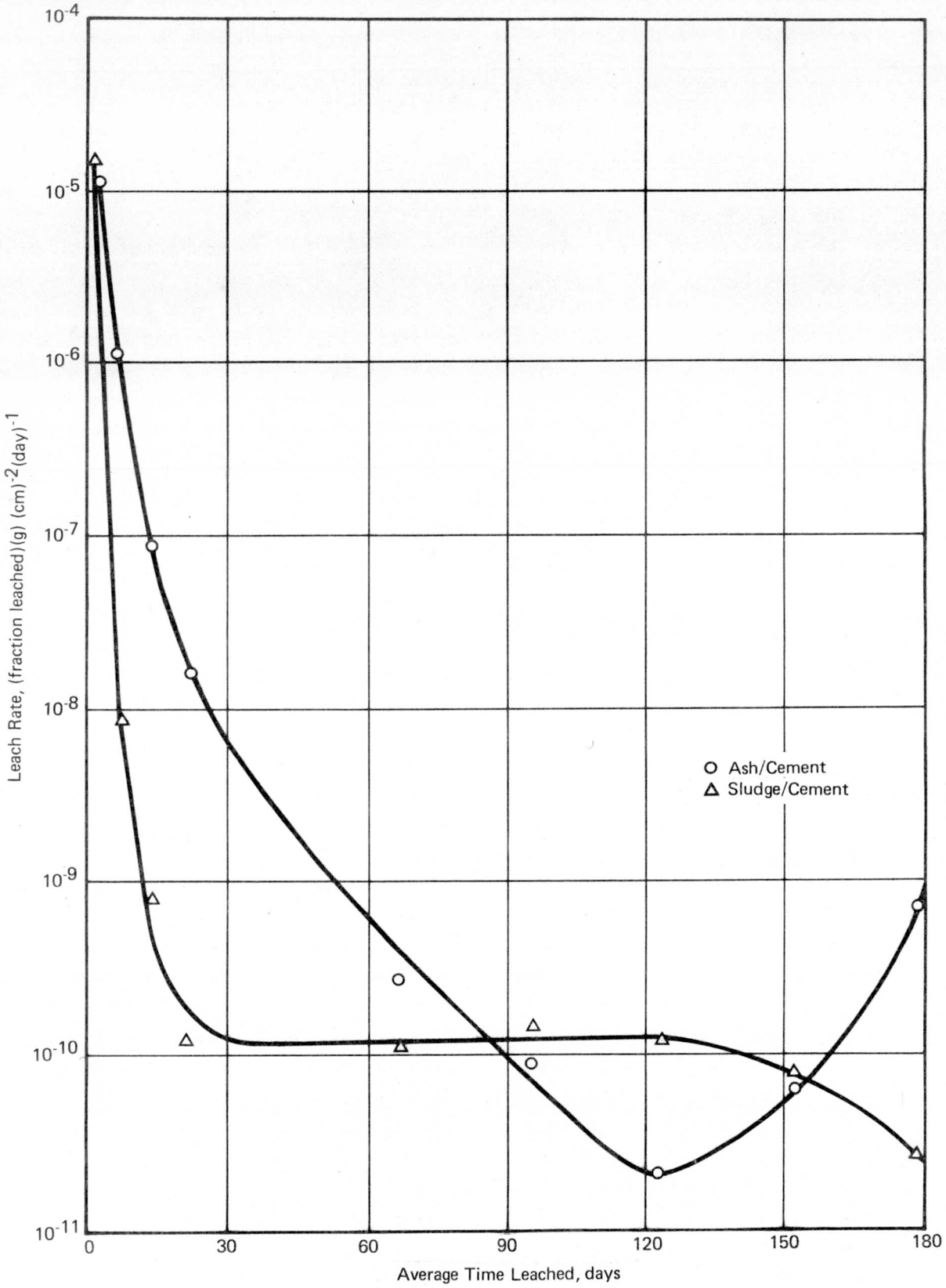

FIGURE 3 - Comparison of leach rates of sludge/cement and ash/cement at ambient temperature.

press must be protected by a Halon fire extinguishing system. Because of the low level of radioactivity handled in the gloveboxes, radiation shielding will not be required.

Equipment procurement and installaition

Equipment for the production waste pelletization line (PWPL) is now being selected and ordered from vendors. Eight vendors were contacted in regard to purchasing the continuous feed, rotary calciner. Only two bids were received for this piece of equipment. The bids of $45,800 and $65,000 were unacceptable because (1) physical size was too large, (2) costs far exceeded available funds, and (3) delivery time was too long (five months) to meet the schedule. Plans are now to purchase a batch type calciner consisting of a metal retort heated in a muffle furnace.

The investigation of pellet presses for the PWPL is continuing. It appears now that either an existing press will be modified or a custom preformer pellet press will be purchased. May 1 has been set as a deadline for press selection.

Two surplus stainless steel gloveboxes have been obtained and will be used for the PWPL. Preliminary engineering drawings have also been prepared for the calcination glovebox and pellet storage/loading glovebox. The grinder, sizing apparatus, and scales have tentatively been selected and will be purchased early in May 1980. It will, however, require several months of installation of the gloveboxes and associated equipment for the completion of the PWPL system.

References

Lewis, E. L. and R. F. Herbert, Jr., Pelletized Waste Form Demonstration Program: April-September 1979, MLM-2670 (December 7, 1979), 23 pp.

Lewis, E. L., Pelletized Waste Form Demonstration Program: Final Design Criteria for Waste Pelletization, MLM-2697 (March 28, 1980), 7 pp.

Distribution

EXTERNAL

TIC, UC-70 (313)
H. N Hill, DOE, Dayton Area Office (2)
T. C. Jones, DOE, Albuquerque Operations Office
A. Sopp, DOE, Albuquerque Operations Office (2)
G. B. Levin, EG&G
R. S. Lowrie, Oak Ridge National Laboratory
R. Y. Lowrey, DOE, Albuquerque Operations Office
G. Oertel, DOE, Division of Waste Management (2)
R. I. Chitwood, DOE, Division of Waste Management
R. D. Walton, Jr., DOE, Division of Waste Management
B. Rawles, Office of Nuclear Waste Isolation Library
R. K. Flitcraft, Monsanto Research Corporation
T. C. Johnson, Nuclear Regulatory Commission
Monsanto Library, St. Louis

INTERNAL

B. M. Alexander
W. R. Amos
W. H. Bond
V. E. Castleberry
W. T. Cave
J. W. Doty
K. V. Gilbert
R. F. Herbert
C. W. Huntington
L. M. Klingler
B. R. Kokenge
E. L. Lewis
J. R. McClain
R. A. Neff
D. L. Prosser
L. B. Stevens
J. E. Todd
R. E. Vallee
Publications
Library (15)

Published by Information Services:
Marjorie F. Hauenstein, Editor