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Volume Reduction System for Solid and Liquid TRU Waste from the Nuclear Fuel Cycle: July-September 1977

Don F. Luthy and William H. Bond

February 6, 1978

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MOUND FACILITY
Miamisburg, Ohio

operated by
MONSANTO RESEARCH CORPORATION
a subsidiary of Monsanto Company

for the
U. S. DEPARTMENT OF ENERGY

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

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Abstract

Laboratory equipment is being assembled for the investigation of unusual particulate and gaseous radioactive material in the incinerator offgas when commercial wastes are incinerated. This equipment will constitute a bench-scale incinerator system with monitoring equipment to effect the investigation. A literature search was made to determine the current technology used in removing the expected effluents from gas streams.

A series of controlled-feed incinerator runs was performed to determine the mass balance of chloride in the Cyclone incinerator system. Approximately 74% of the chloride present in the feed material was found to be in the scrubber solution, 8% in the flue gas, and the remaining chloride was distributed in the ash and retained in the system.

A conceptual design was prepared and modifications were begun on a glove box which is to be used for the demonstration phase of incinerator ash immobilization. Concrete and cold-pressed pellets are being studied and compared for ash immobilization.

Effects of burning beta and gamma contaminated waste

The identification and removal of radioactive gases and particles from the incinerator offgas, different from those encountered in burning plutonium contaminated wastes, have been major concerns of the commercial incinerator project since its inception. This problem is being attacked in a two-front assault: an intensive literature search to remain abreast of developments in related endeavors and a laboratory-scale, experimental incinerator to develop new technology.

The objectives of the selection of an ash immobilization technique and product characterization activities are to demonstrate a chosen process, characterize the product of the process, and compare characterization criteria of the product to the criteria developed in Task II of the TRU Waste Cyclone Drum Incinerator and Treatment System project.

Bench-scale incinerator

Work was started on a bench-scale incinerator system, which will be roughly 1/20 the size of the Cyclone Incinerator. This laboratory system will be used in experiments to determine necessary modifications to the Mound Cyclone Incinerator to effectively remove radioisotopes other than plutonium-238 from the offgas stream. With such modifications, the incinerator will be capable of burning low-level radioactive wastes generated by commercial facilities. Basically, the laboratory incinerator consists of a combustion chamber, deluge tank, mist eliminator, air filter tubes, blower, scrub-liquid storage tank, centrifugal pump, water filter cartridge, and two heat exchangers. Necessary equipment has been ordered or is presently on hand. Required modifications to some equipment are in progress.

Literature search

An extensive literature search was undertaken in preparation for the experimental program. Technology currently available in the field of air pollution control was determined from the literature, as well as specifications and performance characteristics of each technology. On the basis of this literature search, a deluge tank was chosen for the incinerator system because the wet-scrubbing system offered the advantages of low cost, resistance to plugging, and highly efficient particle removal with simultaneous neutralization of acid gases with the use of an alkaline scrubbing solution.

Different chemicals in the scrubber solution may aid in radioisotope removal as well. For example, small amounts of reducing agents such as phosphite, hypophosphite, sugar, $NaNo_2$, NO, NO_2, or formaldehyde can effectively reduce the volatility of ruthenium. Caustic solutions and small amounts of thiosulfate can also aid in the removal of elemental iodine from the offgas stream.

Several more-efficient methods for removing iodine and methyl iodide are available, each with its own advantages and disadvantages. The three most commonly used adsorbents for compounds of iodine are impregnated activated charcoals, silver-exchanged zeolites, and amorphous silicic acid impregnated with silver nitrate. These adsorbents differ widely in price, susceptibility to poisoning by species such as No_x and CO_2, resistance to acids, efficiency under varying conditions of temperature and humidity, and regenerability. There are indications that zeolites and activated charcoal may also be useful in controlling NO_x emissions.

For efficient ruthenium removal, silica gel, hydrous zirconium oxide gel, ferric oxide, and zirconium oxide hydrate-dithionate exchanger were most highly recommended. All the above information will be extremely useful in designing an experimental program for the bench-scale incinerator.

Chloride mass balance

The experiments described in this section were performed for the TRU Waste Cyclone Drum Incinerator and Treatment System project and were also reported in the quarterly report for that project [1].

Experiments were made to more clearly define the disposition of chlorides generated by the combustion of polyvinyl chloride (PVC) in the cyclone incinerator. Known quantities of PVC were added to drums of wastes containing no other significant sources of chloride. The PVC was mixed thoroughly with the other waste in the drums and constituted 25% of the total weight of each drum. The PVC used was determined by analysis to contain 43% by weight of chloride.

Eleven runs were made. For the first five burns, a solution of sodium carbonate served as the scrubber solution. For the remaining six, a potassium hydroxide solution was used.

Before the first run and after each of the following runs, the scrubber solution stream was sampled for chloride, carbonate/hydroxide, sodium, and sulfate. Additional samples were taken between burns when it was necessary to replace water lost by evaporation.

Data obtained during the investigation are given in Table 1. It can be seen that

Table 1 - DISPOSITION OF CHLORIDES GENERATED BY COMBUSTION OF PVC

Run	Base added To Scrub Sol'n	Cl^- in Fuel	Cumulative Cl^- Burned	Vol. Flue Gas (ft^3) Saturated ∿160°	Cl^- in Flue Gas (Analysis)	Cumulative Cl^- in Flue Gas	Volume Scrubber Sol'n	Cl^- in Solution (Analysis)
	9216 g Soda Ash						910 ℓ	409.5 g
1		1075 g	1075 g	16,368	4.7 g			
						4.7 g		
							908 ℓ	1362.0 g
2		860 g	1935 g	11,904	5.4 g			
						10.1 g		
							900 ℓ	2070.0 g
3		860 g	2795 g	9,884	5.1 g			
						15.2 g		
							900 ℓ	2790.0 g
							Added H_2O	
							1115 ℓ	2787.5 g
4		860 g	3655 g	8,928	4.7 g			
						19.9 g	1072 ℓ	3108.8 g
5		860 g	4515 g	8,472	3.2 g			
						23.1 g	1072 ℓ	3752.0 g
	1342 g KOH						888 ℓ	257.5 g
6		860 g	860 g	11,904	4.7 g			
						4.7 g		
							869 ℓ	995.9 g
7		860 g	1720 g	12,648	4.1 g			
						8.8 g	809 ℓ	1618.0 g
	2015 g KOH						941 ℓ	1787.9 g
8		860 g	2580 g	11,160	4.1 g			
						12.9 g	939 ℓ	2159.7 g
	5372 g KOH						968 ℓ	2226.4 g
9		860 g	3440 g	8,472	320.8 g			
						333.7 g		
							942 ℓ	2637.6 g
10		860 g	4300 g	14,880	67.6 g			
						401.3		
							939 ℓ	3474.3 g
11		860 g	5160 g	12,648	8.1 g			
						409.4 g	934 ℓ	4483.2 g

nearly 17% of the chloride burned is unaccounted for. This is a common experience in attempts to perform a material balance on chloride in a large system. Some chloride was probably lost through chemical attack of the system. Other chloride was probably lost because it was attached to solids removed from solution by the liquid filter. Analysis of this sludge was not performed, but it would be reasonable that the chloride concentration of the sludge would be similar to that of the ash.

The amount of chloride remaining in the flue gas after scrubbing was quite low for the majority of the runs. For runs 9 and 10, however, the analytical results show the flue gas chloride to be relatively high. The reasons for this are not known. There are two plausible explanations: sampling-analytical error and, most likely, contamination of the sample scrubber solution. The very dilute nature of the chloride/scrubber solution would make minute contamination significant.

The scrubber solution was permitted to drop into the acid range during runs 7 and 8. The flue gas chloride content was low for both of these burns, but it has been speculated that the drop in pH or subsequent addition of KOH to raise the pH may have triggered a chloride release on the following run, or which at least appeared on the following run analysis.

Since the present scrubber system has been in place, no chloride samples taken before or after this series of runs have had a flue gas chloride content nearly as high as that given for Run #9. Periodic monitoring of the flue gas for chloride content will continue.

Ash immobilization

Demonstration

A conceptual design for the modification of an existing glovebox was presented and approved during the quarter; an Engineering package was prepared and modifications were started. 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.

Concrete vs. pressed pellets

The major effort the past quarter was the comparison of concrete and cold pressed pellets as waste immobilization forms. Portland type 1A and Luminite[a] cement were mixed with cyclone incinerator ash and pressed into pellets or cast as concrete. All samples were 0.5-in. diameter cylinders. Water was used as a binder for both forms. Table 2 shows the mixtures that were used. Water used is not expressed as a weight percent; water content is proportional to the weight percent cement used in the respective mixtures and is expressed as a water/cement ratio. Pellets were pressed at a pressure of 25,000 psi.

Crush strength

Tables 3 and 4 show the crush strength of cast concrete and pressed pellet samples. Each value listed is an average of two samples. The samples were crushed in a FORNEY model FT-21 compression tester. The compression tester has a lower gauge-detection limit of 400 lb. Samples of concrete mixtures C, D, E, and F had crush

[a]Trademark of the Atlas Cement Company

Table 2 - PELLET MIXTURES AND WATER CONTENT

Mix	Cement (wt %)	Ash (wt %)	Water Content: Pressed pellets Water/cement ratio	Water Content: Concrete Water/cement ratio
A	70	30	0.10	0.55
B	60	40	0.12	0.80
C	50	50	0.16	0.98
D	40	60	0.19	1.52
E	30	70	0.29	1.83
F	20	80	0.70	2.76
		Bone Char (wt %)		
G	60	40	-	0.48
H	50	50	-	0.50
		Sludge (wt %)		
I	65	35	-	0.43
J	58	42	-	0.58

Table 3 - AVERAGE CRUSH STRENGTH OF CAST CONCRETE SAMPLES

Mix	Crush Strength of Portland Cement (psi)			Crush Strength of Lumnite Cement (psi)		
	Dry Cure	Wet Cure	800°C 1 hr	Dry Cure	Wet Cure	800°C 1 hr
A	3622	3205	3547	3725	3665	3657
B	3519	3228	3508	3659	3640	3619
C	a	a	a	3591	3670	3604
G	3638	3654	3498	3620	3549	3580
H	3587	3619	3538	3633	3554	3510
I	3636	3649	3582	-	-	-
J	a	-	-	3681	3630	2507

[a]Failed to reach lower limit of compression tester.

Table 4 - AVERAGE CRUSH STRENGTH OF PRESSED PELLET SAMPLES

Mix	Crush strength of Portland Cement (psi)			Crush Strength of Lumnite Cement (psi)		
	Dry Cure	Wet Cure	800°C 1 hr	Dry Cure	Wet Cure	800°C 1 hr
A	3690	4698	3569	3553	4425	3527
B	3665	4440	3604	3545	4079	3527
C	3672	3980	3604	3571	4008	3579
D	3672	3900	3614	3609	3829	3604
E	3662	3838	3619	3614	3786	3614
F	3984	3723	3985	3617	3641	3614

strengths below the 400-lb detection limit. "Dry" cured samples were allowed to cure at room temperature and atmosphere, whereas "wet" cured samples were cured in a high humidity atmosphere. Concrete samples were cured for a minimum of 28 days, and pressed pellets were cured for a minimum of seven days. Wet and dry cured samples of each mixture were sintered at 800°C for 1 hr to prove nonflammability. Crush strengths of heated samples were determined. Data listed in Tables 3 and 4 for heated samples are for those that were dry cured only; however, wet cured samples behave in a like manner.

Loading rate

Loading rate in this report is defined as the grams of ash immobilized per cubic centimeter of the total matrix. Geometric measurements were used to determine volumes (Table 5). Loading rates are virtually identical for both portland and Lumnite cement. Loading rates can be increased by pressing at pressures greater than 25,000 psi. Additional pellet pressings at higher pressures are planned to investigate the further reduction of fines clinging to the surface of the pellets.

Mechanical strength

Cylindrical cast concrete and pressed pellet samples were placed in stainless steel containers which were clamped into a model BT Burrell Wrist-Action Shaker*. The shaker was set at the maximum 10° shake angle to vibrate the samples in a circular motion inside the fixed container. The shaking action was continued for 100 hr, and the percent weight loss was determined (Table 6).

*Trademark of Burrell Corporation.

Wet cured samples of concrete mixtures were placed directly into the stainless steel containers; consequently, a large portion of the weight loss is from water evaporation. Wet cured pellet samples were dried at room temperature for 72 hr. As a result, the disparity between wet and dry cured samples is not as evident. All the samples experienced a slight rounding of the edge that contacted the bottom of the stainless container, and abrasion occured on the walls of the samples. Visual examination showed no cracks in any of the samples.

Leach rates

Concrete and pellet samples of nonradioactive mixtures A through J were suspended in distilled water for leach rate determinations. The leach rate (Table 7) is expressed as a percent weight loss derived by boiling the leachant to dryness and weighing the residue. The leachant was changed weekly. However, the results in Table 7 are expressed as monthly totals. All samples listed in Table 7 were fabricated using portland cement. Initial data indicate that samples fabricated with Lumnite cement have higher leach rates.

Water content

Radiolysis of water contained in packaged radioactive wastes causes gas production and possible pressure problems. These problems can be minimized by using pressed pellets as a waste form for incinerator ash immobilization. The water/cement (w/c) ratios illustrated in Table 2 for concrete and pellet mixtures A through F show that a much greater amount of water is required for concrete immobilization

Table 5 - LOADING RATES

Mix	Ash/Matrix Loading in Cast Samples (g/cm^3)	Ash/Matrix Loading in Pressed Pellets (g/cm^3)
A	0.41	0.62
B	0.51	0.85
C	-	1.00
D	-	1.17
E	-	1.32
F	-	1.50

Table 6 - COMPARATIVE MECHANICAL STRENGTH AS DETERMINED BY WEIGHT LOSS DURING 100-HOUR SHAKE TEST

Mix	Wt Loss from Cast Concrete (%)		Wt Loss from Pressed Pellets (%)	
	Dry Cure	Wet Cure	Dry Cure	Wet Cure
A	0.928[a]	8.730[a]	1.78	0.708
B	1.658[a]	13.840[a]	1.31	0.663
C	-	-	1.32	0.954
D	-	-	2.280	0.745
E	-	-	3.70	0.801
F	-	-	5.062	0.786
G	2.640	10.37	-	-
H	0.664	10.86	-	-
I	1.05	13.58	-	-

[a]Average of three samples. Remaining samples are of a single determination.

Table 7 - COMPARATIVE LEACH RATES AS DETERMINED BY WEIGHT LOSS

Mix	Weight Loss from Cast Concrete (%)				Weight Loss from Pressed Pellets (%)			
	Dry Cure		Wet Cure		Dry Cure		Wet Cure	
	1 month	2 month	1 month	2 month	1 month	2 month	1 month	2 month
A	1.81	0.31	1.86	0.62	2.04	0.755	0.44	
B	3.56	0.454	3.62	0.96	1.92	0.62	0.57	
C	-	-	-	-	1.37	0.50	0.48	
D	-	-	-	-	1.67	0.65	0.42	
E	-	-	-	-	2.0	0.38	0.41	
F	-	-	-	-	2.32	0.39	0.11	
G	2.57	0.80	2.76	2.08	-	-	-	
H	2.09	0.68	3.68	0.87	-	-	-	
I	2.84	0.84	3.52	1.06	-	-	-	
J	-	-	-	-	-	-	-	

than is required for pellets. Pressed pellets require 48 hr of curing to reach their maximum strength. Cast concrete requires several days. After the 48-hr cure, pellets can be heated to 110°C for 1 hr thus reducing the water content, as determined by weight differences, to less than 1% by weight. Moisture contents of pellets will be determined by analytical methods next quarter.

Bone char/sludge

Work has continued on the immobilization of bone char and sludge as illustrated in Tables 2,3,6, and 7. Bone char and sludge have been mixed with cement and pressed into pellets on a limited basis, to date, but this work is expected to be expanded the next quarter.

Pilot plant

Plans were drawn and construction began on a glovebox line for the pelletization of incinerator ash/cement mixtures using a small automatic press. The completion date is set for January, 1978. Bone char/cement mixtures will also be pelletized. The expected flow is shown in Figure 1.

Milestones

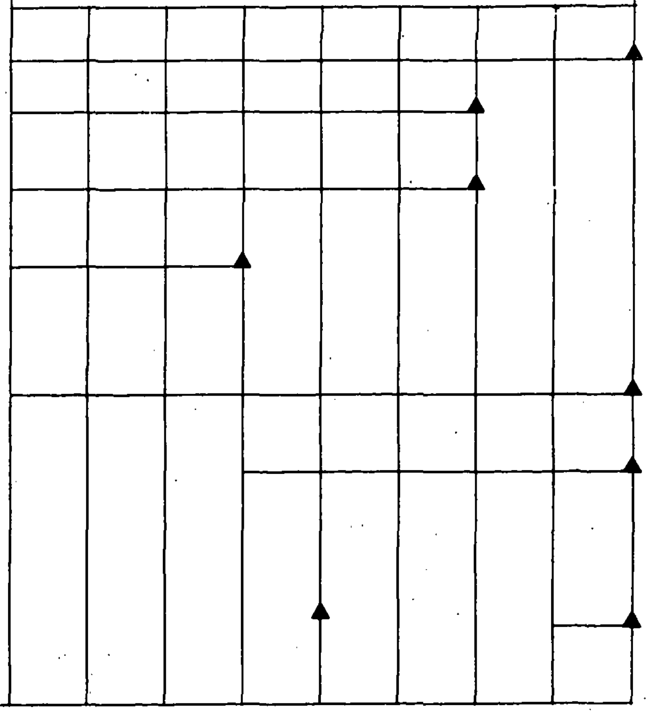
The milestone chart for this quarter is given in Table 8.

Reference

1. D. F. Luthy and J. W. Doty, TRU Waste Cyclone Drum Incinerator and Treatment System: July-September 1977, MLM-2496

Table 8 - MILESTONE CHART

ACTIVITY	FISCAL YEAR	
	FY-1977	FY-1978
PHASE I		
a. Interfaces with Commercial Plants		
b. Adaptation of ERDA Cyclone Design to Commercial Wastes		
c. Selection of Ash Immobilization Technique & Product Characterization		
d. Select Compaction Technique for Empty Waste Containers		
PHASE II		
e. Process Description & Design Criteria for Cyclone Incinerator		
f. Process Description & Design Criteria for Ash Immobilization & Waste Container Compaction Processes		
PHASE III		
g. Project Reports		



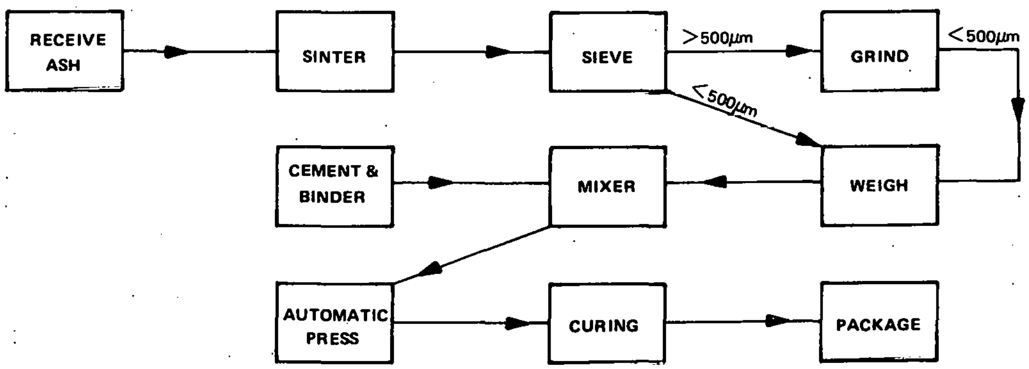


FIGURE 1 - Pelletization process flow sheet.

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