VOLUME REDUCTION OF REACTOR WASTES BY SPRAY DRYING

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ABSTRACT

Three simulated low-level reactor wastes were dried using a spray dryer-baghouse system. The three aqueous feedstocks were sodium sulfate waste characteristic of a BWR, boric acid waste characteristic of a PWR, and a waste mixture of ion exchange resins and filter aid. These slurries were spiked with nonradioactive iron, cobalt, and manganese (representing corrosion products) and nonradioactive cesium and iodine (representing fission products). The throughput for the 2.1-m-diameter spray dryer and baghouse system was 160-180 kg/h, which is comparable to the requirements for a full-scale commercial installation. A free-flowing, dry product was produced in all of the tests. The volume reduction factor ranged from 2.5 to 5.8; the baghouse decontamination factor was typically in the range of 10^3 to 10^4. Using an overall system decontamination factor of 10^6, the activity of the off-gas was calculated to be one to two orders of magnitude less than the nuclide release limit of the major active species, Cs-137.

INTRODUCTION

The nuclear power plant industry presently produces large volumes of low-level liquid radioactive wastes. The costs of disposal, including transportation and burial of these wastes, have been increasing tremendously in the last few years and are expected to continue to increase in the years to come. There is a large incentive to reduce the volume of these wastes in order to minimize these costs. The most common method that is used to volume-reduce low-level liquid wastes is to use a commercially available evaporator which accepts very dilute solutions and concentrates them to 12-25 wt. %. These concentrated solutions are then processed in a solidification system to produce a solid matrix for transportation. The leading methods that are used for solidification are incorporation in a matrix of cement, asphalt, or polymer. Solidification is necessary because liquid waste may not be buried. However, an alternative to a solidification technique, which appears to have major cost advantages, would be spray drying of the concentrated solutions to simply evaporate all the water content and produce a dry powder. Present regulations allow the transport of dry powder in high-integrity containers to a burial site. At the burial site, these containers would be buried in the standard waste burial location.

Rockwell International is well experienced in spray dryer technology for flue gas scrubbing and in the handling of radioactive materials. This provides a great advantage in producing a new system for the processing of low-level liquid radwaste. The system uses a spray dryer to evaporate all the water content from the liquid radwaste and a baghouse to trap the solids. The required throughput for processing low-level liquid radwaste is small compared to other spray dryer applications. Thus, a 2.1-m spray dryer is approximately the right size for a full-scale commercial application of drying low-level liquid radwaste. Rockwell International has a mobile spray dryer-baghouse system, which consists of a 2.1-m spray dryer and a fabric filter, which is ideally suited for the testing of low-level liquid radwaste. In this test report, the mobile system was used for drying simulated liquid radwaste, as follows: sodium sulfate solution, boric acid solution, and a resin-filter aid slurry.

The spray dryer concept has the potential advantage of its low-temperature application for drying. A spray dryer evaporates water at a low enough temperature that the volatilization of radionuclides does not occur, and the production of NO_x and SO_x from the drying of resins in an oxidizing atmosphere is minimized. The objective of this test program was to verify by testing that the radioactive species in concentrated reactor liquid wastes would be retained in the dry product from a spray dryer-baghouse volume reduction system and to verify that nitrogen and sulfur would not be oxidized from ion exchange resins.

DESCRIPTION OF THE MOBILE SPRAY DRYER SYSTEM

Spray Dryer and Baghouse

The Mobile Spray Dryer System consists basically of a 2.1-m-diameter spray dryer and a pulse-jet baghouse. The spray dryer is a standard Bowen model constructed of carbon steel. It is equipped with a 7.5-kW Bowen Model AA-6 spray machine and a 15-cm-diameter Type DH centrifugal atomizer. Atomizer speed is normally maintained at 22,000 rpm. From the spray dryer, the exhaust gases with their entrained solids are piped directly to the baghouse collector. The baghouse is a pulse-jet design containing 64 outside-collecting polyester bags, each 15 cm in diameter and 3 m long. The bags are suspended in an 8 x 8 array. Cleaning is accomplished by a reverse pulse of compressed air initiated by a solenoid signal directed to one row of bags at a time. Each cleaning pulse is 20 msec in duration at 15-sec intervals. An 18.6-kW New York blower induced-draft fan is used to pull hot gas from an excess-air natural gas burner through the spray dryer/baghouse system. A photograph of the mobile spray dryer system is shown in Fig. 1.

Sampling locations for gas analyses were at the spray dryer inlet, the spray dryer outlet, the fabric filter inlet, and the fabric filter outlet. Sulfur

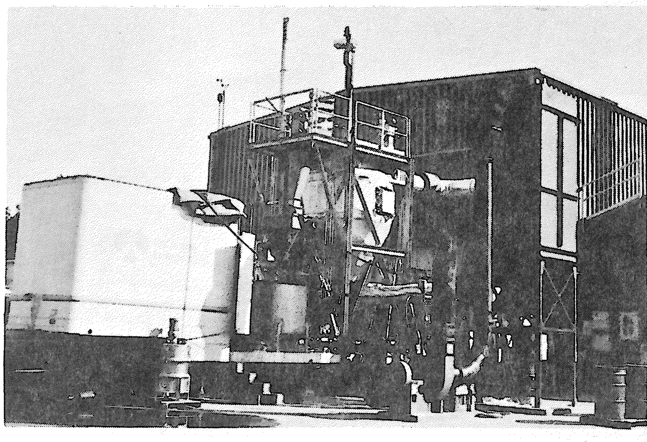

Fig. 1. Photograph of the Mobile Spray Dryer System

dioxide measurements were performed with a Teledyne spectrophotometric analyzer. Nitric oxides were measured with a Monitor Labs chemiluminescent analyzer. The temperatures at various points in the system were monitored with Type K (Chromel-Alumel) thermocouples, whose outputs were shown on digital displays and recorded on a multipoint strip chart recorder. The gas flow rates were determined by standard pitot tube traverse flow measurements. Pressures were measured with Magnehelic differential pressure gauges and standard manometers. Particulate sampling was done using an EPA Method V particulate sampling system.[1] Gas and particulate sampling was done during the testing to verify low concentrations of gas pollutants (NO_x and SO_2) and low radionuclide carryover. (The NO_x and SO_2 monitoring would not be done on a commercial system.)

A simplified flow diagram of the liquid volume reduction system is shown in Fig. 2. The simulated liquid wastes were prepared in a feed tank of 1900-liter capacity. A Moyno slurry pump was used to pump the feed solution to the top of the spray dryer. Hot gas for the drying was produced by an excess air natural gas burner. The gas flow rate was 60 std[a] cubic meters per minute at 450 K. Under these conditions, 2.2 liter/min (2.8 kg/min) of feed solution were dried. The temperature of the exit gas from the spray dryer was 350 K. The dried product from the spray dryer entered the baghouse where it was collected at the bottom in 200-liter drums. The outlet gas from the baghouse traveled down an off-gas duct to an induced draft fan and was then released through a stack.

The simulated corrosion and fission products can be expected to be collected in two places. The majority of these elements would be taken out with the solids from the fabric filter. Those elements passing through the fabric filter would then be collected on a HEPA filter which would provide final cleanup, and the off-gas would then be released. In the tests that were made, a sample was taken of the solids passing through the baghouse and collected on a glass fiber filter. The gas passing through the glass fiber filter was then passed through two chilled water scrubbers. Since the salts that were added to simulate the corrosion products and the fission products are all water soluble, it is expected that any of these five elements passing through the

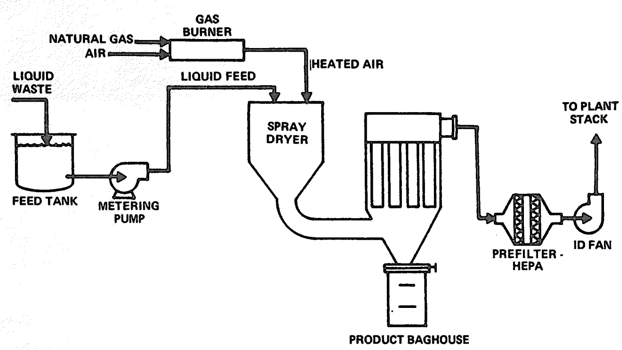

Fig. 2. Simplified Flow Diagram of Liquid Volume Reduction System

glass fiber filter would be collected in the two chilled scrubbers. The chilled scrubbers were concentrated to give a sample size of ~20 g, and the filter papers were extracted with water. These samples were sent to a commercial analytical laboratory for analysis using a spark-source mass spectrometer. This analytical technique can provide a sensitivity to 0.5 ppb. Baghouse decontamination factors were then calculated on the basis of these analyses.

RESULTS AND DISCUSSION

Simulated Radioactive Wastes

Three representative solutions/slurries that were typical of the composition of low-level radioactive wastes from nuclear power plants were prepared for these tests. The three wastes were nominally 20 wt. % Na_2SO_4 solution, 12 wt. % H_3BO_3 slurry, and a resin-filter aid slurry. These slurries were spiked with small concentrations of nonradioactive elements which simulated the presence of corrosion products and fission products that would be expected in a radioactive low-level waste.[2] Corrosion products were simulated using manganese, cobalt, and iron nitrate salts. The fission products were simulated using cesium nitrate and sodium iodide. The concentration of these elements in the feed corresponded to ~40,000 μCi/cc of solution. This concentration of trace fission products was used so that sufficient sample would be obtained for analytical purposes to provide measurement of decontamination factors. In general, the range of activity in low-level waste varies from about 1.0-40,000 μCi/cc.

The description of the three wastes as mixed is given in Table I. The simulated BWR waste is the sodium sulfate waste in which sodium sulfate, trisodium phosphate, and trace elements were added. Trisodium phosphate was used in this waste and in the PWR waste to simulate detergent which would come from cleaning solutions used to clean up spills and to decontaminate surfaces. The simulated PWR waste consisted of boric acid with the other trace elements. The resin filter aid waste consisted of a mixture of Ecodex anion resin, cation resin, anion precoat filter aid (Ecodex P-202-H), and a cation precoat filter aid (Ecodex X-203-H). The precoat material is a mixture of filter aid with resins and is received in what might be called a wet condition, i.e., it contains about 70 wt. % bound or contained water. Technical-grade anhydrous sodium sulfate was used for the BWR waste. The actual simulated BWR waste consisted of a 20.6 wt. % slurry of sodium sulfate. The

[a]Standard conditions are 293 K and 101.3 kPa.

Table I. Waste Compositions

Component, kg	Simulated BWR Waste	Simulated PWR Waste	Resin-Filter Aid Waste
Na_2SO_4, anhyd.	176	0.0	0.0
H_3BO_3	0.0	90.7	0.0
PAO anion resin*	0.0	0.0	6.0
PCH cation resin*	0.0	0.0	7.0
P-202-H precoat filter aid*	0.0	0.0	59.1
X-203-H precoat filter aid*	0.0	0.0	55.8
$Na_3PO_4 \cdot 12H_2O$	9.1	7.94	0.0
$CsNO_3$	0.5335	0.4440	0.2524
$Co(NO_3)_2 \cdot 6H_2O$	1.7957	1.4964	0.8501
$Mn(NO_3)_6$ (51.7 wt. % solution)	0.0	1.9099	1.0844
$Fe(NO_3)_3 \cdot 9H_2O$	2.2675	1.8901	1.0847
NaI	0.4291	0.3576	0.2031
H_2O	664	652	296

*Ecodex brand, Ecodyne Corp., Union, New Jersey 07083

concentration of trace elements in this slurry was 0.0426 wt. %. Approximately 860 kg of simulated BWR waste were prepared and dried. The simulated PWR waste consisted of a 12.0 wt. % slurry of boric acid. The boric acid was granular technical grade material. No difficulty was found in dissolving this material to a saturated solution (about 6 wt. %) in the feed tank. In this slurry, the concentration of trace elements was 0.0400 wt. %. The resin filter aid waste was prepared using 128 kg of wet resins and filter aid material. However, this filter aid material contains a large quantity of bound water and so only 37 kg of bone-dry material were actually mixed with water. This corresponds to a 9 wt. % resin-filter aid slurry on a bone-dry basis. Concentration of trace materials in this slurry was 0.0402 wt. %. Approximately 430 kg of this slurry were dried.

Mass Balance

The overall mass balance for spray drying of the three simulated wastes is given in Table II. The processing of the simulated BWR waste, the sodium sulfate solution, had an overall mass balance of 94%; 183 kg of solids were fed in and 172 kg were collected. The lost solids may correspond to feed material that held up in the ducting and on the fabric filter. At the end of the BWR test, the spray dryer was scraped clean of any solid sediment that accumulated on the walls. The mass balance for the processing of the PWR waste and the resin waste was 99.6%. In general, it is not expected that any material would be lost during the spray drying operation. In the processing of an actual radioactive material, care should be taken that all glands between ductwork are carefully sealed, solid feedlines should be designed with capability for purging the line with a back-flushing stream, and a dryer large enough to prevent wall deposits should be used.

Off-Gas Pollutants

One of the objectives of this test program was to verify that in the processing of these wastes, no off-gas pollutants in the form of SO_2 or NO_x are emitted. The results of these pollutant measurements are shown in Table III. The results of these measurements verified the expected advantage of the spray dryer in minimizing pollutant concentrations. When corrected for baseline concentrations, SO_2 was found to be ~0-2 ppm in concentration. The

Table II. Overall Mass Balance

Stream	Simulated BWR Waste	Simulated PWR Waste	Resin-Filter Aid Waste
Dry solids in feed (kg)	183	98.0	39.8
Dry solids collected (kg)	171.8	83.0	54.4
Percent mass balance*	94%	99.6%	100%

*Assumes extra product weight from resin test is added to PWR waste

NO was found to be ~13 ppm in the flue gas from the burner, and an additional 3 ppm of total NO_x was formed from the resin processing. These values were entirely in line with the expectation that nitrogen and sulfur in the resin are not oxidized in the spray drying process. In addition, this means that no specific off-gas cleanup for SO_2 or NO_x is required on the spray drying system.

Table III. Off-Gas Composition

Gas Species	Simulated BWR Waste	Simulated PWR Waste	Resin-Filter Aid Waste
SO_2, ppm	0	0	2
NO, ppm	0	0	3
NO_x, ppm	0	0	3
H_2O, vol %	5.4	6.8	7.0

In the processing of liquid wastes, the water in the wastes is evaporated and travels in the gas stream. In the standard EPA procedure for measuring particulate concentrations,[1] this off-gas moisture content is measured and is used in correcting the gas flow measurements to dry conditions. The results of the off-gas moisture content measurements are also shown in Table III.

Decontamination Factors

The simulated corrosion and fission products, Cs, I, Co, Mn, and Fe, were tracked through the spray dryer-baghouse system in order to estimate the decontamination factors for radioactive elements in the system.

Using the known baghouse inlet concentrations and the results of filter paper and chilled scrubber samples, the decontamination factors for the five elements were calculated. These results are shown in Table IV. In general, the decontamination factor for the baghouse ranged from 10^3 to 10^4. The addition of a HEPA filter would improve the decontamination factor by a factor of 10^3 or greater; the expected overall decontamination factor for the spray dryer-baghouse system with HEPA filters is 10^6 or greater.

The predicted activity of the off-gas from the spray dryer-baghouse system was calculated for the conditions used in this test program, assuming an overall decontamination factor of 10^6. For typical low-level waste activities in the range of 0.1-10 μCi/cc, the predicted off-gas emission activity is one to two orders of magnitude beneath the emission limit for Cs-137. Waste streams having a higher initial activity can be processed by diluting them with less active streams or by adding an additional HEPA filter to the system.

Table IV. Baghouse Decontamination Factors (D.F.)* for Spray Drying of Simulated Low-Level Liquid Radwaste

Corrosion and *Fission Products*	Simulated BWR Waste	Simulated PWR Waste	Resin-Filter Aid Waste
Cs	1.4×10^4	8.1×10^4	6.6×10^3
I	5.9×10^2	$<3.5 \times 10^2$	$<2.6 \times 10^2$
Co	4.6×10^4	2.5×10^4	8.5×10^4
Mn	†	2.9×10^4	3.6×10^4
Fe	1.7×10^3	1.5×10^3	2.2×10^3

*Baghouse D.F. = $\frac{\text{Baghouse inlet concentration of radionuclide}}{\text{Baghouse outlet concentration of radionuclide}}$

†Manganese decontamination factor not measured for simulated BWR waste

Volume Reduction Factors

In the processing of low-level liquid waste, an important parameter for economic calculations is the volume reduction factor. Table V presents the volume reduction factors which were calculated for these tests. These factors correspond to the initial volume of liquid waste feed divided by the final volume of the solid dried product. The dry product from spray drying can be collected from the baghouse in standard 0.20-cubic-meter drums. This powder is then compressed at 550 kPa using equipment similar to conventional equipment for compressing radioactive solid wastes in drums. The densities of the product solids (pressed at 550 kPa) from the simulated BWR waste, the simulated PWR waste, and the resin-filter aid waste were 0.78, 0.61, and 0.55 g/cm^3, respectively. The values in Table V are based on a 25 wt. % sodium sulfate solution, a 12 wt. % boric acid slurry, and a 9 wt. % resin slurry (based on bone-dry solids). The volume reduction factors for the three wastes as pressed powders are 2.5 for the BWR waste, 4.5 for the PWR waste, and 5.8 for the resin-filter aid waste.

Table V. Volume Reduction Factor for Simulated Low-Level Liquid Radwaste

Quantity	Simulated BWR Waste	Simulated PWR Waste	Resin-Filter Aid Waste
Composition	25 wt. % Na_2SO_4	12 wt. % H_3BO_3	9 wt. % resins and filter aid (based on bone-dry solids)
Liquid density (g/cm^3)	1.24	1.12	1.05
Vol. of 1 kg of liquid (cm^3)	800	890	950
Density of solids pressed at 550 kPa (g/cm^3)*	0.78	0.61	0.55
Vol. of solids from 1 kg of liquid (cm^3)	320	197	164
Volume reduction factor	2.5	4.5	5.8

*Corresponds to normal compaction pressure used for compressing radioactive solid waste in drums.

CONCLUSIONS

The spray dryer-baghouse system was demonstrated at full-scale for the processing of low-level liquid reactor waste. A throughput of 160 to 180 kg/h of liquid wastes was dried in this system. No pretreatment of the waste, such as pH adjustment, was necessary to process these wastes. A dry free-flowing powder was produced from all the wastes, which can then be collected in drums, compressed, and stored or shipped to burial sites. In addition, the system has the following advantages:

1) The technology is well established, and the equipment has been proven in many years of commercial drying applications.

2) Volume reduction of all liquid wastes, solutions, slurries, sludges, and resins, is achieved by evaporation to dry solids containing the radionuclides.

3) The dry product can be used as direct feed to any of the waste immobilization processes currently envisioned.

4) Energy is used efficiently via heat supplied by direct contact of hot gas with the liquid waste.

5) The low operating temperature of the system results in good decontamination factors for radionuclides and no release of NO_x or SO_x from resins.

6) The capital investment is relatively low due to the simplicity of the system.

REFERENCES

1. Federal Register, Vol 42, No. 160, Aug. 18, 1977

2. H. W. Goodbee and A. H. Kibbey, "Unit Operations Used to Treat Process and/or Waste Streams at Nuclear Power Plants," Nuclear and Chemical Waste Management, Vol 2, pp 71-88, 1981