POLYETHYLENE ENCAPSULATION OF MIXED WASTES: SCALE-UP FEASIBILITY

P. D. Kalb, J. H. Heiser, and P. Colombo
Brookhaven National Laboratory
Upton, NY 11973

ABSTRACT

A polyethylene process for the improved encapsulation of radioactive, hazardous, and mixed wastes has been developed at Brookhaven National Laboratory (BNL). Improvements in waste loading and waste form performance have been demonstrated through bench-scale development and testing. Maximum waste loadings of up to 70 dry wt% mixed waste nitrate salt were achieved, compared with 13 - 20 dry wt% using conventional cement processes. Stability under anticipated storage and disposal conditions and compliance with applicable hazardous waste regulations were demonstrated through a series of lab-scale waste form performance tests. Full-scale demonstration of this process using actual or surrogate waste is currently planned. A scale-up feasibility test was successfully conducted, demonstrating the ability to process nitrate salts at production rates (up to 450 kg/hr) and the close agreement between bench- and full-scale process parameters. Cored samples from the resulting pilot-scale (114 liter) waste form were used to verify homogeneity and to provide additional specimens for confirmatory performance testing.

INTRODUCTION

Currently, hydraulic cement is the most widely used method for stabilization of radioactive, hazardous, and mixed wastes. Use of this conventional stabilization technique for mixed waste nitrate salts at Department of Energy (DOE) facilities has resulted in inefficient waste loadings (less waste per drum, wasteful of limited financial and disposal resources) or poor quality waste forms subject to premature deterioration. Nitrate salts are produced or stored in large volume at several DOE facilities including Rocky Flats Plant (RFP), Westinghouse Hanford Company (WHC), Idaho National Engineering Laboratory (INEL), Los Alamos National Laboratory (LANL), Oak Ridge National Laboratory (ORNL) and Westinghouse Savannah River Company (WSRC). Difficulties with cement stabilization of nitrate wastes are attributed to chemical interactions between the waste and binder that interfere with proper cement hydration. Encapsulation of nitrate salts and other mixed wastes in polyethylene (a process developed at Brookhaven National Laboratory) does not rely on a chemical reaction for solidification and, thus, is not subject to the type of failure mechanisms experienced by cement systems. Polyethylene is an inert thermoplastic material, which when heated above its melting point (120°C), mixed with waste to form a homogeneous mixture and allowed to cool, forms a monolithic solid waste form (1).

BENCH-SCALE DEVELOPMENT

Several processing techniques were considered for encapsulation of wastes in polyethylene, including batch heating vessels, wiped film evaporators, and screw extruders. Extrusion was selected based on consideration of processing ease, quality control, and the use of a proven or available technology. Single screw extruders have been used effectively in the plastics industry for over fifty years. This process simultaneously heats, mixes, and conveys the waste and binder, so that a homogeneous, molten mixture is extruded into a container for cooling. A monolithic solid waste form is ensured once the mixture cools below the melting temperature of polyethylene. A simplified schematic diagram of a single-screw extruder is shown in Fig. 1.

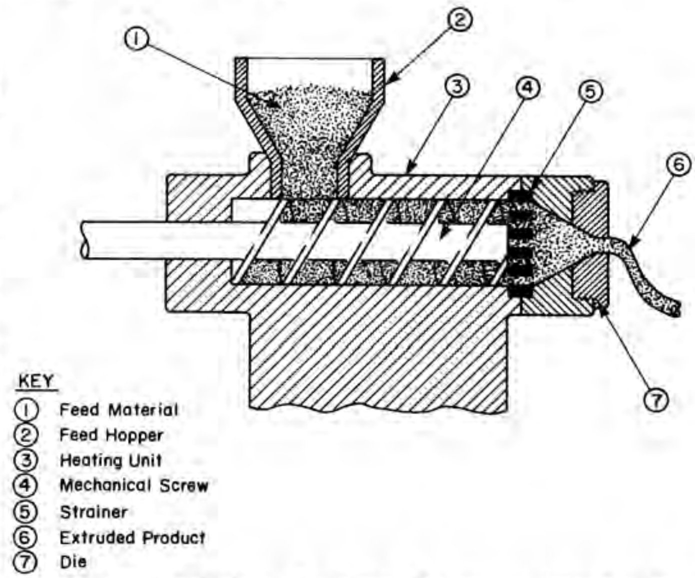

Fig. 1. Sectional view of a simplified screw extruder. The sketch depicts flow of material from the hopper to the output die, where it is extruded in a molten state.

Formulation development work was conducted using a bench-scale 32 mm (1.25 inch) polyethylene extruder, with a maximum output capacity of about 16 kg/hr (35 lb/hr). Density and particle size differences led to segregation of waste and binder when using a stock feed system (static hopper), creating a heterogeneous mixture. Thus, the system was modified using separate, dynamic feeders for waste and binder in order to overcome these difficulties and provide a means of precise control of waste/binder ratios. Figure 2 is a photograph of the bench-scale extruder modified for waste encapsulation.

Maximum waste loadings of up to 70 dry wt% nitrate salt waste were achieved, compared with 13 - 20 dry wt% using conventional cement processes. Application of polyethylene encapsulation in place of cement at Rocky Flats Plant, which has generated up to 1×10^6 kg/yr of nitrate salt waste, could result in 68% fewer drums for storage, transport and disposal. This volume reduction can lead to annual cost savings estimated between $1.5 and $2.7 million (2). Similar improvements have been achieved for other waste streams

Fig. 2. Photograph of laboratory-scale extruder with separate dynamic feeders for waste and binder.

including sludges, evaporator concentrates, incinerator ash, and ion exchange resins. Differential scanning calorimetry was conducted to confirm the absence of potentially deleterious interactions between the polyethylene binder and nitrate salt waste (3). Long-term durability of polyethylene waste forms under anticipated storage and disposal conditions and compliance with applicable hazardous waste regulations were confirmed through a series of lab-scale waste form performance tests (4).

SCALE-UP FEASIBILITY

Plans were initiated to demonstrate this system using production-scale equipment, in conjunction with Rocky Flats Plant, upon completion of bench-scale formulation and testing. Estimates of necessary production capacity were made based on data from RFP. Sizing of extruder equipment varies over a wide range, from bench-scale equipment (diameter of screw between 19 mm and 32 mm, 0.75 in. and 1.25 in.) to very large machines with screw diameters up to 200 mm (8 in.), or more. Typically, lab-scale extruders can process around 9 - 14 kg/hr (20 - 30 lbs/hr), while production-scale machines can process hundreds or thousands of kg/hr. For the technology demonstration, a 114 mm. (4.5 in.) extruder, with maximum output capacities in the range of 900 kg/hr (2000 lb/hr) was selected. A production-scale extruder is shown in Fig. 3. Parameter data generated during bench-scale investigations were reviewed to develop a set of required design specifications. A survey of potential vendors was then conducted. Equipment designs were examined to ensure that processing and monitoring requirements could be met using conventional, "off-the-shelf" equipment.

Prior to final selection of equipment, a production-scale feasibility test was conducted in order to:

- demonstrate system feasibility using simulated waste,
- examine correlation between processing parameters needed for scale-up and those generated during bench-scale studies,
- confirm rated production capacities for waste-binder combinations,
- investigate effects of screw design on production capacity and product quality,
- perform quality assurance testing on scale-up waste forms,
- conduct waste form performance tests for cored specimens taken from the scale-up specimen and compare results with lab-scale test data.

The production-scale feasibility test was conducted using a 114 mm (4.5 in.) extruder at laboratory facilities provided by Davis-Standard (Pawcatuck, CT), a manufacturer of extruder equipment. Their facility is designed for feasibility testing. It is equipped with state-of-the-art monitoring and control systems for data collection and process control, as well as a wide selection of extruder and screw designs to accommodate diverse user needs. Personnel from Brookhaven National Laboratory and Rocky Flats Plant attended the demonstration; the staff at Davis-Standard provided technical assistance.

Two Accu-Rate (Whitewater, WI) Model 610 dry material feeders were purchased and calibrated prior to the feasibility test in order to maintain an accurate waste loading of sodium nitrate salt. A slightly more conservative waste loading of 60 wt% nitrate salt was selected for the scale-up feasibility test (compared with the maximum loading of 70 wt%) due to the many variables under consideration. Optimization of waste loading under full-scale conditions will be performed as part of the planned Technology Demonstration. Calibration curves for the polyethylene and sodium nitrate feeders are shown in Fig. 4. These feeders are larger versions of the feeders used in bench-scale studies. They were installed above the extruder feed throat, in place of a standard static feed hopper. Figure 5 is a simplified process flow diagram of the full-scale polyethylene encapsulation system.

Initial trials resulted in excessive foaming due to air entrainment, caused by the physical properties of the simulated granular salt waste and length of the extruder barrel. This problem was remedied quickly by removing the extruder screw and replacing it with one designed to vent gases midstream. As shown in Table I, process settings similar to those developed at BNL were successfully duplicated. Maximum output rates were not attempted, but an output peak of 1,577 kg/hr (3,477 lb/hr) was attained at 65 RPM and steady-state rates in excess of 454 kg/hr (1000 lbs/hr) were easily achieved. A 114 liter drum (30 gal.) of encapsulated sodium nitrate was filled in about 25 minutes. Only minimal shrinkage was observed upon cooling, indicating a lack of significant voids in the waste form.

Testing of the 114 liter (30 gal.) waste form began by cutting the sample along an axial plane to check for void formation and physical examination of homogeneity. Several small voids were present (the largest measures about 25 mm x 6 mm x 6 mm deep), but no major voids were observed (see the photograph of the cut specimen, Fig. 6). The interior of

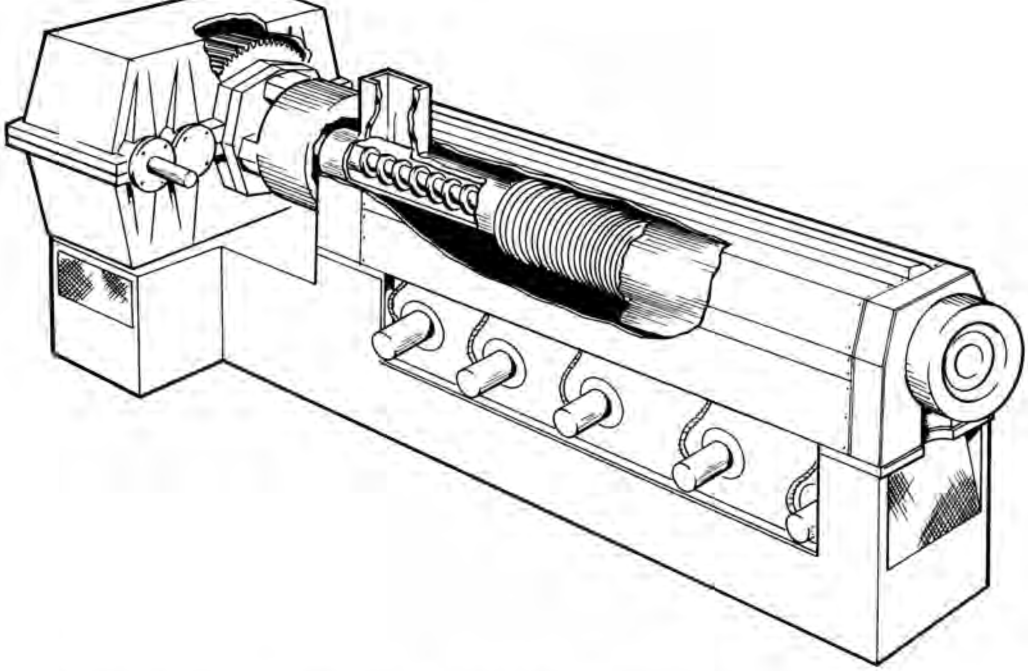

Fig. 3. Production-scale extruder for polyethylene encapsulation of mixed wastes.

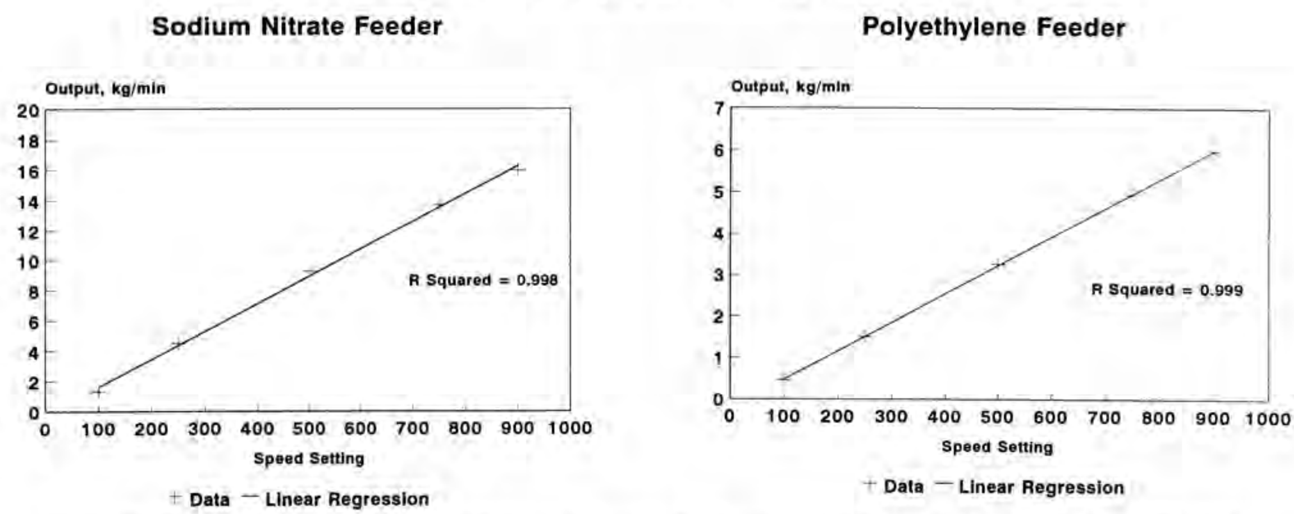

Fig. 4. Calibration curves for the polyethylene and waste feeders used in the production-scale feasibility test.

the waste form appeared well-mixed with salt granules distributed uniformly throughout. It had an even, opaque, white color from the nitrate salt. In contrast, polyethylene with no waste has a distinctive translucent color. No sections were visible in which the salt appears highly concentrated or absent. One half of the waste form section was cut further, into a total of 81 rectangular core specimens nominally measuring 51 mm x 51 mm x 101 mm (2 in x 2 in x 4 in). All specimens were weighed and measured, and apparent densities calculated. Overall average density was 1.345 ± 0.014 g/cm^3. The small variability in density of cored specimens (< 1%) indicates: 1) feed and mixing processes functioned properly, 2) product segregation did not occur during cooling/solidification and, 3) a homogenous waste form product was produced.

In addition to the inspection and density measurements, cored specimens were subjected to several waste form performance tests to provide further quality assurance data for the full-scale system, and to compare with similar data generated from tests of waste form specimens produced with laboratory-scale process equipment. Testing of cored specimens included compressive yield strength, thermal cycling, radiation stability, and biodegradation. Results are presented in Ref. 2. These tests were performed using 6 replicate specimens chosen from various sections throughout the waste form. Variation among compressive yield strength replicates was low (± 2.0% for control samples, ±3.6% for thermal cycled samples, and ±4.0% for irradiated samples, ±11.1% for biodegradation samples; errors were calculated at the 95% confidence level) indicating that mechanical properties are uniform

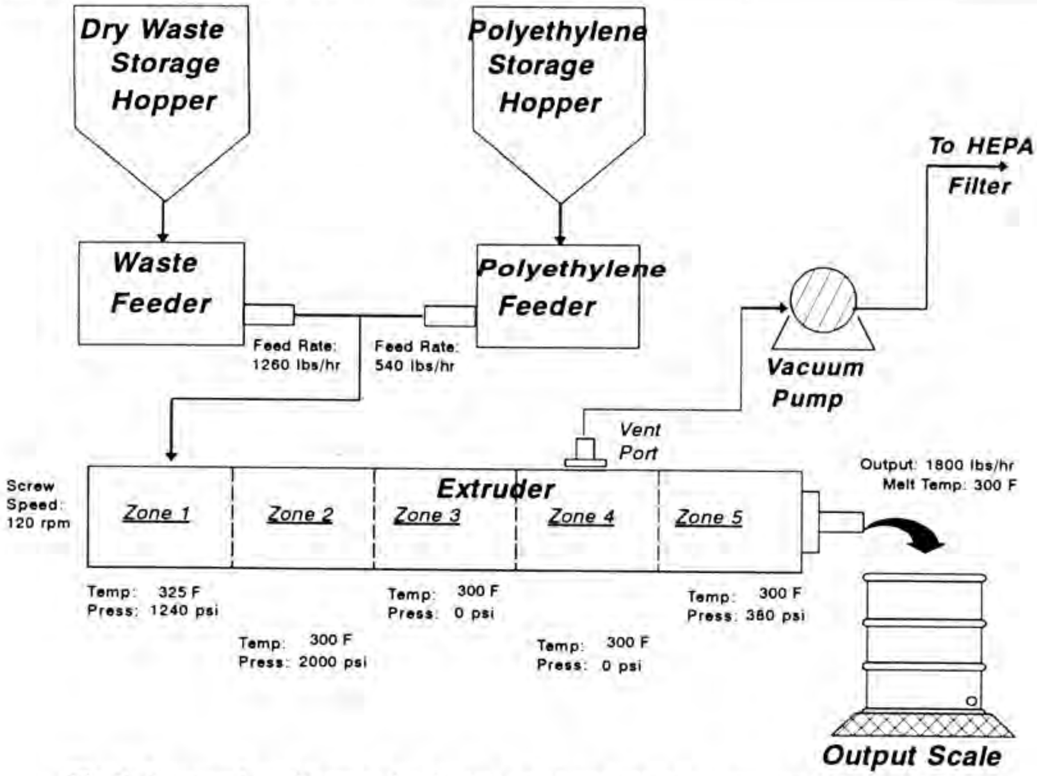

Fig. 5. Process flow diagram for the full-scale polyethylene encapsulation system.

TABLE I
Parameter Settings and Test Data for Polyethylene Encapsulation Production-Scale Feasibility Test

Melt Temperature, °C (°F)	149 (300)
Melt Pressure, MPa (psi)	2.6 (380)
Max. Screw Speed, RPM	65
Steady-state Output, kg/hr (lb/hr)	>454 (1,000)
Max. Output, kg/hr (lb/hr)	1,577 (3,477)
Horsepower at Max Output	354

throughout the scale-up waste form. Results for the compressive yield strength are in close agreement (within 8%) with data from lab-scale samples.

Results of the production-scale feasibility test for the polyethylene encapsulation of nitrate salt wastes may be summarized in the following points:

- Polyethylene encapsulation of at least 60 wt% nitrate salt wastes can be accomplished successfully, using a production-scale 114 mm (4.5 in.) extruder at steady-state rates of at least 454 kg/hr (1000 lb/hr).
- Comparison of bench- and production-scale process data confirms that process scale-up is feasible and that information generated during research and development phases of this project can be applied during the Technology Demonstration phase.
- Quality assurance testing of the 114 liter (30 gal.) waste form produced during the full-scale feasibility test demonstrates that a homogenous waste form with excellent properties can be produced using off-the-shelf production equipment.

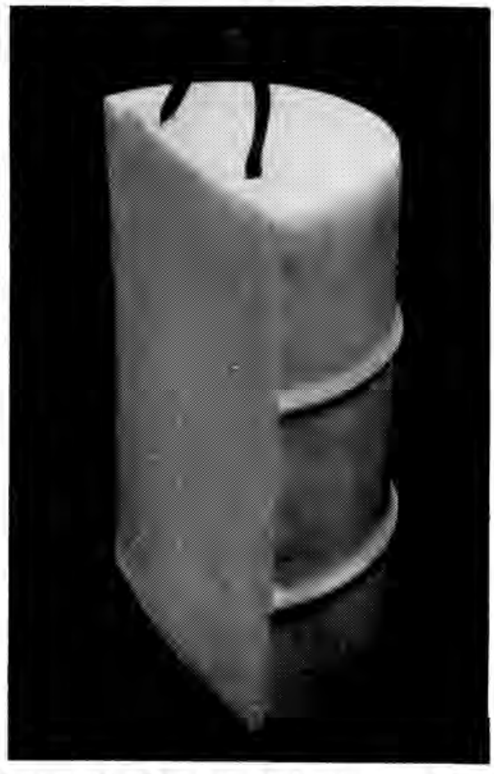

Fig. 6. Photograph of the polyethylene/sodium nitrate waste form produced during the production-scale feasibility test, showing the outer surface and cross-sectional view.

- Close agreement of results from waste form performance tests of specimens produced using bench-scale and full-scale systems, indicates the validity and importance of bench-scale research and development , prior to scale-up and demonstration of new technologies.

TECHNOLOGY DEMONSTRATION

A full-scale technology demonstration of polyethylene encapsulation of nitrate salt wastes is planned based on the results of research and development activities and the information gathered as a result of the full-scale feasibility test. Actual nitrate salt waste or a surrogate waste that closely resembles actual waste in physical and chemical composition, will be used. Pre-treatment requirements (e.g., dryer technology) will be examined to determine the impact on processing and performance. Quality assurance testing will be conducted to provide final confirmation of product quality prior to implementation of polyethylene encapsulation at DOE facilities.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge Sandra G. Lane for her assistance in editing this manuscript.

REFERENCES

1. P.D. KALB and P. COLOMBO, "Polyethylene Solidification of Low-Level Wastes, Topical Report," BNL-51867, Brookhaven National Laboratory, Upton, NY, (October 1984).
2. P.D. KALB, J.H. HEISER, and P. COLOMBO, "Polyethylene Encapsulation of Nitrate Salt Waste: Waste Form Stability, Process Scale-Up, and Economics, Technology Status Topical Report," BNL-52293, Brookhaven National Laboratory, Upton, NY, (July 1991).
3. J.H. HEISER, E.M. FRANZ, and P. COLOMBO, "A Process for Solidifying Sodium Nitrate Waste in Polyethylene," *Environmental Aspects of Stabilization and Solidification of Hazardous and Radioactive Wastes,"* ASTM STP 1033, P. Coté and M. Gilliam, eds., American Society for Testing and Materials, Philadelphia, PA (1989).
4. P.D. KALB, J.H. HEISER, and P. COLOMBO, "Long-Term Durability of Polyethylene for Encapsulation of Low-Level, Hazardous, and Mixed Waste," BNL-46359, Presented at the I&EC Special Symposium, American Chemical Society, Atlanta, GA (October 1 - 3, 1991).