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WASTE MANAGEMENT TECHNOLOGY DEVELOPMENT AND DEMONSTRATION PROGRAMS AT BROOKHAVEN NATIONAL LABORATORY

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ABSTRACT

Two thermoplastic processes for improved treatment of radioactive, hazardous, and mixed wastes have been developed from bench-scale through technology demonstration: polyethylene encapsulation and modified sulfur cement encapsulation. The steps required to bring technologies from the research and development stage through full-scale implementation are described. Both systems result in durable waste forms that meet current Nuclear Regulatory Commission and Environmental Protection Agency regulatory criteria and provide significant improvements over conventional solidification systems such as hydraulic cement. For example, the polyethylene process can encapsulate up to 70 wt% nitrate salt, compared with a maximum of about 20 wt% for the best hydraulic cement formulation. Modified sulfur cement waste forms containing as much as 43 wt% incinerator fly ash have been formulated, whereas the maximum quantity of this waste in hydraulic cement is 16 wt%.

INTRODUCTION

The Department of Energy (DOE) has generated large volumes of low-level radioactive (LLW), hazardous, and mixed waste as a result of its research and defense activities over the last 50 years. These include a broad range of waste types encompassing diverse chemical and physical properties. The total volume of DOE LLW alone, (either buried or disposed) through 1987 is estimated at 2.4×10^6 cubic meters, and these wastes continue to be generated at an estimated annual rate of 1.5×10^5 cubic meters [1]. Sources of commercial LLW and mixed wastes include nuclear power fuel cycle activities (60%), and industrial/institutional sources (40%) such as hospitals, universities, and radionuclide manufacturers. About 1.5×10^6 cubic meters of commercial LLW has been generated, and the current generation rate is estimated at 5.5×10^4 cubic meters/year. Figure 1 compares current sources of both DOE and commercial LLW.

Much of this waste requires solidification/stabilization (S/S) before final disposal to reduce the mobility of contaminants into the accessible environment. The most common practice at DOE and commercial facilities is to solidify waste using hydraulic cement such as portland cement. Historically, cement processes (also known as grouting) were the first methods for S/S of wastes. They continue to be widely used primarily because they are relatively inexpensive (material costs of $0.10/lb or less), readily available, and easy to process. Cement solidification processes are limited however, because cement hardens by means of a chemical hydration reaction that is susceptible to interferences with the waste. For example, many inorganic salts and heavy metals present in LLW and mixed wastes are known to inhibit cement hydration [2]. These

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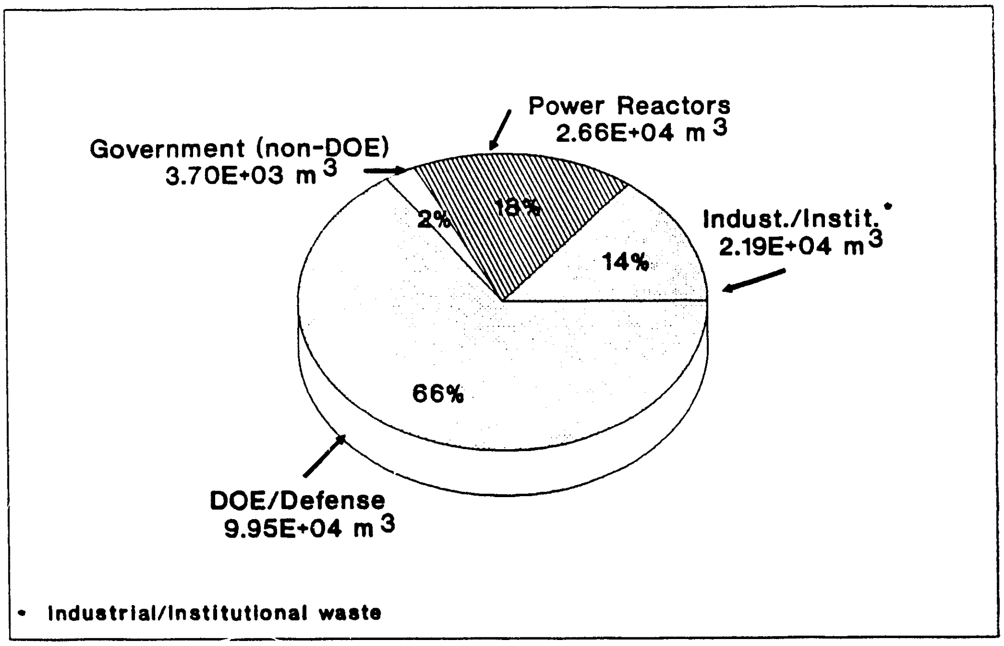

Figure 1 Comparison of DOE and commercial low-level radioactive waste volumes generated annually (1987 data) [1].

interactions reduce the amount of waste that can be incorporated and/or result in poor quality waste forms that do not successfully withstand disposal conditions. Low material costs for hydraulic cement are offset by poor waste loading efficiency (amount of waste per drum) and potentially poor performance of waste forms in disposal. In addition, small variations in waste chemistry over time can necessitate frequent process modifications and quality assurance testing. This seemingly straightforward process can actually prove complex and labor-intensive.

Thus, work conducted at Brookhaven National Laboratory, sponsored by the U.S. Department of Energy Office of Technology Development (DOE OTD), is focusing on the development and demonstration of new and innovative techniques for encapsulation of "problem" wastes. In keeping with OTD's policy of fostering "faster, better, cheaper, and safer technologies," the objectives of this effort are to develop materials and processes that:

- have the potential to encapsulate "problem" LLW and mixed wastes where current practices are inadequate,
- minimize the potential for release of toxic materials to the environment,
- result in durable waste forms that can withstand anticipated conditions during storage, transportation, and disposal,
- are simple to operate, easy to maintain, and economical.

Process development efforts at BNL begin with bench-scale research and development that progresses in logical stages:

1) Waste characterization—Waste composition is analyzed and physical properties relevant to treatment and processing are identified.

2) Investigation of Potential Materials and Processes—A large field of potential systems is examined and the list of preferred candidates is narrowed based on known properties and behavior.

3) Waste-Specific Treatability Studies—Feasibility is investigated based on compatibility of the waste and binder. Preliminary formulations are developed and tested according to broad acceptance criteria (e.g., process results in a monolithic solid waste form with minimum strength characteristics).

4) Formulation Development—The most promising candidates progress to this next phase of R & D in which waste/binder ratios are optimized to provide the best combination of loading efficiency and waste form performance (see below). Often, these two factors represent a trade-off and final formulations must reflect a balance between meeting minimum performance criteria and producing an economical process.

5) Performance Testing—Optimized waste form formulations are subjected to a complete set of waste form property and performance tests to provide a means of comparison among potential S/S options. Performance testing also provides necessary data for predicting long-term behavior of waste forms in storage and disposal. These tests can reveal areas where potential performance improvements are achievable by means of additional waste treatment, modifying formulations, or use of specific additives. In these cases, performance testing and formulation development comprise an iterative process.

6) Economic Feasibility—Overall system cost-effectiveness is examined and compared with conventional and alternative technologies.

Bench-scale systems shown to have potential technical and economic benefits are candidates for process scale-up and technology demonstration activities. The ultimate goal of full-scale technology demonstration is accomplished through the following steps:

1) Site Selection—Appropriate waste generating sites are selected based on types and volumes of waste generated. Cooperation of site personnel is solicited.

2) Feasibility Assessment—Scale-up feasibility is confirmed by means of pilot- or full-scale testing using simulated wastes. Resulting process data are compared with laboratory data and engineering estimates to corroborate scale-up of process parameters. Quality assurance testing of products is conducted to verify proper metering, mixing, and overall product performance.

3) Equipment Acquisition and Installation—Site-specific needs are considered prior to final equipment specification and acquisition.

4) Technology Demonstration—Upon completion of installation, start-up, calibration, and preliminary testing, the technology demonstration is held under actual plant conditions. To provide maximum impact, personnel from throughout the DOE complex, related regulatory agencies, and from the commercial sector are invited to attend.

5) Process Evaluation—Data collected during the technology demonstration is then reviewed to ascertain compliance with quality assurance (QA) requirements and to compare with bench-scale data. Waste form properties of the resulting product are also tested against QA and performance criteria.

6) Technology Transfer—All necessary information for the successful implementation of developed technologies is transferred to target sites within the DOE complex.

SELECTION OF BINDER MATERIALS

Research and development efforts have encompassed waste streams that are common within DOE (e.g., nitrate salts), the commercial sector (e.g., evaporator concentrates, ion exchange resins) or both (e.g., sludges, incinerator ash). Potential S/S binder materials surveyed are listed in Table 1. Because of the inherent problems with using hydraulic cement for solidifying and stabilizing waste discussed above, two thermoplastic materials were selected for further development: low-density polyethylene and modified sulfur cement. These materials can be melted, mixed with waste to form a homogenous mixture and then allowed to cool, resulting in a monolithic solid waste form. Contaminants are immobilized by micro-encapsulation. Since no chemical reaction is required for solidifying, they are compatible with a wider range of waste types and can encapsulate more waste per drum than conventional processes. Process temperatures are relatively low (melting temperature of both materials is ~ 120°C) so volatilization of contaminants is negligible.

Table 1 Potential Encapsulation Materials

Cements	Thermoplastic
Portland	Bitumen
Masonry Cement	Polystyrene
Cement-Sodium Silicate	Polypropylene
Pozzolanic	Polyethylene
High Alumina	Modified Sulfur Cement
Blast-Furnace Slag	
Polymer Modified Gypsum	
Polymer Impregnated Concrete	
Glass	**Thermosetting**
Soda-Lime	Vinyl-Ester Styrene
Phosphate	Polyester Styrene
Slag	Water Extendable
	Polyester

Polyethylene is an organic polymer of crystalline-amorphous structure available in a wide range of densities and molecular weights. These properties, in turn, affect basic material properties such as melt temperature, viscosity, hardness, and permeability. Polyethylene has been shown to withstand conditions that may be encountered in disposal including harsh chemicals, radiation, microbial degradation, freeze-thaw cycling, and saturated conditions [3]. Modified sulfur cement was developed by the U.S. Bureau of Mines about 20 years ago as a means of utilizing by-product sulfur for construction [4]. The supply of sulfur by-products from flue gas de-sulfurization and petroleum refining is growing. More than 5 x 10^6 tons of waste sulfur are projected to be produced annually by the year 2000. Modified sulfur cement is made by reacting elemental sulfur with organic modifiers that increase its stability by suppressing unstable phase transformations. Results of testing have shown that modified sulfur cement is also durable under anticipated disposal conditions [5].

PROCESS DEVELOPMENT AND PERFORMANCE TESTING

Formulation and process development studies have been performed for polyethylene and modified sulfur cement encapsulation of a wide range of waste types. These studies determine the maximum waste loadings for each binder, while still maintaining adequate waste form performance [6,7,8]. Existing waste form performance criteria established by the Nuclear Regulatory Commission (NRC) for demonstrating long-term durability of commercial low-level radioactive waste forms and by the Environmental Protection Agency (EPA) for the toxic leachability of hazardous wastes were applied. Maximum waste loadings achieved for polyethylene and hydraulic cement encapsulation are compared in Figures 2 and 3 (weight % and volume, respectively). Figures 4 and 5 present similar data for modified sulfur cement. Significant improvements in waste loading are attained in each case using these thermoplastic binders when compared with conventional cement systems.

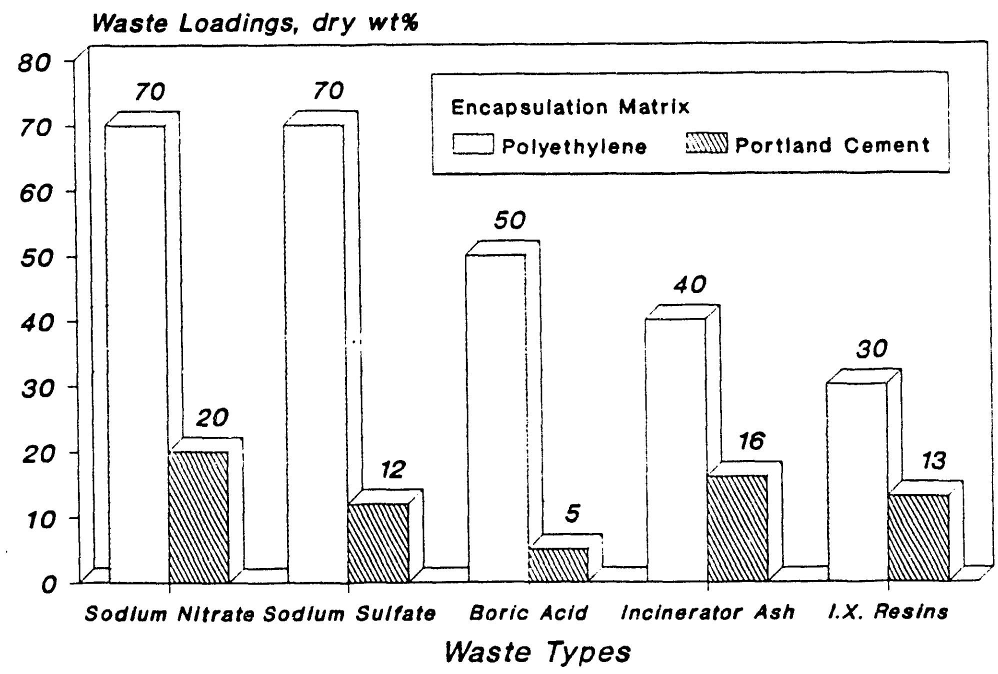

Figure 2 Maximum waste loadings (wt%) for polyethylene encapsulation compared with hydraulic cement processes.

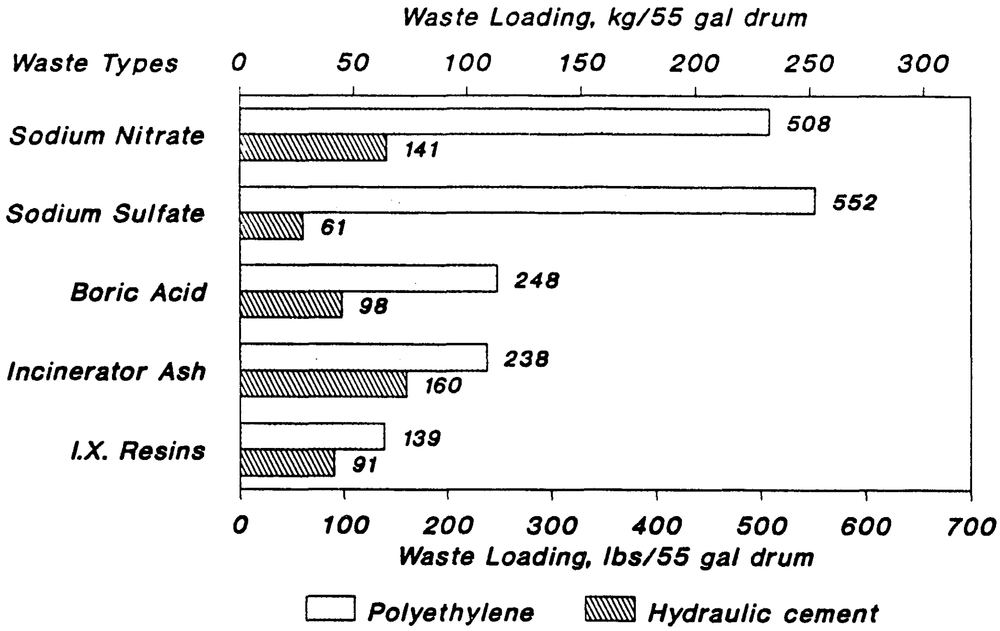

Figure 3 Maximum waste/drum for polyethylene encapsulation compared with hydraulic cement processes.

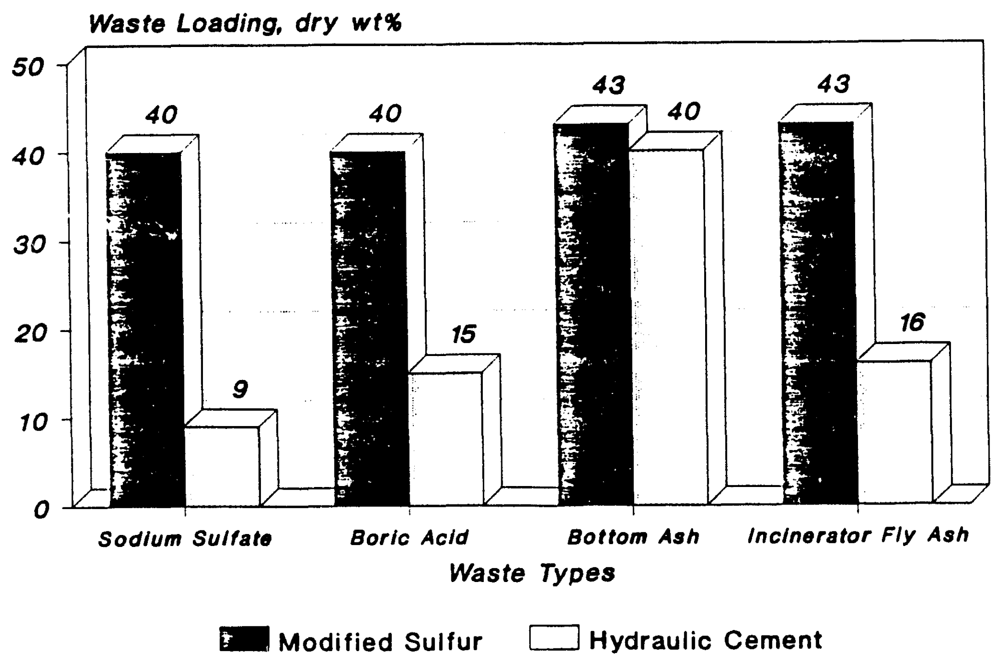

Figure 4 Maximum waste loadings (wt%) for modified sulfur cement compared with hydraulic cement processes.

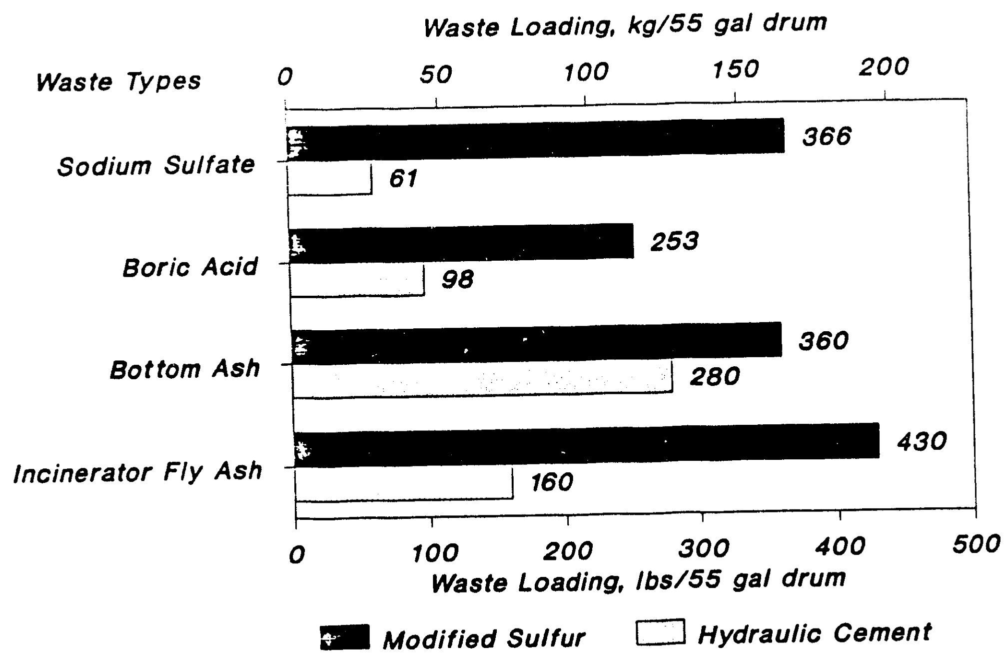

Figure 5 Maximum waste/drum for modified sulfur cement compared with hydraulic cement processes.

PROCESS SCALE-UP AND DEMONSTRATION

On satisfactory completion of bench-scale R & D, scale-up feasibility was investigated for polyethylene encapsulation of nitrate salt wastes. A production-scale feasibility test was conducted using a commercial-grade polyethylene extruder with a 4.5 in. (114 mm) diameter screw and an output capacity of about 2,000 lbs/hr (900 kg/hr). Technical grade sodium nitrate was used to simulate actual mixed waste nitrate salts at a waste loading of 60 wt%. Figure 6 is a process flow diagram for the production-scale test indicating typical parameters. A 30 gallon drum (114 liter) of encapsulated "waste" was filled in about 25 minutes for an average flow rate of about 72 gal/hr (273 l/hr). The resulting product was sectioned to inspect for potential void formation, verify homogenous mixing and provide test specimens for additional confirmatory performance testing. Results of the feasibility test and performance testing are presented in Reference [3], but can be summarized in the following points:

- Polyethylene encapsulation of at least 60 wt% nitrate salt wastes can successfully be accomplished using a production-scale extruder,
- Bench- and production-scale process data are in close agreement,
- QA/performance testing of the 30 gal. waste form demonstrates that a homogenous product with excellent performance properties can be produced using off-the-shelf production equipment.

Based on these results, a production-scale extruder was procured for a technology demonstration to be held at BNL during this fiscal year. The demonstration will be conducted using either actual mixed waste from a DOE facility or surrogate waste that closely approximates actual waste in both chemical and physical composition. This demonstration will be open to all interested parties including those from DOE, NRC, EPA, and the commercial sector.

Scale-up activities for the modified sulfur cement process are continuing and demonstration of production-scale feasibility is planned by the end of FY-1992. The focus of this demonstration will be treatment of mixed waste incinerator fly ash generated at both DOE and commercial facilities.

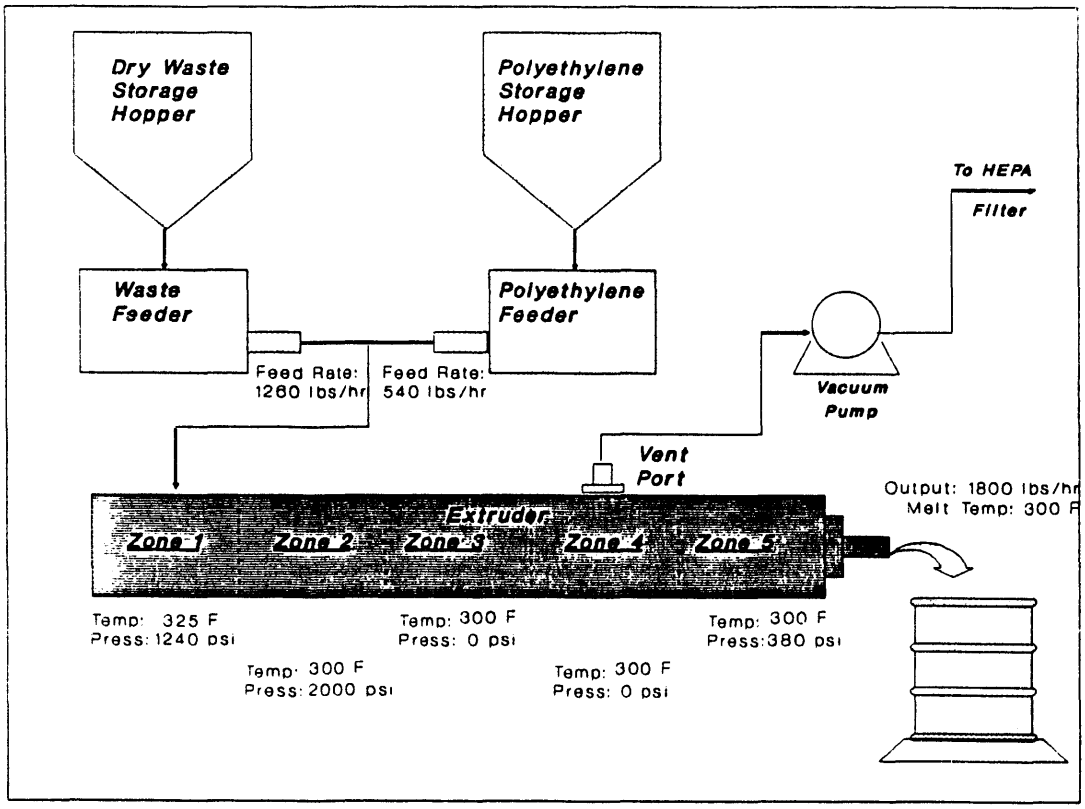

Figure 6 Process flow diagram for full-scale polyethylene encapsulation system.

SUMMARY AND CONCLUSIONS

Waste management technology development and demonstration efforts conducted at BNL are aimed at providing improved methods of treating radioactive, hazardous, and mixed wastes. In keeping with DOE OTD policy, processes must be "better, faster, cheaper, and safer" than conventional technologies. Two systems developed to date (polyethylene and modified sulfur cement encapsulation) have been shown to provide better waste form performance under long-term disposal conditions and improved waste loadings, on a cost-effective basis.

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