ENGINEERING DEVELOPMENT OF PROCESSING METHODS FOR REMOTELY HANDLED TRANSURANIC SLUDGE AT OAK RIDGE NATIONAL LABORATORY*

J. B. Berry, E. L. Youngblood, J. T. Shor,
J. F. Walker, T. L. White, and A. J. Mattus
Chemical Technology Division
Oak Ridge National Laboratory
Oak Ridge, TN 37831-6046

ABSTRACT

The Waste Handling and Packaging Plant (WHPP) is identified as a key element in the U.S. Department of Energy's transuranic (TRU) waste program for both remotely handled (RH) and special case (SC) waste. The mission of the facility is to retrieve, receive, repackage, certify, and ship TRU waste to the Waste Isolation Pilot Plant (WIPP) located near Carlsbad, New Mexico (1).

RH slurry will be removed from underground storage tanks and processed to produce a waste form that meets the WIPP waste acceptance criteria. A development facility is being constructed at the Oak Ridge National Laboratory (ORNL) to provide engineering data needed for the design of slurry processing systems. Surrogate waste is being used during testing of equipment. This paper discusses progress on developing unit operations for slurry processing in the WHPP.

INTRODUCTION

ORNL, from its inception in the early 1940s, has operated numerous facilities which have generated radioactively contaminated liquid waste. During much of this period, these wastes have included significant levels of TRU elements because of early operation of nuclear fuel reprocessing pilot plants, as well as the preparation and use of radioisotopes. Through 1984, the liquid waste was concentrated by evaporation, converted to a grout, and disposed of on the Oak Ridge Reservation via hydrofracture in shale formations ~1100 ft (~335 m) below the surface. The use of hydrofracture was terminated after 1984, and liquid radioactive waste concentrate has been accumulated and stored since that time.

The volume of stored liquid radioactive waste is maintained near the safe-fill limit for the storage tanks by implementing a grout solidification campaign on an as-needed-basis. The solidified waste (non-transuranic) has been stored on site, pending resolution of waste form performance criteria and disposal requirements. Operational constraints are experienced due to the large inventory of stored liquid radioactive waste. It is recognized that a lack of adequate storage volume jeopardizes ORNL's ability to ensure continued conduct of research and development activities that generate liquid radioactive waste, because an unexpected operational incident could quickly deplete the remaining storage volume. Planned solidification campaigns and additional concentration by evaporation of the liquid radioactive waste will be used to maintain a safe-fill limit until a permanent disposal method for the waste can be implemented.

The tanks that provide the storage capacity of the active liquid radioactive waste system contain significant volumes of TRU sludges that have been designated remote-handled transuranic (RH-TRU) waste because of associated quantities of other radioisotopes, including Sr-90 and Cs-137. A method for disposing of the TRU waste is being developed and will be implemented by the construction (2) of WHPP.

The WHPP is a proposed $245 million 1993 line-item project. The facility will prepare RH-TRU slurry, as well as RH-TRU solid waste, for shipment to the WIPP. (A discussion of the RH-TRU solid waste handling aspects of WHPP are beyond the scope of this paper.) The design of the WHPP includes remote operation and maintenance of processing equipment in a specially-designed hot cell. A simplified flowsheet of the preliminary conceptual design for slurry processing is shown (3) in Fig. 1. Engineering development has been directed toward improving the slurry processing technology. Slurry processing includes the following steps: (a) sludge mobilization; (b) evaporation and solidification by conventional technology; and (c) evaporation and solidification by microwave technology. Waste chemistry characterization and waste product certification is necessary to support engineering development. This paper discusses engineering development progress to date in each of the above areas.

WHPP DEVELOPMENT FACILITY

The WHPP Development Facility (WDF) is being constructed at ORNL to provide engineering data needed for the design of the slurry processing systems that will be

* Research sponsored by the Office of Defense Waste and Transportation Management, U.S. Department of Energy, under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

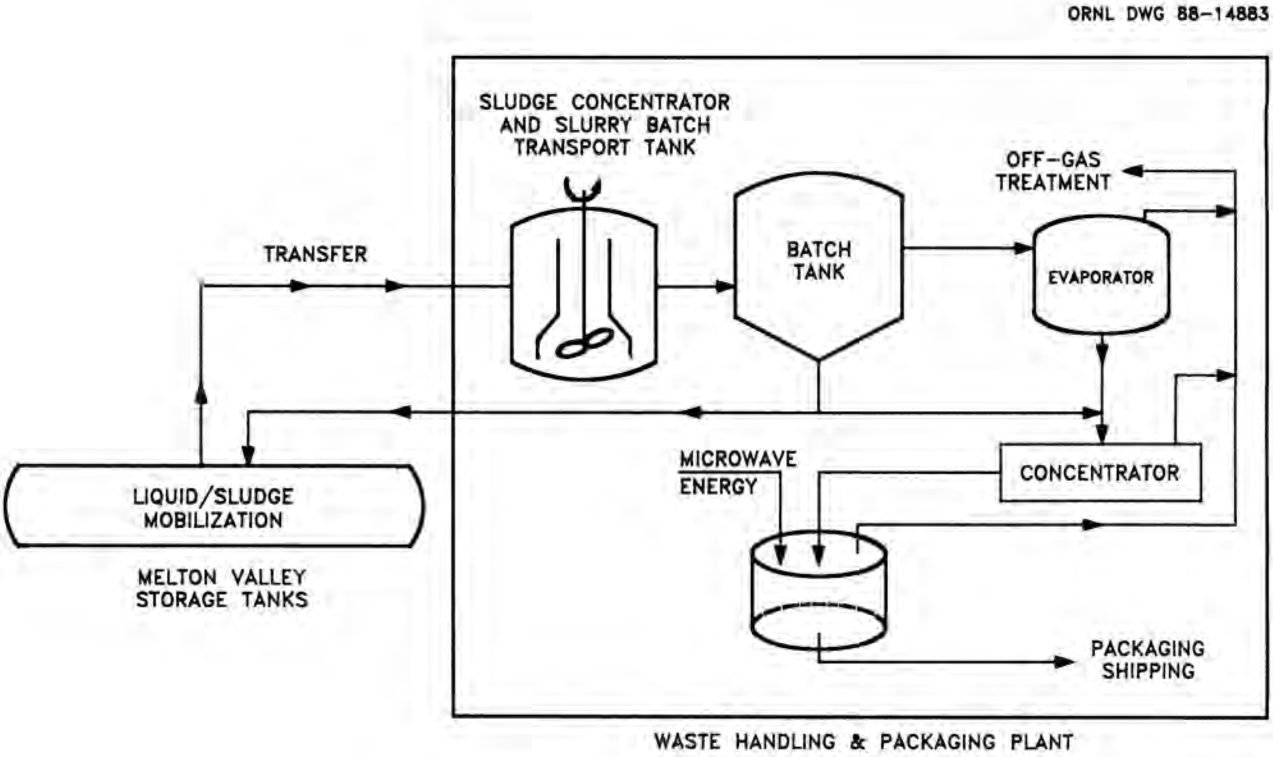

Fig. 1. WHPP Slurry Processing Conceptual Flow Diagram as Included in the Conceptual Design Report.

installed in the WHPP. The WDF will be used to select and evaluate the methods and equipment that are suitable for mobilization, transport, and solidification of the TRU sludge contained in the Melton Valley Storage Tanks (MVSTs). Tests will be conducted using a nonradioactive surrogate sludge that simulates the chemical and physical properties of the MVST sludge.

The WDF will include a 25,000 gal (94.6 m^3) mockup of the MVSTs and supporting equipment, such as tanks, pumps, and instrumentation, for testing and evaluation of methods of sludge mobilization. The system will be initially set up for testing fixed point jets (water or supernate) and jets attached to a scavenging robot for sluicing the sludge from the tank. The WDF will also include base-line equipment (tanks, pumps, condensers) for development and testing of processes for concentration and drying of the sludge and supernate. Processes that will be evaluated for sludge solidification include combinations of thin-film evaporation, microwave drying, and extrusion. The facility will also be used for development and evaluation of instrumentation critical to the certification of the final waste product.

Process flowsheets have been developed; preparation of specifications for instrumentation has been initiated; the 25,000 gal (94.6 m^3) model tank has been installed; and installation of utilities such as water, steam, and electricity has begun. Equipment requiring long delivery time has been ordered.

SLUDGE MOBILIZATION AND TRANSPORT DEVELOPMENT

A common problem of long-term storage of non-acidified liquid radwastes is the formation of sludges (4) of high curie content. This is true of the MVSTs at ORNL which have all been found to contain bottom sludges of various depths, consistencies, and compositions. The objectives of the sludge mobilization development task are threefold: to remove the waste sludges from the tanks, transport them to the proposed WHPP, and there to prepare a mixture of sludge and supernate which would produce a solidified product which meets the WIPP waste acceptance criteria.

The MVSTs are 60 ft (18 m) long horizontal cylinders 12 ft (3.6 m) in diameter with one access manhole 15 ft (4.6 m) from the tank center, a geometry that poses a significant challenge to sludge mobilization. Two techniques to mobilize and remove sludge in this geometry have been under study in a one-sixth linear scale model of the MVSTs. The first involves the near emptying of the tank of supernate and the introduction of a high pressure solid stream spray nozzle into the tank through the man-way. The nozzle is manipulated to direct the slurry of liquid and solids towards the manhole area where it is pumped from the tank. This was the method used successfully in the Gunite Tank Cleaning Program (5), although the shape of the Gunite tanks was different. The second technique is more developmental and involves the use of an industrially available robot which travels along the bottom of the tank pulling its own suction line and which carries a very high pressure water jet to break apart extremely hard sludge. Both techniques will be tested in the WDF. The sludge mobilization flowsheet of the WDF is presented in Fig. 2.

A highly instrumented experimental pipe loop has been built to study the flow of simulated slurries which are formulated based on data from samples of MVSTs sludges. The simulated slurries were formulated based on the hindered settling velocities and rheological properties of the sludges. These properties, in turn, depend on the state of particle aggregation, size, and density. The simulated slurries were designed to be more of a "worst case" than the actual sludges. A Kingston fly ash and water slurry has been found to be a slightly more rapidly settling than the soft MVST sample and, therefore, a good surrogate for slurry transport studies. Pipe flow data, head loss through fittings, and minimum transport velocity data have been obtained. Instrumentation is being evaluated for its sensitivity to process parameters such as density. A typical plot of pressure drop versus flow rate with concentration as a parameter is shown in Fig. 3. Preliminary results are consistent with Thomas' observation (6) that slurries exhibiting low Bingham plastic yield stresses [less than 240 dynes/cm2 (24 Pa)] can be correlated using the Moody plot of Fanning friction factors. The basis of the Thomas correlation is the use of a modified slurry viscosity in the Reynolds number which is applied to the Moody plot and which does not further involve the diameter of the tube as a parameter.

CONVENTIONAL EVAPORATION DEVELOPMENT

The Preliminary Conceptual Design Report for WHPP calls for evaporation of the water from the sludge which is mobilized from the MVSTs. (See Fig. 1.) The concentrated sludge will be transported to 55-gal (0.21 m^3) drum liners, where the sludge will solidify upon cooling. Two technologies are being considered for concentration of the sludge from the MVSTs: (a) conventional evaporation and (b) microwave energy. Development efforts will determine if the WHPP will contain both an evaporator and a microwave system or if the process will rely on one or the other technology. The evaporation product will be a stable waste form which meets the waste acceptance criteria for WIPP.

The primary objectives of the conventional evaporation development task are to determine the feasibility of using commercially available evaporation equipment to concentrate the MVSTs sludges and to evaluate the various evaporator options. To accomplish this, a surrogate feed has been used for vendor demonstration tests (7). These tests have utilized facilities at various manufacturers to determine the applicability of commercial equipment for concentrating the surrogate waste. Equipment which is most promising will be evaluated in pilot tests at the WDF. To date vendor demonstration tests have been conducted using: (a) a vertical thin-film evaporator, (b) a horizontal thin-film evaporator, and (c) an extruder.

The agitated thin-film evaporator utilizes a rotor to spread a turbulent thin layer of solution over the heated surface. Vendor tests of both vertical and horizontal units indicate that the thin-film evaporators can be used to produce either a thick paste or a powder. Use of a thin-film evaporator may eliminate the need to evaporate the liquid and solid streams separately and may replace the kettle evaporator specified in the Preliminary Conceptual Design Report. Thin-film evaporators will be further evaluated in the WDF.

A twin-screw extruder/evaporator was evaluated as a process candidate for drying and melting MVSTs waste to meet waste form criteria. Tests were structured to evaluate the versatility of the extruder to (a) dehydrate the surrogate waste and produce a powdered/granular product which was melted in a second extruder and (b) dehydrate and melt the slurry in a single step. The extruder proved capable of operating in both modes. In each mode the process produced a product which met the waste form criteria. The primary disadvantages of the extruder include (a) cost, (b) multiple steam domes, and (c) close tolerances of screws. A decision to include or exclude the extruder from the WDF testing has not been made.

The conventional evaporation flowsheet of the WDF is presented in Fig. 4. All major components for this system have been ordered, and construction has been initiated.

MICROWAVE PROCESS DEVELOPMENT

Microwave processing of surrogate RH-TRU sludges and liquids is being investigated as an alternative to conventional evaporation technology (8). Results of bench-scale tests using a 1 kW 2450 MHz oven showed that the surrogate can be dried and then melted in a single process. Acoustic emission feedback control was developed to regulate the amount of boiling in the surrogate. A 1/3-scale applicator (6 kW at 2450 MHz) for in-can evaporation and melting has been fabricated. Both batch and continuous processing modes will be investigated. The applicator will employ a wide range of process controls in a PC-based data acquisition and control system. The 1/3-scale microwave system flowsheet is shown in Fig. 5.

A full-scale applicator capable of processing a 55-gal (0.21 m^3) drum of surrogate has been designed and will operate at 60 kW and 915 MHz in the WDF. Data from the

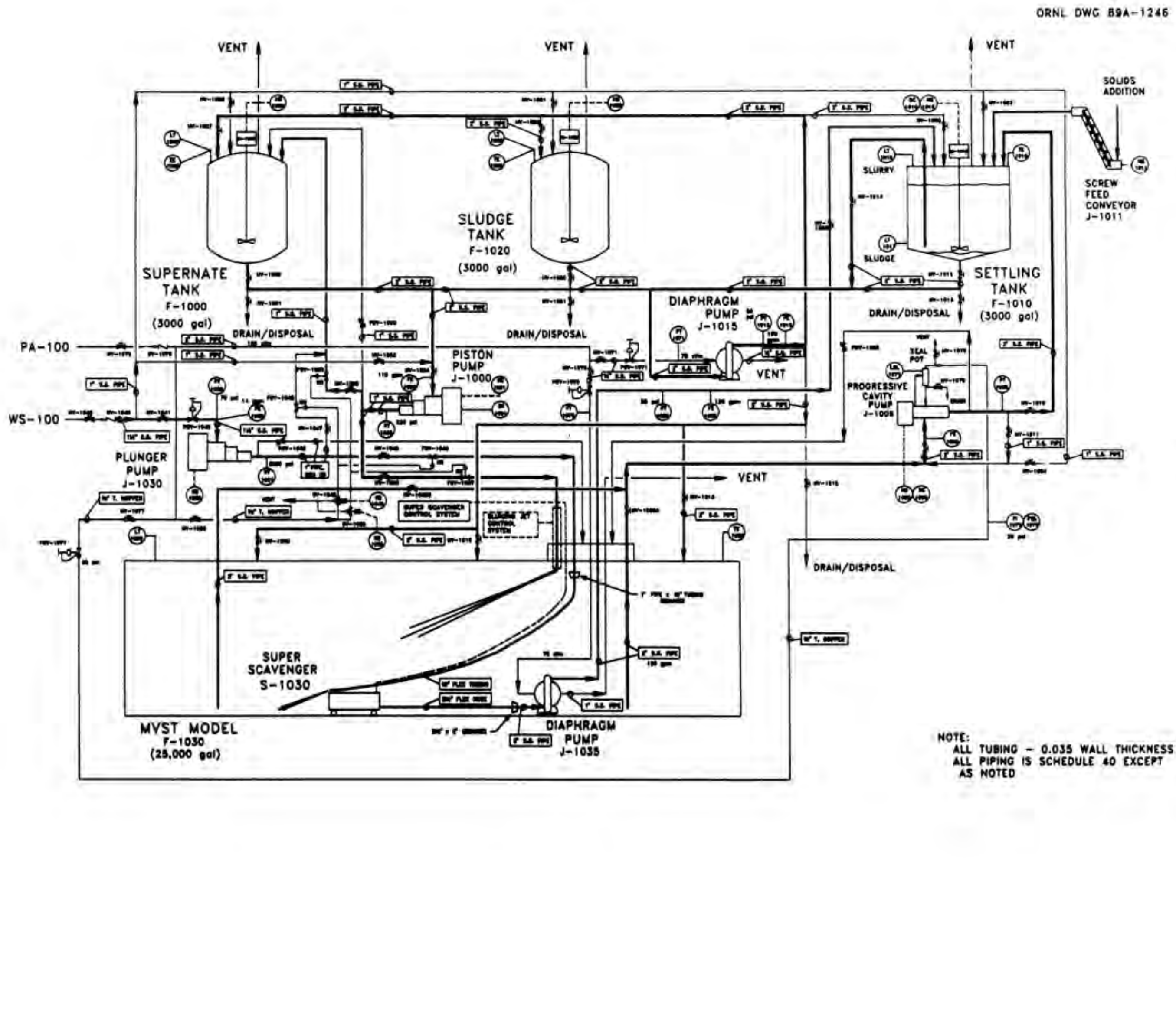

Fig. 2. Sludge Mobilization Flowsheet to be Installed at the WHPP Development Facility.

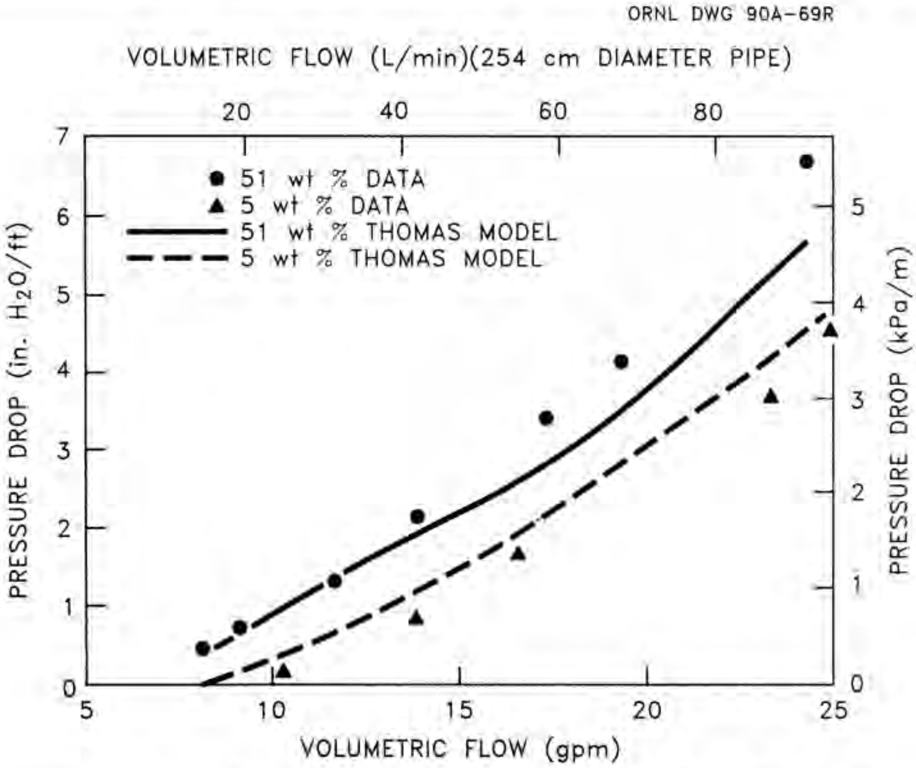

Fig. 3. Comparison of Thomas Model Predictions with Slurry Flow Data.

1/3-scale experiment will be used to optimize the design of the full-scale equipment.

WASTE CHARACTERIZATION AND PRODUCT CERTIFICATION

The liquid, low-level waste contained in the MVST's is composed of an alkaline, sodium nitrate-based waste with a TRU sludge on the bottom of each tank. A blend of the sludge and nitrate-based aqueous waste will be evaporated by either a low or high temperature process to form a salt monolith for shipment to WIPP. In preparation for this disposal, laboratory tests have been run on salt products generated from either the low or high temperature processes to observe product properties for WIPP certification. The WIPP waste acceptance criteria covers many (9) parameters. The parameters paramount to WHPP performance with MVSTs material are the absence of free liquid and particulate matter ≤200 mesh (≤74 μm) in size.

Laboratory testing has been performed on salt products developed from a non-radiological surrogate waste which represents the contents of the MVST's. An extensive sampling of tanks campaign is in progress; data are being collected throughout 1990. The surrogate used in testing to date has been formulated based upon: chemical, radiological, and physical waste characteristics of tank W-26. As more waste characterization data are available, different surrogates will be used in product certification testing to observe the effect of varying chemical constituents on the final waste form.

Laboratory and full-scale processing scenarios include (a) evaporating liquids at 110 to 125°C, or (b) evaporating and melting the salt at 310 to 340°C. Salt products developed by both methods have been studied to ensure that the product meets WIPP waste acceptance criteria and does not exhibit undesirable properties. The primary tests utilized to study salt product properties are shown in Table I.

Comparatively, some select results to date show that both high and low temperature salt products are highly

TABLE I

Laboratory Tests of TRU Salt Products

Physical stability (expansion/contraction)
Hygroscopic properties at 64 and 84% relative humidity
Chemical segregation
Residual Water
Particle sizing
Thermal gravimetry
Bulk density
Potential for NOx generation
Radiolysis and criticality potential

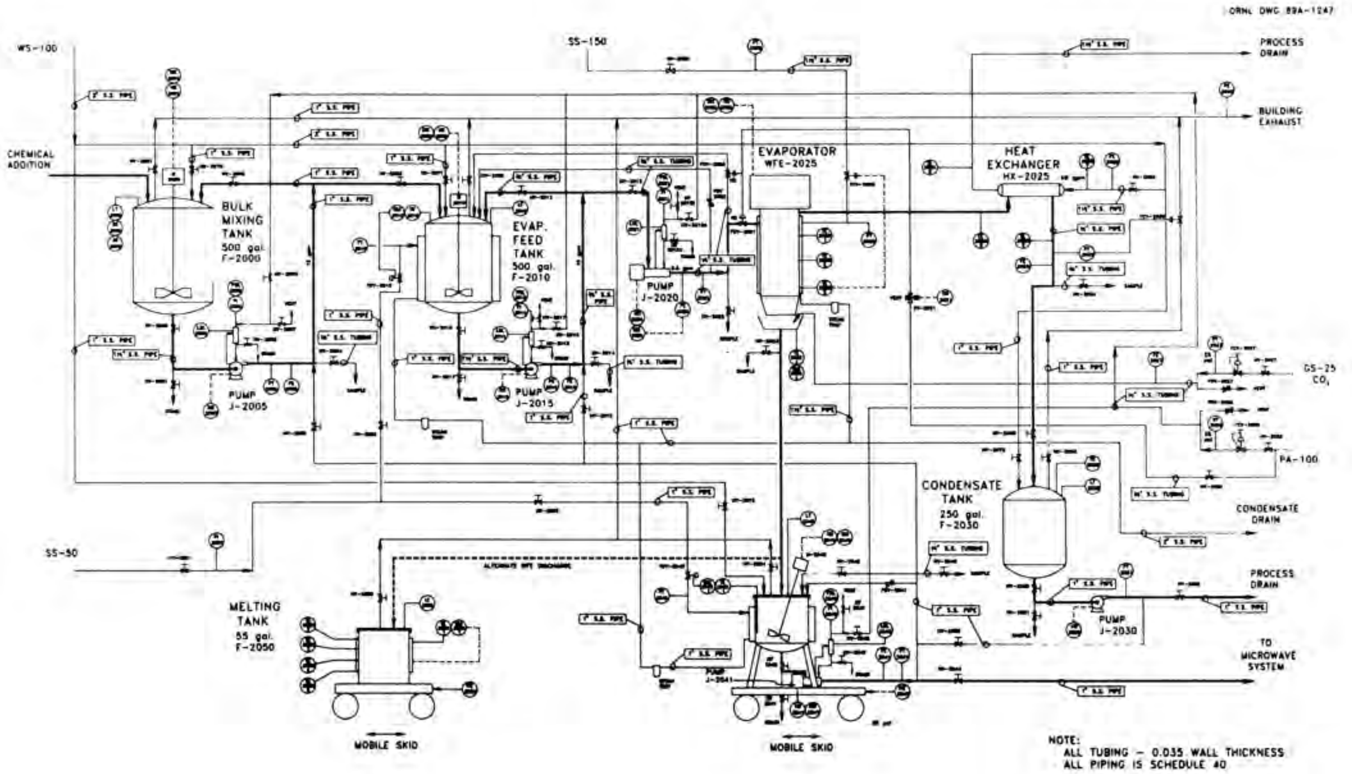

Fig. 4. Conventional Evaporation System Flowsheet to be Installed at the WHPP Development Facility.

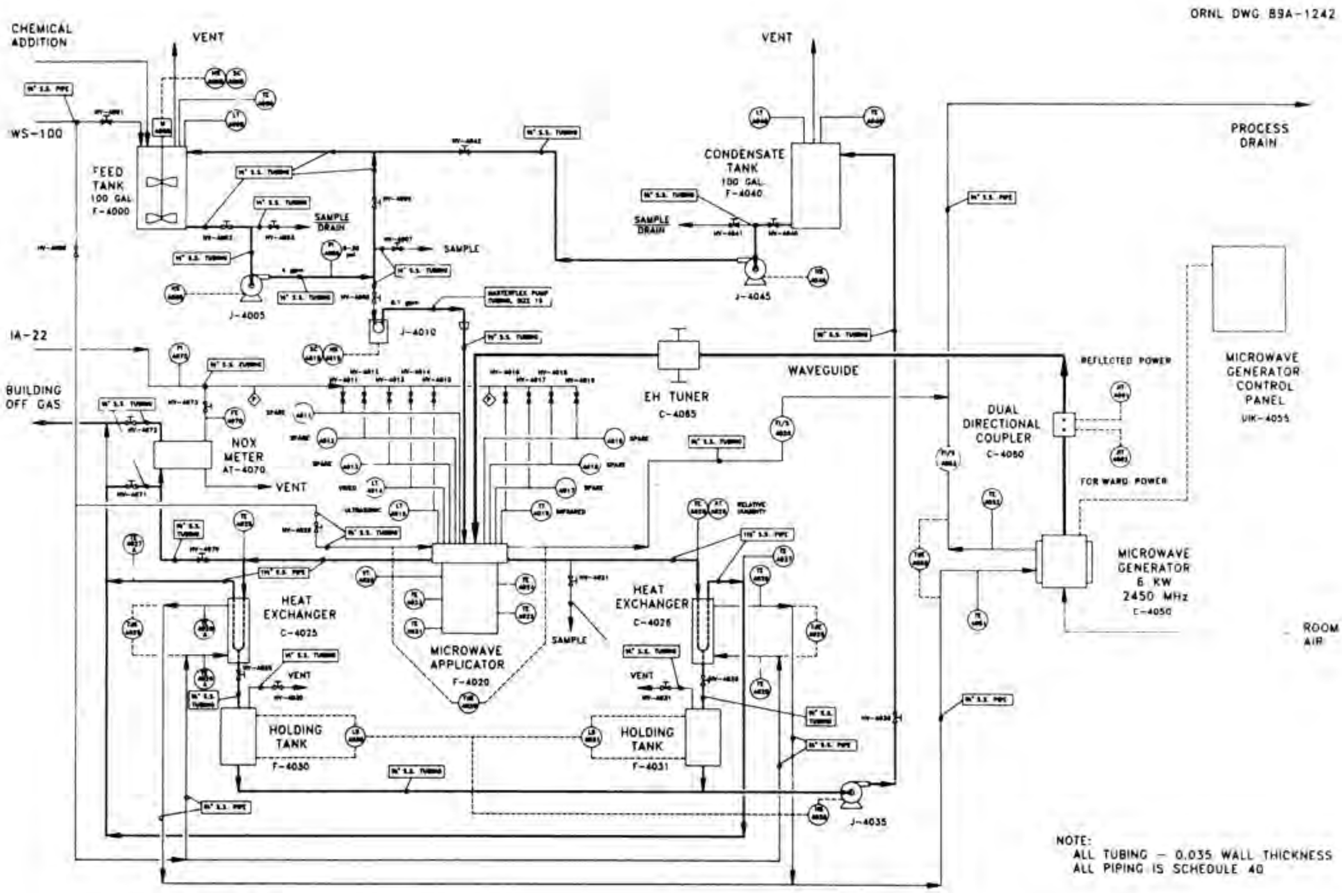

Fig. 5. Prototype 1/3-Scale Microwave Evaporation System.

hygroscopic due primarily to the presence of sodium hydroxide and calcium or magnesium chloride. The melted product presents a much lower salt surface area to humid air and therefore absorbs water at a lower rate when the residual water content is the same in both types of product. The melted salt product also presents a better opportunity for a higher volume reduction since, based upon bulk densities, the melted product is between 15 to 30% denser.

SUMMARY

A permanent disposal method for ORNL RH-TRU liquid waste is needed. The WHPP will be constructed to prepare RH-TRU slurry for disposal at WIPP. A development facility is being constructed and will be used to conduct extensive tests prior to selecting the unit operations that will be installed in the WHPP. Engineering development for slurry processing technology includes sludge mobilization and transport and slurry evaporation and solidification by conventional and microwave techniques. Engineering development is being supported by waste chemistry characterization. An in-depth understanding of waste form performance with regard to the WIPP waste acceptance criteria and transportation requirements will ensure that the waste product can be certified.

ACKNOWLEDGEMENTS

The authors wish to express their appreciation to the following individuals for supporting engineering development efforts: V. L. Fowler, R. C. Cummins, Wiratmo Suyatno, Ben Lie Pen, R. J. Vedder, and W. D. Box.

REFERENCES

1. D. W. TURNER, et al., "An Overview of the ORNL Waste Handling and Packaging Plant," ORNL/TM-Draft (In Preparation), Martin Marietta Energy Systems, Inc., Oak Ridge National Laboratory (Jan. 1990)

2. L. E. MCNEESE, et al., "Overall Strategy and Program Plan for Management of Radioactively Contaminated Liquid Wastes and Transuranic Sludges at the Oak Ridge National Laboratory," ORNL/TM-10757, Martin Marietta Energy Systems, Inc., Oak Ridge National Laboratory (Dec. 1988).

3. Preliminary Conceptual Design Report for the Waste Handling and Packaging Plant, X-OE-453, Martin Marietta Energy Systems, Inc., Oak Ridge National Laboratory (May 1989).

4. MANSON BENEDICT, et al., "Nuclear Chemical Engineering," McGraw-Hill, New York, 1981, p. 576.

5. H. O. WEEREN, "Sluicing Operations at Gunite Waste Storage Tanks," ORNL/NFW-84/42, Martin Marietta Energy Systems, Inc., Oak Ridge National Laboratory (Sept. 1984).

6. D. G. THOMAS, "Significant Aspects of Non-Newtonian Technology", Progress in International Research on Thermodynamic and Transport Properties, ASME, New York, 1962, p. 669.

7. R. J. VEDDER, "Selection of an Agitated Thin-film Evaporator for Processing a Radioactive Waste at ORNL," ORNL/CF-89/289, Martin Marietta Energy Systems, Inc., Oak Ridge National Laboratory (Oct. 1989).

8. T. L. WHITE, et al., "Status of Microwave Process Development for RH-TRU Wastes at Oak Ridge National Laboratory," ORNL/TM-Draft (In Preparation), Martin Marietta Energy Systems, Inc., Oak Ridge National Laboratory (Jan. 1990)

9. DEPARTMENT OF ENERGY, Joint Integration Office, "TRU Waste Acceptance Criteria for the Waste Isolation Pilot Plant," WIPP-DOE-069, Rev. 2 (Sept. 1985).