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STATUS OF MICROWAVE PROCESS DEVELOPMENT FOR RH-TRU WASTES AT OAK RIDGE NATIONAL LABORATORY\*

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ABSTRACT

The Oak Ridge National Laboratory (ORNL) Waste Handling and Packaging Plant is developing a microwave process to reduce and solidify remote-handled transuranic (RH-TRU) liquids and sludges presently stored in large tanks at ORNL. Testing has recently begun on an in-drum microwave process using nonradioactive RH-TRU surrogates. The microwave process development effort has focused on an in-drum process to dry the RH-TRU liquids and sludges in the final storage container and then melt the salt residues to form a solid monolith. A 1/3-scale proprietary microwave applicator was designed, fabricated, and tested to demonstrate the essential features of the microwave design and to provide input into the design of the full-scale applicator. The microwave fields are uniform in one dimension to reduce the formation of hot spots on the microwaved wasteform. The final wasteform meets the waste acceptance criteria for the Waste Isolation Pilot Plant, a federal repository for defense transuranic wastes near Carlsbad, New Mexico.

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## Introduction

The ORNL Waste Handling and Packaging Plant (WHPP) (1) at Oak Ridge National Laboratory (ORNL) is a facility that will process and repackage remote-handled transuranic (RH-TRU) liquid and solid wastes from ORNL and solids from other sites for shipment to the Waste Isolation Pilot Plant (WIPP) (2). The liquid RH-TRU wastes, stored in stainless steel tanks, consist of a watery supernatant/sludge mixture containing mostly  $\text{NaNO}_3$  with the remaining compounds consisting of a complicated soup of chemicals that contain the RH-TRU wastes. The WHPP liquids processing flow sheet calls for evaporation of the free water in the wastes followed by melting of the salt residues. The cooled melt will form a solid monolith because of the binding action of the  $\text{NaNO}_3$  on the remaining solids. The final waste volume will be much less than the original waste volume because of water removal and consolidation/melting of the salt residues. The wasteform satisfies the WIPP waste acceptance criteria (WAC) (3) which state that no free particulates or liquids should be present in the final wasteform (currently the WIPP WAC allow soluble salts containing TRU waste to be stored). The temperature of the melt must be limited to prevent the destruction of the nitrates into  $\text{NO}_x$  gases which would necessitate extensive offgas scrubbing systems for the WHPP.

In the conceptual design stage, the WHPP process required RH-TRU liquid evaporation and melting to be performed by conventional wiped-film evaporators or extruders. More detailed descriptions of the development of the conventional WHPP processes are reported elsewhere (4). In this concept, microwaves would be used only to maintain the temperature in a drum during filling operations. Last year, however, we showed that microwaves can be used to process the RH-TRU liquids over the entire temperature range in a batch microwave oven (5). The wasteform satisfied the WIPP WAC and was similar to wasteforms produced by conventional evaporation technologies. Because of this success, we are developing a more aggressive microwave processing flow sheet that has

the potential to greatly simplify the conceptual WHPP liquids processing flow sheet by replacing the wiped-film evaporator/extruder/microwave drum heater with a single microwave in-drum process. Another possibility is using a conventional evaporator, such as a wiped-film evaporator, to concentrate the liquid, which would then be melted by the in-drum microwave process.

### Microwave Versus Conventional Processes

The microwave energy heats the RH-TRU liquid waste directly because the oscillating electric fields directly couple to the molecular bonds of the chemicals in the waste, causing frictional heating (6). This direct heating eliminates the need for separate heating elements or heat transfer surfaces, which are required by the wiped-film evaporator and extruder. The wiped-film evaporator and extruder also require moving parts to wipe a thin film across a heat transfer surface to heat the waste. These moving parts are subject to wear, especially if small hard particles are present in the waste (from past ORNL records, some of the RH-TRU storage tanks may contain hard particles such as grout and other hard deposits). Frequent maintenance of the wiped-film evaporator or extruder would be very complicated in a remote-handled hot cell environment. The microwave process that we are developing contains no moving parts and is designed to heat the liquid waste in the final storage container, thus eliminating the need to transport hot chemicals from the heated casings of the wiped-film evaporator or extruder into the storage container.

The wiped-film evaporator and extruder produce a more homogeneous product than the microwave process because of the mixing action of their mechanical process movers (a rotor/blade in the case of the wiped-film evaporator and twin co-rotating screws in the case of the extruder). However, there is no WIPP requirement on wastefrom homogeneity at present.

The microwave process is much less developed than the conventional technologies, and the goal is to develop a full-scale microwave process so that all technologies can be at the pilot plant stage before the WHPP is built. Finally, the direct heating of the microwave energy must be carefully controlled to avoid process upsets such as splattering, arcing, and thermal runaway.

## System Description

The 1/3-scale microwave system is shown in Fig. 1. The surrogate RH-TRU liquid is stored in a mixing tank and pumped through a slurry transport loop to keep solids from clogging the loop. A smaller metering pump taps off a small portion of the loop flow near the applicator in order to control the amount of surrogate that is fed to the applicator. The line from the metering pump to the applicator is kept as short as possible to avoid clogging. The metering pump is reversible so that surrogate in the short line can be drawn back out of the applicator feed pipe. The applicator feed pipe is located above, and along the axis of, the stainless steel waste container (18 cm diameter by 23 cm deep, 4.6-L usable volume). The applicator is powered by a 6-kW, 2450-MHz microwave generator. Forward and reflected microwave power are monitored by a dual-directional waveguide coupler (50 dB coupling, 30 dB directivity) so that net absorbed microwave power can be measured. An E-H tuner matches the applicator to the waveguide system to maximize the power absorbed by the applicator. The proprietary applicator couples the microwave energy to the waste container via an array of slots machined in the waveguide. The microwave electric fields are controlled in the applicator by carefully choosing the length, width, and orientation of the slot array. The microwave fields produced by the slots form a unique electric field pattern in the container that minimizes the likelihood of arcing at the walls while maximizing the uniformity of the microwave heating. The slots are purged with air to keep water vapor and particulates out of the slots and waveguide system. The offgases from the waste are removed in a symmetrical fashion by openings around the sides of the applicator.

The openings are connected by a manifold, which feeds a pair of heat exchangers to remove water vapor from the offgases. The distillate is collected and stored for later analysis and/or recycling to the mixer tank. An NO<sub>x</sub> meter is connected to the offgas system to monitor the amount of NO<sub>x</sub> gases produced during processing. A humidity/temperature probe is being procured to monitor the drying of the wasteform during processing.

Nine diagnostic ports have been located in the top of the microwave applicator, with each port having its own air purge. Two large ports are 2.3 cm I.D. and seven smaller ports are 0.6 cm I.D. One of the two large ports contains a color TV camera which is connected to a time-lapse video recording system to document the applicator performance and physical appearance of the microwaved wasteform during processing. The other large port contains an infrared sensor connected to a programmable temperature controller that continuously monitors the surface temperature of the wasteform during processing. The temperature controller produces an output signal proportional to the difference between the actual surface temperature and a preprogrammed temperature versus time profile stored in the controller's memory. This output signal is connected to a silicon controlled rectifier (SCR) controller in the generator which controls the microwave power level. This results in a closed-loop feedback control system to regulate surface temperature. A rigid borescope for illumination and viewing of the wastes and a thermocouple are attached to two of the seven small ports. Five spare ports are available for future diagnostics. An ultrasonic level control was to be implemented on the applicator to measure the waste level in the container, but we plan to defer procurement pending evaluation of a more reliable alternative technology. Other future upgrades include a PC-based data acquisition and control system to develop automated process control strategies. During earlier bench-scale work in a modified microwave oven (7), we developed a proprietary acoustic emission feedback control to regulate the degree of boiling in the surrogate. This was required because

temperature control alone is not always sufficient to preclude splattering of liquids. We plan to implement this technology on the 1/3-scale applicator in the near future.

### Initial System Performance

Early testing of the applicator involved verifying the microwave design. Some testing was done at milliwatt power levels to optimize the geometry of the applicator slots. Also, probe measurements were made of the electric field profile across the container diameter to verify the correct microwave field distribution. Further testing at the 1-kW power level was done using a sandwich of thermal paper between thin sheets of plate glass. The microwaves heat the plate glass directly, and the glass is in good thermal contact with the thermal paper. The thermal conductivity is poor in the plane of the glass so that microwave heating patterns diffuse relatively slowly in the plane compared to normal to the glass plane. The thermal paper is exposed first where the heating rate is highest and produces a dark image. The thermal paper technique produces a two-dimensional map of the microwave heating profile.

For actual surrogate experiments we used the analysis of tank W-26 shown in Table I. The surrogate contains a fine SiC powder which is used to simulate the strong microwave-absorbing properties of  $U_2O_3$ . Uranium was not used in this surrogate because its use would have classified the surrogate as low-level radioactive mixed waste and would have precluded testing in the present location. Also, use of uranium oxide would have made the task of optimizing the applicator very difficult. When fully mixed, the surrogate has the appearance of dilute chocolate milk. The solids settle out of solution to form a fine brown sludge and a clear supernatant in about 36 hours.

At this writing, we have just started initial microwaving of RH-TRU surrogate. What follows is a description of one of the early runs. The container is filled to a depth of 5 cm with surrogate and the microwave power is adjusted to the 1-kW level. The surface

temperature rises quickly while the subsurface temperature stays cool. This is because the surrogate has a high electrical conductivity that limits microwave penetration. Continued heating results in bubbling at the surface. A white scum floats on the dark brown liquid and is associated with the bubbling. This scum may be  $\text{NaNO}_3$  crystals that have been formed at the microwave/liquid interface. As the surface temperature reaches  $110^\circ\text{C}$ , the bubbling region forms a ring around the center of the surface. This ring corresponds to a peak in the desired microwave heating profile. At 40 minutes into the run, the bubbles become smaller and the white scum begins to dry out, creating white salt crystals. As heating proceeds the white salt crystals turn a dark brown color. This color change occurs near the melting temperature of  $\text{NaNO}_3$  ( $308^\circ\text{C}$ ). The temperature is increased to  $350^\circ\text{C}$  and at 60 minutes the feed is introduced into the center pipe at a  $10\text{-mL/min}$  flow rate. The first drops hit the center of the pool, vaporize, and cool the center to form a ring of  $\text{NaNO}_3$  salt crystals. The center area boils vigorously. The dark pool of molten  $\text{NaNO}_3$  liquid persists around the light ring of  $\text{NaNO}_3$  crystals. Continued feeding produces a crater in the middle consisting of boiling surrogate surrounded by a ring of  $\text{NaNO}_3$  deposits, followed by a dark pool of molten  $\text{NaNO}_3$  all the way to the container wall. When the feed is interrupted with the power on, the entire container reheats to molten  $\text{NaNO}_3$ . Upon cooling, the melt solidifies with no free liquids or particles. Some arcing was observed in the waveguide system, but no arcing was observed in the waste and no  $\text{NO}_x$  gases were generated. Initial operations have demonstrated the key features of the microwave design, but more work needs to be done to establish process repeatability and reliability. Optimization of the 1/3-scale equipment will help in the design of the full-scale equipment during the next fiscal year. Current work is focusing on evaluating the possibility of continuously processing the slurry or operating in a semi-batch mode where salt layers are built up in stages. One possible operating mode for the WHPP is to preheat and concentrate the slurry using a wiped-film evaporator whose output product would be a

120°C salt slurry. The preheated slurry could then be fed into the microwave applicator and heated to the final process temperature.

## Summary

Testing on a 1/3-scale microwave applicator to concentrate and melt RH-TRU liquids has just begun. Initial results show that the microwave heating is distributed on the surface of the surrogate liquid in a broad ring around the center of the container. Plans call for implementing a computer data acquisition and control system, a humidity probe, and an acoustic emission sensor to limit boiling.

Table I

## W-26 Surrogate Formulation for Microwave Processing

<u>COMPOUND</u>	<u>GRAMS PER LITER OF SURROGATE SOLUTION</u>
H <sub>2</sub> O	851
NaNO <sub>3</sub>	370
KNO <sub>3</sub>	12.9
Al(OH) <sub>3</sub>	6.96
NaCl	6.60
SiC (surrogate for U <sub>2</sub> O <sub>3</sub> and ThO)	2.82
CaHPO <sub>4</sub>	2.19
Ca(OH) <sub>2</sub>	1.77
Na(OH)	1.27
Wyoming Bentonite clay	1.06
CaCO <sub>3</sub>	0.94
Mg(OH) <sub>2</sub>	0.38
Fe <sub>2</sub> O <sub>3</sub>	0.38
Sr(NO <sub>3</sub> ) <sub>2</sub>	0.036
CsNO <sub>3</sub>	0.022

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