CONF-900466--54

FIRST RESULTS OF IN-CAN MICROWAVE PROCESSING EXPERIMENTS FOR RADIOACTIVE LIQUID WASTES AT THE OAK RIDGE NATIONAL LABORATORY*

T. L. WHITE, E. L. YOUNGBLOOD, J. B. BERRY, AND A. J. MATTUS
Oak Ridge National Laboratory, P. O. Box 2009, Oak Ridge, TN 37831-8071

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. Conductivity cell measurements suggest that the microwave energy heats near the surface of the surrogate over a wide range of temperatures. 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.

CONF-900466--54

DE90 012168

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 $NaNO_3$ 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 $NaNO_3$ 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 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

---

*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.

DISCLAIMER

wiped-film evaporator/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 wasteform 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 the stainless steel waste container (18 cm diam 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 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 recyling to the mixer tank. An NO_x meter is connected to the offgas system to monitor the amount of NO_x 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 ID, and seven smaller ports are 0.6 cm ID. 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

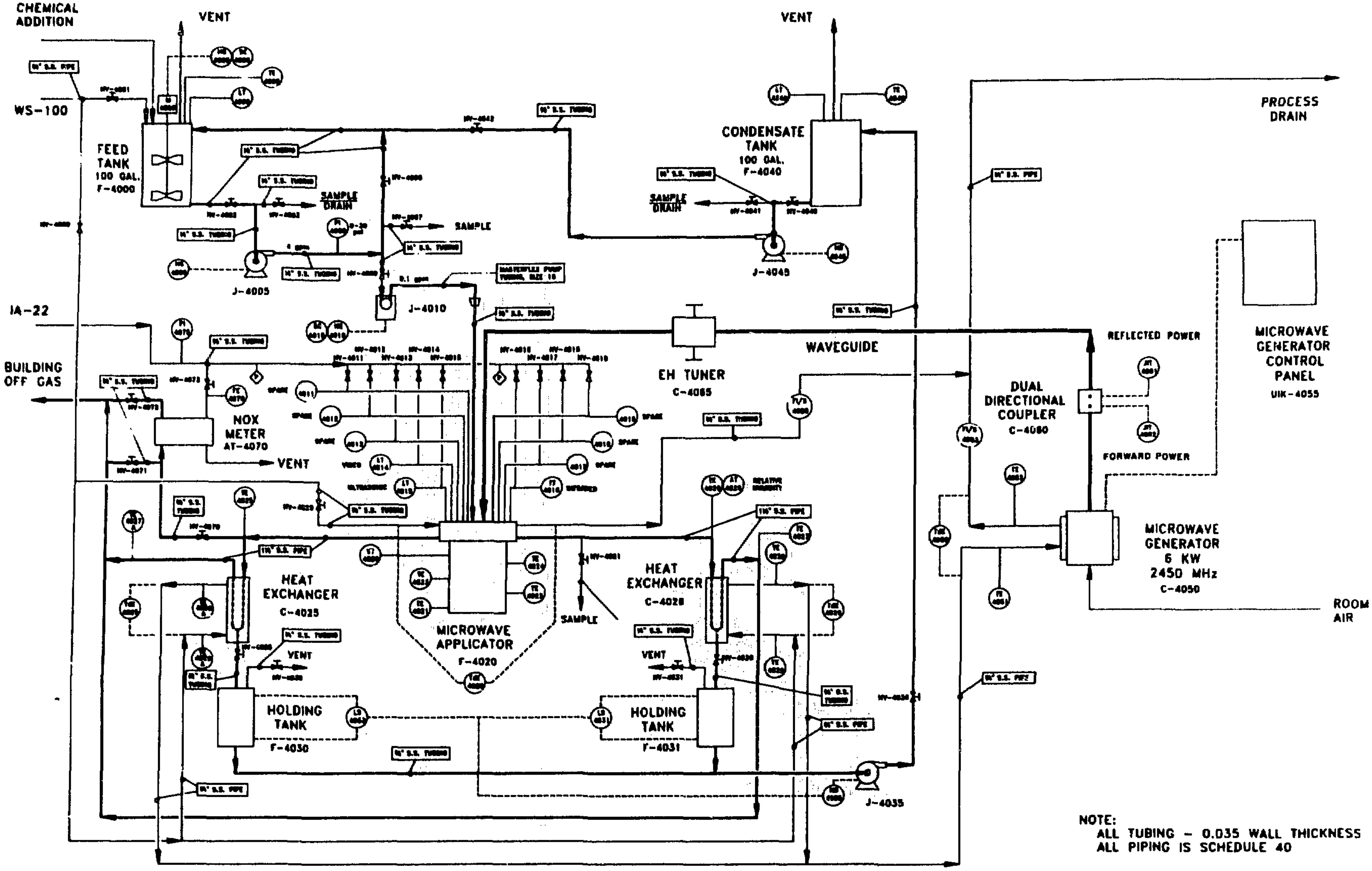

Fig. 1. 1/3-scale microwave in-can processing experiment.

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 contract 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 $NaNO_3$ crystals that have been formed at the microwave/liquid interface. As the surface temperature reaches 110°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 $NaNO_3$ (308°C). The temperature is increased to 350°C and at 60 minutes the feed is introduced into the feed pipe at a 10-mL/min flow rate. The first drops hit the center of the pool, vaporize, and cool the center to form a ring of $NaNO_3$ salt crystals. The center area boils vigorously. The dark pool of molten $NaNO_3$ liquid persists around the light ring of $NaNO_3$ crystals. Continued feeding produces a crater in the middle consisting of boiling surrogate surrounded by a ring of $NaNO_3$ deposits, followed by a dark pool of molten $NaNO_3$ all the way to the container wall. When the feed is interrupted with the power on, the entire container reheats to molten $NaNO_3$. 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 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.

TABLE I
W-26 Surrogate Formulation
for Microwave Processing

Compound	Grams per liter of surrogate solution
H_2O	851
$NaNO_3$	370
KNO_3	12.9
$Al(OH)_2$	6.96
NaCl	6.60
SiC (surrogate for U_2O_3 and ThO)	2.82
$CaHPO_4$	2.19
$Ca(OH)_2$	1.77
Na(OH)	1.27
Wyoming Bentonite clay	1.06
$CaCO_3$	0.94
$Mg(OH)_2$	0.38
Fe_2O_3	0.38
$Sr(NO_3)_2$	0.036
$CsNO_3$	0.022

AC CONDUCTIVITY MEASUREMENTS

A series of bench-scale tests was undertaken to better understand where the microwave power was being absorbed in the surrogate, as a function of surrogate temperature. The microwave penetration depth δ is given by

$$\delta(T) = \frac{1}{\sqrt{\pi f \mu_o \sigma(T)}} \qquad (1)$$

where

$\pi = 3.14159$
f = frequency
μ_o = permeability of free space
$\sigma(T)$ = electrical conductivity
T = temperature

A conductivity test cell was fabricated and is shown in Fig. 2. A stainless steel beaker is filled with surrogate and placed on a hot plate. Inside the beaker is a centered, stainless steel center electrode with a circular plate of area "A" at the end. The plate is immersed in the liquid surrogate and is separated from the bottom of the beaker by a gap "d." An alumina cylinder surrounds the center electrode so that the currents produced by the circular plate are confined to the gap. An a.c. voltage "V_{cell}" is applied across the gap, and a cell current "I_{cell}" is measured. At frequencies less than a few hundred kHz, the conductivity is given by

$$\sigma(T) = \frac{I_{cell}(T)}{V_{cell}(T)} \cdot \frac{d}{A} \tag{2}$$

where both the voltage and current are functions of cell temperature. The temperature of the liquid surrogate is measured by a type K thermocouple.

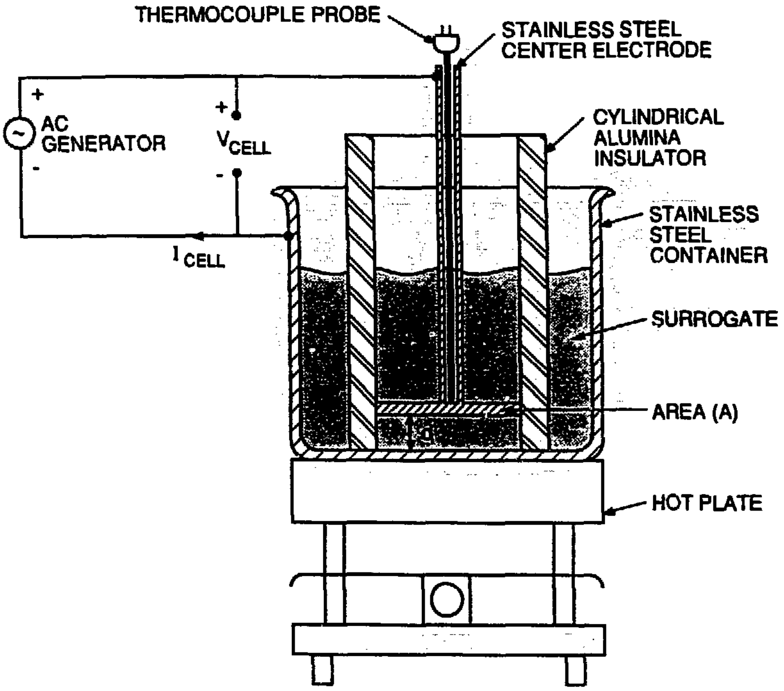

Fig. 2. Conductivity cell test setup.

The results of a typical run at 100 kHz are shown in Fig. 3. The conductivity data can be divided into several distinct regions. At room temperature and up to 120°C, the slurry has a fairly high conductivity of 2.8 S/m. Substituting this value into Eq. 1 with f = 2450 MHz gives a microwave penetration depth δ = .61 cm. Above 120°C, the residues begin to dry out, and the conductivity plummets to only .007 S/m (δ = 12 cm). However, continued heating begins to melt some of the surrogate components, and the conductivity rises back to nearly the same values it has at room temperature. When the hot plate is turned off at 360°C, the surrogate cools down, and the conductivity drops to the very low values indicative of a very dry wasteform. Several runs were made at 60 Hz, 1 kHz, and 10 kHz, and the conductivity data had nearly the same behavior as it had in the 100 kHz run. This behavior offers some justification for extrapolating the 100 kHz conductivity data to 2450 MHz. The data indicate that the microwaves penetrate into the surrogate best during the drying out phase around 120°C and only heat near the surface at the lower and higher temperature extremes. This conclusion is also supported by the observation of surface boiling below 120°C and by thermocouple profiles that show that the highest temperatures are near the surface of the liquid.

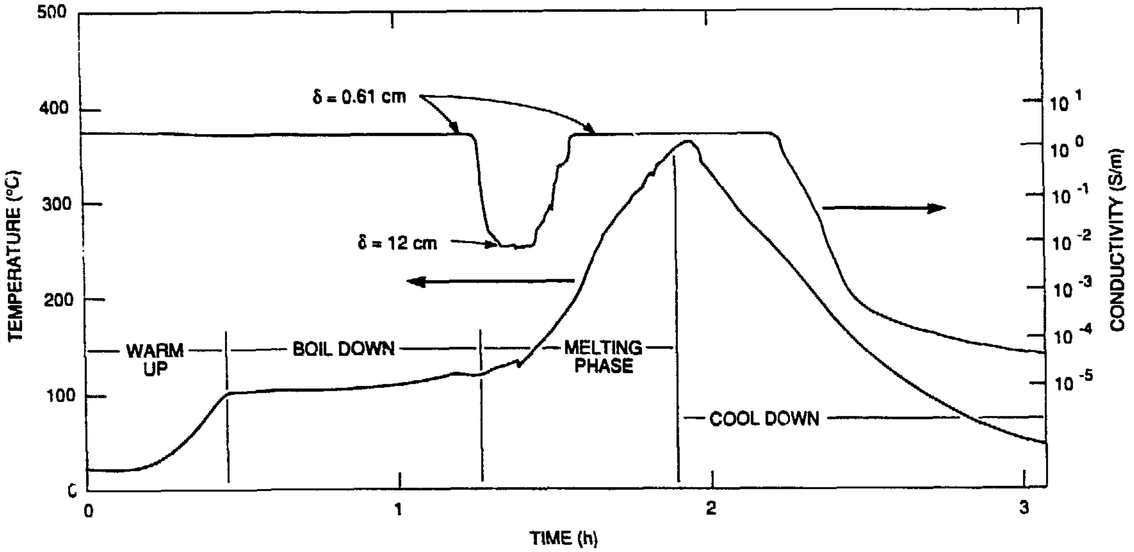

Fig. 3. Conductivity cell data at f = 100 kHz.

SUMMARY

Testing on a 1/3-scale microwave applicator to concentrate and melt RH-TRU liquids has just begun. Initial results show that the microwaved wasteform meets the repositiory waste acceptance criteria. Plans call for implementing a computer data acquisition and control system, a humidity probe, and an acoustic emission sensor to limit boiling.

REFERENCES

1. D. W. TURNER, J. B. BERRY, J. W. MOORE, and D. L. LENNON, "An Overview of the ORNL Waste Handling and Packaging Plant," Waste Management '90, Tucson, Arizona, February 25-March 1, 1990, Arizona Board of Regents (1990).
2. J. B. TILLMAN, "Meeting the Challenge of Radioactive Waste Disposal at the WIPP," Waste Management '89, Tucson, Arizona, February 26-March 1, 1989, Vol. 1, p. 209, Arizona Boards of Regents (1989).
3. "TRU Waste Certification Compliance Requirements for Remote-Handled Wastes for Shipment to the WIPP," WIPP-DOE-158, UC-70, September 1985.
4. J. B. BERRY, E. L. YOUNG- BLOOD, J. T. SHOR, J. F. WALKER, T. L. WHITE, and A. J. MATTUS, "Engineering Development of Processing Methods for Remotely-Handled Transuranic Sludge at Oak Ridge National Laboratory," Waste Management '90, Tucson, Arizona, February 25-March 1, 1990, Arizona Board of Regents (1990).
5. T. L. WHITE, R. D. PETERSON, and A. J. JOHNSON, "Microwave Processing of Remote-Handled Transuranic Wastes at Oak Ridge National Laboratory," Waste Management '89, Tucson, Arizona, February 25-March 1, 1989, Vol. 1, p. 263, Arizona Board of Regents (1989).
6. W. H. SUTTON, ed., Microwave Processing of Materials, Proceedings of the 124th Symposium of the Materials Research Society, p. 33, Publishers Choice, Mars, Pennsylvania (1988).
7. Ref. 5, op. cit.