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OAK RIDGE NATIONAL LABORATORY

MARTIN MARIETTA

CRADA Final Report
for
CRADA Number ORNL93-0190

HEAT TRANSFER ENHANCED MICROWAVE PROCESS FOR STABILIZATION OF LIQUID RADIOACTIVE WASTE SLURRY

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MTI

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FINAL REPORT

for

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HEAT TRANSFER ENHANCED MICROWAVE PROCESS FOR STABILIZATION OF LIQUID RADIOACTIVE WASTE SLURRY

by

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ABSTRACT

The objective of this CRADA is to combine a polymer process for encapsulation of liquid radioactive waste slurry developed by Monolith Technology, Inc. (MTI), with an in-drum microwave process for drying radioactive wastes developed by Oak Ridge National Laboratory (ORNL), for the purpose of achieving a fast, cost-effective commercial process for solidification of liquid radioactive waste slurry. Tests performed so far show a four-fold increase in process throughput due to the direct microwave heating of the polymer/slurry mixture, compared to conventional edge-heating of the mixer. We measured a steady-state throughput of 33 ml/min for 1.4 kW of absorbed microwave power. The final waste form is a solid monolith with no free liquids and no free particulates.

PURPOSE AND BACKGROUND

Among the emerging technologies required to better execute cleanup and contain costs are new techniques for reducing the volume of radioactive waste sludges so that the water does not accompany the wastes in the burial scenario. Also critical is the solidification of the dehydrated waste into a leach resistant waste form which has high physical integrity and stability, and which will retain these properties over many years of disposal. An example of such liquid radioactive waste solutions which must be processed in such a manner is the liquid generated as a result of scrubbing the off-gas from a radioactive waste incinerator. Another example is the sludge wastes contaminated with heavy metals and radionuclides which are collected in radioactive waste drains. Previously, such wastes have been solidified by mixing them with Portland cement. This process hydrates the water and yields a stable solidified solid, albeit with a substantial penalty due to increased disposal volume.

MTI has developed a proprietary process for solidification of liquid radioactive waste which incorporates a heat transfer polymer. This approach provides an energy efficient technique for dewatering and ultimately solidifying liquid radioactive waste. The MTI polymer process requires a mixer with a sweep arm to wipe the heated polymer from the circumference of a conventionally-heated cylindrical vessel. Waste slurry is added to the heated polymer until the desired waste loading is achieved. A setting agent is added and the batch is poured into the final storage container. This process is disclosed under U.S. Patent Application No. 07/982,654, entitled "Process for Treating an Aqueous Waste Solution". Since the polymer is a poor thermal conductor, direct microwave heating of the mixed polymer material along with conventional heating of the vessel circumference is believed to be superior to either form of heating alone. This combined or "hybrid" heating scheme should result in enhanced throughput and faster startup times compared to conventional heating alone. These factors can reduce the overall cost per pound of the polymer waste form.

The objective of this CRADA is to couple the MTI polymer technology with state-of-the-art microwave technology developed by Oak Ridge National Laboratory (ORNL), for the purpose of achieving a cost-effective commercial process. In this technology developed by ORNL, scientists have achieved promising results in a proprietary process for delivering microwave energy to radioactive wastes. The technology is patented under U.S. Patent No. 5,324,485, entitled "Microwave Applicator for In-Drum Processing of Radioactive Waste Slurry". This process was developed by Martin Marietta Energy Systems, Inc. (Energy Systems), which manages and operates ORNL, in it's Contract No. DE-AC05-84OR21400, with the DOE. ORNL has been able to demonstrate that this process can achieve the following:

1. an azimulthally uniform circular mode microwave energy field which can be used to heat a liquid which is contained is a metallic waste drum;

2. the circular mode microwave energy field permits an unrestricted operation, even when the drum is fitted with a center shaft mixer blade or a sweep arm mixer blade associated with conventional mixing equipment.

This method of processing liquid radioactive waste will provide an unprecedented efficiency in conversion of electrical power into thermal energy. The waste produced will be highly concentrated, thus reducing disposal volume. The waste form will provide superior performance compared to conventional grouting, which suffers from unexpected reactions, uncertain long term stability, and quality control problems.

In radioactive processing applications, microwave heating is simpler and more maintainable than other conventional heat sources. This is due to the direct-heating nature of microwaves on dielectric materials. Fast efficient coupling of microwave energy to a material can be achieved depending upon the material's "loss tangent". No heating elements are required and no heat transfer surfaces are required due to direct heating. Generators can be isolated from radioactive processing areas by wave guides and windows, thus facilitating maintenance. Microwave heating is versatile. A wide range of processing temperatures are available in a single system. From rapid drying due to selective heating of water (~100^o C), to dehydration (100 - 600^o C) to sintering / vitrification (600-1200^{o}C). There are two basic processing options. The first option is to process the waste in a final storage container (in-drum) the second option is to process the waste in a dedicated vessel with waste transferred to a final storage container. This option may require a process mover with moving parts unless the waste is a flowing vitreous material. A wide variety of input waste stream candidates can be process with microwave energy. They are:

1. mixed-waste sludges
2. .combustible non-metallic wastes
3. incinerator ash
4. contaminated soils
5. heterogeneous solids (excluding metal fragments)

DESCRIPTION OF EXPERIMENT

The 6 kW, 2.45 GHz, 5 L, 7 in. diameter in-drum system used for this CRADA is shown in Fig. 1. This system is a 1/3-scale prototype for ORNL pilot plant studies designed to process sodium nitrate sludges to 350° C in 55-gal. drums. This facility supports the proposed ORNL Remote-Handled Transuranic Waste Processing Facility.

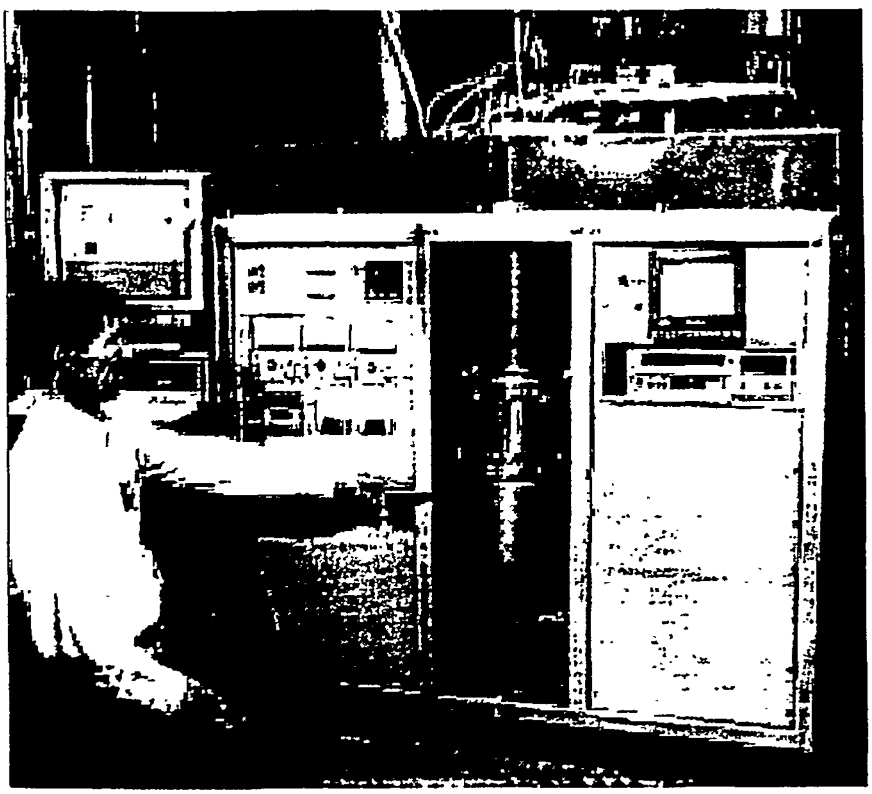

Fig. 1. The 6-kW, 2.45-GHz, 5-L, 7-in. diameter in-drum system.

This system has a 6-kW 2.45-GHz microwave generator to supply the microwave energy to a 5-L stainless steel mixer. The microwave energy is coupled to the mixer from above using a proprietary applicator. A 5-L MTI-furnished mixer was connected to the bottom of the applicator. Absorbed power in the waste is measured by taking the difference between the measured forward and reflected power as measured my directional couplers. The slurry is fed into the top center of the mixer by a flow-calibrated peristaltic pump. The slurry was a dilute solution of NaCl and water. A proprietary polymer resin was loaded into the mixer prior to each run. The top surface temperature is measured by an infrared sensor. The tuning position of the drum is a measure of the drum location required to minimize reflected power. The microwave energy produces a ring-shaped surface heating pattern or "mode" on the top of the mixer as shown in Fig. 2. The fields are azimulthally uniform and maximum (red color) half-way between the center and the edge of the mixer diameter. The microwave fields are small (purple color) at the walls and at the center of the mixer.

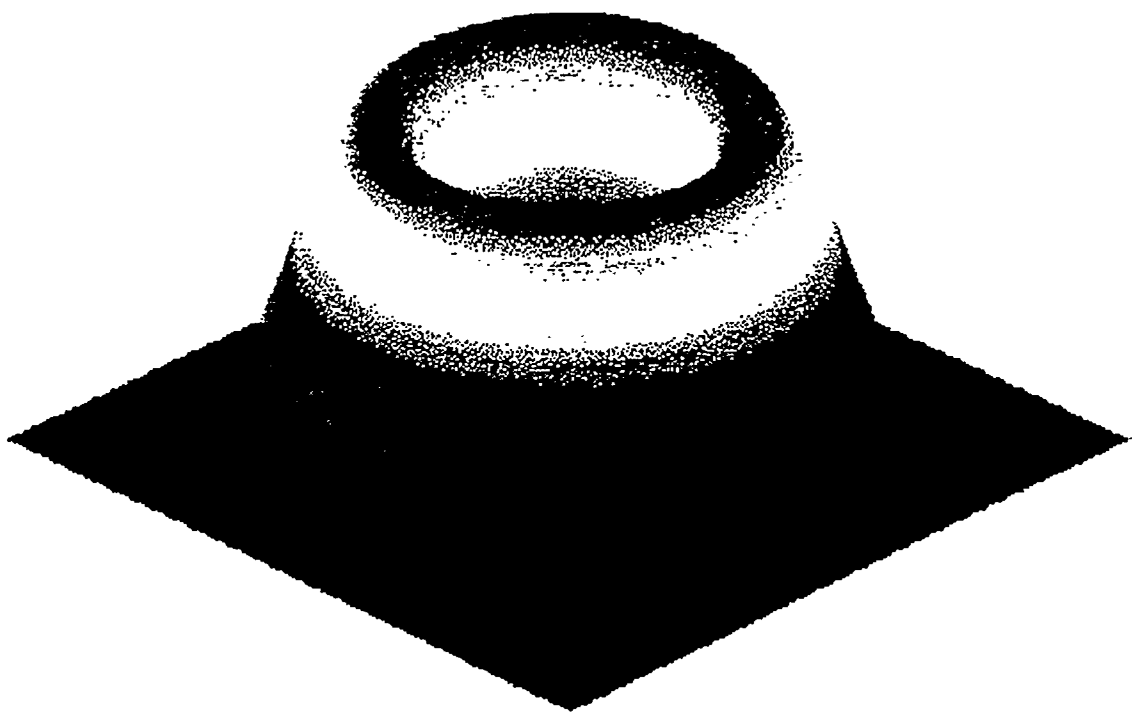

Fig. 2. The microwave "mode" used in the in-drum system.

The slurry is fed on the top of the mixer by a central feed tube, the mixer is started and microwave power applied. The water is directly heated by the microwaves and the salt is transferred from the water to the resin until a sufficient concentration of salt has built up in the resin. The mixer is removed and a hardening agent is added to the concentrate. The mixer is operated briefly to mix the hardening agent with the resin and the mixture is pored into and final storage container. Hardening takes place within a few minutes. The entire process is videotaped by a miniature video camera and light source located above the applicator. A single-color infrared pyrometer measures the surface temperature. Off-gasses are removed by a vent located above the applicator and a condenser removes water vapor form the off-gasses.

EXPERIMENTAL RESULTS

Data from the third and final run is shown in Fig. 3. During the first 30 min. the absorbed power is 1.4 kW and the input flow rate is adjusted to 33 ml/min. to just balance the rate of evaporation of water from the NaCl slurry. This flow rate is in reasonable agreement with theoretical calculations based solely on the energy required to supply the heat of vaporization energy for water. The input flow is shut off after 30 min. and heating continues to fully remove all traces of water from the system. The infrared temperature is about 150^{o} C and the temperature spikes are caused by changes in surfaces emmissivity due to vortices formed during the high-speed mixing process.

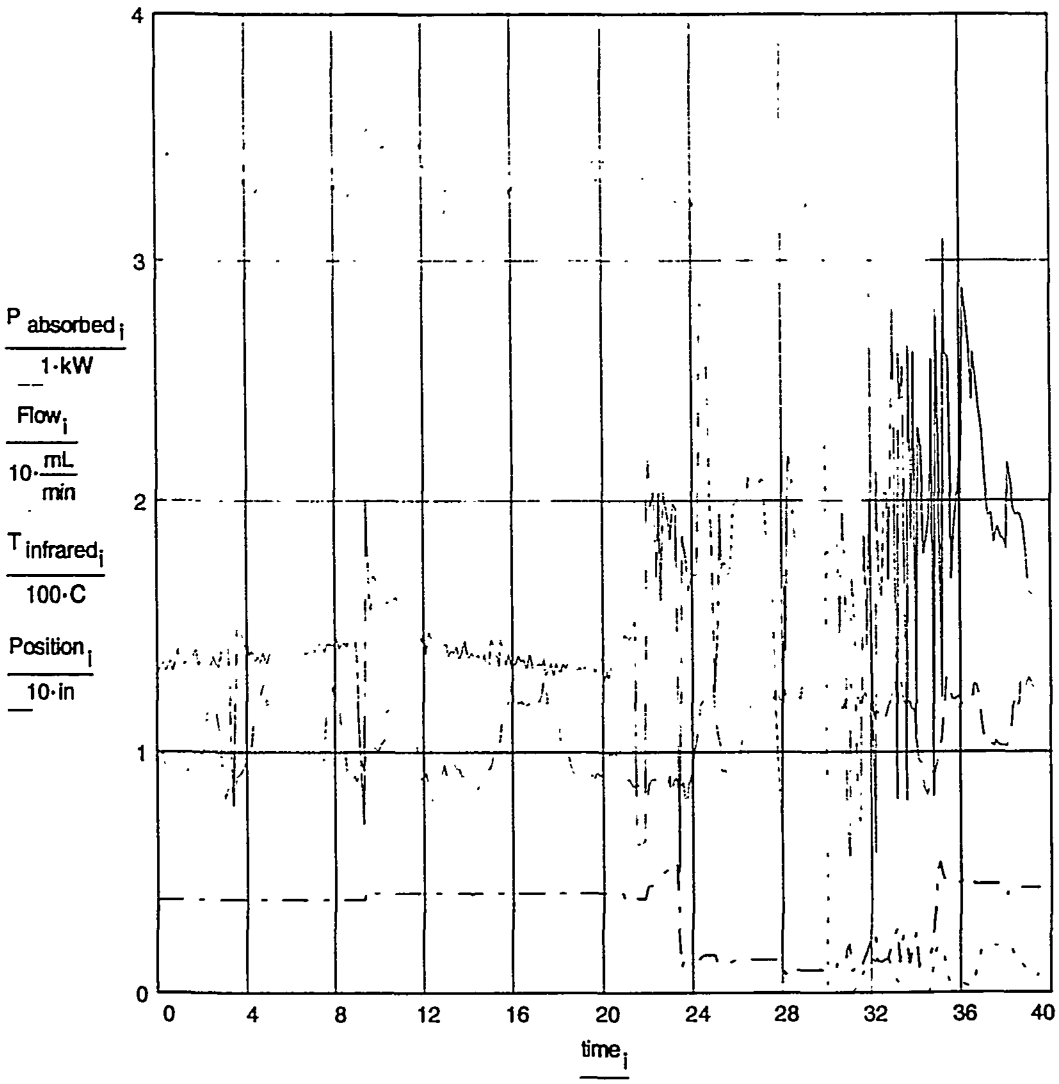

Fig. 3. Absorbed power, flow, infrared temperature and tuning position versus time (all parameters normalized as shown).

The 40 min. microwave processing time is approximately four times faster than heating the circumference of the mixer with conventional heat. This is due to the fact that the polymer is a thermally insulating, viscous fluid that is difficult to edge heat quickly without "scaling" or breaking down the polymer bonds at high heat inputs. Microwaves heat the water preferentially due to the microwave transparency of the polymer. Therefore no heat transfer problems arise and no scaling of the mixer walls occurs. The visual appearance of the run is shown in Figs. 4 through 7.

Fig. 4. Initial filing of the mixer at 0 min. into the run.

Fig. 5. Heating at 5 min. into the run.

Fig. 6. Heating at 15 min. into the run.

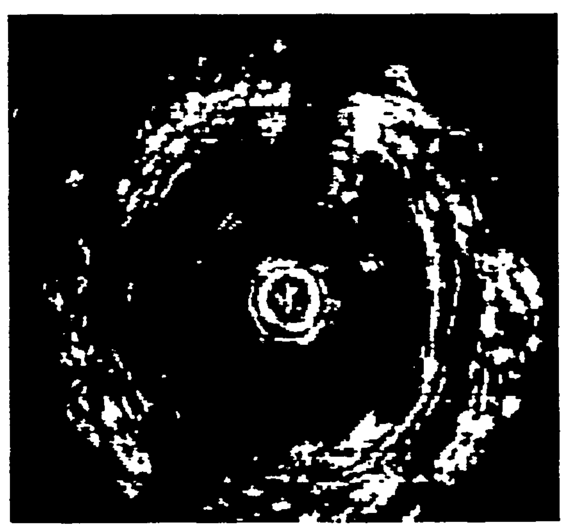

Fig. 7. Final concentration of the hot polymer at 40 min. into the run.

The final solidified product is shown in Fig. 8. No TCLP tests were performed on these samples but conventionally-heated samples have been shown to pass these tests.

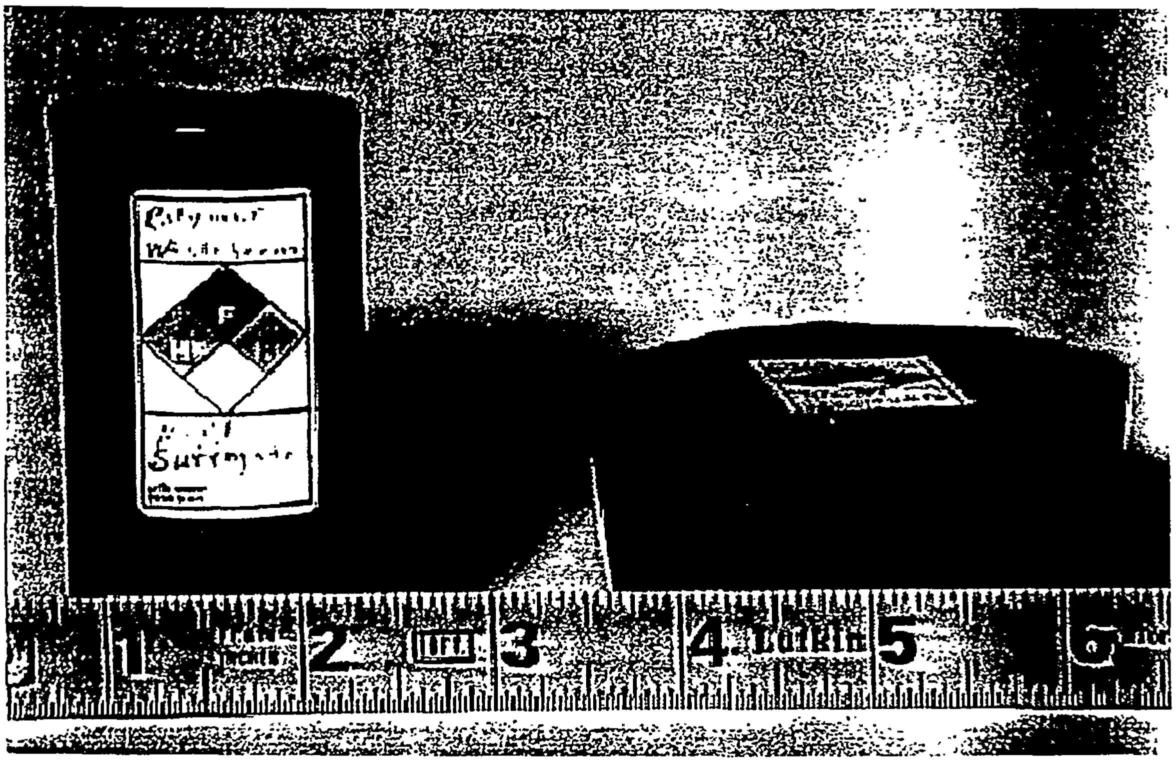

Fig. 8. Samples of the solidified polymer waste form.

INVENTIONS DEVELOPED

A top-entry mixer would be more relevant for true in-drum processing than the bottom-entry MTI furnished mixer. In addition, the MTI mixer blade was not designed for microwave compatibility. Accordingly, A microwave-compatible mixer blade and drive system (not shown) were fabricated for follow-on studies on the in-drum system. Due to a number of delays this new mixer was never tested.

CONCLUSIONS

The objective of this CRADA is to couple the MTI polymer technology with state-of-the-art microwave technology developed by Oak Ridge National Laboratory

(ORNL), for the purpose of achieving a cost-effective commercial process. In this respect the CRADA were only partially met. While we have shown technical feasibility of the process and improvements over conventional heating of polymers, the high cost of the polymers relative to other waste disposal options, and limited throughput of a scaled-up microwave device relative to other waste disposal options may limit commercial application. Follow-on funding of the CRADA into full-scale operation was not forthcoming and so more detailed engineering and cost data were not obtained.

This process would be useful for a low-volume waste stream where the entire processing could be done on a mobile unit at the customer site. Microwave-heated mixing of polymers and other hard-to-heat materials may have applications in the chemical processing industry as well.

We have enjoyed working with the staff at MTI and have found this CRADA an extremely useful tool for interacting with private industry. We would do so again should the opportunity arise.