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MICROWAVE ENERGY FOR POST-CALCINATION HIGH-LEVEL NUCLEAR WASTES

MASTER

G. Gombert, S. J. Priebe, J. R. Berreth

**Exxon Nuclear Idaho Company, Inc.
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ABSTRACT

High-level radioactive wastes generated from nuclear fuel reprocessing require treatment for effective long-term storage. Heating by microwave energy is explored in processing of two possible waste forms: (1) drying of a pelleted form of calcined waste and (2) vitrification of calcined waste. It is shown that residence times for these processes can be greatly reduced when using microwave energy rather than conventional heating sources, without affecting product properties. Compounds in the waste set to the glass frit additives couple very well with one 2.45 GHz microwave field so that no special microwave absorbers are necessary.

III. PREVIOUS STUDIES

A. Microwave Process Application

In a previous study, microwave energy was found to be potentially advantageous for either drying pellets or melting glass to produce a final radioactive waste form by remote processes. Microwave heating has several potential advantages. The intrinsic properties of microwaves allow them to be channeled from a non-radioactive area to a radioactive processing cell with only minimal (about 0.2 dB/meter) power loss. Virtually all of the microwave equipment can be kept outside of the radioactive contamination zone, leaving only the hollow waveguide and the containment vessel (applicator) in the radioactive environment.

I. INTRODUCTION

In many of the current and prospective phases of radioactive waste treatment at the Idaho Chemical Processing Plant (ICPP) such as calcining, concentration by evaporating, drying of pelleted waste, or waste vitrification, heat is required. Microwave energy is considered in this paper as a source of the process heat in the latter two cases because of its potential for remote application for a radioactive process and its inherent quality of "volume" instead of "surface" heating.

II. MICROWAVE ENERGY

Microwave power consists of electromagnetic (E-M) radiation of relatively short wavelength. The frequency used for the studies reported here is 2450 MHz, which has a 12-cm free-space wavelength. A microwave heating system consists of three main parts: the microwave generator, the applicator and the connecting waveguide. The 6 kW variable generator used here included a 60 Hz to dc power source, a magnetron with isolator to generate the microwaves, reflected power dummy load, and a 100 impedance matching tuner. The applicator is simply a vessel to contain the microwaves and the material to which they are to be applied. The microwaves are routed from the generator to the applicator through a hollow rectangular waveguide.

The greatest advantage of microwave heating comes in that the process material is heated and not the process. Product temperatures are not limited by the temperature of the heat source as they are by conduction, convection, or radiation. The absorbent temperature is limited only when its rate of heat loss is equal to its rate of absorption. For an absorbent material, adequately insulated, temperature has no upper limit using even the least powerful generator. Practical limitations are mostly due to the materials' ability to couple with and absorb the microwaves, the maximum temperature allowable in the applicator, power available and the generator efficiency (about 75% for most magnetrons).

B. Microwave Breakdown

As in large dc electric field strengths, but by a slightly different mechanism, gases can undergo electron avalanche breakdown when operating at microwave frequencies with high-intensity electric fields. Calculations were done to determine the effects of ionizing radiation and off-gases on the level at which microwave breakdown occurs.

The calculations showed that the intensities of ionizing radiation and the size of microwave pellet cover would tend to only decrease the breakdown field strength. Volatile materials such as water, fluorides, and NO_x would not significantly reduce the breakdown field strength from that of clean air conditions. It is recommended, however, that off-gases should be swept from the microwave field to prevent them from becoming predominant or

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breakdown will occur at significantly reduced field strengths.

IV. DRYING OF A PELLETTED WASTE FORM

A. Laboratory-Scale Drying Tests

Laboratory experiments showed that the moist or "green" waste pellets as produced at the ICPP lose 15-25% of their total weight during the drying step.³ Six to seven percent of the pellet weight is lost as NO_x , most of the balance is water. Microwave drying requires 10-20 min versus the 80-120 min required using hot air. Figure 1 shows the general shape of drying rate vs time for four levels of power in a commercially available household microwave oven. Figure 2 shows the wt% water left in the pellet versus time for exposure to the same four levels of power. As can be expected, the rate of water loss is highest when the water content is highest, and both asymptotically approach zero. The higher power values yield not only higher drying rate peaks but reach those peaks in shorter times. This initial drying rate must be limited to prevent the pellets from exploding or expanding.

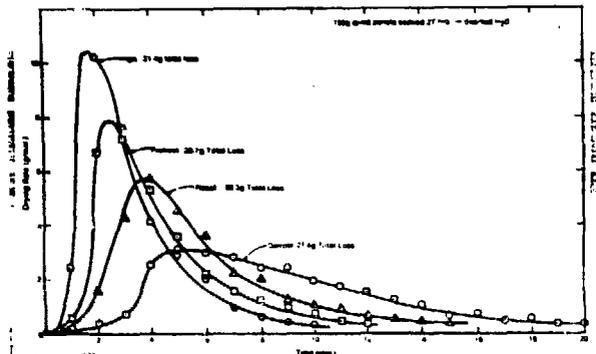


Figure 1. Microwave Drying Rate vs. Time for ICPP Pellets

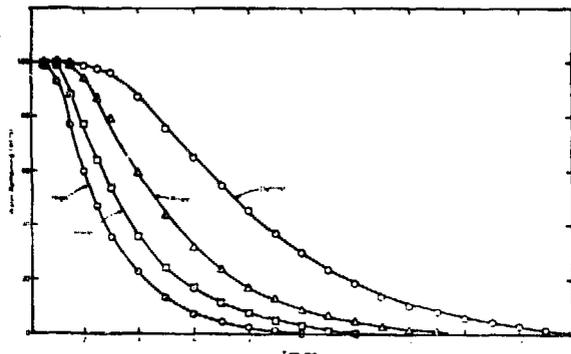


Figure 2. Water Remaining (Wt%) vs. Exposure Time to Microwaves for ICPP Pellets

B. Pilot-Scale Microwave Dryer

Successful rapid drying of the pellets in the laboratory without detrimentally affecting the final product prompted design of a pilot-scale dryer (see Figure 3) for use in a pelletizing pilot plant.⁴ The dryer/applicator was sized such that for the frequency range deviations of the magnetron, the number of possible modes was maximized. Each mode develops standing waves and nulls; i.e., lines of higher and lower field strength where reflected waves are reinforced or cancelled by wave interference respectively. In the presence of a mode-stirrer, a revolving conductive fan blade, the prevailing modes alternate, thereby changing the wave-null pattern in the cavity and yielding fairly homogeneous heating of the sample. Generally, the larger the cavity the more modes that are possible, but certain dimensions lead to more modes than others even though they may yield the same applicator volume.

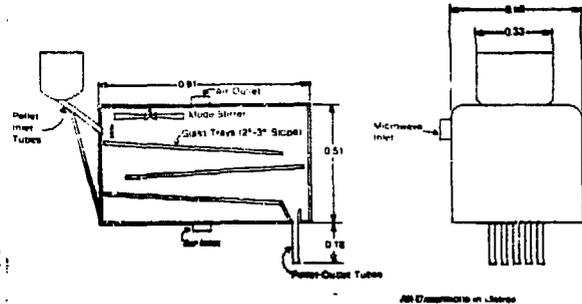


Figure 3. Schematic of Microwave Applicator

The pellet dryer was sized to maximize the number of possible modes in the desired volume, giving a 51 cm x 56 cm x 91 cm cavity. The dryer has two microwave inlets from the generator which may be operated from 0-6 kW at 2.45 GHz. Pellets, 0.5-1.0 cm in diameter, are vibrated down three flights of glass trays tilted at 2-3° to give a residence time of 15 minutes which is sufficient to dry 25 kg of pellets per hour. Pellet inlet and outlet are one inch tubes which allow gravity flow of pellets, but contain the microwaves in the applicator. The volatiles given off during drying such as H_2O and NO_x are removed by sweeping the applicator with a preheated ($\sim 200^\circ\text{C}$) stream of air.

Care must be taken in the applicator design. There can be no metallic projections into the cavity as they absorb energy. Applicator internals must be microwave transparent. Materials such as glass and Al_2O_3 ceramics must be used. Also entries into the microwave applicator must be designed to prevent microwave leakage.

Operation of the microwave drier will begin as soon as checkout of the pilot-plant system is complete. Tests will include residence time versus throughput and required power, and evaluation of the applicator physical construction. Microwave drying

In this case is advantageous for at least three reasons: (1) heat is applied from a generator located in a nonradioactive area, (2) drying is faster than by conventional methods, and (3) microwaves uniformly dry the whole pellet whereas radiant heating tends to initially form a brittle crust on the surface of the pellet that may crack as the inner volatiles heat and expand against this shell.

V. MELTING OF CALCINED WASTE

A. The Glass Melting Process Using Microwaves

Another waste form being considered is immobilizing ICPP calcines in glass containing 20-33 wt% calcine.⁵ The glasses are a combination of vitrifying materials (frit) and the calcine (see Table I) all melted at 1050 to 1100°C. The mixture of calcine and frit is melted for up to eight hours to ensure homogeneity before pouring into a final storage canister. Joule heating, as proposed, uses electrodes under the glass level that pass current through the molten pool, using the resistance of the glass to produce heat. The possible advantages of microwave over joule heating for glass melting are: (1) simple melter startup, (2) no dependence on glass electrical conductivity, (3) no electrodes, and (4) no limit on operating temperature due to electrode materials. To use joule-heating, some supplementary means must be used to raise the temperature of the glass to a point where it becomes electrically conductive. With microwaves, the energy absorbent glass will heat from ambient temperatures. Using joule heating, the rate of heat production is a function of the current and the square of the path resistance. The melting rate and temperature of the glass melt are therefore functions of glass resistivity, so power application controls temperature, and the temperature change does not affect power efficiency. With microwave heating, the power source may be remotely located away from process contamination, and the electrodes are eliminated.

TABLE I

Composition of Simulated ICPP
Zirconia Calcine and Glass Frit 51

ICPP Calcine Component Wt%		Glass Frit 51, (GF51) ^a Component Wt%	
CaF ₂	50-56	SiO ₂	66
ZrO ₂	21-37	B ₂ O ₃	8
Al ₂ O ₃	13-17	Na ₂ O	24
B ₂ O ₃	3-4	CuO	2
Misc.	2-5		

^aA frit developed to vitrify ICPP zirconia calcine.

Disadvantages include dependence on glass absorptivity and microwave breakdown in very high radiation fields. Although the absorptivity of the glass is not temperature dependent it does depend on composition. Therefore microwave absorptivity

materials must be included in the glass. This is not a well understood phenomenon. There are indications that oxidation state may play an important part in microwave energy absorbance.

Additives to enhance microwave absorbance can be separated into three categories. First, those additives whose molecules couple in a lesser manner with the electric field of the microwave E-M field to generate heat. Water and NH₃ are examples, each having dipole molecules with coupling to 2.45 gigahertz radiation such that the dipole "flips" in response to the alternating field direction to generate heat.

Second, additives having magnetic domains that are able to couple to the magnetic portion of the 2.45 gigahertz E-M field. Certain soft ferrites have a magnetic domain flipping-rate capable of responding at gigahertz rates. Since each domain "flip" results in a hysteresis loss in material, heat is generated.

Finally, additives such as carbon black and certain salts can make a completely non-interacting substance somewhat conductive such that electric current is generated through the volume of the resultant mixture by the electric field of the 2.45 gigahertz E-M waves. The distributed current through the distributed "resistance" causes heating of the substance.

Conversely if the absorptivity is too high, indications are that the microwave energy is absorbed in a thin surface layer of the glass. The glass must then be adequately conductive to melt the bottom layer of the glass pool. "Skin effect," as this is called will vary of course with the material, temperature, and microwave frequency. In simulated commercial waste at the melt temperature, the skin depth appears (by visual observation of a heated sample) to be only about 1 centimeter. For non-melted simulated commercial waste powder, the skin depth apparently is many centimeters. The electrical conductivity of the hot, melted waste obviously is much larger than that of unmelted powder leading to the difference in skin depth effect.

The initial impression from testing of microwave heated melts is that after melting has actually been achieved, the skin-depth effect causes further heating to resemble surface or radiant heat behavior rather than volume type heating. In pellet drying, though, the pellet size is such that typical microwave volume heating should result.

B. Laboratory-Scale Melting Tests

Experiments using microwave energy to melt glass have been reported.^{7,8} The laboratory-scale microwave applicator built at the ICPP to vitrify simulated calcined waste was of simple design for exploratory experiments, so it was designed as large as possible in the allowable space to give flexibility in the applicator wave modes. A mode stirrer was included in the cavity. The melter was built from a two-foot diameter stainless steel

pipe one foot high, with a welded top and bottom and a removable top closure to allow placement of glass crucibles. Testing was primarily directed toward qualitative measurement of the absorptivity of calcine, glass frit components, and microwave absorbent glass additives. Two types of calcines were tested, ICPP zirconia calcine, and a calcined waste composition proposed by Allied General Nuclear Services (AGNS). Additives included three commercially available ferrites, all of the second type described above. They were BaO-6Fe₂O₃ (#106) with a silicate sintering agent, SrO-6Fe₂O₃ (#201) also with the sintering agent, and BaO-6Fe₂O₃ (#307) alone.

Table II shows the mixtures tested, time to melt, power setting, total exposure time, and results. The results are qualitative, but point out an important trend. None of the individual components exposed in these tests melted, although CuO and Ferrite #201 showed slight sintering. Conversely, when either of the calcines was mixed with the glass frit with or without one of the ferrites, melting did occur. It would appear that not only is the individual component absorptivity important, but the interaction of the mixture is also important, probably due to change of oxidation states, and may not necessarily be implied by the single constituents. Future tests will have to more thoroughly investigate this point. Perhaps most important is that melting temperatures were easily reached from ambient startup.

TABLE II

Results of Microwave Melting Tests on Glass Components

Material	Time to Melt/Power	Exposure Time	Comments
Calcine (ICPP)	Never/3 kW	15 min	Slight warming
SiO ₂	Never/7 kW	15 min	No change
Na ₂ B ₄ O ₇ -5H ₂ O	Never/1 kW	15 min	Steam expansion
Na ₂ SiO ₃	Never/1 kW	15 min	No change
CuO	Glowing 5 min/1 kW	15 min	Partially fused
Ferrite #106	Never/1.5 kW	15 min	No change
Ferrite #201	8 min/1.5 kW	15 min	Partially fused
Ferrite #307	Never/1.5 kW	15 min	No change
ICPP+GF51	4 min/2 kW	30 min	Uniform melt except surface
ICPP+GF51+106	15 min/2 kW	30 min	Clear melt some striation
ICPP+GF51+201	5 min/2 kW	30 min	Like 106 fewer striations

TABLE II (continued)

Material	Time to Melt/Power	Exposure Time	Comments
ICPP+GF51+307	17 min/2 kW	30 min	Like 201 fewer striation, free ferrite chunks
AGNS+GF51	4 min/2 kW	13 min	Much unmelted calcine
AGNS+GF51+106	4 min/2 kW	35 min	Some unmelted calcine
AGNS+GF51+307	6 min/2 kW	35 min	Much unmelted calcined

C. Future Testing

The power required to melt the glasses needs to be more accurately measured. Absorbivity tests need to be done on prospective glass tank refractories as well as on glass mixtures. Measurements to determine the effects of off-gases and high-radiation fields on microwave breakdown must be done. Measurements to judge breakdown effects and to compare volatility to joule-heating experiments are also needed.

VI. CONCLUSIONS AND RECOMMENDATIONS

Experiments show that microwave power appears practical for drying pelleted waste and has potential for melting waste glasses. Planned pilot-scale experiments will verify microwave pellet drying. Laboratory-scale melting tests indicate adequate microwave absorptivity in the glass components to promote melting. Further testing remains to draw any conclusions on glass quality and process characteristics.

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