MICROWAVE WASTE PROCESSING TECHNOLOGY OVERVIEW

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ABSTRACT

Applications using microwave energy in the chemical processing industry have increased within the last ten years. Recently, interest in waste treatment applications process development, especially solidification, has grown. Microwave waste processing offers many advantages over conventional waste treatment technologies. These advantages include a high density, leach resistant, robust waste form, volume and toxicity reduction, favorable economics, in-container treatment, good public acceptance, isolated equipment, and instantaneous energy control. The results from the "cold" demonstration scale testing at the Rocky Flats nuclear weapons facility are described. Preliminary results for a transuranic (TRU) precipitation sludge indicate that volume reductions of over 80% are achievable over the current immobilization process. An economic evaluation performed demonstrated cost savings of $11.68 per pound compared to the immobilization process currently in use on wet sludge.

INTRODUCTION

Microwave technology has been used in the food and chemical industries since early 1970, with the majority of the work concentrated in the areas of drying and the vulcanization of rubber. (1) High temperature technology has been developed by the Japanese for converting plutonium nitrate, recovered from spent fuel reprocessing, to plutonium oxide for nuclear fuel production and for solidifying of waste incinerator ash. (2,3) Laboratory scale vitrification of calcined high-level nuclear wastes using microwave energy was done by the Idaho National Engineering Laboratory in 1979. High-level wastes were mixed with a composite of fluxing agents in ceramic crucibles and placed in a microwave cavity. The resulting glass was poured into a metal storage container and allowed to solidify. (4) Significant work with solidification of inorganic wastes was performed at the Rocky Flats Plant since 1985. Wastes successfully processed include precipitation sludge, incinerator ashes, nitrate salts, and soils. The incinerator ashes included Fluidized Bed, catalyst ash, and ash from shredded HEPA filters. Both types were mixed with fluxing agents and a silica source, and heated using microwave energy. Nitrate salts were heated to their melting points to form a salt cake. Oak Ridge National Laboratory has performed additional salt solidification work with outstanding results.

There are three primary mechanisms involved in heating with microwave energy. Type I is characterized as the vigorous vibration of a dipole molecule due to the oscillation of the electromagnetic field. The vibration causes frictional heat to build up between the molecules which elevates the temperature of the material. The simulated and actual sludge used in these tests contain metal oxides which are normally electrically neutral; however, when placed in an electromagnetic field they become dipolar. Type II heating involves substances that are magnetic in nature and couple with the magnetic component of the microwave field. The oscillation of the magnetic component of the field results in hysterisis loss within the material which generates heat. Type III heating takes place when an electrically conductive material, such as carbon black, is a component of the material being heated. A current is generated throughout the material by the electric component of the microwave field. The material is heated by the current flow through the material. (2)

The process developed at the Rocky Flats Plant is an in-drum melting system designed to isolate the molten waste in the shipping container during processing. This processing system offers advantages over other thermal processes. Greater energy transfer efficiency is achieved due to direct coupling between the microwave energy and the waste. Since the waste is being directly heated, energy control is instantaneous. Because the material does not have to flow, viscosities of the melt can be higher than glasses, allowing much higher waste loadings, and greater densities. Synthetic minerals also offer a leach resistant, stable matrix especially suited for hazardous and radioactive wastes. The high temperatures also ensures destruction of soluble materials such as nitrates. The process equipment is also more easily maintained because the heat is contained in the disposable drum which is insulated from the rest of the process equipment.

This paper describes the work performed at the Rocky Flats Plant on precipitation sludges. Bench scale development, using both simulated and actual wastes, pilot scale, and demonstration scale testing have all resulted in the continual advancement of this technology.

COLD BENCH SCALE TESTS

The acidic and basic wastes from the plutonium recovery areas at the Rocky Flats Plant (RFP) are treated in a hydroxide co-precipitation process to remove plutonium and americium. The metals are co-precipitated along with ferric and magnesium hydroxides. The resultant slurry is passed through a rotary drum vacuum filter, precoated with diatomaceous earth (diatomite) filter media, to remove the solids from the waste stream. A thin layer of filter cake is continuously cut from the drum filter, producing a wet sludge. A Portland cement/diatomite mixture is continually added to the sludge as it falls into the waste container to absorb free liquids.

Microwave solidification of precipitation sludge was tested using simulated and actual TRU sludge at bench scale, and simulated sludge at pilot and demonstration scale. "Cold" Bench scale testing, using simulated sludge, was completed in 1987. "Hot" bench scale testing was completed in 1989.

Results from the "cold" bench scale tests indicated that volume reductions of up to 80% are achievable by continuously feeding dried sludge into a waste container while applying microwave energy. An economic evaluation was completed showing that volume and weight reductions of up to 87% are achievable over the immobilization process currently in use on the wet sludge.

The equipment used in the bench scale tests included: 1) a 6kW, 2.45 GHz microwave generator, 2) an aluminum cavity, 3) a turntable, 4) WR-284 waveguides, 5) a reflected power meter, 6) an infrared (IR) thermometer and 7) a screwfeeder. Two methods of sludge addition to the microwave cavity were studied, batch and continuous. In the batch fed method, an initial charge of 2 kg of dry sludge was added to an insulated 8 liter stainless steel container; subsequent 2.5 kg additions were made to the container after the initial charge melted. Temperatures of the melt ranged from 1000°C - 1300°C. The average density of the batch fed casts was 1.21 g/cc. This increase in density over the dry sludge translates into an average volume reduction of 74.8%.

Continuous feeding resulted in higher densities and greater volume reductions than the batch fed method, due to the offgases ability to migrate through the sludge more easily. The batch fed casts had large void spaces where gases had been trapped. Nine tests were performed continuously feeding sludge through a screwfeeder into the container. The average density of the casts was 1.92 g/cc with an average volume reduction of 81.1%. Table I is a summary of data collected from the solidification tests.(5)

TABLE I

Cold Bench Scale Microwave Solidification

Trial #	Density (g/cc)	Wt.Red. (%)	Vol.Red. (%)	Comment
8	1.00	21.1	75.7	Batch Fed
11	1.32	11.6	75.0	Batch Fed
14	1.25	15.3	75.1	Batch Fed
31	1.9	8.7	81.5	Continuously Fed
33	1.9	10.5	82.0	Continuously Fed
35	1.9	11.3	82.5	Continuously Fed
36	2.3	4.0	83.4	Continuously Fed

HOT BENCH SCALE TESTS

Bench scale tests using TRU radioactive waste were completed at the Rocky Flats Plant. The microwave cavity was *housed in a glovebox* while the generator remained outside the contaminated area separated by a window in the waveguide. Both teflon and quartz were previously tested as window material. However, quartz was chosen for actual testing because of its high operating temperatures.

Ten test runs were completed using the batch fed method. The average density was 1.68 g/cc, with an average volume reduction of 54.4%. This differs from the batch fed, "cold" bench scale volume reduction and average density of 74.8% and 1.21 g/cc due to the increase in bulk density of the simulated waste from 0.40 g/cc to 0.65 g/cc for actual waste. Table II summarizes the results of the "hot" bench scale tests.

TABLE II

TRU Bench Scale Microwave Solidification

Trial #	Density (g/cc)	Wt.Red. (%)	Vol.Red. (%)	Waste Loading (%)
1	1.37	43.0	51.0	66.0
2	2.30	18.6	71.0	55.0
3	2.13	0.6	68.5	32.0
4	1.68	43.9	64.3	60.0
5	1.36	--	55.9	40.0
6	1.76	17.0	65.9	60.0
7	1.10	25.0	35.0	70.0
8	1.22	20.4	50.8	70.0
9	2.17	23.1	72.4	60.0
10	1.67	21.0	64.0	60.0

Weight reductions for the test runs averaged 21.3%. The majority of the weight loss can be attributed to the loss of water of hydration and offgas production; the sludge contains up-to 21 wt% sodium/potassium nitrate. A mixture of diatomaceous earth and anhydrous borax was used as the fluxing agent for solidification. Waste loadings of 32-70 wt% were tested along with a variety of flux ratios. (6)

PILOT SCALE TESTS

The objective of the pilot scale system was to determine if scale-up from bench scale system to near production capacities was possible. The first generation pilot system used a 30 kW, 915 MHz microwave generator and a 30 gallon, carbon steel drum as the resonant cavity. Simulated waste was used in the process. Thirty gallon drums were used because the drummed waste is supposed to be stored in the Waste Isolation Pilot Plant, and shipped in the TRUPACT. It was dictated that the waste be overpacked in a 55 gallon drum with a rigid liner. Thirty gallon drums are the only commercially available drums that fit inside the liner and 55 gallon drum.

The microwave applicator is divided into two sections; an upper access chamber and a lower load chamber. The access chamber is used to connect the resonant cavity (drum) and the microwave input. This section also allows entry into the applicator through a door and provides for venting and an intake HEPA filter. Waste is also introduced into the drum through the access chamber via a screw feeder. The chamber was fabricated from 304 stainless steel and measured 52" X 52" X 39". The drum mated with the access chamber by protruding through the floor, so that the upper rim of the drum was level with the chamber floor. A gap was maintained between the drum wall and the chamber. An energy choke was built into the side of the opening so that the combination of the drum wall and the choke attenuated the energy, precluding microwave leakage outside of the access chamber/resonant cavity. This arrangement allows the drum to rotate freely without the use of seals, and peripheral equipment to remain outside the microwave field.

The load chamber housed a lift table/turntable, insulation cage, microwave leakage meter, and the temperature control elements. The drum was placed on the turntable and lifted until the rim of the drum engaged the access chamber choke. The drum was then continuously rotated to uniformly feed waste into the drum. Rotation also distributes the energy to the waste uniformly. The initial charge of sludge was added to the drum to initiate a melt. The charge melted in approximately 30 minutes. Additional 12 kg charges were added upon complete melting of the previous charge. Between charges continuous addition, at a lower rate, was done to preclude melting of the material at the end of the screwfeeder. Nine samples were processed through the pilot system using this feed method and are summarized in Table III.

TABLE III

Cold Pilot Scale Microwave Solidification

Trial #	Fin.Wt. (kg)	Wt.Red. (%)	Vol.Red. (%)
3*	139	33.0	76.1
4*	116	32.0	74.7
5	45	18.6	----
6	148	13.1	77.2
7	183	12.2	82.5
8	118	34.7	75.1
9	54	15.5	76.1

* Drum Failure

Samples three and four ended with severe melt-through of the drum using the temperature control method described in the bench scale systems. Modifications to the control system were implemented to improve the operation and to safeguard against elevated melt temperatures in contact with the drum wall. Originally, as in the bench scale tests, an infrared thermometer was used to monitor the surface temperature of the melt. However, because of the differing properties between 2450 MHz and 915 MHz waves (wavelength and penetration depth) this method was not sufficient. The modifications to the temperature control system included a clam shell insulated cage with Type K thermocouples on the inside wall of the insulation. The thermocouples were staggered three inches apart with the lowest three inches from the bottom of the drum. As the drum turned the thermocouples measured the temperatures at various levels of the drum wall. Drum wall temperatures were controlled to approximately 900°C by adjusting the microwave energy level.

Samples 6, 7, 8 and 9 were run using the modified control system described above. The input power was maintained at 21 kW with an average feed rate of 24.5 kg/hr. The average volume reduction was 77.7%, with densities averaging 1.85 g/cc. Generally, all four trials behaved similarly during processing. (6)

DEMONSTRATION SCALE SYSTEM

The objective of the Demonstration Scale System is to obtain the operational data necessary to install a production scale process using an advanced design. The equipment consists of a 50 kW, 915 MHz microwave generator, second generation design melting cavity, based on the pilot scale design, WR-975 waveguides with a quartz window, traversing lift/turn table and insulation cage.

Simulated waste, produced by a lab scale rotary vacuum filter, is used in operation of the process. Wet sludge is cut from the filter drum at approximately 13.6 kg/hr. The sludge is dried using a 50 kW, 915 MHz microwave dryer.

The cavity design is based on the pilot scale system. Thirty gallon, unpainted, steel drums are used as the melting receptacles. The upper resonant cavity couples with the drum through a rotary choke. The choke allows rotation of the drum without microwave leakage into the drum load chamber. A sludge transfer screw penetrates the load chamber and drum choke directly above the drum. Dried sludge, from the microwave dryer, is conveyed into a storage hopper; silica (diatomite) and sludge are metered into the transfer screw and conveyed into the drum. A traversing system is used to move the drum into the load chamber. A lift/turntable is also included on the traversing system. After a drum is placed on the lift/turntable and an insulating jacket closed around it; it is lifted until the drum engages the rotary choke with approximately 1000 pounds of force. The turntable oscillates, turning the drum 360° before reversing. The process is monitored using spring loaded, type K thermocouples that protrude through the insulation and contact the drum wall.

Thriteen test runs were completed to test the microwave generator, thermocouples, feed system and control system. These tests were processed at 30 and 40 kW and 60 wt% waste loading. The second through fifth runs resemble a synthetic low grade iron ore (taconite), with a bulk density of approximately 3.2 g/cc. The material exhibits conchoidal fracturing when broken and contains the proper iron/silicate content to be classified as taconite. Table IV summarizes the data collected, to date, for the demonstration scale system.

TABLE IV

Cold Demonstration Scale Microwave Solidification

Trial #	Power (kW)	Flow Rate (kg/hr)	Density (g/cc)	Comment
1	30	30	2.2	CS
2	20	19	2.3	CS
3	30	27	2.7	CS
4*	40	40	---	CS
5	30	23	3.0	CS
6*	40	26	---	CS
7	30	23	1.7	CS, Continuous
8	30	18	1.9	CS,Continuous
11	40	27	2.2	SS
12	30	20	1.9	SS
13	40	21	2.3	SS

* Drum Failure

Trials 1 through 6 were performed in carbon steel drums while continuously adjusting the operating parameters (input power and flow rate) during the runs. The adjustments were made to maintain the surface of the material in a molten state. Although the highest product density, 3.0 g/cc, was obtained using the method of operation, a greater risk of drum failure was observed. Trials 7 and 8 were again performed using carbon steel drums, however, the operating parameters were fixed at the start of the run and maintained at constant values throughout the trial. This method of operation reduced drum degradation, however, the observed product densities were significantly lower than the previous trials. During Trials 11 through 13, 304 stainless steel drums replaced the carbon steel drums as melting receptacles for the operation. This allowed more rigorous operating conditions, thus, increasing the product density with only limited sensitization of the drum's interior surface. Additional investigations are planned to further define the optimum operating conditions.

COST ANALYSIS

An economic analysis was performed to compare the costs associated with four methods of sludge immobilization. All of the methods studied eliminated the potential for free liquids and excessive amounts of particulate as mandated in the WIPP-WAC. These included: 1) addition of a portland cement/diatomite mixture, 2) solidification using microwave technology, 3) cementation with Portland cement, and 4) polyethylene solidification.

Table V contains the results of the analysis, all costs are based on a production rate of 178,840 pounds of wet sludge per year. The wet sludge contains 66 wt% water and 34 wt% sludge on a dry basis. Dry sludge contains 20 wt% NO_3. All of the processes, except the base case, assumed a dried product input.

The diatomite/Portland cement process currently used in the old waste processing facility was used as the base case to compare the solidification processes. Other assumptions made for the analysis included: 1) Material costs for cement, diatomite, and polyethylene were estimated to be $0.10, $0.20, and $0.60 per pound, respectively. 2) The cost of shipping was based on $5000 per round trip for a TRUPACT. 3) The TRUPACT load limit is 7,000 pounds or 14 drums. 4) Storage costs are based on $300 per cubic foot. 5) microwave and polyethylene processing are in 30 gallon drums and need to be overpacked with 55 gallon drums (for shipment to WIPP). 6) Drums were estimated to cost $35 for 30 gallon and $60 for 55 gallon. 7) Drums weigh 20 lbs/30 gallon and 60 lbs/55 gallon. Equipment and installation costs were not a part of this analysis.

CONCLUSIONS

The microwave solidification process offers distinct advantages over other thermal and solidification technologies. These advantages include high temperature, in-drum processing; production of high density synthetic minerals, which translates into greater volume reductions than other solidification processes; greater energy efficiency; and more easily maintained equipment. Results from testing indicate that volume reductions of up to 87% over the current process at Rocky Flats are achievable. The process produces a synthetic taconite with a density of 3.0 g/cc which allows for a much more leach resistant product than most solidification processes. Cost savings of $11.68/lb of wet sludge produced are also possible compared to the present immobilization system used at the plant.

TABLE V
Economic Analysis

	PC/Dia.	Cementation	Polyethylene Solid.	Microwave Solid.
Sludge Wt.	178,840	60,810	60,810	60,810
Density (g/cc)	1.1	1.4	1.4	3.0
Waste Loading	40%	30%	60%	60%
Weights				
Water	---	60,810	---	---
Cement	134,130	81,080	---	---
Diatomite	134,130	---	---	40,540
Polyethylene	---	---	40,540	---
Net Weight	447,100	202,700	101,350	89,190
No. of Drums	996	460	288	212
Wt. of Drums	59,760	27,600	23,040	16,960
Gross Weight	506,860	230,300	130,150	110,390
Costs ($)				
Cement	13,410	8,110	---	---
Diatomite	26,826	---	---	8,110
Polyethylene	---	---	24,320	---
Drums	59,760	27,600	27,360	20,140
Shipping	355,710	164,290	102,860	75,710
Storage	2,205,900	1,018,790	637,850	469,530
Total Cost	2,661,610	1,218,790	792,390	573,490
Cost Reduction ($/lb sludge)		$8.07	$10.45	$11.68

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