Conf-890490--1

"PREPRINT EXTENDED ABSTRACT"
Presented Before the Division of Environmental Chemistry
American Chemical Society
Dallas, Texas, April 9-14, 1989

MICROWAVE PROCESSING OF RADIOACTIVE MATERIALS - I*

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CONF-890490--1

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DE89 005130

INTRODUCTION

This paper is the first of two papers that reviews the major past and present applications of microwave energy for processing radioactive materials, with particular emphasis on processing radioactive wastes. The second paper will be presented at a later meeting.[1] Microwave heating occurs through the internal friction produced inside a dielectric material when its molecules vibrate in response to an oscillating microwave field. For this presentation, we shall focus on the two FCC-approved microwave frequencies for industrial, scientific, and medical use, 915 and 2450 MHz. Also, because of space limitations, we shall postpone addressing plasma processing of hazardous wastes using microwave energy until a later date.

Microwave heating of radioactive materials has the following important advantages over conventional heating.

1. Radiant heating and joule heating of radioactive dielectric materials require local heating elements near the highly radioactive, usually corrosive material, whereas microwave heating requires no local heating elements because the microwave heat is absorbed directly. With microwave heating, the radioactive material becomes its own heating element. No radiant heating elements are required, and this makes microwave heating much more reliable and easier to maintain in a radioactive material processing application where contamination of process equipment and maintenance exposures can be kept to a minimum.

2. Because microwave energy is absorbed directly by a large class of materials, microwave heating has the considerable advantage of much higher efficiency and faster process control, as compared with conventional radiant heating. For radiant heating, the entire oven must be brought to a working temperature, which requires more time and energy than the microwave method. This is because radiant energy heats most materials while microwaves do not effectively heat most metals or special low-loss ceramics, glasses, and plastics. Microwaves, however, can effectively heat other materials such as clothing, gloves, shoe covers, concrete, metal oxides, nitrates, sulfates, filtering media (nonmetallic), water, carbon, standard glasses, and many other dielectric materials.[2,3] This allows for considerable efficiency improvement over conventional radiant heating by careful selection of the materials in a microwave oven design. Because of this material selectivity, microwave ovens

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*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.

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can heat materials to high temperatures while the microwave cavity remains relatively cool.

3. Microwave power can be transmitted through waveguides from generators that can be located safely outside a radioactive hot cell area where routine generator maintenance can be performed in a hands-on environment. Waveguide windows can effectively isolate the generator from the hot cell.

4. Microwave processes frequently generate the least amount of waste product, as compared with other chemical or mechanical processes, thus minimizing storage and transportation costs.

5. Microwave heating is effective for both wet and dry materials and thus has the potential to supplement or even replace conventional evaporation steps in slurry processing flowsheets, greatly simplifying the overall process.

Several issues unique to microwave processing must be addressed before plant facilities can be designed.

1. The depth of penetration of the microwave power into the material must be measured to determine whether the microwaves penetrate into the interior of the material to produce bulk heating or they heat only the surface because of a small penetration depth. The microwave penetration depth is a function of material composition, temperature, and frequency of the microwave energy. The user can, therefore, exercise some control over the penetration depth by choosing the appropriate heating frequency (915 MHz or 2450 MHz) for the application. This information is crucial to understanding the effect of material geometry on the uniformity and controllability of microwave heating.

2. Container materials that are compatible with the radioactive material being processed must be investigated; in particular, ceramic and glass materials that are microwave transparent are highly desirable because they allow the microwave power to heat the radioactive material from all directions, thus improving efficiency and processing speed over the levels attained with metal containers. Metal containers, however, can be designed with rounded surfaces that minimize arcing; they would offer the advantages of low cost and resistance to thermal shock. Depending on the details of the process under consideration, corrosion resistance of the container material will also need to be addressed.

3. Because microwave heating is direct, fast, and very efficient, the possibility exists for thermal runaway if the local heating rate in the material exceeds the rate at which heat diffuses through the material.[4] This tends to produce hot zones in the material, which because of their higher temperatures, absorb more microwave power (microwave absorption usually increases with temperature in most materials). This leads to thermal runaway where hot zones become even hotter because of thermally enhanced microwave absorption and become thermally detached from the bulk of the material. This may lead to local boiling in liquids or slurries and cracking in solids. Thermal runaway can be prevented by (1) carefully controlling the microwave power while continuously monitoring the temperature of the material and (2) providing a uniform microwave illumination of the process material.

4. Finally, bench- and pilot-scale microwave processing experiments must be designed to be truly microwave compatible and produce good heating uniformity. Microwave arcing must be prevented when microwave equipment operates at the highest powers and smallest heating loads. This is achieved by eliminating sharp metallic edges and points in the microwave cavity which tend to draw microwave arcs. Smooth radiused edges and well-rounded corners are preferred in a high-power microwave environment. Also, some means to increase the inherent nonuniformity of microwave heating is essential for good process control. This can be accomplished by using one or more of the following: (a) a rotating metal fan (mode stirrer) to

spatially move the complex microwave heating patterns in time, (b) a rotating turntable or conveyor belt to move the product at a set speed through the heating patterns, (c) multiple microwave feeds, and (d) microwave tubes that can be electronically frequency modulated.

APPLICATIONS

We shall review four applications of microwave power to processing of radioactive materials and also look at current development efforts in this area. The first application involves using microwave energy to melt glass frit mixed with high-level nuclear waste.[5] The vitreous mixture, upon cooling, solidifies into a leach-resistant wasteform suitable for long-term storage. The pilot scale equipment is shown in Fig. 1. The glassfrit, in the form of cylindrical glass-fiber plugs, is fed through a 10-cm diam tube by a plunger. A simulated high-level waste in the form of a solution is absorbed by the glass-fiber plugs and dried by the application of 2.5 kW of microwave power at 2.45 GHz. The dried plugs now proceed to the vitrification cavity where they fall into a ceramic crucible and are melted by 5 kW of microwave power at 2.45 GHz. The vitrification cavity contains a mode stirrer, essential to improve the uniformity of the microwave heating and the ceramic crucible is well insulated by ceramic fiber tubes and bricks. The melt is continuously drained into a glass storage vessel where cooling takes place. The main advantages of the microwave vitrification process are:

1. No electrodes are required such as in joule heated ceramic melters now used in this application.[6]

2. The heating can be made more uniform than in joule-heated melters by using any of the techniques mentioned in the introduction.

3. The microwave process is self-starting at room temperature since the glass can absorb microwave power throughout its temperature range. Joule-heated melters are not self-starting since the dc conductivity of glass is very poor and current will not pass between electrodes at room temperature. Auxiliary SiC electrodes are required for this purpose in joule-heated ceramic melters.

4. The microwave process has a low thermal inertia compared with conventional melters because the only material heated by the microwaves is the crucible and glass frit. The low thermal inertia allows for more precise temperature control.

5. The glass-fiber plugs serve as a filter media to trap off-gas particulates and return them to the melt thus reducing the particulates that the off-gas system has to handle.

One of the potential disadvantages of the process is that at melting temperatures, the microwaves do not fully penetrate the melt volume due to the stronger absorption at these temperatures. This would result in surface heating rather than volume heating. However, this situation can be remedied by using lower frequency microwave sources (915 MHz) which penetrate more deeply and/or limiting the melt thickness to less than the microwave penetration depth.

The second application involves microwave evaporation of corrosive radioactive solutions. Highly acidic radioactive solutions are frequently encountered in nuclear fuel recycling processes. In order to reduce corrosion associated with complicated multiple tube heat exchangers employed for evaporating these solutions, a novel idea using microwave heating of a fine spray has been described[7] and is shown in Fig. 2. Radioactive liquid waste is sprayed into fine drops and injected via a spray tower into an evaporator casing. The casing is also fed by a microwave generator connected by a waveguide system, impedance matching network, and waveguide window. The microwaves heat the fine spray directly, and the liquid is evaporated into a gaseous phase which is separated from the liquid phase using a conventional phase-separator tower. The invention has the following advantages:

1. Conventional heat exchangers consisting of many small tubes are eliminated by a simple structure with potentially greater reliability.
2. Scale formation on the sides of the evaporator casing does not degrade performance as in conventional evaporators since the microwave heating is direct.
3. The evaporator casing wall can be made very thick to increase service life without affecting the performance of the evaporator.
4. The liquid waste material consists of many fine drops which have a large total surface area, and they are also is easily penetrated by the microwave energy leading to a high evaporation rate.

The possible problem areas to be investigated are potential plugging of the spray nozzles, maximum allowable droplet size versus microwave power level, and corrosion of the evaporator casing and phase-separator.

Small bench-scale studies are underway on a microwave-assisted process[8] for detoxification of hazardous waste. TCE, a mixture of trichlorethylene and trichloroethane, is a major organic constituent in many of the DOE hazardous and mixed-waste streams and for that reason was chosen for the study. The detoxification method chosen was to oxidize the TCE absorbed on active carbon by air in the presence of catalysts with the direct heating of the carbon by 2.45 GHz microwave energy. A known weight of active carbon together with catalysts was placed in a quartz sample tube located in a microwave oven (Fig. 3). The TCE vapor was absorbed on the predried carbon bed, weighed, and then microwave power was applied with air flow over the carbon bed. The carbon bed was weighed after the run and gas samples were periodically collected from the product gas stream and analyzed by mass spectroscopy. The total conversion of TCE into degradation products, i.e., HCl, CO, and CO_2, etc. was greater than 80%. The temperature of the bed was found to be about 355°C with about 120W of microwave power and with moist air flowing over the bed at a rate of 100 ml/min. Microwave energy was also used in an attempt to regenerate the carbon bed although complete removal was not achieved.

A commercial system[9] developed for drying and solidification of concentrated radioactive and toxic liquid wastes is shown in Fig. 4. Concentrated liquid is fed to a drum through a hood which also has a waveguide feed. 6 kW of 2.45 GHz microwave power is applied to the drum contents and evaporates the free water in the concentrated liquid at the rate of 5 to 7 ℓ/h. The process can operate in a batch mode or in a continuous feed mode where the inlet flow is balanced by the distillate flow. The salt resuide can be further heated to achieve even greater volume reductions if desired. A waveguide window prevents steam from entering the waveguide system. The manufacturer claims the following advantages for the microwave drying system:

1. No mechanical moving parts.
2. No heat exchanger surfaces.
3. Automatic evaporation rate control.
4. Final product directly packed in the final storage container.
5. Efficiently high volume reduction.
6. Low cost storage.
7. Applicable to various drum systems.
8. Feasible for stationary or mobile use.

The manufacturer applies the microwave power to the waste with a simple open waveguide which produces a half-power beam width of only 75°. This produces localized heating of the liquid waste only at the liquid surface under the waveguide opening. Heat transfer to the rest of the waste volume occurs rather slowly via convection. When the water is removed from the waste-form the dried salts have very poor thermal conductivity which requires that the microwave power be limited to prevent thermal runaway of the salt cake directly under the waveguide feed. In addition, as the level in the drum rises due to continuous feeding, the heated zone moves closer to the waveguide feed thus further serving to localize the heating under the waveguide. A better solution would be to improve heating uniformity using any of the techniques mentioned in the introduction which should improve throughput. The manufacturer states that the process is limited to 200° C, however, about 350° C is required to melt nitrates commonly found in radioactive salts for the highest volume reduction ratios and to form a solid monolithic structure without free particulates.

CONCLUSIONS

Microwave processing of radioactive materials is proceeding here and abroad, where work is somewhat more advanced. Some of the factors that have limited the application of microwaves to industrial processes here in general are unfamiliarity of personnel with microwave technology, lack of comprehensive data on microwave properties of materials, and the fact that only a few domestic companies[12,13,14,15] are active in microwave process engineering. To overcome these disadvantages, the microwave process must demonstrate superiority over alternatives in one or more of the areas of speed, efficiency, condition of final product, remote maintenance, reliability, process flexibility, and minimization of process wastes. In addition, end users must work closely with microwave process engineers who understand what constitutes good microwave practice when designing bench- and pilot-scale experiments.

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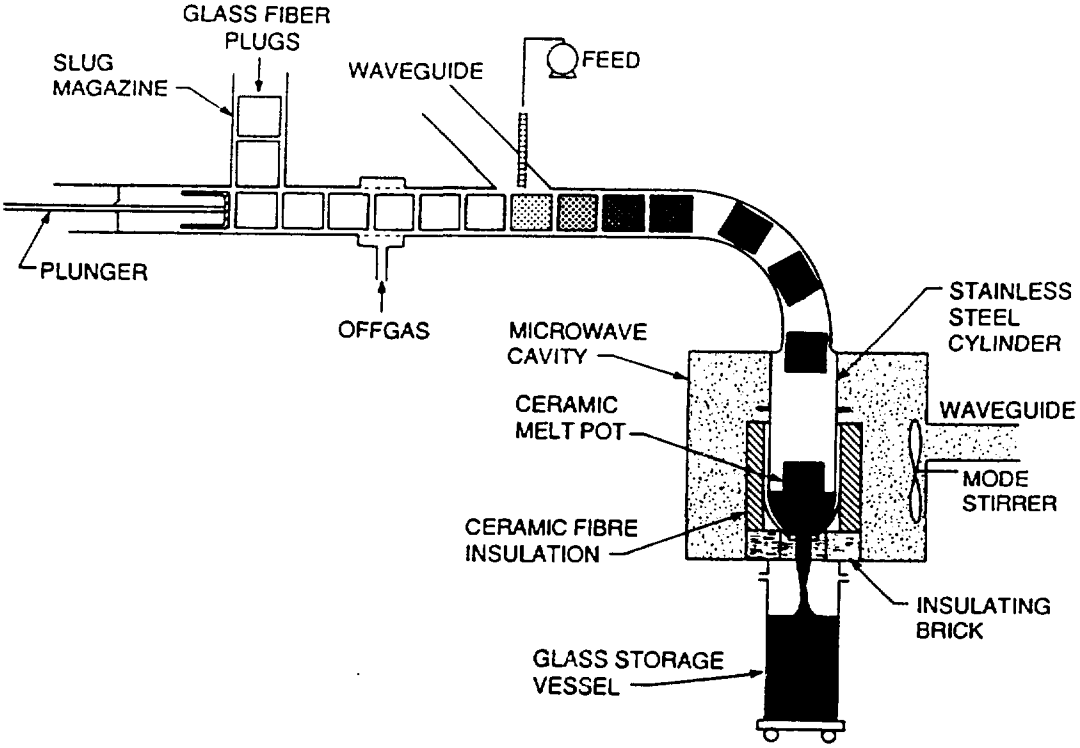

Fig. 1 The pilot-scale microwave vitrification equipment. Adapted with permission from M. P. Simpson and M. S. Morrell, Electronics and Power, 28(9), 613 (1982).

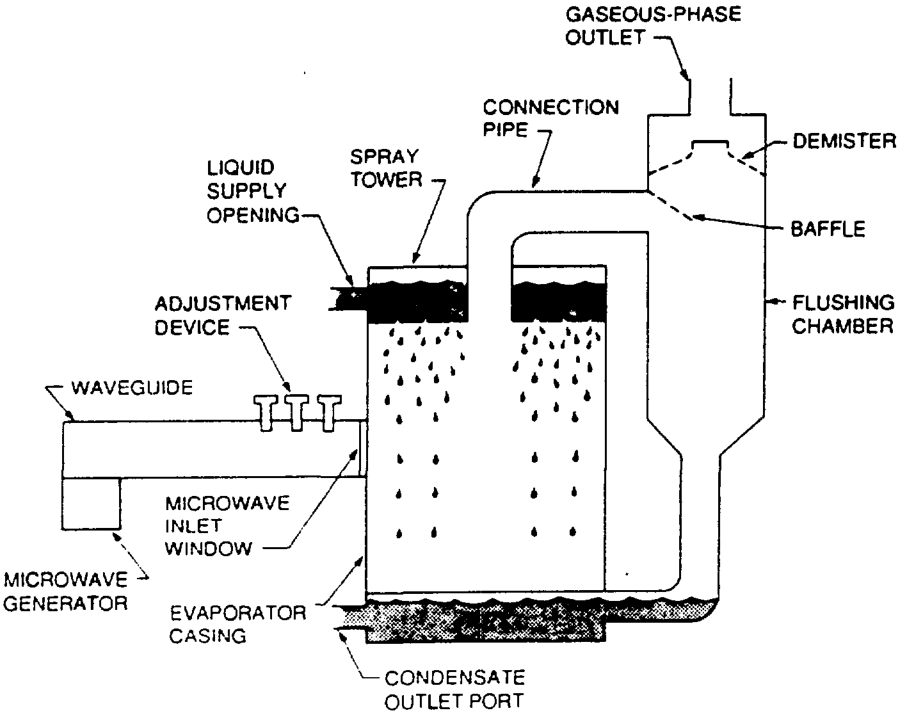

Fig. 2. The microwave spray evaporator. Adapted with permission from S. Taura et. al., Japanese patent 60-153, 567 Mitsubishi Heavy Industries Co., Ltd. 599 (1987).

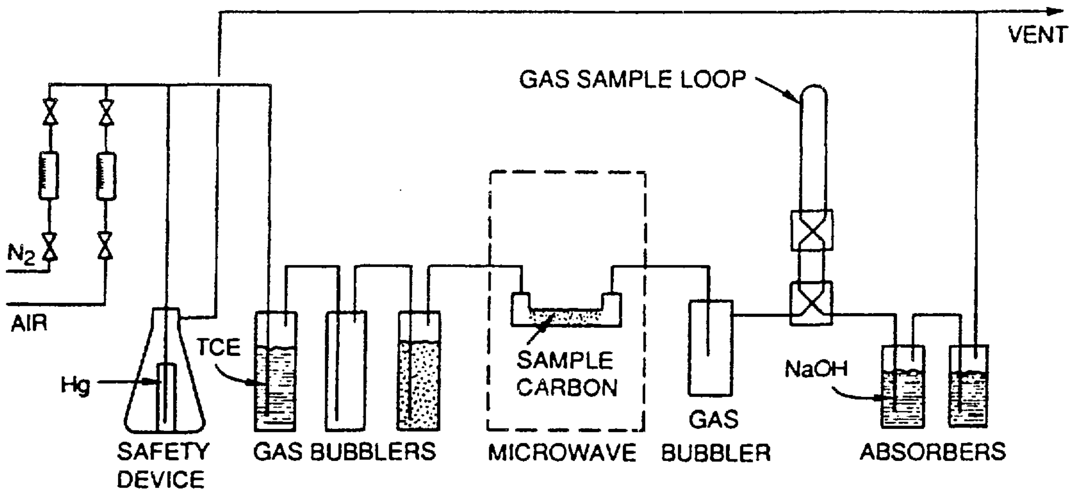

Fig. 3. The microwave-assisted hazardous wastes detoxification equipment. Adapted with permission from R. Varma et al., Argonne National Laboratory Report, ANL-87-20, 4 (1987).

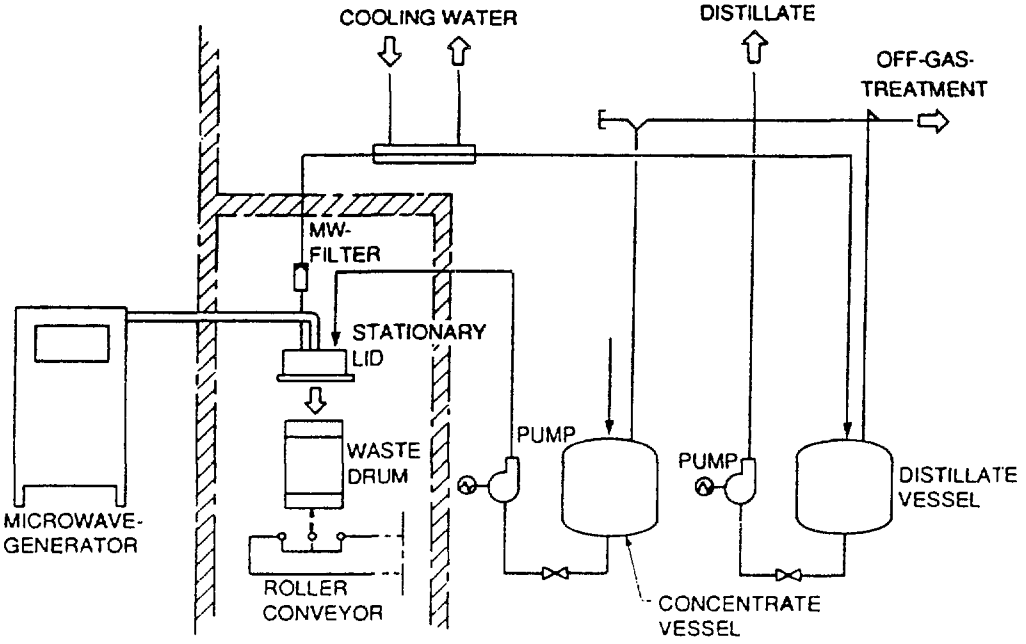

Fig. 4. The microwave in-drum evaporation equipment. Adapted with permission from Kraftanlagen Heidelberg product literature, 2 (1988).