

Microwave In-Drum Drying

***A New Volume-Reduction
Process for Radioactive
and Toxic Liquid Waste***

By Christian
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Liquid radioactive wastes are generated during the regular operation, downtime, or shutdown processes of nuclear power plants and other nuclear facilities. In nuclear power plants, these liquids are generally enriched using a circulation evaporator to a dry substance content of up to 20 weight percent. These concentrates must undergo additional drying/concentration processes prior to being accepted for final disposal.

The concentrates must be dried in a separate drying plant and placed in a receptacle (e.g., a 200-liter drum) for the final disposal. Common techniques include using a rotary film evaporator; other techniques include evaporation at reduced pressure by external heating. The concentrates must be enriched up to a dry substance content of approximately 80 wt% to get a solid salt block for final disposal. The hardening of the hot waste product to a solid salt block occurs during the cooling process. The residual (20 wt%) water is bound in the crystal during the cooling step.

For several reasons, it is safer and more economical to apply the drying procedure directly inside the final disposal container. The most common procedure is to heat up the shell of the drum from the outside with resisting heat tapes. However, heating the outside of the drum leads to a temperature gradient between the drum wall and the drum center. The highest temperature is achieved at the cover of the drum, leading the solidification process to start there and resulting in the formation of a salt layer there. This causes reduced heat conductivity from the out-

A microwave-heated waste drying system can provide a tenfold reduction in liquid waste processing time.

side compared to the inside (liquid solution) and a corresponding increase in the thermal resistance. As a result, the solution in the center of the drum generally does not completely crystallize, leading to longer process times and the need for considerably higher outside temperatures. The outside temperatures, however, are limited by the temperature resistance of the materials and by process conditions. The actual time to dry a drum with an outside heating method is about 85 days.

A NEW SOLUTION

As a result of a long-term development program conducted in conjunction with the German nuclear industry, Linn High Therm has patented a procedure using microwaves to reduce the liquid content in radioactive liquid waste. This process, microwave in-drum drying (MIDD), is an evaporation-driven crystallization process.

In cooperation with the German nuclear industry, Linn built a pilot MIDD plant. Several test runs with defined wastewaters (nonradioactive simulated solutions) have been successfully performed, results of which are presented later in this article.

The most important advantages of the MIDD process

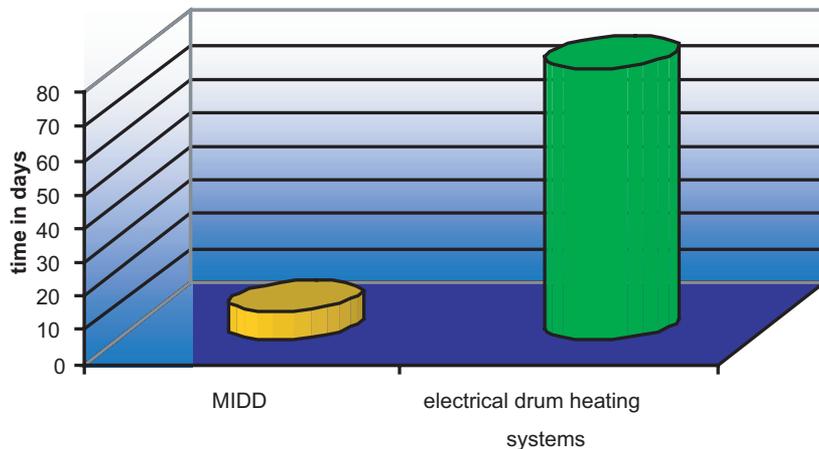


Fig. 1. Comparison of process times for the MIDD system and electrical drum heating systems.

compared to the outside heating process include the following:

- Microwaves that generate heat directly in the solution over the complete volume of the liquid layer.
- Minimal temperature gradients and homogeneous crystallization.
- Process times that are 10 times shorter than the more common electrical heating methods (see Fig. 1).

THE MIDD PROCESS

The MIDD process is a semicontinuous one that starts with feeding an initial amount of liquid waste into a final package (e.g., a drum) while it is being preheated by induction heating. Then the microwave heating system is engaged, and more liquid waste is continuously fed by a dosing pump. During evaporation, the drum and the microwave applicator are kept under a slight vacuum. Exhaust steam is suctioned off by a fan. An aerosol separator removes entrained dust particles and liquid drops. The feed steam is condensed in a water-cooled heat exchanger, and the condensate is collected in a separate vessel. At the end of the process, the dosing pump is stopped, and the microwave heat evaporates the rest of the liquid inside the container. After cooldown of the drum and material, the container is replaced with an empty one, and the cycle can begin again.

Generally, the condensate is discarded, and the solid waste in the drum is ready for direct disposal. During the process, a closed-circuit cooler ensures a constant low temperature of the cold side of the heat exchanger, which guarantees that the exhaust steam is condensed completely. To control the MIDD plant, a Simatic S7 computer is used.

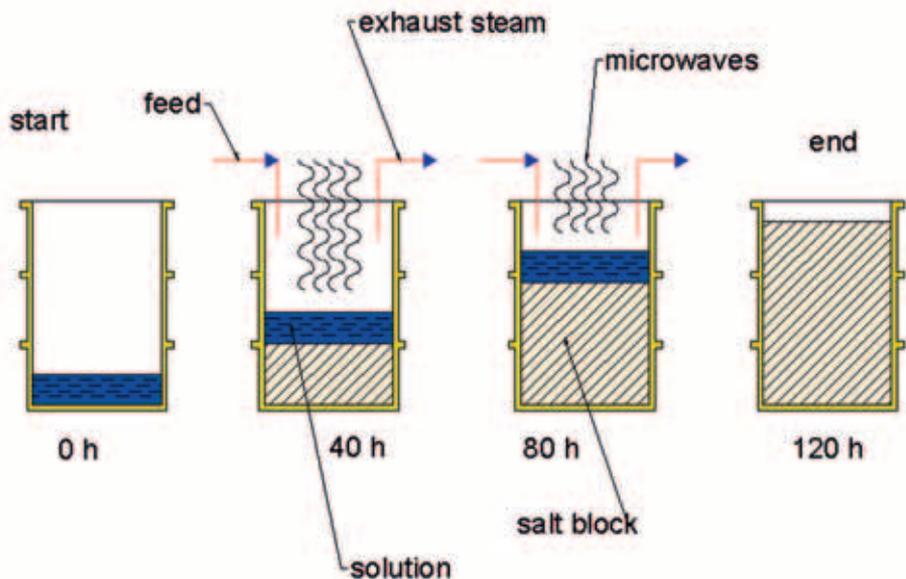


Fig. 2. The MIDD process.

All in- and outgoing mass flows are measured, so that the mass balance of the evaporation process is ensured. In addition, the pressure, the temperature, and the liquid level inside the container are measured continuously. Together with the other control parameters, the thickness of the liquid layer above the dry salt layer inside the container is determined, which is necessary for controlled crystallization. All data are logged in a data-logging unit. The data can either be shown at a 12-inch touch-screen at the control panel or printed from a connected external personal computer and printer. The progression of the MIDD process can be observed at the touch-screen control panel.

TECHNICAL DETAIL

MIDD is a semicontinuous crystallization process (see Fig. 2). Before the start of microwave heating, a certain amount of solution has to be pumped into the recipient drum.

Then, the solution is heated by a microwave process. The microwave power has to be controlled with respect to the increasing boiling point of the liquid during the evaporation process because of rising salt content. At a dry substance content of about 30 wt%, the solution is oversaturated, and the boiling point, as well as the concentration, stays constant. At this point, the content of the solid phase is steadily growing. The thermal balance is reached at approximately 100°C and a reduced pressure of around 900 millibars absolute pressure. After the liquid waste feed has been stopped, the homogeneous liquid surface over the solid product disappears. This causes the product surface to be heated too fast, because of the low thermal conductivity and capacity. The boiling curve is shown in Fig. 3.

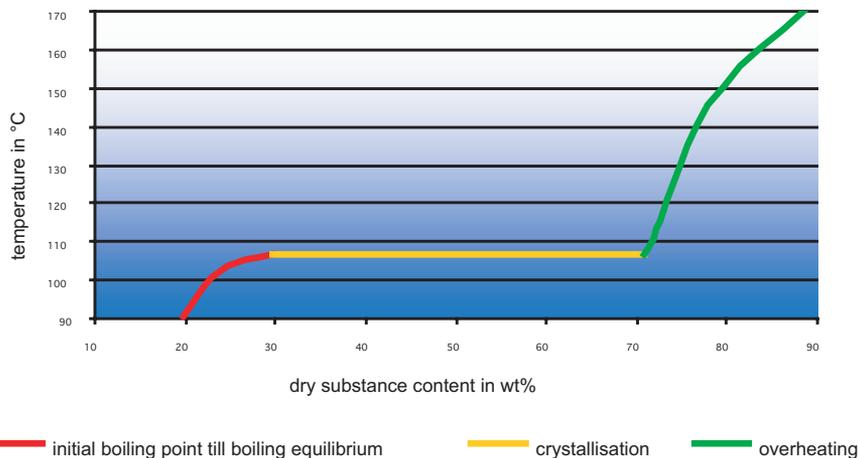


Fig. 3. Temperature diagram.

The 70 wt% dry substance content is calculated from the mass balance. The remaining weight percentage of water is bound as crystal water. The residual moisture content is 0.2 wt%. A pyrometer, installed to prevent overheating of the solution, detects the temperature of the product surface.

The controlled and determining process parameters are the microwave power and the feed flow. The grade of the drying depends strongly on the exact process control. The liquid waste contains salts that form crystal complexes with water at low temperatures. A 17 wt% solution containing aqueous sodium sulfate solution and other additives can be used as a reference solution. Sodium sulfate crystallizes in the Glauber's salt ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$) modification at low temperatures of approximately 32°C. To achieve better drying, the solution should not be cooled below this temperature during the process. The evaporation-driven crystallization goes on continuously, until the drum is completely filled with dry waste product.

PILOT TEST RESULTS

At the MIDD pilot plant, we conducted various tests to optimize the process parameters. We performed the two most important tests with defined concentrates surrogates (recipes originating from circulation evaporators of nuclear power plants). The main part of this solution is sodium sulfate. Some tests were performed with a microwave power of a 6-kW magnetron, while other tests were performed with a 20-kW magnetron. The composition of the solution was about 82 percent H_2O , 16 percent Na_2SO_4 , 1 percent $\text{Fe}_2(\text{SO}_4)_3$, and 1 percent other additives.

In the following, we present all test results as mean values. With the 6-kW magnetron, 1750 kilograms of those aqueous concentrates are dried in about 200 hours, yielding 345 kg of dry salt

and 1405 kg condensed water. The pilot plant was operated in 12- to 25-kg batches without interruption. With the 20-kW magnetron, 1280 kg of those concentrates were dried in 123 hours, yielding 363 kg dry salt and 917 kg condensed water (see Fig. 4).

The results of these tests show that an increase of microwave power leads to a consequent increase of the evaporation rate and feed flow. The mean evaporation rate increased from 6.3 kg/h to 7.8 kg/h, and the feed flow

was increased from 8.4 kg/h to 10.6 kg/h. The small increase is the result of high reflections due to the small volume of the 200-L drum. Also, the type of operation influences the feed flow and the evaporation rate.

Other tests were done with discontinuous and semi-continuous feed supply. These results show that semi-continuous feed supply increases the feed flow and the mean evaporation rate by 20 percent.

The dry substance content of salts from 6-kW trials was higher than those from 20-kW tests, because the 20-kW trial could not be operated continuously. Interruptions caused a temperature decrease to less than 32°C. Water not evaporated was bound as crystal water during these interruptions, and Glauber's salt was formed in the bulk. The 6-kW trial was operated without interruption during the batches, and so a larger portion crystallized into sodium sulfate instead of Glauber's salt than in the 20-kW tests. Therefore, the 6-kW tests showed a larger dry substance content. In other short-term tests, a dry substance content of approximately 82 percent was nearly always achieved.

All data depend on evaporation rate and applicator efficiency. These parameters were measured continuously during the crystallization of the defined liquid waste. The highest evaporation rate was at a temperature of approximately 106°C (boiling equilibrium) and absolute pressure of around 900 mbar. The best applicator efficiency was

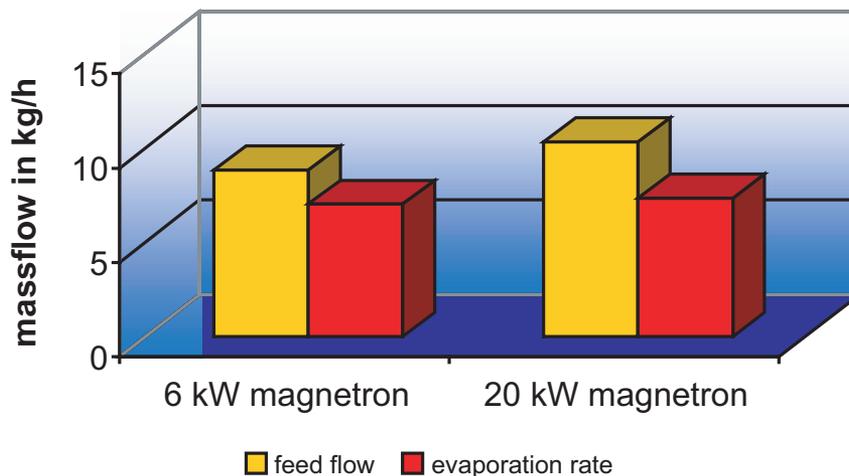


Fig. 4. Comparison of test runs.

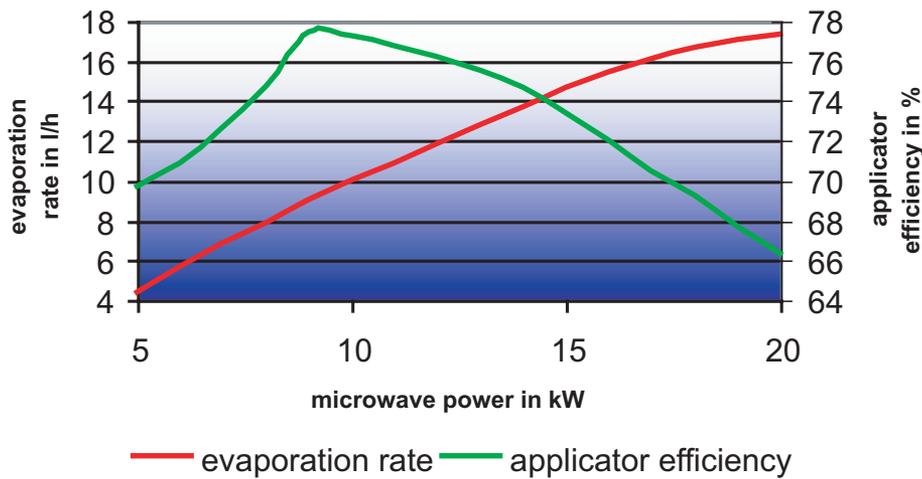


Fig. 5. Evaporation rate and applicator efficiency.

also at the boiling equilibrium. The applicator efficiency achieves a maximum of 77.3 percent at approximately 9 kW microwave power. The efficiency decreased below 69.8 percent when the microwave power was increased above 9 kW. The reason for this behavior is increased microwave reflection, which leads to a decreasing applicator efficiency. The dependence of the evaporation rate (stream of condensate) on applicator efficiency from the microwave power input is shown in Fig. 5.

Depending on the accuracy of the control of the MIDD process, the resulting waste can vary between a salt block and loose dry powder.

COMMERCIAL DEVELOPMENT AND COSTS

As a result of the different tests and further developments to optimize the process, Linn High Therm has created a new MIDD plant model (see Fig. 6). This new prototype is ready for use in the nuclear industry. The MIDD prototype is being adapted to nuclear safety regulations in cooperation with a partner from the German nuclear industry.

From a technical point of view, it was important to achieve the best possible homogeneous electromagnetic field and the best applicator efficiency. Therefore, in the commercial model, the single 20-kW magnetron was replaced by nine 800-W standard magnetrons. All parts of the new plant are made of stainless steel, polytetrafluoroethylene, or silicone. The plant is designed to be cleaned easily, with quick connectors and short distances between the different components, which include the following:

- External power supply (optional).

- Microwave generator.
- Drum adaptations (e.g., 200-L drum, 400-L drum, rectangular containers, concrete containers).
- Remote control panel.
- 20-in. housing container (optional).
- Simatic S7 control system.
- Dosing adaptation systems (e.g., 200-L drum, 400-L drum).

The energy cost using the MIDD system for crystallization of liquid waste would cost around 180 euros per ton of waste in Germany. This cost estimate covers power and energy consumption of the evaporation crystallization of 1.14 cubic meters of watery solution with a 20 wt% content of sodium sulfate in a 200-L drum. The costs of cooling water and pressurized air were converted to energy, because those costs vary among different countries. The mean process time was calculated with an evaporation rate of 8 kg/h with the best applicator efficiency. The lifetime costs of the magnetrons and other components of the MIDD plant were included in the estimate. For companies that produce only small amounts of liquid waste, Linn High Therm will also offer a MIDD plant for leasing. ■

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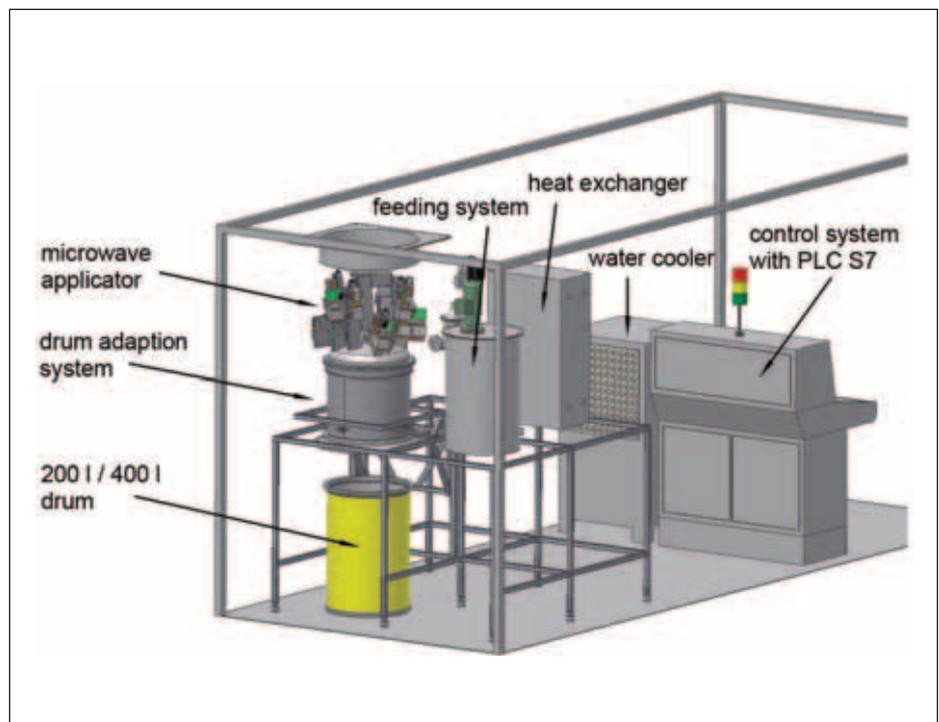


Fig. 6. The commercial MIDD system setup