SOLIDIFICATION OF LOW LEVEL SALT SOLUTIONS WITH MICROWAVES

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ABSTRACT

A new microwave heating process has been developed for drying and solidification of liquid wastes arising from the treatment of liquid and gaseous effluents of nuclear facilities. The resulting waste product is a solid salt block contained in a standard steel drum suitable for interim storage and ultimate disposal. The waste to be treated is injected into a drying chamber through a spray nozzle in the drum lid. The microwave energy is brought in through a window in the drum lid. Heat exchanger surfaces are not at all required. The vapors are released through a microwave filter into the condenser. The condensate is directed to a distillate tank. The plant has no mechanically actuated parts in the contaminated area.

INTRODUCTION

Heating with microwaves has been practiced in medicine and industry for many years. Also in private households, microwave ovens nowadays are already standard equipment.

Microwave heating is suitable for concentrating and drying all kinds of substances containing aqueous and organic solvents. The substances must have a high relative dielectric loss factor, which is a parameter determining the energy dissipated in the dielectric. This method makes use of the fact that the electromagnetic waves which penetrate into a dielectric substance of a high loss factor, are almost completely absorbed by that substance. During this process, the high-frequency energy is entirely converted into heat.

The origin of microwave heating lies in the ability of the electric field to polarise the charges in the material and in the inability of this polarization to follow extremely rapid reversals of the electric field. The polarization vector lags behind the applied electric field ensuring that the resulting current has a component in phase with the applied electric field, which results in the dissipation of power within the material. Parallel to these polarization effects, dielectric heating is ensured due to direct conduction.

These are the reasons why for this heating process does not require a supply of energy from outside via heat exchanger surfaces.

It is important to exactly specify the frequency ranges defined by the term 'microwave'. These frequencies range between 300 MHz and 30 GHz. At frequencies above 500 MHz it is not possible to use the usual wire circuits and that is why the power is transferred in waveguides to the actual consumer i.e. to the material to be processed. This technique is referred to as microwave heating. The principal frequencies allocated for industrial application of microwave heating are 900\MHz and 2450 MHz.

In the last three years we have developed a new microwave heating process for drying and solidification of liquid wastes arising from the treatment of liquid and gaseous effluents of nuclear facilities. The main problem was to attain a maximum power transfer from the generator to the material to be processed. The resulting waste product is a solid salt block in a standard steel drum ready for interim storage and ultimate disposal. This technique is a further step in waste volume minimization.

After first promising results of the research and development work, this process was selected as one of the waste solidification techniques to be applied in the German Spent Fuel Reprocessing Plant under construction in Wackersdorf, Bavaria. There it will be used for the solidification and drying of radioactively contaminated scrubber liquid originating from the dry active waste incinerator.

The advantage of using microwave drying and solidification is that a considerable volume reduction of radioactive waste solutions is attained. As compared to the conventional method of solidifying the waste in cement, this method makes it possible to reduce the volume of the produced waste by more than factor 10. The product is available in solid form and is suitable for ultimate storage. This process therefore contributes a great deal to meeting the requirement of minimizing radioactive waste.

The main design features of the new microwave solidification facility are:

- no heat exchanger surfaces
- no mechanically actuated parts
- automatic evaporation rate control

PLANT DESIGN

The plant consists of the following components:

- A microwave generator, including magnetron, local control panel with performance indicator, and watercooled circulator. Waveguide interconnections

- Impedance matching-transformer for power optimization
- Drying chamber, consisting of a sheet metal drum (200 l, 330\l or 400 l waste drum) and a special lid. The lid is a stationary component. The drum is intended as final packing, and is of commercial standard design.
- Microwave stop filter in the vapor hood
- A condenser for vapor condensation
- A concentrate tank and a diaphragm dosing pump for the concentrate
- A distillate tank and a distillate pump
- A spray nozzle in the concentrate line
- Various metering and control instruments

A view and a flow sheet of the plant are shown in Fig. 1 and Fig. 2.

PROCESS DESCRIPTION

The plant is ready for operation as soon as the drum is coupled to the lid, and when all electrical connections are made. Actual coupling is either a screwed connection, or it is ensured by quick-action clamping of the two flanges. Drying is performed as follows:

Concentrate is injected into the drying chamber through the spray nozzle in the drum lid. At the same time the generator is also put into operation, and the microwave energy is transferred into the drying chamber at the lid coupling.

Alternatively, it is also possible to fill the drum with concentrate before starting the microwave generator. In the course of the drying process, and as soon as a constant distillate flow is obtained, concentrate is continuously dosed at a ratio 1:1, related to the evaporation rate. Towards the end of the batch, dosing is stopped and the still remaining water is eliminated by supplying further energy.

Additional shell heating may heat the drum up to the operating temperature, whereas on the outside it may be protected against heat losses.

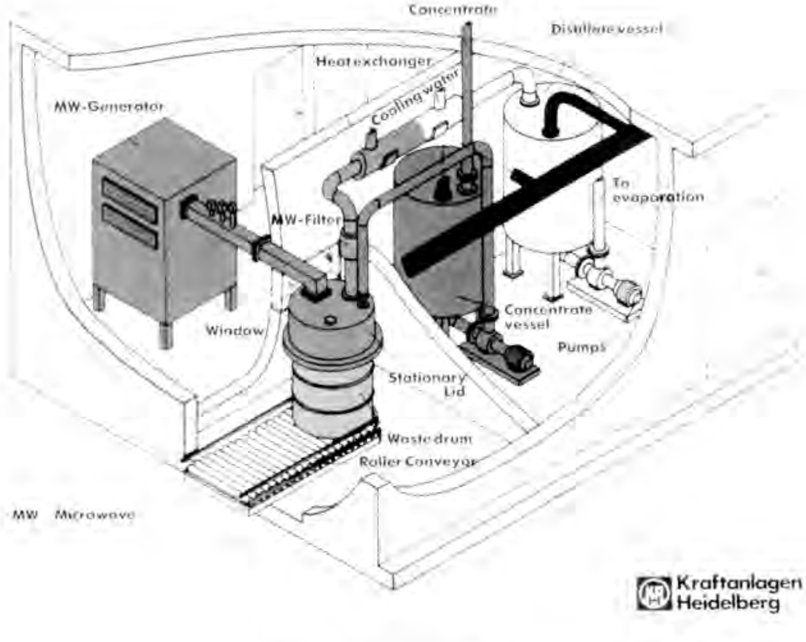

Fig. 1. View of the Plant.

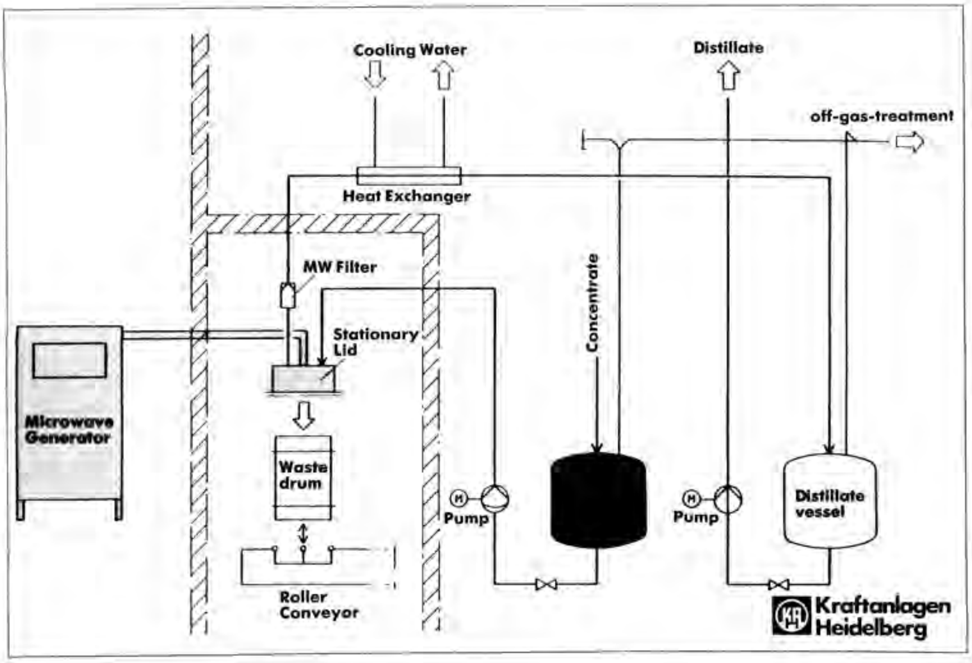

Fig. 2. Flow Sheet of the Plant.

The microwaves ensure heating of the substance to be dried, until the contained water is evaporated. The vapors are released through the microwave filter into the condenser where they undergo condensation. The condensate is directed to the distillate tank.

In an efficient heating system, it is indispensable that the transfer of heating energy from the generator to where it is used, is guaranteed with only a minimum loss. The major feature of a microwave heating system is the unusually high heat transfer coefficient which is restricted by only two factors. First, energy is absorbed by the transmission waveguide walling. But this is only a very small portion of the transmitted power, and is therefore negligible. Second, power is reflected from the load when there is no optimum matching. It is generally known that optimum matching is prerequisite to a maximum transfer of power from the source of power to the load, and as a consequence the load resistance should equal the generator internal resistance. If the load resistance is less than the generator internal resistance, there is a higher current flow and more current is dissipated in the generator, and this may cause damage if excessive. If the load resistance is higher than the generator internal resistance, there is a lower current flow and less power is dissipated in the load. The same argument applies with complex impedances leading to the result that for optimum power transfer the load impedance should be the complex conjugate of the generator impedance. But the load impedance in our microwave heating process is not constant.

Load variations, reflection from the drum walls and the lid, difficult adaptation to conditions in the drying chamber cause a part of the supplied microwave energy to be reflected to the waveguide, and back again to the generator. That is why we installed a circulator and a transformer for optimum matching.

The circulator installed in the generator ensures that by means of a water load the reflected power is converted into heat. This prevents the destruction of the magnetron.

In order to minimize this reflected power, the plant is provided with a triple screw impedance-matching transformer. This transformer consists of three screws fixed in the waveguide at a distance of a half wave length from one screw to the other. The length of the microwaves in free space is approx. 12.4 cm. The screws can be automatically turned and are intended as an impedance corrector.

This enables the control of the phase between forward and return wave to such an extent that the return wave is minimized. In this way the load resistance is matched to the generator internal resistance.

In order to prevent vapors condensing in the waveguide, the lid is provided at the waveguide connection with a window which is permeable to microwaves, but impermeable to steam. This window is designed as a Teflon plate, or is made of borosilicate glass with only a little dielectric loss factor.

The plant is suitable for drying all kinds of salt solutions. With an increasing concentration of the salt solution, the supplied microwave energy causes an elevation of the boiling point of the solution which finally causes the solidification of the salt products in the drum, when all the water is entirely evaporated. However, heating is stopped before the salt attains its melting point. There is no melting of salt inside the drum. The temperature is limited to a maximum of 220C. This is necessary in order to preserve the materials used (window, window seal, flange seal). The level with the highest temperature in the drum rises in accordance with the increasing filling level. This level is the section where the microwaves are actually absorbed in the substance to be dried. Permanent control of this level signals when the drum has reached its highest permissible filling level. Maximum filling level is reached at about 3-4 cm below the drum flange. As soon as this limit is reached, the concentrate dosing pump is switched off and the rest of the water in the drum is evaporated. After a short time, the distillate flow diminishes and finally stops. Now the generator is switched off and the additional heating of the drum is stopped. After a certain cooling time the drum is lowered with the lifting device. Prior to this action the drum flange and lid are disconnected.

The drum and the lid become contaminated inside during drying. Therefore the process is done in a closed compartment. Manipulation is possible through glove ports. However, the constructional design of the plant also allows an arrangement without boxes. In doing so, the individual plant components shall be disposed so as to ensure that a spread of contamination to the outside is avoided, even in open condition.

For instance a sliding gate may be provided to separate the lid from the environment. This gate shall guarantee that, after un-coupling, the drum and the stationary lid do not cause a spread of contamination.

With regard to the desired frequency of 2.45 GHz the generator size is limited to an output of 6 kW. With this power, evaporation rates of 5 to 7 l/h are obtained, depending on the evaporation enthalpy of the salt solution to be dried. The plant can be extended by adding further modules when higher evaporation rates are required. In doing so, several magnetrons (up to 4 units) and circulators shall be provided in the switch cabinet. Then it is possible to supply the energy through 4 waveguides into the drying chamber. In this way it is also possible to ensure evaporation rates of *more* than 20 l/h, depending on the evaporation enthalpy. The plant efficiency may additionally be increased by using commercially available drum shell heaters. The additional heat supplied in this manner increases the evaporation rate accordingly.

TEST RESULTS OBTAINED SO FAR

The plant has so far been operated for approx. 1000 hours. The substances used were simulated solutions from washing loops and simulated low-level radioactive evaporator concentrates. In a continuous run approx. 1400 l of a simulated solution from washing loop was processed. The solution mainly consisted of sodium chloride. The total processing time was about 10 days corresponding with an average flowrate of 6 l/h.

The characteristic data were as follows:

- feed-concentrations of approx.33 weight % salt
- feed-density of approx 1.25 kg/dm^3
- conductivity in the distillate 30 to 120 μ S
- the concentrates were of a drying efficiency of more than 98 % (definition of residual humidity by weighing, and repeated drying)
- the temperature in the vapor chamber was constantly kept at 100°C
- temperatures up to 220°C were measured in the sump (concentrate)
- the maximum evaporation rate was 7.8 l/h, the minimum towards the end of drying was below 0.1 l/h
- the concentration factor was as high as 7
- the density of the final product (salt NaCl) was at about 1.95 kg/dm^3

At present, the preparatory work is underway which is required to enable that the plant is used for drying and solidification of low evaporator concentrates originating from power stations and the fuel cycle. From the power plants a typical composition of the usually acid evaporator concentrate is: Sodium chloride NaCl, sodium nitrate NaNo3, calcium chloride $CaCl_2$, boric acid H_3BO_3, caustic soda NaOH, silicone anti-foam agent, washing agent and water. For the waste coming from the fuel cycle the evaporator concentrate consists mainly of sodium nitrate with considerable amounts of detergents and decontamination agents with an average pH-value of 9 to 10. At the Nuclear Center of Karlsruhe, KfK, the complete installation will be set up and a first pilot test will be carried out with original LAW, produced at the KfK LAW evaporator installation. The current time schedule provides for plant operation being started by June 1988. We will provide further experience concerning the operating behavior, product quality and personnel radiation exposure.

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