Industrial vitrification processes for high-level liquid waste solutions

A technical overview of processes actively being used to treat and solidify liquid wastes in glass

by W. Baehr

The high-level waste (HLW) management programmes in countries pursuing the fuel reprocessing option are based on immobilization of HLW solutions into monolithic forms. Research and development on solidification of concentrated fission product solutions commenced in some countries more than 30 years ago. The initial work was mainly directed to defining a suitable matrix material and only later to developing techniques for industrial scale solidification.

Candidate materials for fission-product encapsulation have ranged from simple denitrated calcines to glasses, crystalline ceramics, or more complex forms, such as pellets coated with durable materials and glass or ceramic beads embedded in inert matrices. During recent years, there has been a growing concensus that glass offers the best compromise regarding properties, ease of fabrication, and experience. Several types of glass have been investigated; however, only silicate or borosilicate glass formulations have been selected.

Worldwide many solidification processes have been developed and demonstrated to a considerable extent but for various reasons are not now employed. The experience from such facilities nevertheless has produced much relevant data for designing current facilities. The solidification facilities presently being actively operated in the world on an industrial scale apply only the vitrification process.

The main steps of vitrification processes are the concentration of the high-level liquid waste (HLLW) solution by evaporation of the water and nitric acid; drying and calcination, which decomposes the nitrates to oxides; and reaction of oxides with glass-forming additives and fusion to produce HLW glass.

Depending on the processes, these steps may be separate or combined; that is, single or multi-stage processes. Drying and calcination operations are combined in one unit — the rotary kiln calcination — in the most established process.

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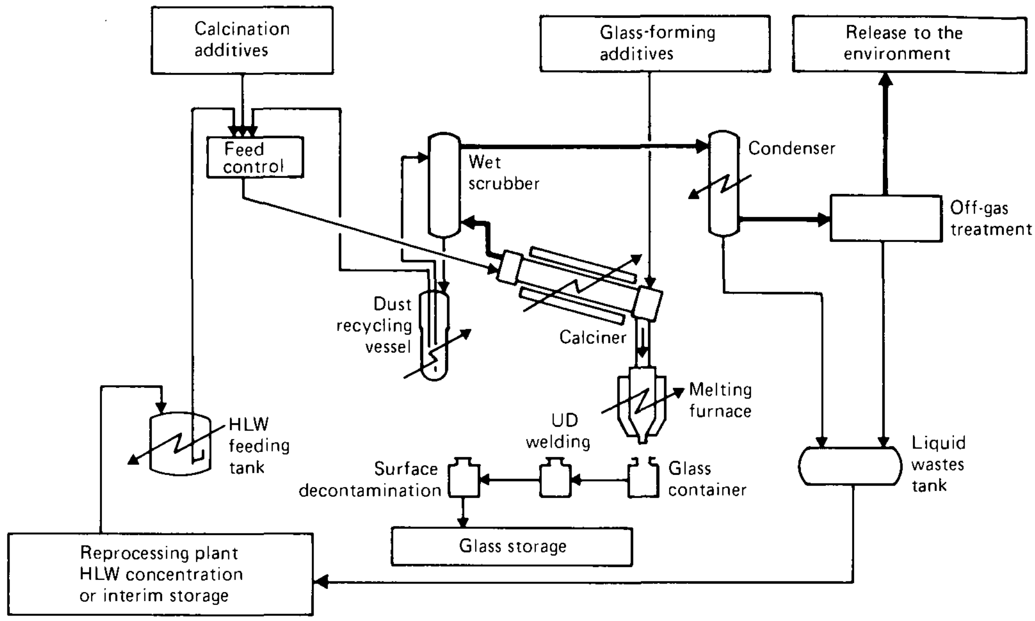
The glass melting process may be carried out basically either in a metallic melter or in a ceramic melter. The metallic melter may be a furnace with induction heating of the metal shell with heat transfer to the glass product. The main advantages of metallic melters are low costs and easy handling. The disadvantage is that the throughput capacity is limited to 30-to-40 litres per hour. Ceramic melters are usually directly heated by submerged power electrodes and achieve a throughput capacity of HLLW greater than 100 litres per hour. However, the ceramic melter process is so far characterized by high costs and comparatively complicated handling.

Two major vitrification processes have evolved for the conversion of HLLW solution to borosilicate glass. One is the well-known French AVM process (Atelier de Vitrification Marcoule), a continuous two-stage vitrification technique which commenced industrial processing of highly active waste in 1978. The other process is the continuous single-stage ceramic melter process, which has been demonstrated under radioactive conditions in the Federal Republic of Germany's Pamela plant located in Mol, Belgium, since 1985.

The AVM process

The AVM process consists of a combination of a rotary kiln and an induction-heated metal glass-melting crucible. *(See accompanying figure.)* The high-level fission product solution is fed with calcining additives into the upper end of the calciner, a slightly inclined tubular kiln which rotates at 30 revolutions per minute (rpm). The kiln is supported at each end by roller bearings. Special gas-tight seals are located at each end, allowing longitudinal expansion and maintenance of tightness. A rabble bar inside the kiln is designed to avoid possible caking. The tube is heated externally by an electric resistance furnace divided into four zones. The first two zones devoted to the evaporation have a heating capacity of 20 kilowatts each and the others 10 kilowatts each. The temperature varies from 225° Celsius at the feed point to a maximum of 600° Celsius.

Marcoule Vitrification Facility



The waste solution flow rate is normally around 40 litres per hour. The outlet from the rotary calciner is connected to the melter. The calcined products flow by gravity into the melter which is heated by medium frequency induction to about 1150° Celsius. The melter is simultaneously fed with the glass frit and the glass is cast at 8-hour intervals. The lower portion of the melter terminates in a casting nozzle which allows the contents to be emptied by melting a normally solid glass plug. The melter throughput is about 15 kilograms per hour.

The off-gases generated in the melter and in the calciner are exhausted through the calciner and treated at first in a hot scrubber. The scrubber traps the entrained particles and dissolves them in a continuous flow of boiling nitric acid. The resulting solution is continuously recycled to the calciner. The off-gases are then processed successively in a recombination vessel for treating the nitrous vapour fraction, two absorption columns, and filters. The gases are then discharged to the ventilation system. Recombined acid from off-gas treatment is recycled.

The glass is poured into a refractory stainless steel canister which, when full, is sealed with a lid by automatic plasma arc welding. It is then decontaminated with a water spray at a pressure of 250 bar and the canister transferred to a ventilated pit in the interim storage facility.

At the end of October 1988, AVM had converted around 1225 cubic metres of fission-product solution into glass with a total activity content of 250 megacuries.* This resulted in the production of 1547 canisters containing about 540 tons of borosilicate glass.

The same process is to be used in two vitrification facilities designed and built by SGN at the La Hague fuel reprocessing site. The two units are practically identical: both include three vitrification lines, each with an evaporation capacity of 60 litres per hour and a glass production capacity rated at 25 kilograms per hour.

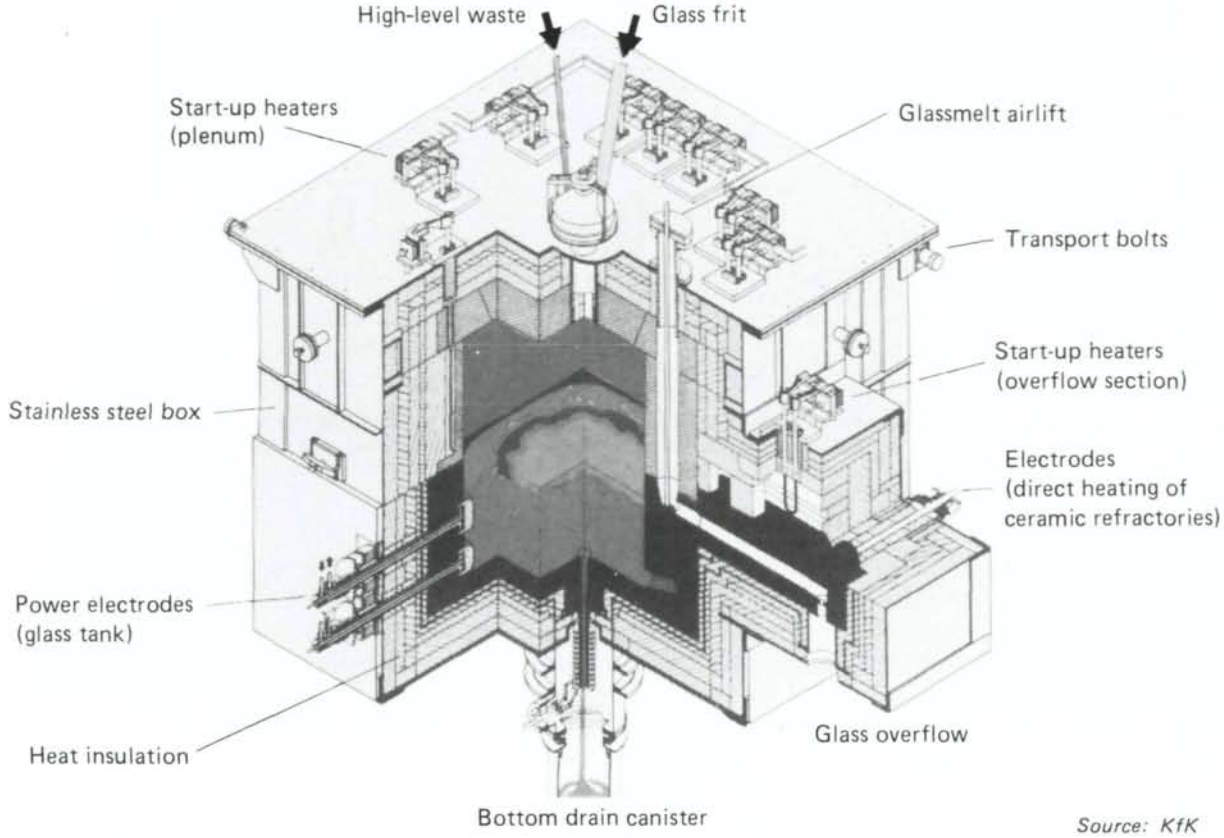
The AVM process has also been adopted in the Windscale Vitrification Plant (WVP) facility at Sellafield in the United Kingdom, which will use related equipment, with two fabrication lines.

The Pamela process

The Pamela vitrification plant is a single-step process. It is based on a liquid fed ceramic melter in which the high-level fission product solution is fed directly — together or separately with the glass forms — into the glass melter where the process steps of evaporation, calcination, and melting occur simultaneously. The principle behind the operation of the ceramic melter is Joule heating, which can be utilized since the glass is a good electrical conductor at high temperatures. An alternating electric current passing between electrodes immersed in the glass generates heat by the Joule effect. Dissipated resistive heat maintains the molten glass and melts incoming material. These melters are constructed from high corrosion resistance refractory materials. The power input is obtained by four pairs of Inconel-690 plate electrodes, placed in two levels of the melter pool at 1150–1200° Celsius. *(See accompanying figure.)*

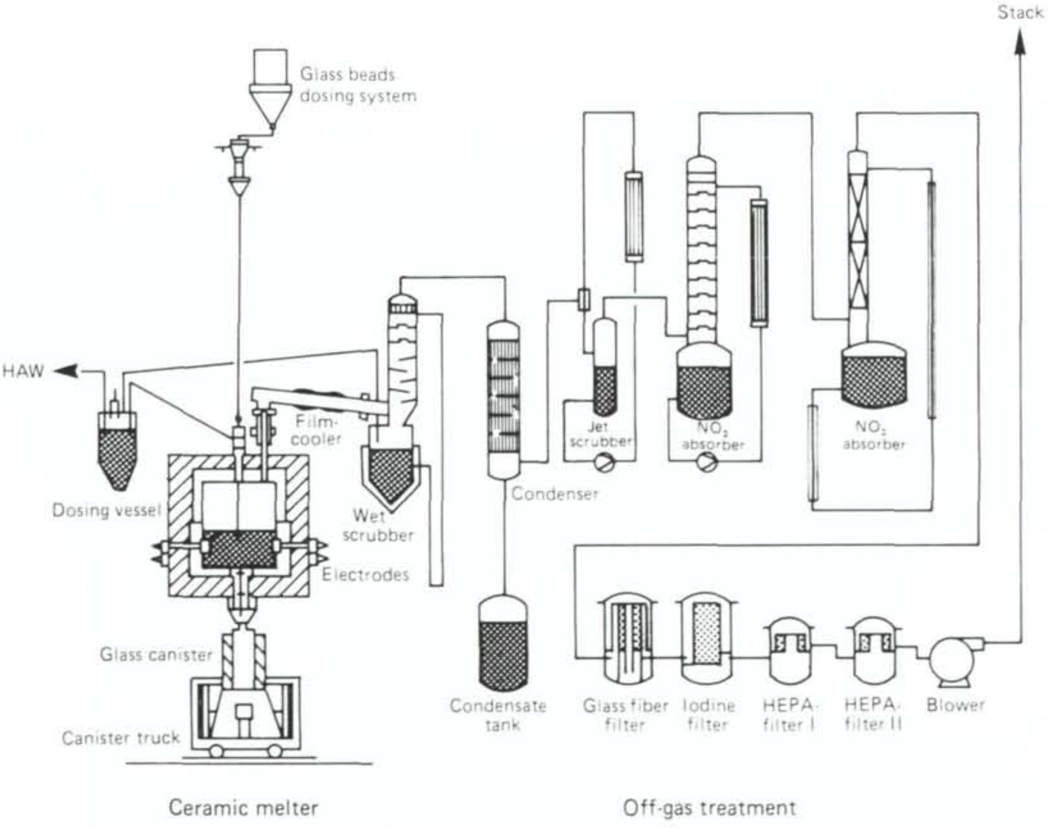
* One curie equals 37 giga-becquerels.

Ceramic-Lined Electric Waste Glass Melter (Pamela process)

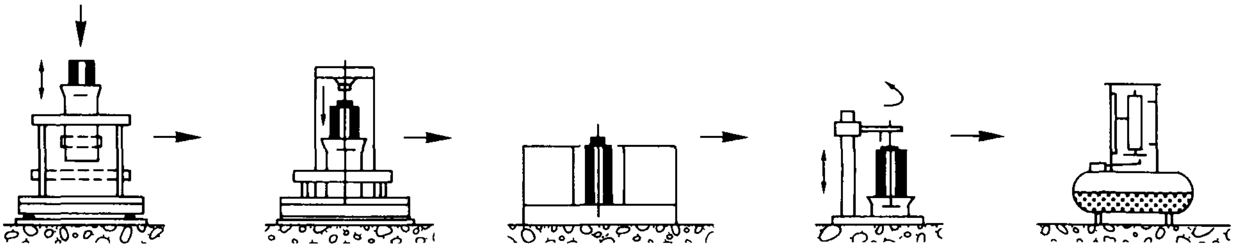


Source: KfK

Pamela Vitrification Plant



Remote handling sequence for filled high-level waste canisters



The figure shows the handling procedures for the glass canister generally implemented in vitrification plants based on both AVM and Pamela processes. After the glass is drained, the filled canister is subjected to a number of operations to prepare it for internal storage or ultimate disposal. The main operation steps are controlled cooling of the filled canister, welding of the lid, leakage testing, and decontamination.

Glass discharge can be accomplished by a bottom drain or by an airlift supported overflow. Two heating circuits are used to raise the glass temperature in the outlet channel of the bottom drainage for the start of the glass flow. Termination of the glass flow requires only that one heating circuit be switched off. A similar heating system is used for the filling of canisters with molten glass through the overflow drainage system. The off-gases of the ceramic melter have to be cleaned in a multi-stage off-gas cleaning system before the exhaust is allowed to be released to the environment. The off-gas cleaning systems of the diverse vitrification processes are basically similar. *(See accompanying figure showing a simplified flow sheet of the Pamela process.)*

Controlled cooling of a canister is necessary to avoid cracks in the glass and deteriorated product quality. Leak testing and decontamination ensures that no contamination is spread from the vitrification facility.

From the start of the Pamela plant in October 1985 until May 1988, approximately 265 cubic metres of highly active waste solutions have been vitrified. This has resulted in the production of 1381 canisters totalling about 265 tons of borosilicate glass and nine megacuries of activity.

Concluding remarks

Over the last decade great progress has been made in techniques for vitrification of high-level liquid wastes.

The French AVM process has been established in safe and successful operation over the years at Marcoule. Two further plants have been constructed at the La Hague site based on the same rotary calciner/metallic melter process. The first unit of the new plants began active operation in 1989 and the second is scheduled to achieve hot operation in 1990. A similar plant is under construction at Sellafield in the United Kingdom and also starts operation in 1990.

The ceramic melter process which was being developed during the 1970s has been chosen by several countries and is in various stages of application. The Federal Republic of Germany has successfully operated a Pamela version at the Belgium Eurochemic reprocessing plant. The USSR has operated a vitrification plant also based on the ceramic melter technology from 1986–88. Presently vitrification plants based on the ceramic melter are under construction at Savannah River and West Valley in the United States and at the Tokai Site in Japan. Plants are in the design stages for the Hanford site in the USA and for Japan. These plants are of large industrial-scale capacity and meet all standard industrial and nuclear criteria.

Over the coming years, the ceramic melter process has to be established in safe and efficient operation alongside the AVM. High-level glass produced by both processes must meet the disposal requirements established by the authorities for HLW repositories.