VITRIFICATION OF HIGH LEVEL WASTES IN BOROSILICATE GLASS
USING THE PAMELA CERAMIC MELTER

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ABSTRACT

In 1976, the PAMELA process for vitrification of high level radioactive wastes began a planned 10-year program of pilot testing in Germany and Belgium. To date 70 m^3 of simulated HLW have been processed, producing 40.7 metric tons of vitrified product. The pilot facility has accumulated substantial valuable experience, allowing for refinements in ceramic melter design and operating conditions. In addition, the feed and product handling systems have been optimized. A 30 kg/hr PAMELA system for processing of "hot" HLW is under construction at the Eurochemic fuel reprocessing center in Mol, Belgium. This system will be fully operational in October 1985. The cost of this process line, installed, de-bugged, and cold tested, is budgeted at DM 147 million or $60 million. This paper will report on the results of pilot scale testing and the design of the production unit now under construction.

WASTE FEED

The liquid high-level waste, which will be treated by the PAMELA-Process is called LEWC (Low Enriched Waste Concentrate). This name refers to the liquid waste solutions which have been produced during the reprocessing of low enriched uranium-fuels (uranium content of 5%). Prior to transfer to the waste storage tanks this waste solution has been concentrated to about 0.5 m^3 LEWC per ton of reprocessed uranium.

Tables I and II show the activity levels of the LEWC-feed, corrected to the year 1985, the expected year of operation of the hot PAMELA demonstration plant.

Nuclides	Tank 1 A (27 m^3)		Tank 1 B (32 m^3)	
	Activity (Ci/Tank)	Spec.activ. (Ci/l)	Activity (Ci/Tank)	Spec.activ. (Ci/l)
α+β total without H-3	2.7 E 6	100	4.8 E 6	150
β-total without H-3	2.4 E 6	90.3 (1985)	4.2 E 6	132 (1985)
H-3	12.7	4.7 E-4	12.7	4.0 E-4
Sr-90	8 E 5	29.5	1.25 E 6	39
Y-90	8 E 5	29.5	1.25 E 6	39
Ru-106	410	1.5 E-2	800	2.5 E-2
Rh-106	410	1.5 E-2	800	2.5 E-2
Cs-134	6.2 E 3	0.23	1.8 E 4	0.56
Cs-137	8.5 E 5	31	1.7 E 6	53
(Ba-137)	7.2 E 5	2.67 E 1	1.5 E 6	47
Ce-144	80	2.95 E-3	365	1.15 E-2
Pr-144	80	2.95 E-3	365	1.15 E-2
Eu-154	700	2.6 E-2	2.1 E 3	6.7 E-2
Co-60	700	2.59 E-2	2.1 E 3	6.6 E-2
α-total	7.9 E 3	0.29	2.1 E 4	0.67
Pu total (α) (1985)	430	1.6 E-2	960	0.03
Am-241	5.42 E 3	0.2	1.25 E 4	0.39
Cm-242	1 E-3	3.7 E-8	3.9 E-2	1.2 E-7
Cm-243/244	2.07 E 3	7.7 E-2	7.98 E 3	0.25
decay heat	1.02 E4W	0.38 W/l	1.85 E4W	0.58 W/l

Table I Activity Data for the LEWC based on Measurements in 1979/80 and calculated for 1985

Component	Tank 1 A (27 m^3)	Tank 1 B (32 m^3)
	Feed	Feed
Density g/cm^3	1.26 at 20°C	1.27 at 20°C
Free nitric acid mol/l	2.8	2.2
Sulfate mol/l	0.08	0.098
Fluoride mol/l	0.54 ≙ 10.2 g/l	0.62 ≙ 11.7 g/l
Na g/l	36	57
Fe g/l	13.3	18
Cr g/l	0.55	2.9
Al g/l	8.25	7.6
Mn g/l	2.05	6.0
Mg g/l	< 1	< 1
Ca g/l	1.21	1.9
Sr g/l	0.4	0.6
Ba g/l	- (0.2)	-
Cer g/l	1.35	2.23
Nd g/l	2.11	3.4
Zr g/l	0.2	0.2
Mo g/l	1.24	1.32
Ru g/l	1.06	1.7
Ag g/l	0.03	0.05
U g/l	0.9	1.3
Pu mg/l	40	91
Rb g/l	0.18	0.28
Y g/l	0.24	0.37
La g/l	0.66	1.02
Pr g/l	0.63	0.96
Sm g/l	0.47	0.72
Eu g/l	0.09	0.13
Gd g/l	0.07	0.11
Tc g/l	0.44	0.68
Rh g/l	0.2	0.31
Pd g/l	0.75	1.16
Cd g/l	0.05	0.07
Sn g/l	0.03	0.04
Sb g/l	0.006	0.009
Te g/l	0.3	0.46

Table II Complete Data of the LEWC-content based on Measurements in 1979/80

VITRIFICATION PLANT

Figure 1 gives a schematic overview of the PAMELA Vitrification Plant by subdividing the plant into three main groups.

These groups are:

- production of glass canisters
- secondary waste treatment
- utilities

Table III shows the main data of the PAMELA Process.

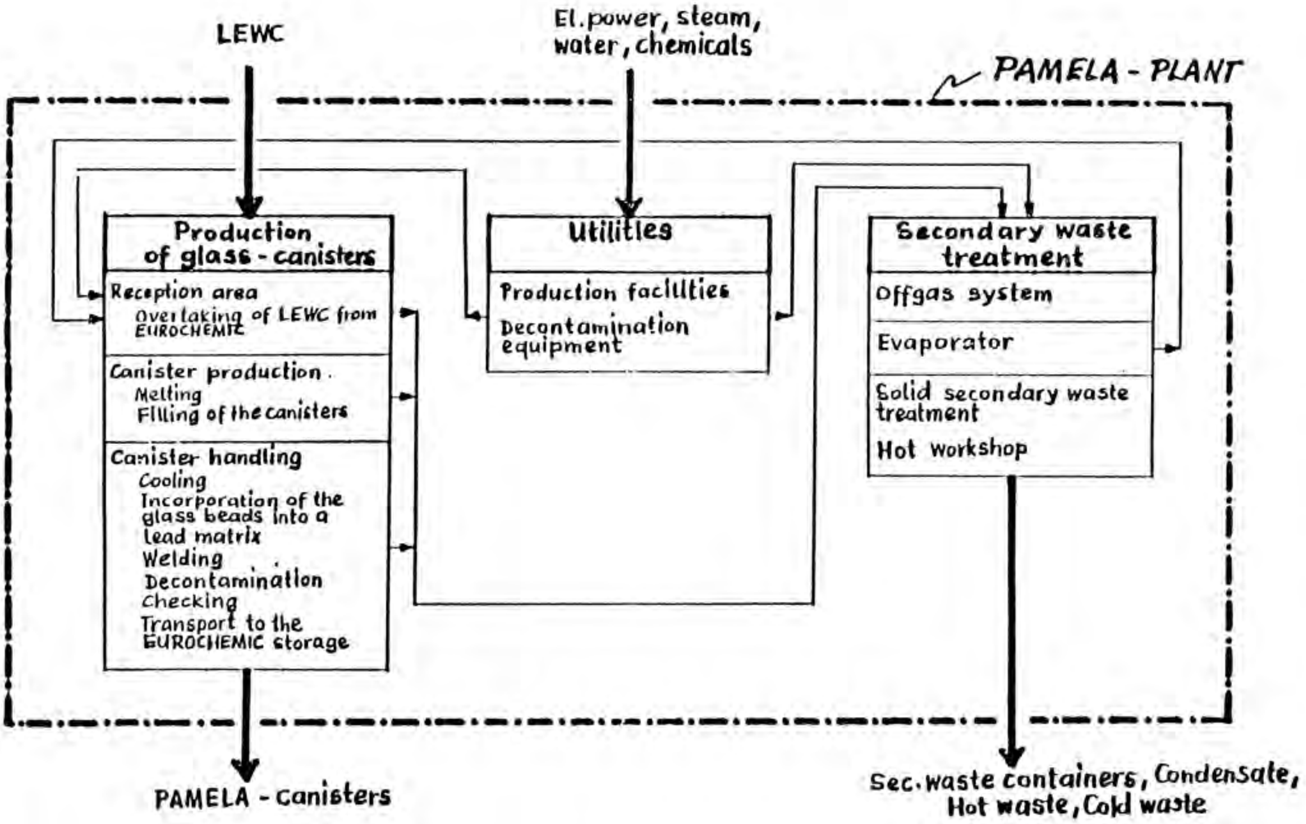


Fig. 1. Schematic Overview of the PAMELA-plant.

Table III. Main Data of the PAMELA-process.

Amount of LEWC	59 m^3
Batch transfer Volume	1,85 m^3
Number of batch transfers	32
Steamjet dilution per batch	8 percent (equal about 150 liter)
Cleaning solution per batch	1 m^3
Total volume received at the Pamela-Plant, per batch	3 m^3
Vitrification time per batch	100 hours
Continuous feed-flow to the melter	30 liter per hour (30 liter per hour feed, equal about 20 liter per hour LEWC-Feed)
Volume reduction: LEWC/glass	2
Glassvolume produced (Min/Max.)	30/67 m^3
Effective hot operation time of the Pamela-Plant	minimum 3200 hours
Glassblock production	discontinuous one canister per 6 hours
Vitromet production	continuous one canister per 3 hours and 20 minutes
Canister dimensions	total height 1200 mm
	outside diameter 298,5 mm
	wall thickness 8 mm
	usable volume 55 liter for the glassblock canister, for filling with 53 liter glass
	45 liter for the VITROMET canister, for filling with 30 liter glass beads

Production of Glass Canisters

Reception

The LEWC stored in two existing storage tanks (27 m^3 and 32 m^3 volume) is transferred batchwise by steamjet to a reception vessel in the PAMELA-Building. Jet steam and rinsing water dilute the LEWC to about 3 m^3/batch. One batch is treated in the PAMELA-Plant within a period of 100 hours.

Canister Production (Fig. 2)

From the reception vessel the LEWC is air-lifted to a mixing vessel. There, glass powder is added batchwise to the LEWC. The feed is homogenized by an air pulser. The homogenized feed is transferred continuously to the melter by a dosing airlift. In the inlet nozzle, additional glassfrit is mixed to the feed slurry.

In the melter the following processes take place concurrently:

- evaporation of the water content in the feed
- denitration of the feed solution
- calcination of the LEWC components to oxides
- melting of the added glassfrit
- incorporation of the waste oxides into the glassmelt at a temperature of about 1150°C

The construction of the melter is shown in Fig. 3.

The melter itself basically consitutes several layers of ceramic material encased in a stainless steel housing.

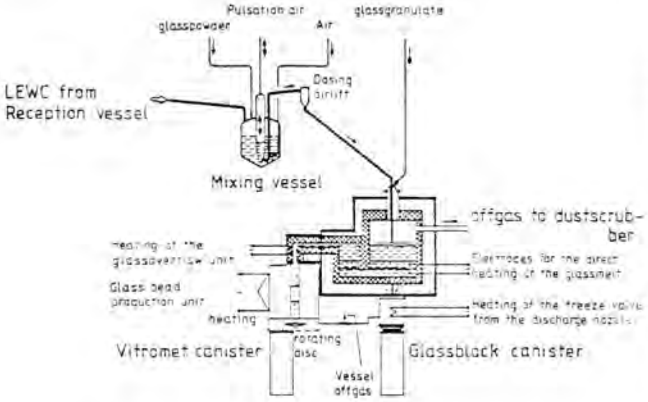


Fig. 2 Melter with Dosing System

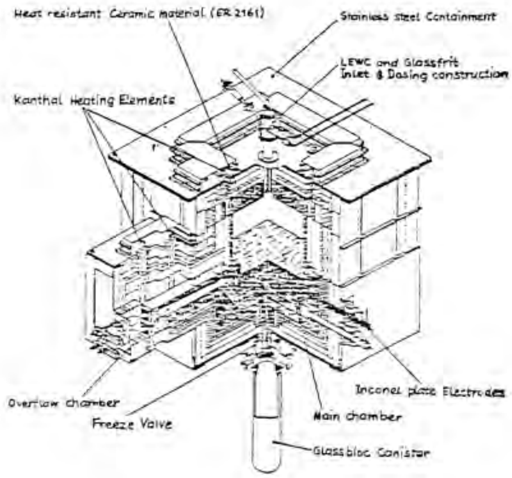


Fig. 3 Isometric drawing of the Ceramic Melter

The ceramic layer in direct contact with the glassmelt is a high temperature resistant ceramic (ER 2161). It is surrounded by a ceramic insulation, which is retained by the stainless steel casing. This casing serves as an airtight containment of the melter.

The melter is divided into two areas:

- the glass production zone or main chamber
- the overflow or secondary chamber

The main chamber has a surface of 0.75 m^2 and has a normal operating volume of 310 liters when the height of the glassmelt is about 0.4 m.

The main chamber is heated by a direct heating system. For this purpose, four pairs of plate type electrodes (Inconel 690) are installed at both sides of this chamber. The installed electric power for this resistance heating is 150 kW. The power density in the submerged electrodes is about 0,8 A/cm^2.

The dome of the melter is equipped with 12 heating elements (Kanthal) with a max. power consumption of 60 kW. They are used to heat up the melter to a temperature of about 750°C before starting up the melting process. Finally they are used to reheat the cooled glass, once the melter was out of operation. At the temperature of 750°C the above mentioned electrode heating system can take over and the heating elements in the dome are shut off.

For the filling of the canisters, two different discharge methods are provided:

- production of glass block canisters by means of a bottom drain valve, which is operated discontinuously.
- production of glass bead canisters (VITROMET) by means of an overflow outlet, which is operated continuously

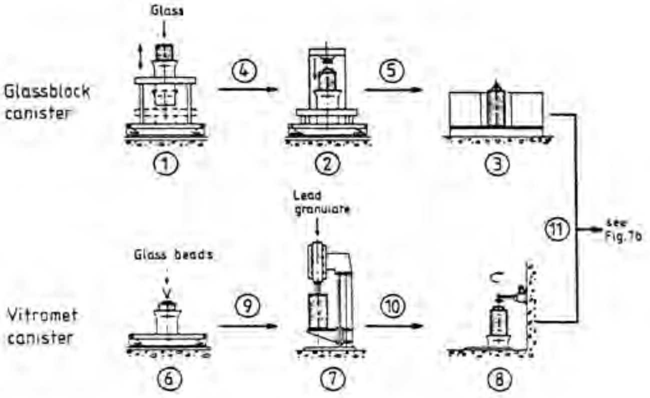
The <u>bottom drain system</u> is operated as a freeze valve, using two totally independent heating systems: an electrical direct heating of the glass in the ceramic part, and an electrical induction heating of the metal part of this drain system. The glass discharge can only take place when both heating systems are switched on. To stop the glassflow (60 liter per hour), after filling the canister, both heating systems are switched off.

The overflow outlet is operated by heating the overflow chamber, where two independent heating systems are installed: the first system consists of three Kanthal heating elements, which heat the ceramic part of the overflow chamber, the second consists of two Inconel plate electrodes, which heat the glassmelt in the overflow directly. Each system itself is capable of heating the overflow system up to temperature of 1200°C.

The glass which flows through the overflow system is collected inside the bead production unit, which is located underneath the overflow system. Caused by an overpressure the glass flows through 10 discharge *nozzles* and falls in form of glass beads on a rotating disc, which is kept at a temperature of about 300°C. By means of a scraper the beads get into a buffer vessel and from here they fall through a discharge pipe into the VITROMET Canister (13.5 liters per hour).

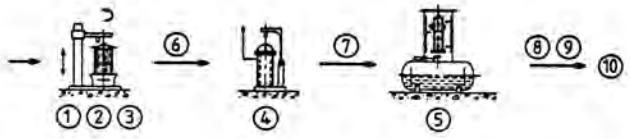
Canister Handling (Figs. 4, 5, 6)

The main steps of the canister handling after filling with glassmelt respectively with glass beads are shown in Figs. 4,5,6.



1. Filling of the glasscanister through the bottom discharge system
2. Closing of the glassblock canister at a temperature of 600°C
3. Cooling of the glassblock canister from 600°C to 50°C
4. Transport by trolley
5. Transport by trolley and crane
6. Filling of the Vitromet canister with glass beads through the overflow system
7. Incorporation of the glass beads into a lead matrix
8. Closing of the Vitromet canister
9. Transport by trolley and crane
10. Transport by crane
11. Transport by crane

Fig. 4 Canister Handling



1. Welding of the cover to the canister
2. Filling of the canister with helium
3. Closing of the mushroom extension on the canister cover
4. Helium leak test
5. Canister decontamination
6. Transport by crane
7. Transport by crane
8. Transport by crane
9. Contamination control
10. Transport to the Eurochemic storage facility

Fig. 5 Canister Handling

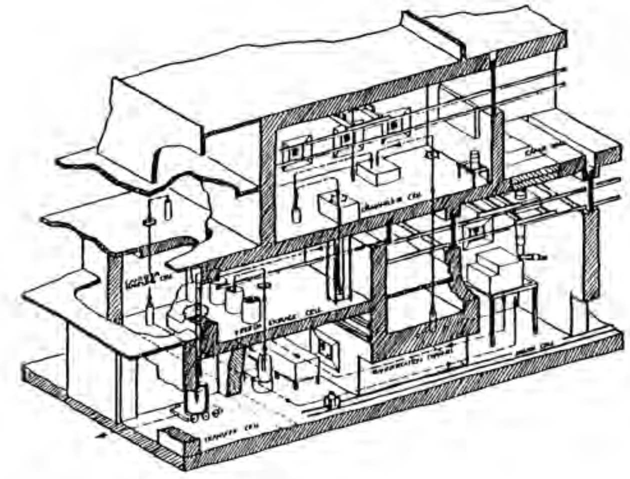


Fig. 6 Canister Handling

Secondary Waste Treatment

Offgas Treatment System

The offgas from the melter is first washed in a dust scrubber, then cooled in a condensor and again washed in a jet scrubber.

Between condensor and jet scrubber the vessel offgas is added to the stream and from here both gas streams are treated together. From the jet scrubber the gas flows through a NO_x-Absorber, two HEPA-Filters, a cooler and a demister. Via a draft fan, the offgas is discharged to the atmosphere through the existing Eurochemic-Stack. The design criteria for this offgas treatment system are:

- 4% of the salts which are added to the melter are entrained with the offgas as calcine
- more than 1000 vpm NO_x are expected in the offgas (the discharge limit is 1000 vpm NO_x)

The question of RuO_4 in the offgas has been greatly discussed. Recent experiments show that only 3 - 4% of the Ru show up in the offgas as RuO_4, the rest being RuO_2. Therefore, scrubbing is considered to be sufficient for the Ru-removal and additional Ru-filters, such as silicagel, have been discarded.

Liquid and Solid Waste

All liquid process effluents are collected and treated by evaporation.

For solid waste, a dismantling cell is provided. The dismantling cell is equipped with necessary tools to cut up even large pieces of process equipment such as the ceramic melter.