

VITRIFICATION OF HIGH LEVEL RADIOACTIVE WASTE - OPERATING EXPERIENCE WITH THE PAMELA PLANT

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ABSTRACT

The construction of the PAMELA vitrification plant at the EUROCHEMIC site in Mol/Belgium was completed in late 1984. After one year of cold test campaigns with simulated waste solution the plant was put into radioactive operation in October 1985. The available operating experience is outlined for the general process technique, liquid-fed ceramic glass melter, off-gas treatment, canister handling, and remote maintenance in the vitrification cell. Production data, decontamination factors, and activity balances are presented. Finally, the schedule for future vitrification campaigns are given.

INTRODUCTION AND OBJECTIVE OF THE PAMELA PLANT

The PAMELA vitrification plant, based on a liquid-fed ceramic melter process, is located at the EUROCHEMIC site in Mol/Belgium. It was constructed from 1981 to 1984 by DWK (Deutsche Gesellschaft für Wiederaufarbeitung von Kernbrennstoffen) with financial support of BMFT (Bundesministerium für Forschung und Technologie). The facility serves primarily as a pilot demonstration plant while vitrifying EUROCHEMIC's high-level liquid waste which accumulated during spent fuel reprocessing from 1966 to 1974. DWK is responsible for plant operations. The process building, shown in Fig. 1, is 58 m long, 30.4 m deep, and 25.5 m high. The PAMELA plant began radioactive operation on October 1, 1985.



Fig. 1. Process building of the PAMELA vitrification plant

There are two types of high level waste solutions stored at the EUROCHEMIC site to be vitrified: approximately 50 m³ of LEWC and 800 m³ of HEWC. The LEWC (Low Enriched Waste Concentrate) contains significant amounts of process chemicals, especially sodium, in addition to the actinides and fission products. The HEWC (High Enriched Waste Concentrate), however, is primarily an aluminium nitrate solution with about one tenth the radioactivity of the LEWC.

The composition and characteristic data for LEWC, the current PAMELA feed, are presented in Fig. 2. The LEWC

contains a significant amount of sulphur. Because of the limited solubility of sulfates in glass, the sulphur concentration dictates the waste glass loading when the LEWC-solution is vitrified. The glass production is controlled so that the waste glass contains 11 wt% of LEWC-oxides. This waste loading level sufficiently suppresses the tendency of the sulfate to separate and accumulate as a sodium salt phase on top of the molten glass pool.

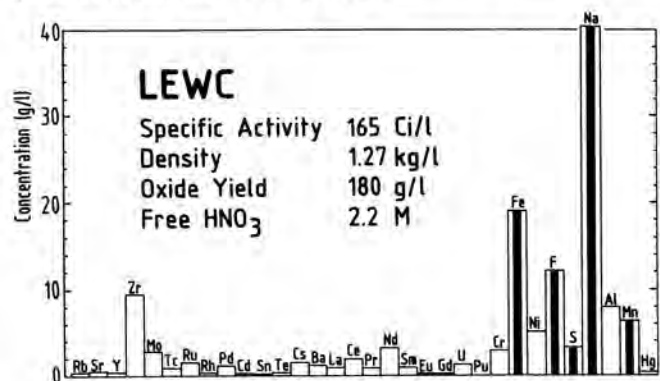


Fig. 2. Composition and some characteristic data of the LEWC-solution currently vitrified in the PAMELA plant (LEWC sample from storage tank 253-1b)

As glass making material, glass frit is used in the PAMELA process. The current glass frit has low sodium oxide content to adjust for the high sodium concentration in the LEWC-solution. The composition of the glass frit and the LEWC glass product are shown in Tab. I together with the data for viscosity and specific electrical resistivity. The design throughput of the PAMELA plant for LEWC vitrification is 30 kg/h waste glass production (4.8 glass canisters/day) corresponding to a feed rate of 22 l/h.

According to the objectives of the PAMELA-project, two different types of final waste glass forms will be produced. Approximately 80 % of the total LEWC waste glass production will be glass blocks in stainless steel canisters, the remainder will be a demonstration of the VITROMET process (glass marbles incorporated in a metallic matrix). The glass canisters and VITROMET have the same outside configuration and container dimension (300 mm in dia., 1200 mm high). The glass canisters contain an average of 150 kg of waste glass, the VITROMET canisters 100 kg. The canisters are stored in a separate storage building near the PAMELA process facility. Interim storage is envisioned to last about 50 years. Final storage is planned to be carried

TABLE I

Composition of the glass frit and the LEWC waste glass produced in the PAMELA plant

Constituent	Glass frit wt. %	Waste glass wt. %
SiO ₂	58.6	52.2
B ₂ O ₃	14.7	13.1
Al ₂ O ₃	3.0	2.7
Li ₂ O	4.7	4.2
Na ₂ O	6.5	5.8 ¹⁾
MgO	2.3	2.0
CaO	5.1	4.5
TiO ₂	5.1	4.5
LEWC-oxides	-	11.0
Viscosity, 1150°C (dPas)	83	50
Electr. resistivity, 1150°C (Ω cm)	5.3	4.3

1) Total Na₂O in the waste glass is 9.1 %

out in a future geological repository in the Federal Republic of Germany.

This paper discusses the process techniques applied in PAMELA, describes the hot operation experiences, and presents results available from the recent radioactive campaigns.

PAMELA PROCESS TECHNIQUE

Building Description

The PAMELA plant is an independent unit equipped with all its own support facilities. The integral volume of the processing building is 32,450 m³ including the shielded area of 4,250 m³, in which the process and handling equipment are installed. The shielded area is constructed from reinforced concrete. The thickness of the walls and ceilings in this area are determined by shielding requirements. The maximum thickness is 1.1 m. The outer portion of the building is of standard construction using brick and concrete walls on a structural steel framework. The location of the process cells and service areas are shown in Fig. 3.

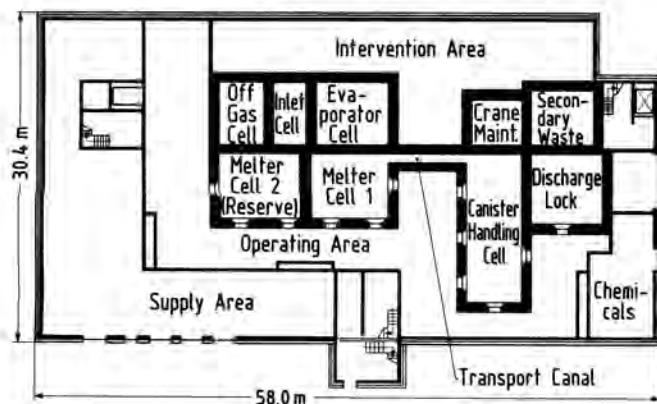


Fig. 3. Ground floor plan PAMELA

In the regulated areas of the processing building, the following materials are used:

- A complete lining of stainless steel where there is a high risk of contamination,
- Stainless steel trays and epoxy-resin coatings for walls and floors where there is a low risk of contamination,
- Epoxy-resin coatings and paint on walls, ceilings and floors in the other areas.

Process Flow Sheet

The basic flow sheet of the vitrification process is shown in Fig. 4. LEWC waste solution and small glass beads of borosilicate glass are fed directly into the melter. On the surface of the molten glass, excess water and free nitric acid are evaporated. The remaining salts are calcined to oxides and incorporated into the glass melt at about 1150°C. The off-gas from the melter is treated by an extensive off-gas system.

Glass is removed from the melter through a bottom drain into canisters (glass blocks) or through an overflow drain to produce glass beads, which in an additional process step are imbedded in molten lead (VITROMET). After filling, the canisters containing glass blocks are transported to a cooling station. After a cooling time of about 2.5 days, the cover lid is welded to the canister. The canisters are then decontaminated in an ultrasonic bath of dilute nitric acid and rinsed with water. A smear test for contamination is made before they leave the building. A shielded canister transporter is used to move two glass containers at a time to the above ground intermediate storage building.

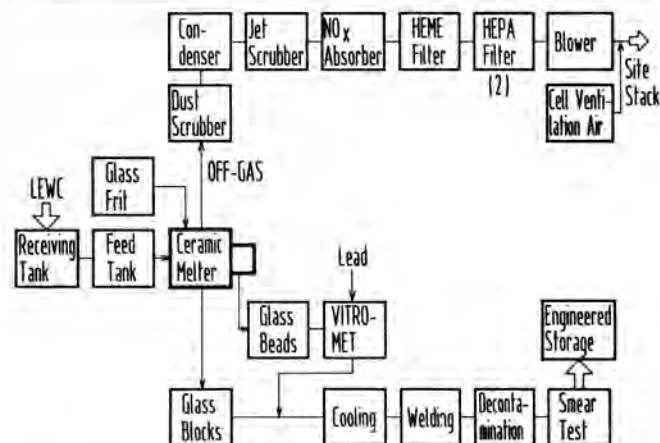


Fig. 4. Basic flow sheet of the PAMELA vitrification process (recycle streams not shown)

Figure 5 shows more detailed the flow, activity, and solids concentration for each of the major process flows including the recycle streams.

DETAILED PROCESS DESCRIPTION

Feed Systems

There are two main process feeds, the LEWC liquid waste stream and glass frit. The LEWC is transferred batchwise from EUROCHEMIC's storage tanks by stream-jets through a shielded overhead pipe line into the PAMELA 3 m³ receiving tank. After each transfer, of approximately 1.5 m³, the tank is agitated with air and sampled for solids

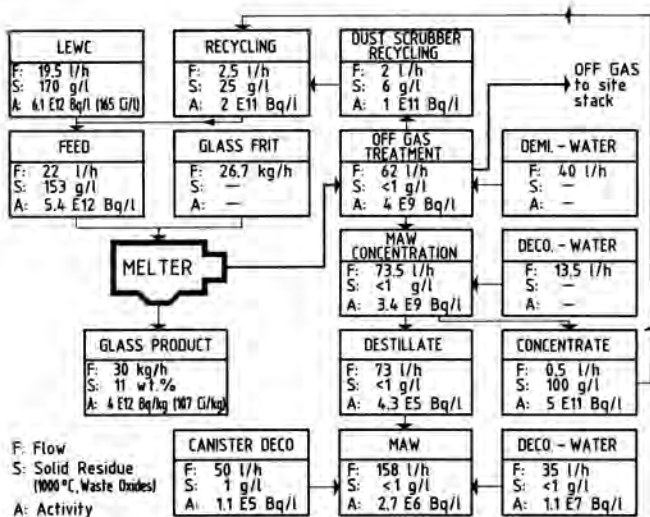


Fig. 5. Summary flows for LEWC waste vitrification including the recycle streams and activity.

The receiving tank material is batch transferred to the feed tank, by a two stage air-lift system, in approximately 10 liter increments. Then it is continuously fed to the melter using an adjustable rate air-lift system. The 1.5 mm diameter glass frit beads, received in 5 kg bags, are loaded into a solids handling system. This solids handling system automatically weights out and feeds six equal increments of glass frit for each 10 liter transfer of LEWC into the process. Based on the LEWC analyses, the weight of the frit additions are adjusted so that the desired oxide loading is obtained in the final glass product.

The major component of the PAMELA process is a joule heated glass melter (Fig. 6). It has outside dimensions of 2 x 2 x 2.6 m (l x d x h). The inside of the melter is lined with a ceramic refractory containing 30 % chromoxide which is resistant to corrosion from the molten borosilicate glass. The melter internal dimensions are 0.88 x 0.88 x 1.16 m (l x d x h) which provides 0.77 m² of glass pool surface area. The normal glass pool operating depth is 0.4 m and contains approximately 310 liters.

Melter

The joule heating for glass melting is provided by four pair of power electrodes, two upper and two lower made from Inconel 690. These electrodes which are below the glass melt line are air cooled to keep the electrode temperatures below 1120°C which reduces electrode corrosion. The electrical input to the melter is controlled by fixing the current to the power electrodes. The power consumption of the melter is then dependent on the resistivity of the glass. For molten glass the resistivity varies inversely with the glass temperature. At maximum rates, the power consumption is 55 KW on the upper pair of electrodes and 20 KW on the lower pair of electrodes.

Since the melter is performing three major functions, evaporation, calcining and glass melting, it is important to know the internal melter chamber temperature at various points. Based on previous experience, four thermocouples, placed in Inconel-690 tubes, are installed through the melter cover to various depths which measure the plenum, calcination and molten glass zones for the normal melter level ranges. These temperature profiles, along with the power consumption and mass balances then provide an understanding of the melter performance and determine the need for changing feed rates or for removing glass product.

In the production of glass blocks, glass product is drained to a canister batchwise. When the glass level in the melter (based on temperature readings) reaches a predetermined quantity, 150 kg or about 60 liters are removed from the melter. This is accomplished through a bottom drain valve which is normally full of solid glass. To establish flow, induction heat is provided to the bottom drain valve from a water cooled medium-frequency heating coil and then additional heating is provided by passing current from the bottom drain tube to the bottom electrodes. The initial fill rate is about 150 kg/hr and averages about 120 kg/hr. The total time from start of heating to a full canister is about 2 hours. The weight of glass in the canister is determined by a strain gage balance located on the canister lifting carriage and these readings are integrated periodically to determine the fill rate. Two gamma detectors are installed to measure the glass level in the canister to confirm the present weighing system. The upper detector is set for maximum fill level and the lower detector is 10 cm below maximum fill.

In the production of glass beads for VITROMET, a continuous overflow system is used. The rate of overflow is controlled by use of a nitrogen fed airlift located in the melter riser. The bead production unit consists of an Inconel 690 pot, heated by a resistance furnace to 1100 °C which divides the continuous glass stream from the melter into drops of glass. These droplets of glass fall on to a rotating turntable where they solidify during one revolution. A scraper transports the beads to a hole in the center of the turntable, where they fall into the vitromet-container. During the time when the container is changed, the continuous stream of glass beads is temporarily collected in a buffer. After cooling down, the void volume between the beads is filled with lead by heating the container to about 400 °C with an induction coil. The rest of the container handling is the same as normal glass block canister.

The melter is operated at a slight vacuum.

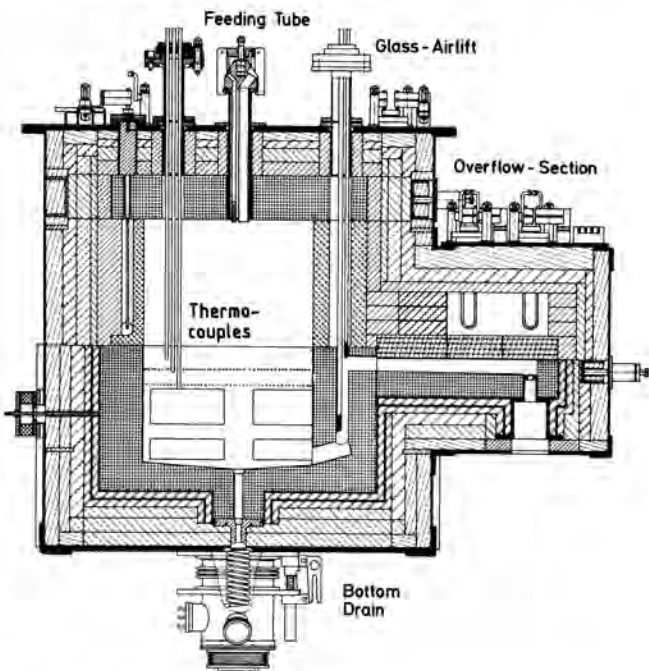


Fig. 6. Joule-heated ceramic waste glass melter

For melter start-up, fifteen auxiliary MoSi₂-heating elements are installed behind the ceramic walls of the melter plenum. The mechanical stability and tightness of the melter are provided by the outer stainless-steel shell. The melter is operated at a slight vacuum.

Off-gas Treatment

The melter off-gas is treated by a series of process equipment as shown in Fig. 7. The initial treatment is in the dust scrubber where the larger particulates are removed. It consists of a pot with a 204 mm diameter column which contains 9 plates on a 175 mm plate spacing. The scrub-solution, held at 80 - 85 °C by an electric heater, is circulated counter-current to the gas flow by a submerged airlift. This temperature is maintained to prevent condensing the water vapor. The solids removed in the scrub-solution are periodically transferred back to the melter feed tank. The solids which deposit in the off-gas tube between the melter and dust scrubber are periodically pushed back into the melter by a mechanical cleaning system. During the cleaning procedure waste feeding into the melter need not to be interrupted.

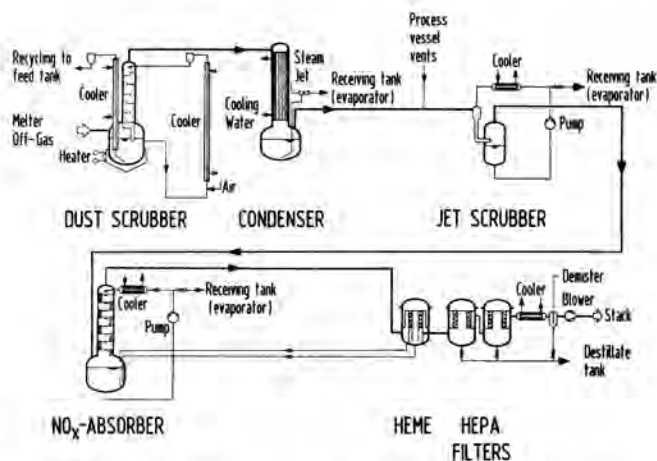


Fig. 7. Off-gas treatment in the PAMELA plant

The off-gas leaving the dust scrubber goes to a condenser where cooling water at 30 °C is used as the condensing media. The condenser is of two part construction; the upper section a tube bundle for condensation and the lower section a vapor disengaging and liquid collection vessel. The condenser contact surface is sufficient to produce an off-gas exit temperature of 40 °C. The condensate, which is an aqueous nitric acid solution, is periodically transferred to the secondary waste evaporation system.

The off-gas leaving the condenser, consisting mainly of NO_x, small particulates and aerosols, is combined with the vents from the radioactive tanks. This mixture is fed to a jet scrubber where it is contacted by a recirculating fluid stream operating at a pressure of 5 bar. The recirculating scrub-solution energy is provided by a pump. Periodic analysis of the scrub solution is made and it is replaced with fresh water when the activity reaches about 0.5 Ci/l. The used scrub solution is sent to the secondary waste evaporation system.

Next the stream is treated to remove NO_x. This is accomplished by passing the off-gas stream through a scrubbing column which has 8 valve trays. The scrubbing media is a 5 to 6 molar nitric acid solution which is circulated by pump from the base of the column to the top tray. This solution is maintained at 15 °C by a cooling water coil to cool the gas and remove the heat of solution. Because of the low solubility of NO, a 0.2 molar hydrogen peroxide solution is added to the scrubber to oxidize the NO to NO₂ which is more soluble, resulting in lower NO_x discharges to the stack. The nitric acid scrub solution is periodically transferred to the secondary waste evaporation system.

The off-gas at this point of the process is essentially air, nitric acid aerosols and fine particulates. The stream is then treated in a fiber glass filter where the nitric acid aerosols are removed. This system acts similar to a wetted filter trapping radioactive particulates in the coalesced aerosols.

The off-gas is then heated to 70 °C to insure that it is above its dew point and passed through two HEPA filters in series to minimize the radioactive discharge. The gas is cooled to 30 °C, sent through an impingement and to the final off-gas blower which discharge it to the stack.

CANISTER HANDLING

The canister used at PAMELA is a stainless steel container shown in Fig. 8. At its nominal fill level of 85 %, it holds 150 kg of waste glass (approximately 60 liters). The canisters are designed so that they may be stacked on top of each other. During storage in the Engineered storage building, the filled canisters will be stacked 5 high.

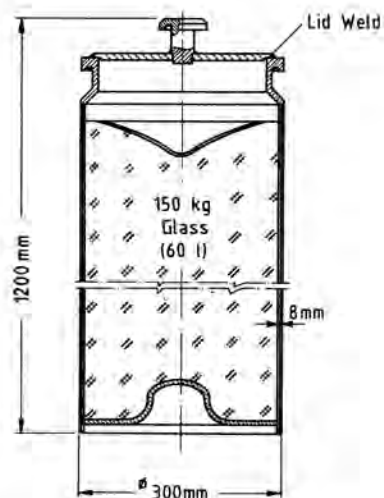


Fig. 8. PAMELA waste glass canister

Special tools were developed to handle the canisters and their overpacks. The melter cell, canister handling cell and smear-test area are designed so that all normal canister transport operations can be carried out using the cranes, power and master-slave manipulator in these cells. The major steps in the canister handling procedure are illustrated in Fig. 9.

New canisters are brought by crane into the handling cell where they are put into heat-insulating overpacks. When a canister is needed in the melter cell, the handling cell crane positions the canister and its overpack in the canister transporter. The canister and its overpack are transported through a tunnel into the melter cell. There they are moved to the canister lifting carriage. This carriage is driven along a rail to position the canister directly underneath the bottom fill valve (or the turntable for Vitromet production). It then lifts the canister up to the bottom valve bellows for filling. The carriage has a strain gage system to determine canister content during filling. After filling, the canister and its overpack are returned to the handling cell for cooling. Here it is allowed to cool for 2 1/2 days (about 60 °C). This slow cooling minimizes glass cracking. It is then removed from the overpack, taken to the welding station and the cover is welded.

Welding is by a TIG welder. Quality of weld is assured by controlling the welding parameters, visual inspection of

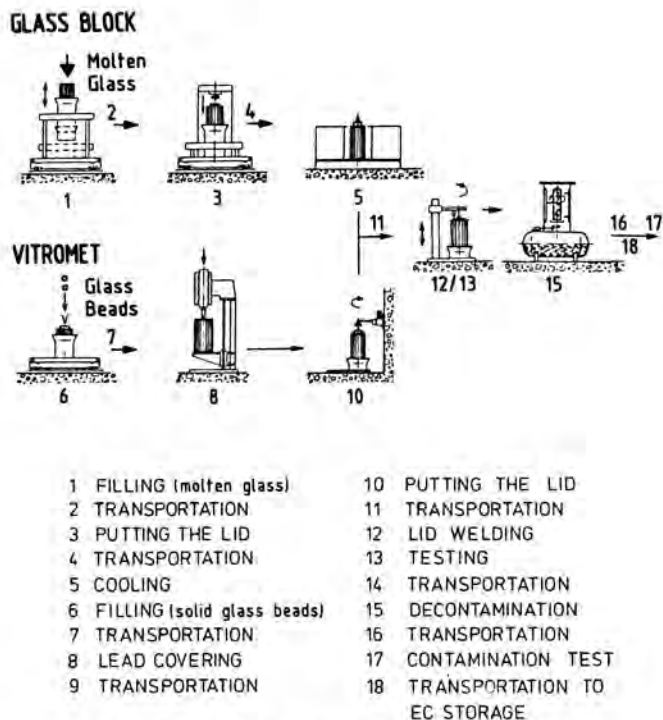


Fig. 9. Canister handling procedure in the PAMELA plant

the weld, and every 50th weld is made on an empty canister which is leak tested.

The welded canister is decontaminated in an ultra-sonic bath containing 1 molar nitric acid. It is then moved to a separate chamber where the canister is rinsed with fresh water prior to lifting it into the smear-test cell located above the decontamination station. The canister is smear tested on the top, bottom and side for contamination. If the surface contamination is less than 10^{-7} Ci/m², the canister is lowered by crane into the shielded transporter for delivery to the Engineered storage building. If the canister fails the smear test, it is returned to the ultra sonic bath for further decontamination.

Process Control

Control of the PAMELA process is accomplished by an automated process control system which allows the operator to control or monitor the majority of the process and its support systems from a central control room. It consists of two tables, each equipped with three high resolution 19" color computer monitors. They are supported by printer systems and a programmer's desk and console. The operator input into the computer is done through a light pen pointed at the desired function on the monitor. Most of the mechanical operations (e.g. transfer of canisters from cell to cell) are controlled from local panel boards situated near viewing windows.

Another important aspect of the PAMELA process control is the analytical laboratory. This facility is located inside the processing building for analyzing the different process streams to provide data for process control and monitoring the emissions in the off-gas and other waste streams. The sampling system is automated with its sampling lines connected to various points in the process. Sampling is controlled from the laboratory although the computer monitors in the central control room indicate when sampling is in progress. As shown in Fig. 10, the samples are received first in the High Activity Lab where they are analyzed and

then diluted and transferred to the Low Activity Lab for additional and more detailed analyses. An inductive coupled plasma (ICP) spectrometer is used to determine the concentrations of 25 elements. A computer controlled gamma spectrometer and a 10 channel alpha-beta low level counter are used for radioactivity measurements. Glass samples can also be taken in the melter cell and pneumatically transferred to the lab for analyses.

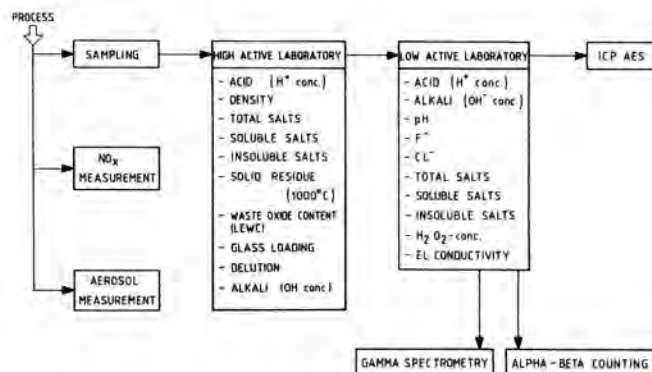


Fig. 10. Scheme of the analytical laboratory

Maintenance Procedures

A photograph of the melter cell is given in Fig. 11. The equipment located in this cell including the melter, the feed tank and dust scrubber are designed for remote replacement. Equipment in other areas is repaired by direct maintenance in an intervention cell if it is not severely contaminated or by removing contaminated or radioactive equipment prior to cell entry.

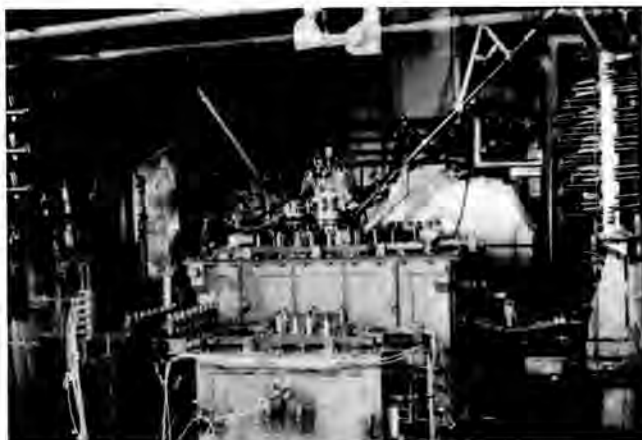


Fig. 11. PAMELA melter cell

Plant Organization

The PAMELA plant organization is shown in Fig. 12. In total 53 people provide the direct management and operating support for the process.

Other site support functions such as guards, medical, low level waste treatment, canister handling and storage beyond the confines of the main process building are provided by the current site landlord, Belgoprocess (formerly EUROCHEMIC).

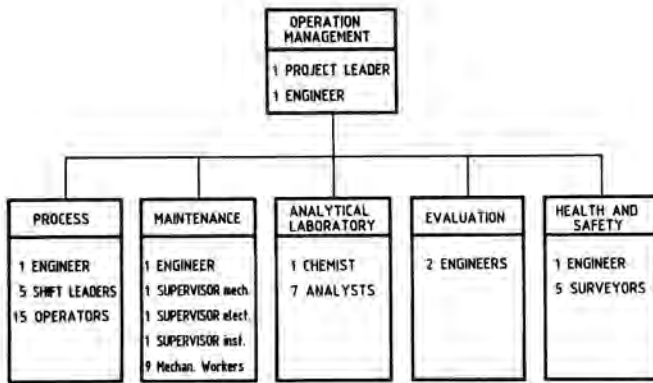


Fig. 12. PAMELA plant organization

RESULTS FROM RADIOACTIVE CAMPAIGNS

The PAMELA plant started hot operations after an extensive cold run period. Prior to hot start up a final test run was performed using a mix of 100 l actual LEWC waste (10,000 curies) and 2400 l simulated LEWC to confirm the decontamination factors of the off-gas treatment. Also remote replacement of the melter and other equipments of the melter cell was demonstrated before the hot operating campaigns began.

Three hot operation campaigns have been scheduled to vitrify the 50 m³ of L E W C waste:

- 1st campaign from October 1 to December 4, 1985 (glass block production)
- 2nd campaign from January 13 to beginning of March 1986 (glass block production)
- 3th campaign from April 1 to end of April 1986 (Vitromet production)

The production results for the first campaign and estimated values for the second campaign, now in progress, are shown in Table II.

First Campaign

In the first LEWC processing campaign 22.7 m³ of LEWC were processed into 30.6 metric tons of glass. The loading of waste oxide into the glass product was 8.6 weight % and was lower than the goal rate. This was because of the higher than expected sodium content in the LEWC solution from storage tank 253 - 1a. To maintain the final glass product sodium oxide at 9.5 wt% the LEWC to glass frit ratio had to be adjusted. Equipment availability, as measured by the time the system was actually processing waste oxides into glass, was 75 %. A two week outage to replace the MF-coil of the bottom drain system was the most significant outage. Other than this the available time was 96 %. The need to replace the water cooled medium-frequency heating coil was caused by a slight untightness of one of the coil connectors.

The total β-gross activity processed in the first campaign was 8.65 E16 Bq (2.34 E6 Ci). A balance for the β-gross activity is shown in Fig. 13. More than 99.89 % of the activity went into the glass product. Most of the remainder went into the lower activity waste streams that will be further processed on the site. Off gas emissions were less than 5.18 E2 Bq (1.4 E-5 Ci). This very low emission rate is

TABLE II

Production results for the 1st radioactive campaign of the PAMELA and estimated values expected for the 2nd campaign

Parameter	1 st Campaign (carried out)	2 nd Campaign (in progress)
Total campaign time	1560 h	-
Feeding LEWC waste	1163 h	-
Down time	397 h	-
Equipment availability	75 %	-
LEWC Storage tank No.	253 - 1a	253 - 1b
LEWC concentration (oxides)	116 g/l	171 g/l
β-gross activity	100 Ci/l	165 Ci/l
Total glass production	30.6 mto	26 mto *)
Average glass rate	26.3 kg/h	-
Average feed rate (incl. recycling streams)	22 l/h	-
Waste glass loading	8.6 wt%	11 wt% *)
Total LEWC waste processed	22.7 m ³	16.7 m ³ *)
Total β-radioactivity processed	8.65 E16 Bq (2.34 E6 Ci)	1.02 E17 Bq *) (2.75 E6 Ci)
Number of waste glass canisters	199	170 *)

*) Goal value

the result of the good operation of the off-gas treatment system.

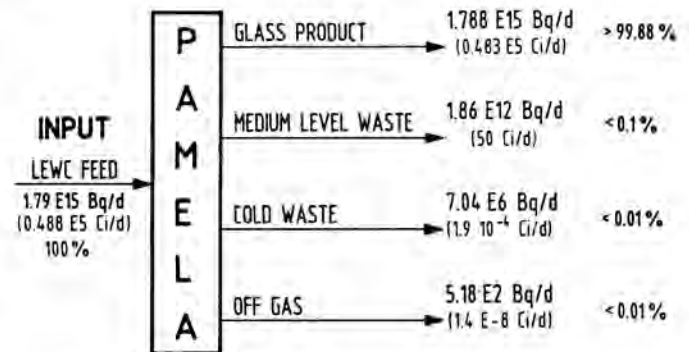


Fig. 13. Balance for the β-gross activity

Off-gas System Performance

Table III summarizes the results for the DF's for α, β, and Cs 137 activity. The jet scrubber proved to be very effective for removal of the remaining α- and β-activity which passed both the dust scrubber and condenser. Its DFs for α-activity, β-activity, and Cs 137 were measured to be more than 30. The glass fibre filter showed also high efficiency. The excellent results for the total feed to stack DFs, determined from a measuring filter in the off-gas stream before the stack, reflect better than forecast DFs for the jet scrubber and the glass fiber filter (HE ME).

TABLE III

Decontamination factors obtained for the first operating LEWC campaign

System	α -total	β -total	Cs 137
Melter	576	63.9	26.5
Wet off-gas treatment I (Dust scrubber, Con- denser, Jet scrubber)	78.5	63.7	61.3
Wet off-gas treatment II (NO _x -absorber)	-1)	25	-1)
Glass fiber filter (HEME)	-	2000	-
HEPA-filters (2)	-	> 2.5 E5	-

Total DF from single DFs	-	> 5.8 E13	-

Total DF feed to stack (direct measurement based on measuring filter before the stack)	1.6 E13	6 E14	6 E14

1) Values not available

The recovery of NO₃ for the process is shown in Fig. 14. About 90 % of the nitrate enters the process with the LEWC, the remaining is introduced as fresh acid primarily for the decontamination of canisters.

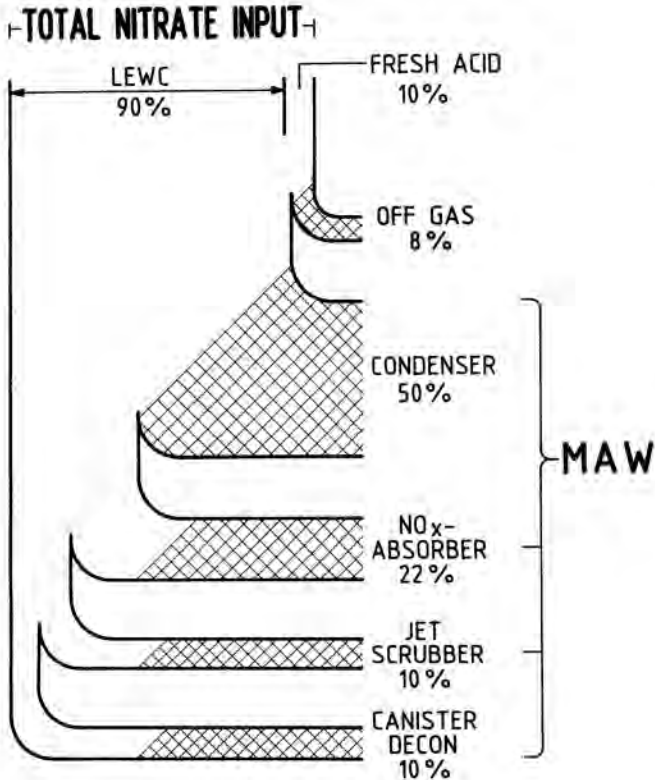


Fig. 14. Balance for the NO_x-removal

Approximately 92 % of the total nitrates are recovered and returned to EUROCHEMIC as medium-level waste which can be incorporated in Bitumen. This stream is generated

from the secondary waste evaporator which reduces the radioactivity of the streams received from the off-gas condenser, jet scrubber, and NO_x scrubber by distilling the nitric acid overhead.

Personnel Exposure Experience

Personnel at PAMELA wear dosimeters to measure their exposure to radiation. The average total dose during operations was about 80 mrem/week, that means an individual dose of less than 2 mrem/week/person. During maintenance periods, when interventions are made for repairs, the total dose was 120 mrem/week or an individual dose of 3.4 mrem/week/person. The average individual dose rate to date is 5 mrem/week/person with the maximum single individual having thus far received 120 mrem. Expressed on a production basis, 0.7 mrem of exposure was experienced per kilogram of glass production. Fig. 15 shows the total dose rate for different operating periods of PAMELA.

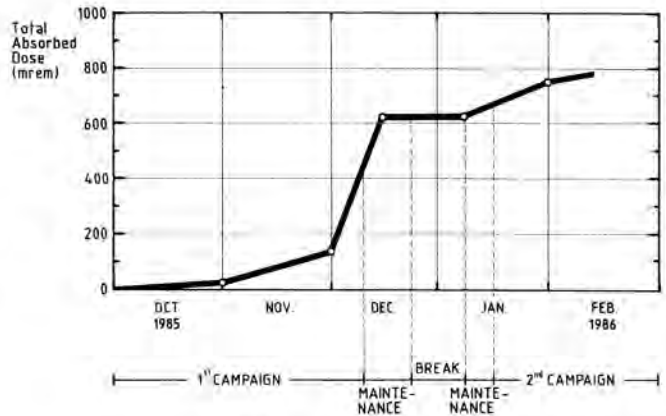
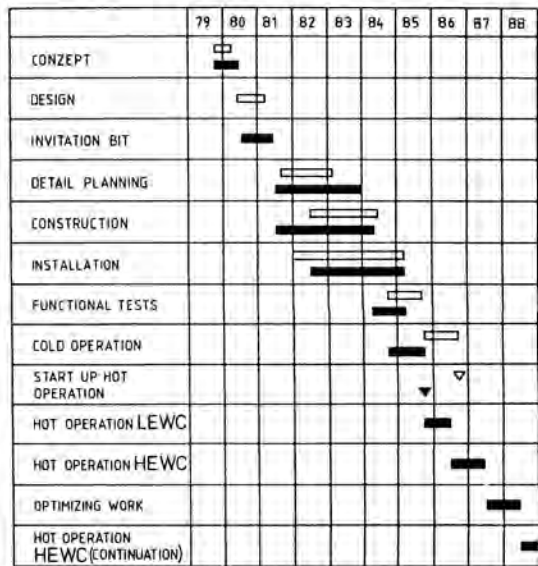


Fig. 15. Total dose rate for different operating periods of PAMELA

Overall PAMELA Project Schedule

PAMELA will complete the processing of the 50 m³ of LEWC by May, 1986. It will then be available to process the 800 m³ of HEWC presently stored on the site. Fig. 16 reviews the overall PAMELA project schedule.



□ Schedule from Oct 1979
■ State 1985

Fig. 16: Overall PAMELA project schedule

SUMMARY

The PAMELA plant has had a successful first hot operating campaign (the 2nd campaign is in progress)

- It has met its schedule for startup
- The feed system for LEWC waste and glass frit respectively performed well
- Stable melter operation has been achieved and controlled by the use of 4 control thermocouples. TV viewing of the melt pool, although desirable, has not been required
- The bottom drain system has been reliable and pouring can be stopped within 1 kg of the desired weight.
- A gamma system has been added to provide a second independent system to measure the glass level in the

canisters. The results show excellent agreement between the weight computed by gamma level detection and the strain gage weighing system

- The off-gas system has demonstrated decontamination factors equal or better than design goals.
- A technique has been developed to clean the off-gas line between the melter and dust scrubber during operation
- TIC-welding of the canister lid did not provide problems
- The canister decontamination is adequate to meet safe handling outside the process building
- The process equipment can be maintained without subjecting the employees to high dose rates.
- The PAMELA plant will be available, after LEWC vitrification is finished, to process the 800 m³ of HEWC stored on the site.