ADVANCED JOULE-HEATED NUCLEAR WASTE GLASS MELTERS

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ABSTRACT

This paper outlines the efforts made at INE to achieve substantial design improvements for a liquid-fed ceramic waste glass melter. Key points of melter improvements are given in comparison with the melter technology presently applied in the PAMELA plant at Mol/Belgium. Attention is drawn to design features which can extend melter lifetime, assure the noble metals-compatibility, and simplify the melter system. The advanced technology will be tested in a new mock-up facility presently under construction. The simulate to be used for the test campaigns corresponds to the composition of the high-level waste stored at the Karlsruhe Reprocessing Plant WAK.

INTRODUCTION

Joule-heated ceramic waste glass melters have been chosen for six high-level waste vitrification projects: In Belgium for the PAMELA plant (operated since 1985), in Japan for the Tokai Vitrification Plant and the vitrification facility at the future Shimokita reprocessing site, in the United States for Savannah River's DWPF-plant, the West Valley Project WVDP, and Hanford's future HWVP-plant. The Tokai plant and DWPF are scheduled for start-up in 1992, West Valley in 1993, and the other facilities towards the end of this decade. The basic technique has been world-wide developed in the past ten years. Its adaptability for a wide range of feed throughput requirements has been proved, beeing 20-30 l/h for the Tokai facility and PAMELA, and up to 240 l/h for DWPF. The process technique has been described in detail elsewhere (1,2,3). Comprehensive experiences and data have been gathered with a number of inactive test facilities operated at the various sites. Plant operational experiences are currently based on processing 550 m^3 of high-level liquid waste (HLLW) to 350 tons of waste glass by the PAMELA plant (4). Radioactive test results are also available at Pacific Northwest Laboratories (PNL/Richland) where 5 tons of glass has been produced within a special test program started in 1984 (5).

The detailed evaluation of the comprehensive process experiences provides an excellent base for substantial improvements of joule-heated melters, indispensable for processing of noble metals-rich HLLW from high burn-up fuel elements. At the Institut f r Nukleare Entsorgungstechnik (INE), a technology programm is to achieve this approach, i.e. to develop a progressed second generation of joule-heated glass melters compared to the INE-developed first melter generation currently used in PAMELA. The advanced melter technology can be applied to solidify the HLLW presently stored at the Karlsruhe Reprocessing Site WAK. This waste represents a prototypical commercial-like HLLW. The paper describes the key points of melter improvements including the mock-up facility used for testing.

DESCRIPTION OF THE NEW MOCK-UP FACILITY VA-WAK

The design and construction of a completely new mock-up facility designated VA-WAK was started early in 1988. A simplified flowsheet of this plant is given in Fig. 1. It was designed with a maximum feed throughput capacity of 30 l/h, thus corresponding in terms of production-scale with the PAMELA plant. The advanced waste glass melter integrated in this facility is designated K-6'. Construction of the whole mock-up is now nearly completed, and operation start-up is scheduled for May 1990.

Long-term runs will be performed using fully simulated waste including the noble metals Ruthenium, Rhodium, and Palladium. The actual composition of the existing high-level WAK-waste is given in Table I together with that of the simulate. The major characteristics of the waste are: a high specific radioactivity of 515 Ci/l (β/γ), a high sodium content, significant concentrations of the noble metals, and a total oxide yield of nearly 100 g/l. The waste glass loading will be 17 wt%, with nominally 1 wt% of noble metals in the glass. Glass frit development work for vitrification of this waste has been performed in recent years and will be continued for optimization, especially regarding waste-form qualification.

The glass melter K-6' is schematically illustrated in Fig. 2. Its outside dimensions are 3.1 m x 2.4 m x 2.7 m (LxDxH) with a total weight of 19 tons. The melter peripherie as well as the standards kept for material fabrication and melter construction allow the option of using this unit in the PAMELA plant. Although the outside dimensions were not changed compared to the present PAMELA melter, the glass pool surface area could be enlarged from 0.74 m^2 to 0.88 m^2. This is advantageous for improving the melter operability and the production rate, respectively.

For routine glass pouring an induction heated bottom drain valve is used. The melter is additionally equipped with a glass overflow system. The two pairs of main power electrodes are arranged in the upper part of the glass tank to release the major portion of the power needed for processing the waste and melting it together with glass frit at

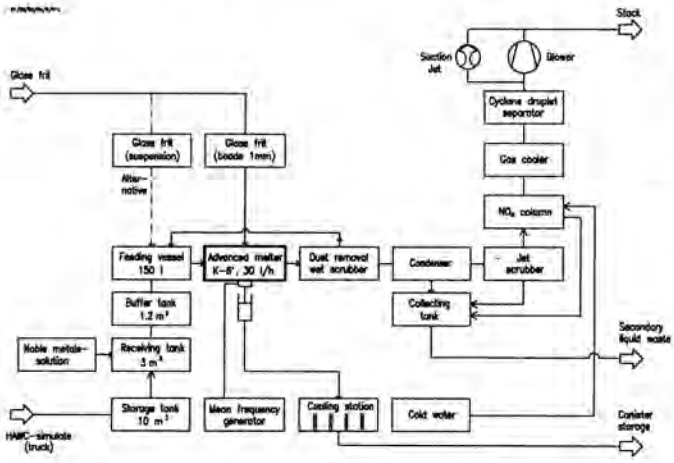

Fig. 1. Simplified Flowsheet of the New Mock-up Facility VA-WAK With the Advanced Waste Glass Melter K-6'.

1150°C. The lower part of the glass tank serves as a settling chamber to the bottom drain to collect precipitates like noble metals for removal with routine glass pouring. Power release in this chamber is just for maintaining glass temperatures between 1050-1100°C, which are sufficient for draining. For this purpose one pair of electrodes is used. It is arranged on opposite walls in the mid-section of the glass tank. An additional possibility for glass heating in the settling chamber is provided by a bottom electrode. It can be alternately fired against the two electrodes in the mid-section of the glass tank.

There were three primary areas of melter design where the efforts for improvements had been focused on. The first one was to achieve an extended service lifetime of the melter; the second was to assure its noble metals-compatibility; the third topic was the simplification of the melter system regarding melter construction, remote maintenance and decomissioning.

EXTENSION OF SERVICE LIFETIME

A large-scale radioactive melter represents a significant capital investment. It can also be an appreciable source of medium-and high-level solid waste when being decomissioned. Those reasons suggest that the overall service life of the unit should be as long as reasonably possible. Operational experiences at INE and PAMELA have indicated that the individual lifetimes of the power electrodes and the glass draining devices may primarily control the overall melter lifetime. Therefore it is most important to draw the attention to these key systems. A detailed evaluation analysis of failure events have been made, especially from PAMELA's long-term radioactive operation, to get the necessary design informations.

Bottom Drain Life Time Experiences From Radioactive Operation

The first melter of the PAMELA plant had to be replaced in 1988 after nearly three years of operation producing 220 tons of waste glass. At the middle of its service time, there were indications that a slight glass leakage had occured at the interface between the bottom drain valve and the surrounding ceramic refractories of the melter bottom. A thin glass film proceeded slowly along the surface of the upper part of the drain valve, as is schematically illustrated in Fig. 3. This part of the valve is firmly integrated into the melter bottom due to safety considerations. Thus the thick-walled Inconel 690 drain pipe is not remotely replaceable, in contrast to all other parts of the bottom drain system. The observed glass leakage was the reason to turn to the glass overflow system for routine glass pouring. The bottom drain was later used for emptying the melter before its replacement.

Electrode Life Time Experiences From Radioactive Operation

Some of the power electrodes suffered severe local corrosion attack. The electrode cooling air, directed to the cooling channels in the back-side of the electrodes, penetrated partly through the bulk glass melt. The observa-tion was indicative that openings were present in the thick-walled electrodes which connected the cooling channels with the glass-contacting electrode surface. The severe local corrosion could be explained by significantly overheated glass in the neighbourhood of the electrodes, which was caused by local excess current densities.

The effects were induced by the formation of a very conductive glass layer of some cm thickness at the flat melter bottom. It became enriched with settled noble metals precipitates. The situation is illustrated in Fig. 4, together with the verified electric potential and current field respectively. The ten times higher conductivity of the layer compared to the bulk glass caused nonuniform current distribution at the electrode surfaces. Areas of excess current densities are indicated in Fig. 4 by A, B and C. The electric field patterns were obtained by a semiquantitive procedure, using the rules for drawing electric potential and current fields (6) togehter with electric operational data of the PAMELA melter. Data of the electric properties of the bottom layer were gained by sampling, analysing, and preparing simulated samples with the same composition for subsequent measurements.

Despite the local damage of some of the power electrodes, and although their air cooling had to be switched off, melter operation could be safely continued, showing the ruggedness of the basic technology. The final decisive reason for the melter replacement was not a final failure of the

TABLE I

Composition of the Radioactive Reference Waste Solution HAWC-WAK and of the Simulate Used for Operation of the Mock-up Facility

Constituent	HAWC-WAK		Simulate
	Element, g/l	Oxide, g/l	Oxide, g/l
Major fission products			
34 Se	0.08	0.11	0.11
37 Rb	0.46	0.50	-
38 Sr	0.9	1.06	1.06
39 Y	0.7	0.89	0.89
40 Zr	6.2	8.38	8.38
42 Mo	4.6	6.90	6.90
43 Tc	1.0	1.57	-
44 Ru	3.0	3.95	3.95
45 Rh	0.83	1.02	-
46 Pd	1.8	2.07	2.07
47 Ag	0.1	0.11	-
48 Cd	0.07	0.08	-
52 Te	0.8	1.0	1.0
55 Cs	3.0	3.18	3.18
56 Ba	2.3	2.57	2.57
57 La	2.0	2.35	2.35
58 Ce	3.1	3.81	3.81
59 Pr	2.0	2.34	2.34
60 Nd	5.3	6.18	6.35
62 Sm	1.2	1.39	1.39
63 Eu	0.16	0.18	-
64 Gd	0.8	0.92	0.92
Actinides			
92 U	6.2	7.31	-
93 Np	0.3	0.34	-
94 Pu	0.2	0.23	-
95 Am	0.15	0.16	-
Corrosion products			
24 Cr	1.3	1.9	1.9
25 Mn	0.17	0.27	0.27
26 Fe	5.2	7.44	7.44
28 Ni	1.2	1.83	1.83
29 Cu	0.8	1.00	1.00
30 Zn	0.1	0.12	0.12
82 Pb	0.1	0.11	0.11
Process chemicals and others			
11 Na	16.0	21.6	21.6
12 Mg	0.2	0.33	0.33
13 Al	0.2	0.38	0.38
19 K	0.4	0.48	0.73
20 Ca	0.4	0.56	0.56
PO_4^{3-}	0.6	0.45	0.45
HNO_3	5.3 M	-	5.3 M
Total oxide yield	-	95.4	78.0
Radioactivity (β/γ)	515 Ci/l	-	-

power electrodes but a corroded wall section in the riser of the glass overflow system. Due to this corrosion the glass transfer rate of the glass-airlift had became too low for canister filling.

Based on the experiences described, design work were carried out to achieve progress in respect to longer service life of the bottom drain and the power electrodes. As a general objective an overall service lifetime of approximately 5 years is envisaged for a high-level waste glass melter.

Approach for Improving the Lifetime of the Bottom Drain

For achieving the corresponding long-term reliability of the bottom drain it is evident that an optimized integration of the drain valve into the melter bottom is needed. The design must assure tightness against glass-leak-

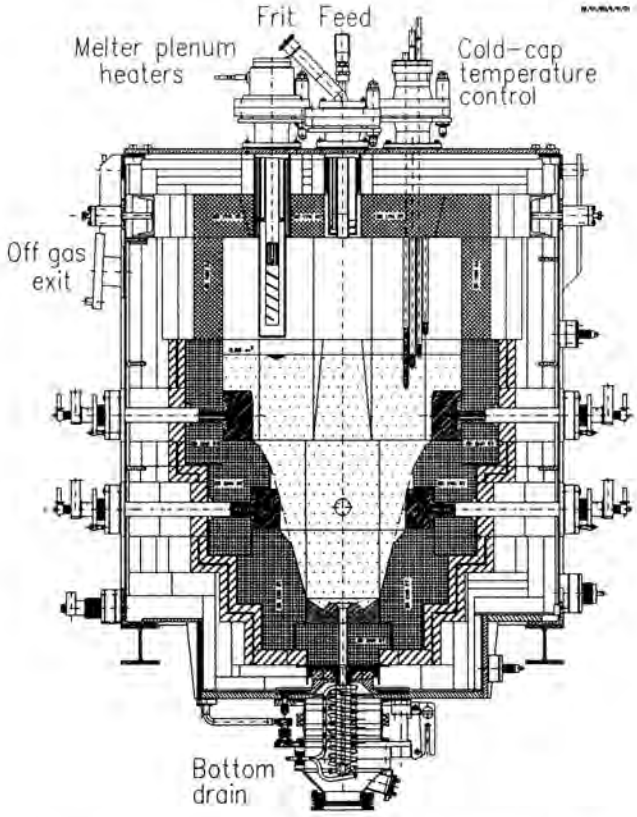

Fig. 2. Schematic Illustration of the Advanced Waste Glass Melter K-6'.

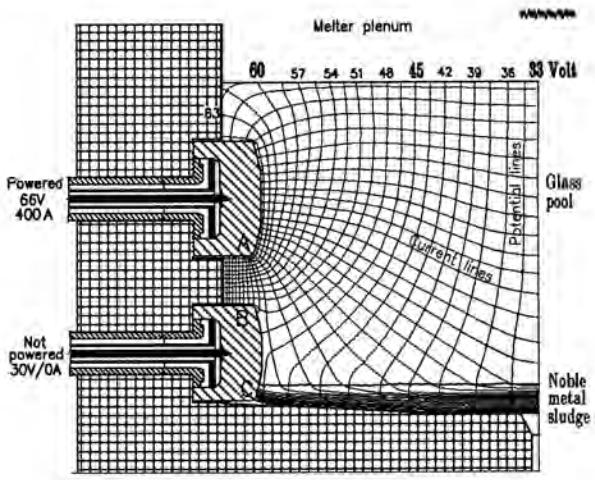

Fig. 4. Verification of the Influence of a Conductive Glass Layer on the Electric Current Field in the First Active PAMELA Melter. A, B and C are Areas Showing Excess Current Densities.

age. For this purpose there are a number of design possibilities. Fig. 5 shows the solution applied for the advanced melter K-6'. A similar design was also used for the second radioactive PAMELA-melter which is sucessfully in operation since August 1988. The features of the advanced design are the special configuration of the upper part of the valve. It acts as glass flow barier. Additional barriers, made of Inconel 690, are connected with the metallic melter shell. A hot glass film will not be able to proceed towards the outside of the melter shell as in the previous design. The

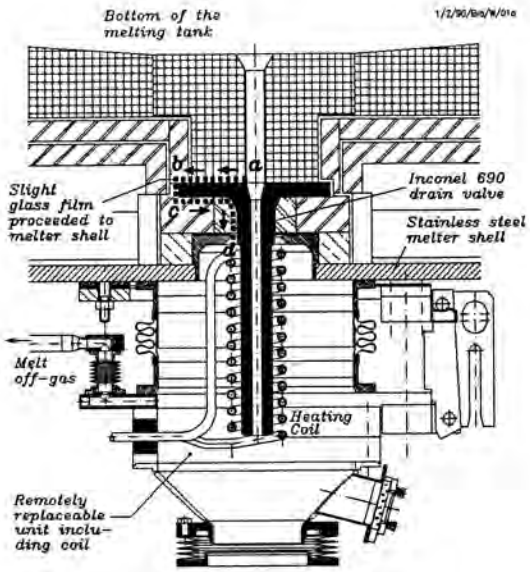

Fig. 3. Bottom Drain of the First Active PAMELA Melter. Glass Leakage Detected at Point d. The thin glass film proceeded most Probably Along the Way a-b-c-d.

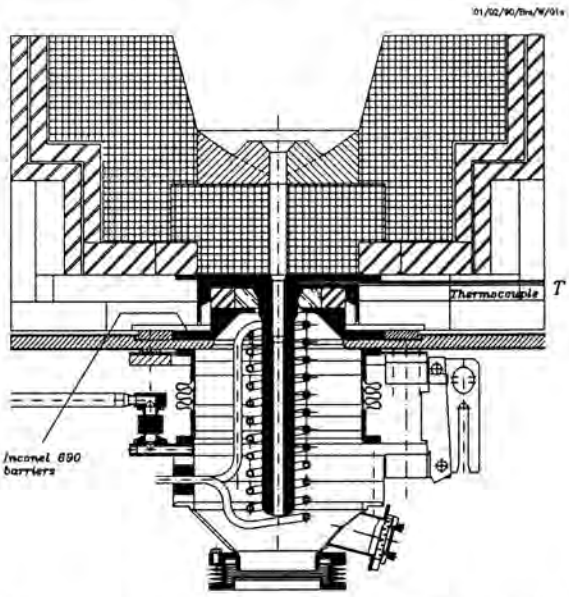

Fig. 5. Advanced Bottom Drain Design for the Melter K-6'. Inconel 690 Parts are Indicated by Black Colour.

barriers are located in the low temperature region of the melter bottom (< 200°C). If a fluid glass film would reach this region it would be frozen and stopped.

Temperature monitoring in the upper portion of the drain valve and inside the flange has also been improved. Remotely replaceable thermocouples have been integrated for more reliable operation, especially to keep the maximum temperature in the valve flange below 1100°C. Furthermore, the glass pouring system is protected against service time shortening caused by ceramic refractory pieces, originating from long-term corrosion of the melter walls. They could potentially cause blockage of the valve channel. The protection has been achieved by the specially designed bottom electrode, and by allowing the glass melt to flow into the valve channel via five independent openings in the bottom electrode.

Improving lifetime of the Power Electrodes

This task implicates particularly carefull considerations not only on electrode design, but also on melt tank configuration, electrode set arrangement, and knowledge about both the electric potential and current field. The common key point is to prevent conditions in the glass tank which can be accompanied with local excess current densities at the submerged electrode surface. A precondition for this goal is, that the melt tank configuration is suitable to promote the removal of settled and conductive precipitates with routine glass pouring. This keeps the melter's glass reservoir electrically homogenous. The current density distribution anticipated for the power electrodes of the advanced K-6' melter in the normal feeding and powering situation is shown in Fig. 6. The results have been obtained by 3D mathematical modelling, which besides the electric field, also included the power density field, the temperature field, and convection pattern in the glass pool. For the case shown in Fig. 6 it was assumed that the main electrode sets and the mid-section electrode pair were powered, and that the walls of the settling chamber were covered with a conductive layer being 5 cm thick and 2.5 times higher conductive compared to the bulk glass melt. The results show that (i) the main electrode set in the upper level of the glass pool is well separated from the electrode pair in the mid-section thus preventing uncontrolled current transfer between each other, and (ii) the current density at the electrode surfaces can be expected rather uniform despite the assumed noble metal rich glass layer, in contrast to the situation shown in Fig. 4 for the present PAMELA melter design.

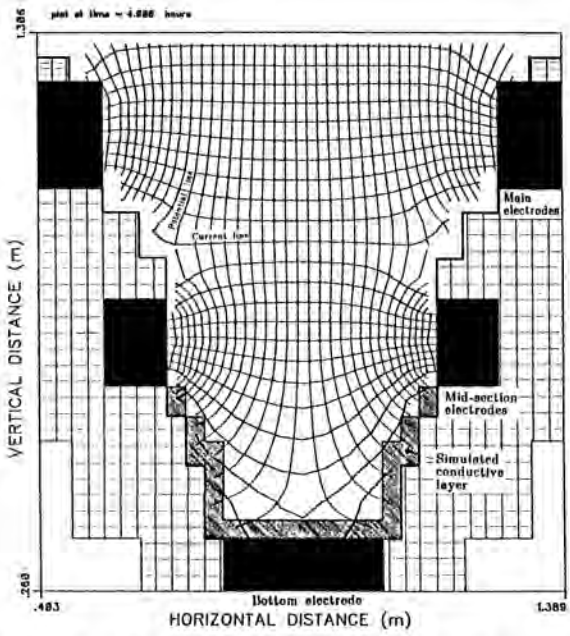

Fig. 6. Electric Potential and Current Field Respectively, Obtained by Mathematical Modelling of the K-6' Melter (Conductive Layer Assumed at the Walls of the Settling Chamber).

The thickness of the conductive layer in the actual melter K-6' is expected to be far below the 5 cm assumed for the model because of the highly sloped walls of the settling chamber. Besides the results shown in Fig. 6 for the main mode of powering, the current field have been calculated also for the case when the bottom electrode is fired against the electrode pair in the mid-section. The results show that the current distribution at the surface of the mid-section electrodes is then less uniform compared with the normal mode of firing. The current densities across the immersed electrode surface differs here by factors up to 3.5. This fact will be taken into account to limit the maximum current allowed for this mode of firing. However, the power necessary for maintaining the desired glass temperatures in the settling chamber was calculated from mathematical modelling to be relatively low. The limit of 0.7 Amp/cm^2 surface current density will not be reached even at those parts of the mid-section electrodes which are expected to have the highest current densities.

NOBLE METALS-COMPATIBILITY

Flat-bottomed melters allow settling noble metals precipitates to form a glass layer at the melter floor which is not only higher conductive but also appreciably higher viscous compared to the bulk glass. The presence of the layer decreases power release in the glass tank and thus feed throughput capacity, affects melter operability by undesired shifting of heat dissipation, and causes nonuniform and uncontrollable current distributions at the surface of the electrodes. The removal of such a viscous layer from a flat melter floor proved to be too incomplete, even by routine glass pouring via a bottom drain. Its typical viscosity and electrical conductivity behaviour compared with the bulk

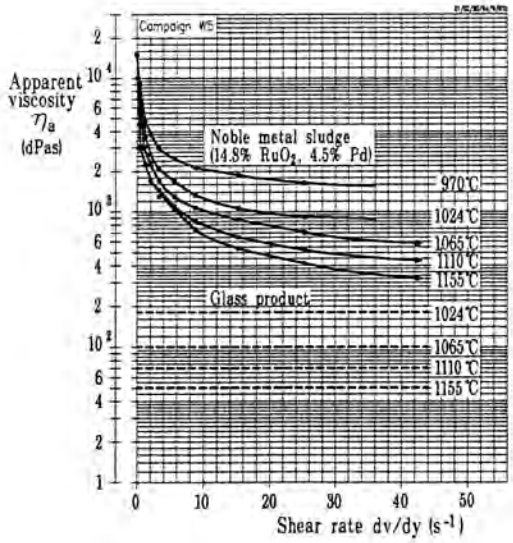

Fig. 7. Viscosity of Noble Metal Enriched Waste Glass in Comparison With the Normal Waste Glass Melt.

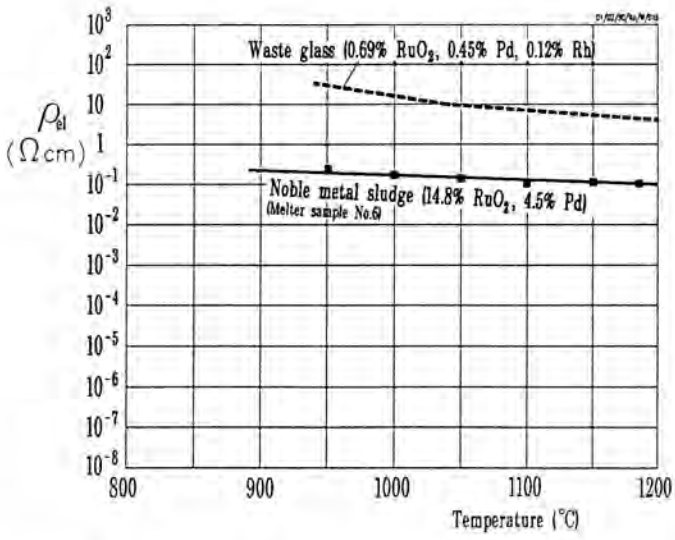

Fig. 8. Specific Electrical Resistivity of Noble Metal-Enriched Waste Glass in Comparison With the Normal Waste Glass Melt.

glass is shown in Figs. 7 and 8. The noble metals enriched glass melt represents a non-Newtonian fluid. Its viscosity depends on shear rate, temperature, and on noble metals concentration. Viscosity and conductivity probably increase with the age of the layer due to kinetically controlled growth and agglomeration of noble metal particles.

A first technical solution at INE to minimize the extent of formation of such a layer, and to remove it with routine glass pouring, was the construction and operation of a melter designated K-W2 with 45° sloped bottom (7). This large-scale melter, having 1.4 m^2 glass pool surface and 72 l/h nominal feed throughput capacity, is installed in the existing industrial-scale mock-up plant V-W1 which has been operated during the past two years. The noble metals removal efficiency of 25% in case of a flat melter floor was increased with this melter to about 65%, and with additional air sparging of the glass pool raised to 85-90%. The removal characteristic of the melter K-W2 is shown in Fig. 9 for Ruthenium as an example. The Ruthenium amount, fed to the melter with the simulated waste solution, is compared with that removed with routine glass pouring. The results have been obtained from two long-term campaigns. The data are based on 3000 analysed glass samples taken from glass pouring stream during canister filling. The results show that a melter with sloped bottom is a step in the right direction to achieve the necessary high removal efficiency for these undissolved waste glass constituents.

The results have been used along with the PAMELA experiences to design the advanced melter K-6' (see Fig. 2). It has vertical walls above the main power electrodes. Between the two power electrode sets, the walls are 60° sloped. The lower part of the melting tank has the function of a settling chamber with 75° wall inclination. The design of this melter has the potential for scale-up from 0.88 m^2 glass pool areas to 2 to 3 m^2 with feed throughput capacities of 100-150 l/h.

There is no reasonable doubt that the task of achieving a noble metals-compatibel melter can be solved. The problem in early development years was more, that no special attention had been payed to the settling behaviour of noble metals precipitates and their effects on melter operability. This was mainly because of the simple fact that the noble metal elements were considered to be, properly speaking, too expensive to introduce them into the simulated solutions for inactive test runs. But due to the experience from recent noble metals experiments at INE and PNC (8) the present stage of melter development regarding the overcome of the noble metals problem is well progressed. Final research and development of the technology is facilitated by the practice now applied at INE to recover the biggest portion of the noble metals contained in the inactive glass product by a special process, and to reuse them for new tests.

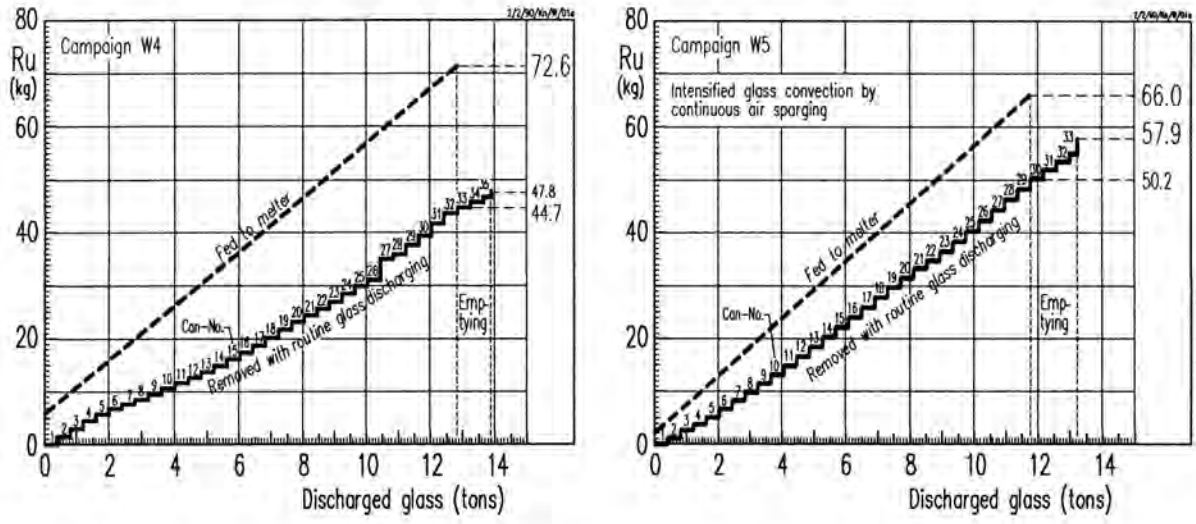

Fig. 9. Removal Efficiency of Ruthenium From the Large-Scale Melter K-W2 With Routine Glass Pouring, Including Emptying the Melter's Glass Reservoir After Each of the Two Operation Campaigns (K-W2 Melter With 45° Sloped Bottom and 1.4 m^2 Pool Surface).

SIMPLIFICATION OF THE MELTER SYSTEM

Melter Construction

Besides longer melter lifetime, simplification is another challenge for an advanced melter system. The construction of the K-6' melter has been simplified especially regarding configuration and assembly of the ceramic refractory blocks composing the glass melting tank. The new configurations allowed easier casting and fabrication of the refractories as well as their more controllable assembly to the melt tank. The melt tank structure is well separated in three sections (settling chamber, mid-section, and upper main pool). This contributes to both a simpler assembly as well as an easier disassembly for later melter cut up and disposal.

Further substantial facilitation of melter cut up could be introduced in future designs. It will be attempted that the melter core can be composed of completely prefabricated subunits. It would allow to use for assembly as well as disassembly for decomissioning a procedure similar to what can be described a modul technique. This would be expected also to be helpful to minimize that fraction of a shutdown melter which must go as high-level solid waste to the repository. An estimation shows that in case of the K-6' melter this fraction would be less than 2 wt% of the produced glass. This number is based on a melter lifetime of 5 years with 500 tons of total glass production, and the weight of the melter core being 10 tons.

Melter Peripherie and Remote Maintanence

The K-6' melter peripherie was substantially reduced by combining the transfer of electric current to the power electrodes with that for providing the electrode cooling air. For this purpose thick-walled copper tubes were used connecting the power electrodes with the remote couplings. The couplings were specially designed at INE for this twofold purpose. The electric current supply for melter plenum heaters has also been optimized. This task is now carried out by flexible wires equipped with remote couplings. It particularely contributed to the simplification of the melter peripherie by eliminating a large number of electrically insulated current supply bus bars fixed on the upper side of the melter.

CONCLUSIONS

With the design features of the melter K-6' it is anticipated that essential melter improvements have been introduced, referred to the first INE-developed melter generation being used in PAMELA. The progress regards extended lifetimes of both the power electrodes and the bottom drain system, furthermore noble metals-compatibility, and simplification of the melter structure. The latter one makes not only construction of the melter core easier, but will also be helpful to facilitate melter cut up and disposal. There is still more potential in future designs to make melter decomissioning as simple as possible and minimize that fraction of melter to be disposed of as high-level solid waste.

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